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J.P. Pelz (Ph.D. Thesis)

December 1987

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Flicker (1/f) Noise in Copper Films
due to Radiation-Induced Defects

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December 1987
Flicker (1/f) Noise in Copper Films due to Radiation-Induced Defects.

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Abstract

Changes were measured in the 1/f noise and resistivity in polycrystalline Cu films due to defects induced by 500 keV - 1.1 MeV electron irradiation or 1 MeV Kr+ ion irradiation. The Cu films were irradiated while maintained at 90 K on a cold stage of an electron microscope, and all noise and resistivity measurements were made in-situ. Irradiation with 500 keV electrons increased the voltage noise level in the films by more than an order of magnitude, while the resistivity increased by at most 10%. When the films were annealed at progressively higher temperatures, both the 1/f noise and the resistivity were reduced; however, at lower annealing temperature, the fractional reduction in the induced noise was substantially more than in the added resistivity. These results suggest that a large fraction of the induced noise may be generated by certain "mobile" added defects that are more readily annealed than the majority of the added defects. The temperature dependence of the noise after irradiation and partial annealing indicated that the induced noise was thermally activated in a manner consistent with the Dutta-Dimon-Horn model. Isolated defects created by 1.1 MeV electron irradiation were found to produce substantially higher noise levels than clustered defects resulting from 1 MeV Kr+ irradiation. Isolated In and Be impurities in Cu are known to trap radiation-
induced interstitial defects. Measurements on a number of films indicate that the induced 1/f noise is not sensitive to the type and quantity of interstitial traps. A simple "local interference" model is presented, which uses calculations by Martin to estimate the 1/f noise magnitude generated by moving defects. This model can account for the induced noise, provided one assumes a sufficient fraction of the added defects to be mobile. Several simple models are examined concerning the identity of the defects responsible for the induced noise. A model which attributes the noise to the motion of vacancy-type defects close to surfaces, grain boundaries, or dislocations is most consistent with the experimental results and theoretical considerations, though none of the models considered provides an entirely satisfactory explanation of the induced 1/f noise.
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I. INTRODUCTION

It is well known that a wide variety of metal films biased with a constant current exhibit low-frequency voltage fluctuations in excess of the equilibrium Johnson noise.\(^1,2\) The spectral density \(S_v(f)\) of these voltage fluctuations often scales roughly with the inverse of the frequency \(f\), and is usually referred to as "Flicker" or 1/f noise. In practice, \(S_v(f)\) rarely scales exactly as 1/f. The designation "1/f" is generally applied to noise which scales as \(1/f^m\), where the frequency exponent \(m\) has a value close to one.\(^3\) Noise of this form is quite a general phenomenon in a number of very different physical systems.\(^2\) It has been observed in semiconductors,\(^4\) ionic solutions,\(^5\) charge density wave materials,\(^6\) as well as in a number of devices.\(^7-13\)

There has been considerable interest in 1/f noise over the past several decades. Early experimental results seemed to indicate a common spectral shape and "normalized" magnitude to the noise in very different physical systems,\(^14\) spurring theoretical attempts to formulate a "universal" explanation for the noise. However, more recent experimental work has made it abundantly clear that the noise level can vary by many orders of magnitude from system to system,\(^15\) and that the shape of the noise spectrum often differs significantly from a true 1/f form.\(^7-12\) In a recent review Weissman\(^2\) has emphasized that quite diverse mechanisms are likely responsible for the noise observed in various systems.

It has however become apparent in recent years that two general properties are common to the 1/f noise observed in a variety of physical systems. The 1/f spectrum in a number of systems appears to
result from a superposition of Lorentzian spectra, with a broad
distribution of characteristic times.\textsuperscript{1,7-12} Each Lorentzian is thought
to be due to a particular microscopic process, which is often thermally
activated. Dutta, Dimon and Horn,\textsuperscript{16} expanding on earlier ideas of Du
Pre\textsuperscript{17} and Van der Ziel,\textsuperscript{18} showed that such a superposition of
Lorentzians could provide a simple, self-consistent explanation of the
1/f noise observed in a number of systems.

It also appears that atomic / defect motion is responsible for the
low-frequency noise observed in a variety of systems.\textsuperscript{9,11,12,19} The
case for defect involvement is particularly strong for metal films.
Eberhard and Horn\textsuperscript{20} showed that annealing freshly-evaporated noble
metal films reduced the 1/f noise while it increased crystallite size.
Fleetwood and Giordano\textsuperscript{21} also found that annealing substantially
lowered noise levels and sample resistivity in AuPd films. Zhigal'skiy
et al.\textsuperscript{22} found that noise levels in metal films depended on internal
stresses, while Fleetwood and Giordano\textsuperscript{23} showed that that externally
applied stress could cause large increases in sample noise. Since
defect motion is known to accompany strain relaxation, these results
support the hypothesis that defect motion is a source of 1/f noise in
metals. Scoffield et al.\textsuperscript{15} studied room temperature noise in a variety
of metal films and found that, when appropriately normalized, the
"cleanest" (largest residual-resistivity ratio) films generally had the
lowest noise. From measurements on Al films doped with Cu and Si, Koch
et al.\textsuperscript{24} found a correlation between "noise" activation energies
inferred from the temperature dependence of $S_{V}(f)$, and activation
energies for electromigration failure. These authors proposed that
defect motion along grain boundaries, known to be responsible for
electromigration failure in Al films, was also responsible for the observed 1/f noise. Black et al. measured symmetry properties of resistivity fluctuations in thin films, and found them to be non-scalar in a number of metals. These results were interpreted in terms of rotations of symmetry-breaking defects. Garfunkel and Weissman studied noise statistics in very small volume metal resistors in order to estimate conduction-electron scattering cross sections of the individual microscopic processes responsible for the noise. For most metals (except Nb) they found cross sections under $10^{-15}$ cm$^2$, consistent with expected cross sections for point defects in metals. Several theoretical models have also been proposed to explain the noise magnitude in terms of defect motion.

In order to provide a more complete description of 1/f noise in metal films, several open questions must be addressed. More direct experimental evidence should be obtained to link 1/f noise to defects. In particular, the specific types of defects responsible for the noise should be identified. Yet to be determined is the physical mechanism by which a defect motion causes a voltage fluctuation in metal films, as well as the expected magnitude of such fluctuations. Theoretical proposals for this mechanism should be formulated and compared with experimental determinations of the 1/f noise magnitude due to measured numbers of mobile defects. Ultimately, one would like to understand the dynamics of the "noisy" defects: why certain defects are mobile, what determines their activation energies, etc.

In this thesis we address several of these questions. We report the results of a number of experiments performed to study the dependence of
1/f noise in Cu films on a particular class of defects: those created by high energy electron and ion irradiation. The Cu films described here were irradiated while cooled below room temperature inside a high-voltage Transmission Electron Microscope (TEM), and the sample noise and resistance were measured in-situ. Since the change in average sample resistance is proportional to the added defect concentration, changes in the noise could be directly related to the number of added defects.

It is found that the 1/f noise level in polycrystalline Cu films maintained at 90 K can be increased by more than an order of magnitude by adding sufficient numbers of defects induced by 500 keV electron irradiation, while the sample resistivity increases by about 10%. This demonstrates directly that structural defects are a source of 1/f noise in metal films. Both the noise and the resistivity return nearly to initial values when defects are removed via thermal annealing, although the noise is reduced more rapidly at lower annealing temperatures than is the resistivity. The noise level in the irradiated films increases strongly with sample temperature in a manner consistent with the thermal activation model of Dutta et al. Isolated defects created by 1.1 MeV electron irradiation are found to produce substantially higher levels of noise than clustered defects resulting from 1 MeV Kr$^+$ ion irradiation. It is known that In and Be impurities in Cu can trap interstitial defects created during irradiation. Noise measurements from a number of Cu films (including films doped with In and Be) indicate that the irradiation-induced 1/f noise is not sensitive to the type and quantity of interstitial traps.

The initial work described here was done using an Hitachi HU-650
TEM operated by the National Center for Electron Microscopy at Lawrence Berkeley Laboratory (LBL). Later work was done using the Kratos AEI-EM7 1200 keV TEM and Ion-Beam Interface at the High-Voltage Electron Microscope / Tandem Facility at Argonne National Laboratory (ANL). Parts of this work have been presented elsewhere.\textsuperscript{19,29,34,35}

In the course of this thesis, we make reference to the extensive literature concerning radiation damage in metals. This field of study has a very long, rich and controversial history; even basic interpretations of certain experiments remain contested after 30 years.\textsuperscript{36,37} The analysis of many experiments requires detailed knowledge of irradiation conditions and sample properties (e.g. sample dimensions, purity, initial defect structure, crystalline orientation, etc.) Much of the relevant work to date has been done on freely suspended bulk materials of high crystalline quality. We note here that the complex nature of our samples (thin films, small crystallites, probable oxidation at surfaces and grain boundaries, the presence of a substrate) makes quantitative comparison with previous work on bulk materials uncertain, though qualitative similarities are evident and will be used to interpret our results. We do however caution against attempts to over-interpret the data presented here in terms of detailed concepts of defect structures and dynamics established for bulk, crystalline Cu.
II. BACKGROUND INFORMATION

II.A. Properties of 1/f Noise in Metal Films

In metal films, the excess voltage spectral density $S_V(f)$ generally scales roughly with $I^2$, where $I$ is the sample bias current.\textsuperscript{1} It is generally agreed that the noise is due to intrinsic resistance fluctuations in the film, present even in the absence of a bias current.\textsuperscript{38,39} We may therefore write

$$\delta V_E(t) = I \delta R(t) = I(\ell/wd) \delta \rho(t),$$

(2.1)

where $\delta V_E(t)$ is an excess voltage fluctuation, $\delta R(t)$ and $\delta \rho(t)$ are the corresponding fluctuations in the resistance $R$ and spatially-averaged resistivity $\rho$ in a sample with dimensions $\ell, w, d$, and $t$ is time. Equation (2.1) implies

$$S_V(f)/V^2 = S_R(f)/R^2 = S_\rho(f)/\rho^2$$

(2)

where $S_R(f)$ and $S_\rho(f)$ are the spectral densities of the resistance and resistivity fluctuations, respectively.

In a number of metals, the noise magnitude has been found to scale inversely with the sample area $A = \ell w$, indicating that the spatial coherence length of the fluctuations is less than the smallest sample widths\textsuperscript{15,40} (15 nm - 500 nm). This upper limit is consistent with earlier direct measurements of the coherence length in chromium and bismuth,\textsuperscript{41} and gold.\textsuperscript{42} The measured noise magnitude generally decreases with thickness, but it remains unclear whether the noise can be generally attributed to either a bulk\textsuperscript{43,44} or surface\textsuperscript{24} effect. This determination is severely complicated by large sample-to-sample variations in noise magnitude,\textsuperscript{40} difficulties in preparing very thin continuous films and in measuring noise on thick films ($>1\mu m$), and
the strong dependence of film morphology and grain structure on film thickness.

A convenient way to characterize measured 1/f noise in metal films, in a manner which removes explicit reference to bias current and sample dimensions, is by the frequency exponent \( m \),

\[
m \equiv \partial \ln S_V(f) / \partial \ln f,
\]

and the dimensionless parameter \( \alpha \),

\[
\alpha \equiv N f_0 S_V(f_0)/V^2 = N f_0 S_R(f_0)/R^2 = N f_0 S_p(f_0)/\rho^2.
\]

Here \( N \) is the number of atoms in the sample and \( f_0 \) is a reference frequency (typically ~ 1 Hz) chosen within the experimental bandwidth. Since \( m \) is in general not strictly equal to 1, \( \alpha \) depends weakly on \( f_0 \). Equation (2.4) is equivalent to an expression popularized by Hooge, except in that he defines \( N \) as the number of free charge carriers in the sample.

Scofield et al. have pointed out that the parameter \( \alpha \) may not be directly indicative of the magnitude of the microscopic noise source, if \( \rho \) happens to depend strongly on sample parameters not directly connected to the noise. As an example, we consider models which attribute the noise to changes in the conduction-electron scattering cross sections of particular "noisy" defects. Conduction-electron scattering due to phonons and/or "quiet" defects may be processes independent of the "noisy" defects, but will nevertheless affect \( \rho \), and hence \( \alpha \). In order to avoid this problem, Scofield et al. have proposed an alternate normalization of the noise in terms of a parameter \( \rho^* \), defined by

\[
\rho^* = N f_0 S_p(f_0) - \rho \rho^2.
\]

In this thesis, we will have occasion to use both normalizations.
II.B. Radiation Damage in Copper

What follows is a brief summary of radiation damage in Cu, with particular emphasis on those areas which are relevant to the present study. Very similar radiation damage behavior occurs in a number of other metals as well.31

II.B.1. High-Energy Electron Damage

It is well known that bombardment by high-energy electrons, ions or neutrons creates31 defects in bulk crystalline Cu. An in-coming particle of mass \( m \) and kinetic energy \( E \) can transfer an energy \( T \) to a host lattice atom of mass \( M \) through a collision/recoil process. The maximum recoil energy \( T_m \) of the host atom is given in the non-relativistic limit as

\[
T_m = \frac{4EmM}{(m + M)^2}.
\]  

(2.6)

A relativistic approximation good for incident electrons (with \( m \ll M \)) is

\[
T_m = \frac{2E(E + 2mc^2)}{Mc^2},
\]  

(2.7)

where \( c \) is the speed of light. This maximum recoil energy corresponds to a direct (head-on) collision; glancing angle collisions result in a lower energy transfer.

When sufficient energy is transferred to the recoiling atom in a metal held at sufficiently low temperature (\( T < 20 \) K for Cu) stable defects in the form of "Frenkel Pairs" (vacancy-interstitial pairs) can be formed.31 The vacancy is simply a missing lattice atom (with some
surrounding lattice relaxation) while the most stable interstitial complex is the <100> "dumbbell" (see Fig. 2.1(a)). A Frenkel Pair is formed by a "replacement collision sequence" (see Fig. 2.1(b)), in which an incident electron displaces an atom (creating a vacancy), which displaces an adjacent atom, which replaces another, etc. until the sequence ends with the creation of an interstitial. The Frenkel Pair is stable at low temperatures if the vacancy and interstitial are separated by more than about three atomic distances. The electrical resistivity $\rho$ of the Cu increases with Frenkel Pair concentration by an amount $\Delta \rho = \rho_F \cdot C$, where $C$ is the fractional concentration of Frenkel Pairs and $\rho_F = 2.75 \times 10^{-4}$ Ωcm.

The "threshold energy" $T_d$ necessary to create a stable Frenkel Pair is in general a strong function of the recoil direction relative to the crystal lattice. In Cu the minimum threshold energy $T_{d,\text{min}}$ is about 19 eV for recoil in a <100> direction, which corresponds (via Eq. (2.7)) to a minimum incident electron energy $E_{\text{min}} = 390$ keV. In pure, bulk polycrystalline Cu one ideally expects the defect-production rate to be negligible for $E < E_{\text{min}}$, but to increase rapidly for $E > E_{\text{min}}$. In practice, small levels of "sub-threshold" damage are often observed for $E < E_{\text{min}}$. This damage is not fully understood, but is thought to be connected to the presence of light-atom impurities or initial defect-structure (e.g. dislocations).

For incident electrons of modest energy ($E < 10$ MeV) the interaction cross section and maximum recoil energy are sufficiently small to ensure uniform generation of spatially uncorrelated Frenkel Pairs throughout samples sufficiently thin to attenuate the electron beam insignificantly. In Cu, these defects are translationally
Fig. 2.1 Frenkel Pairs in fcc metals. (a) "dumbbell" $<100>$ split-interstitial in fcc metals. (b) Replacement collision sequence leading to production of Frenkel Pair. Figure from Ref. 45.
immobile at low temperatures \((T < 20 \text{ K})\) but can undergo thermally-activated migration at elevated temperatures. This is evidenced by a number of added resistivity "recovery stages" observed in annealing experiments.\(^{31}\) Recovery in the "Stage I" temperature range \((0 - 60 \text{ K})\) is thought to correspond to interstitial motion. Close-pair recombination is observed at lower annealing temperatures, while for \(T > 40 \text{ K}\) the remaining interstitials migrate freely until they recombine with vacancies (thus removing Frenkel Pairs), cluster with other interstitials, or are trapped at suitable sites (impurity atoms, dislocations, grain boundaries, etc.). Recovery in the "Stage II" range \((60 \text{ K} - 220 \text{ K})\) is thought to be related to the thermally activated release of interstitials from traps.\(^{31}\) The most prominent feature in the Stage III range \((220 \text{ K} - 330 \text{ K})\) is strong recovery step attributed to the onset of free migration of monovacancies.\(^{37}\)

The bulk of the current experimental evidence and analysis support the above description, known as the "1-interstitial model." However, a small number of researchers hold an alternate "2-interstitial" interpretation\(^{36}\) in which the Stage I recovery is attributed to the motion of a metastable \(<110>\) "crowd-ion" interstitial, Stage II corresponds to trap release and thermal conversion of \(<110>\) to \(<100>\) interstitials, Stage III recovery is due to motion of \(<100>\) interstitials, while vacancy motion occurs at higher temperatures. In contrast to other authors,\(^{49}\) we do not believe that any results to be described here can be used to distinguish one model from the other and we will make no statements as to their relative merits. We will describe our results in terms of the "1-interstitial model" because it is currently the more widely accepted view, but a parallel
"2-interstitial" interpretation is certainly possible.

For irradiations done in the Stage II temperature range where interstitials are mobile but vacancies are not, an interstitial can recombine with its own vacancy ("correlated" recombination), migrate and recombine with another vacancy, or be trapped at an impurity atom, dislocation, etc. The interstitials migrate so quickly at these temperatures that the concentration of free interstitials at any time is sufficiently small that the chance of interstitial clustering is negligible. The accumulation of vacancies and trapped interstitials under such conditions has been successfully described by the "unsaturable trap" model. One considers the rate of increase $\frac{dC_i}{d\phi}$ of the fractional concentration $C_i$ of trapped interstitials with total electron dose $\phi$:

$$\frac{dC_i}{d\phi} = g \left[ \frac{C_t}{C_t + \left( \frac{r_v}{r_t} C_v \right)} \right].$$

Here $g = \frac{dC_{f1}}{d\phi}$ is a generation term for the freely migrating interstitial concentration $C_{f1}$ (i.e., those which escape correlated recombination), $C_t$ and $C_v$ represent the fractional concentration of traps and vacancies, respectively, while $r_t$ and $r_v$ are the trap and vacancy effective trapping radii. The term in parenthesis is simply the probability that a freely-migrating interstitial is captured by a trap rather than a vacancy. As the vacancy concentration $C_v$ increases with total electron dose, the freely migrating interstitials have an increasing tendency to recombine with a vacancy rather than be trapped, causing the quantity $\frac{dC_i}{d\phi}$ to decrease.

Since vacancies and interstitials are removed in pairs, we have $C_i = C_v$, and Eq. (2.8) may be re-written as
\[
\frac{dC_i}{d\phi} = g \left[ \frac{C_t}{C_t + (r_v/r_t)C_i} \right]. \tag{2.9}
\]

In the limit where \( g \) and \( r_t \) are independent of \( C_i \), this equation can be integrated to yield

\[
C_i = C_t^* \left[ (1 + 2g\phi/C_t^*)^{1/2} - 1 \right] \tag{2.10}
\]

where \( C_t^* \equiv (r_t/r_v)C_t \) is an effective trap concentration. It is known that the added sample resistivity due to interstitials is roughly independent of whether they are isolated in the bulk or located at traps.\(^{52}\) Thus Eq. (2.10) may be written

\[
\Delta\rho = Q[(1+2g\phi/C_t^*)^{1/2} - 1] \tag{2.11}
\]

where the "trapping term" \( Q \equiv \rho_P \cdot C_t^* \). According to this model, \( \Delta\rho \) should increase linearly with \( \phi \) for small electron dose, then change to a \( \phi^{1/2} \) dependence for \( g\phi \gg C_t^* \). This general behavior is well confirmed experimentally.\(^{31,52}\)

One can also re-write Eq. (2.9) as a reciprocal damage rate,

\[
\frac{d\phi}{d\Delta\rho} = (\rho_P)^{-1} \left[ 1 + \Delta\rho/Q \right]. \tag{2.12}
\]

One can thus determine the parameters \( g \) and \( Q \) from a plot of \((d\phi/d\Delta\rho)\) vs. \( \Delta\rho \). In deriving Eq. (2.11) we assumed that \( g \) and \( r_t \) were independent of \( C_i \), and hence of \( \Delta\rho \). In fact, \( g \) is a weak function\(^{53}\) of \( C_i \) (as well as \( C_t \)), and the trapping radius \( r_t \) can increase with \( C_i \). Thus Eqs. (2.10) and (2.11) are only approximately valid, and Eq. (2.12) is a non-linear function of \( \Delta\rho \). Careful measurements of the reciprocal damage rate \((d\phi/d\Delta\rho)\) vs. \( \Delta\rho \) have in fact been used to estimate how the parameter \( g \) as well as the trapping radii \( r_t \) and \( r_v \) vary with impurity (trap) concentration and with sample
For the purposes of this thesis, we will use the initial slope of \( \frac{d\phi}{d\Delta p} \) as a means to estimate the approximate trap concentration in different Cu films.

II.B.2. High-Energy Ion Damage

Due to their relatively larger mass, ions can deliver much more energy to a lattice atom than an electron with equivalent kinetic energy. With reference to Eqs. (2.6) and (2.7), one can easily check that the maximum primary recoil energy resulting from a collision is \( T_m = 70 \text{ eV for } 1 \text{ MeV electrons, } T_m = 60 \text{ keV for } 1 \text{ MeV protons, and } T_m = 1 \text{ MeV for } 1 \text{ MeV Kr.} \) Because ions can transfer energy via long-range Coulomb interactions as well as direct collisions,\(^55\) an "average" primary recoil energy \( T_{1/2} \) will be less than the maxima listed above (\( T_{1/2} = 1 \text{ keV for } 1 \text{ MeV protons, } -50 \text{ keV for } 1 \text{ MeV Kr}) \) but are nonetheless substantially larger than recoil energies delivered by electrons.

When the primary recoil energy is large, multiple defects can be produced in spatially localized "displacement cascades."\(^55\) The size of a cascade in general increases with increasing \( T_{1/2} \), but the size saturates at large \( T_{1/2} \) due to the formation of multiple subcascades. If irradiations are performed at low sample temperature (with no defect thermal migration) one finds that nearly all the Frenkel defects which are created (as calculated by computer codes) are retained in the metal as long as the primary recoil energy is small (\( T_{1/2} < 1 \text{ keV} \)). However, when \( T_{1/2} \) is large, spontaneous recombination within the cascade region reduces the efficiency for retaining defects. For recoil energy sufficiently large that subcascade formation is probable, only about
one-third of the Frenkel Pairs are retained.55

In large cascades, many of the defects which do not spontaneously recombine will collapse into clusters. At elevated temperatures many of the remaining defects will undergo thermally activated correlated reactions within the cascade, but a fraction will escape clustering and recombination within the cascade and migrate freely through the lattice. Measurements of Radiation-Induced Segregation in ion-irradiated metal alloys maintained in the temperature range 650 K - 900 K have been used to estimate the relative efficiency for producing freely migrating defects55 as a function of the average primary recoil energy $T_{1/2}$. Such measurements on a Cu (1 at.% Au) alloy indicate that, relative to the value for $T_{1/2} \approx 700$ eV, the relative efficiency for producing freely-migrating defects drops from 100% to around 5% as $T_{1/2}$ is increased from about 700 eV to 50 keV. Measurements with other fcc metal systems (including pure Cu), other techniques, and different irradiation temperatures agree with these efficiencies55 to within a factor of 3. The above efficiencies are normalized relative to the total defect production. If instead they are normalized relative to the number of defects which survive the spontaneous post-cascade recombination (which annihilates 2/3 of the defects created by irradiations with large $T_{1/2}$), the relative efficiency drops from 100% to the order of 15% with increasing $T_{1/2}$. Thus for these higher recoil energies, most of the surviving defects will be in the form of large clusters, and only a small fraction will have the opportunity to migrate and react as isolated interstitials and vacancies.
III. EXPERIMENTAL METHODS

III.A. Sample Preparation

Samples for the LBL experiments were prepared as follows: 99.99% pure Cu pellets were thermally evaporated in a $5 \times 10^{-7}$ Torr vacuum to produce 100 nm thick polycrystalline Cu films on room-temperature $<100>$ Si wafers, which had been oxidized to have a 350 nm surface layer of amorphous SiO$_2$. Standard photolithography and wet chemical etching were used to produce $90 \times 4 \mu m^2$ central structures with five large contact pads (Fig. 3.1(a)). The wafers were diced into 9 mm squares (one structure per square) and the samples were annealed at 400°C for 1 h at a pressure below $10^{-5}$ Torr. Transmission Electron Microscopy later revealed continuous films with occasional small pinholes and typical crystallite size in the range 100 - 200 nm.

A 10:3 mixture of HNO$_3$ and HF acids was used to etch a small hole into the wafer from the reverse side. The rest of the wafer was protected by layer of Parafilm. As the etch rate is faster in the Si than the SiO$_2$, it was possible to interrupt the etching when the acid had reached but not penetrated the SiO$_2$ layer on the front side. In this way a freely suspended SiO$_2$ "window" roughly 300 µm in diameter was produced within 50 µm of each Cu central structure (see Fig. 3.1(a)). This window (partially transparent to electrons) allowed us to position the beam of the TEM on the central structure. The SiO$_2$ is grown under compression and buckles when the Si substrate is removed. In order to keep the Cu central structure on a flat surface and to improve thermal grounding, the structure is positioned to the side of the window over a region of finite Si-substrate thickness.
Thin Cu film sample geometry (top view).
(a) Samples used at LBL. (b) Samples used at ANL. The five-probe central structures were each located within 50 \( \mu \text{m} \) of freely-suspended SiO\(_2\) "window." The "window" was used to position the electron beam so that it covered the entire five-probe central structure.
Samples for the ANL experiments were prepared somewhat differently. We first cut 3 mm disks from an oxidized Si wafer using an ultrasonic mill, and then etched each disk to produce a "window." The front surface of each disk was carefully cleaned with a "piranha" etch ($\text{H}_2\text{O}_2 : \text{H}_2\text{SO}_4$, in a 1:5 ratio) before it was attached to a larger Si wafer for further processing. Photolithography on 3 mm disks proved difficult but not impossible. After an HMDS treatment, a disk was centered on the rotation axis of a photoresist spinner, and Shipley 1450J photoresist was applied and spun at high speeds (8000 rpm - 9000 rpm). High spin speed was necessary in order to produce a uniform photoresist thickness at the disk center and to minimize the size of an excess photoresist "bead" which formed around the disk edge. Care was taken not to expose small features directly onto the window since adhesion of the photoresist to the buckled SiO$_2$ was not always good.

The photoresist was processed into a liftoff pattern as shown in Fig 3.1(b), with total area 480 $\mu$m x 4 $\mu$m$^2$. Thermal evaporation of a high-purity (99.9999%) source Cu was then used to form - 100 nm polycrystalline films onto the substrate disks. The lift-off was performed with acetone. A lift-off process was used instead of an etch to minimize exposure of the Cu films to air, water, and chemicals. Several samples were made with Cu doped with small amounts of In and Be. One set of CuIn films was made by using a conveyor-belt arrangement to sequentially drop alloy pellets into a hot Ta boat. CuIn and CuBe were also made under much cleaner conditions by using electron-beam evaporation of dilute alloy sources in a vacuum of 2 x $10^{-8}$ Torr. All the ANL samples were annealed in the manner described for the LBL samples, and were stored in a dry N$_2$ desiccator.
III.B. Irradiation Apparatus

A Hitachi HU-650 TEM operated by the National Center for Electron Microscopy was used for all the experiments at LBL. Prior to insertion of the sample, the electron beam was aligned at low magnification (~500) and the electron flux was measured via a Faraday cup located below the camera plane. The integrated electron dose $\phi$ during an irradiation was determined by multiplying this measured flux by the total irradiation time. Spatial and temporal non-uniformities in the electron beam intensity introduced approximately a ± 20% relative uncertainty in $\phi$. The absolute dose calibration was uncertain by roughly a factor of 2 due to uncertainties in magnification and Faraday cup collector efficiency.

A modified Kratos AEI-EM 1200 keV TEM was used for all irradiations performed at ANL. This microscope is interfaced to a 2 MeV National Electrostatics tandem ion accelerator and a 300 keV Texas Nuclear ion accelerator to allow in-situ ion irradiations.

The electron dosimetry of this TEM has been described in detail elsewhere and is summarized below. Before a sample was inserted into the TEM optical axis, the electron beam was aligned and adjusted for maximum uniformity. The electron flux was monitored continuously during each irradiation by a Faraday cup located in the viewing chamber of the TEM, positioned next to the image of the sample. The absolute magnification at the Faraday cup position, as well as its aperture area and collection efficiency have previously been measured to an accuracy of better than a few percent. The Faraday cup current was monitored by an electrometer, which outputs a voltage proportional to
the cup current. This voltage was fed into a voltage-to-frequency converter connected to a scaler. The scaler output was monitored by a microprocessor, which recorded on disk the integrated electron dose.

For the samples used during 90 K irradiations, the part of the electron beam used for dosimetry passed through the freely-suspended amorphous SiO$_2$ "window" before reaching the Faraday cup. Some of the electrons are scattered by the window, and thus the Faraday cup measured only a relative electron flux. The transmission efficiency of 1.1 MeV electrons (typically around 50 %) was measured at room temperature for each sample and used to determine the absolute electron dose. On samples Cu01 and Cu05, both used to measure low-temperature damage rates, the sample "windows" were broken to allow an unobstructed electron path to the Faraday cup.

III.C. Sample Cold Stages

For the LBL experiments, the 9 mm square samples were mounted on a custom-built single-tilt liquid-nitrogen (LN$_2$) cold stage which could operate over a temperature range 90 K - 300 K. A small LN$_2$ reservoir held outside the TEM cooled a Cu rod extending to the TEM optical axis, where the sample, a wire-wound heater, and a platinum-resistance thermometer (PRT) were mounted. The thermal time constant of the system was roughly 5 m which insured sufficient temperature stability for accurate noise measurements. Cu leads were attached to the 5 contact pads by use of pressed In disks. The leads each had a room-temperature resistance of the order of 0.1 Ω, and thus contributed minimal Nyquist noise as compared with the sample. Connection was made to the leads via a vacuum feedthrough on the cold stage body.
For the ANL experiments, the 3 mm sample disks were mounted on a single-tilt liquid-He cold stage, designed by W. King\textsuperscript{58} for making in situ resistivity measurements on samples irradiated with the Kratos TEM located at ANL. A liquid-He transfer tube extends from an external He dewar to a point inside the cold stage several cm from the TEM optical axis, where He flow cools a small Cu sample block. The exhaust He gas is pumped through a valve by an external mechanical pump, and temperature regulation is achieved with use of a PRT in the sample block and feedback control of the exhaust valve. The stage was slightly modified for noise measurements to have 5 contact leads, which were attached to the sample via 50 μm gold wire and small pressed-In contact pads.

The cold stage was designed to achieve low base temperatures (~ 8 K) and rapid temperature cycling over a range 8 K - 200 K. The thermal time constant of the Cu sample block becomes quite small below 70 K due to the rapid decrease in heat capacity and rapid increase in thermal conductivity in Cu with decreasing temperature. Although the feedback system could maintain the average temperature to better than 0.1 K over the entire temperature range 8 K - 300 K, small fluctuations in sample temperature made low-level sample noise measurements unreliable below 70 K, and introduced some extra low-frequency system noise (with roughly a $1/f^2$ spectrum) for 70 K < T < 150 K. Analysis of noise data under such conditions will be described later.

III.D. Resistivity Measurements

In the LBL measurements, a four terminal digital voltmeter was used to obtain the relative resistivity of the samples maintained at 90 K to
better than 0.1%. Uncertainties in sample dimensions produced a ±30% error in the absolute resistivity, which was about $10^{-6}$ Ωcm at 90 K compared with $3 \times 10^{-6}$ Ωcm at room temperature.

The apparatus for measuring sample resistivity at ANL has been described in detail elsewhere. A set of microprocessor-controlled relays directed a constant current (typically 1 mA) through the sample, connected in series with a 10 Ω standard resistor. The sample resistance was determined by a microprocessor-controlled digital voltmeter which alternately measured the voltage across the sample and across the standard resistor. The values of the resistance measured with the current flowing in each direction through the sample were averaged in order to reduce the effects of thermal e.m.f.'s in the lead wires. Several such resistance measurements were then averaged together to suppress low-frequency noise in the voltmeter preamplifier. The sample resistance (~10 Ω) was stable provided the sample was cooled below 10 K, and could be measured to better than 1 part in $10^4$. This translates to a relative uncertainty in sample resistivity of about ±0.03 nΩcm. The uncertainty in the absolute resistivity is about ± 15 %, due to uncertainties in the sample dimensions. This is discussed below.

In order to determine sample resistivity, one multiplies the four-point resistance measurement by the sample geometry factor $G_F \equiv \frac{w d_e}{l}$. Here, $w$ and $l$ are the measured sample width and length, and $d_e$ is the "electrical" sample thickness, which may differ from the physical thickness $d_m$, measured, for example by a stylus profiler. A non-conducting oxide layer, for example, would cause a difference between $d_e$ and $d_m$. 
In previous damage-rate measurements made by King and co-workers\textsuperscript{59} at ANL, an "effective" geometry factor $G_F^*$ was determined by the equation

$$G_F^* = \left[ \frac{\rho_b(H) - \rho_b(L)}{R_H - R_L} \right], \tag{3.1}$$

where $\rho_b(T)$ is the known bulk resistivity of pure Cu at a temperature $T$, $R_H$ and $R_L$ are the measured sample resistance at an elevated temperature $H$ (\approx 300 K), and low temperature $L$ (\approx 8 K), respectively. This method was used by King and co-workers because the small aspect ratio of their thin-film samples made accurate measurement of $\lambda/\omega_d m$ difficult. Using this geometry factor, they measured damage rates for electron energies around 1 MeV which are nearly equal to damage rates measured in bulk materials.\textsuperscript{46}

In the ANL experiments, we also take $G_F^*$ as the geometry factor, both to be consistent with the methods of King and co-workers, but also because we believe it provides a more consistent estimate of the true $G_F$ than does the expression $G_F^\# = \omega d_m / \lambda$. Any systematic effects which produce a difference between $G_F^*$ and the true $G_F$ (e.g. a softened phonon spectrum in the films relative to bulk Cu) are expected to affect all the samples in roughly the same way. On the other hand, rather large experimental uncertainties in $w$ (\pm 10\%) and $d_m$ (\pm 10\%), will result in unsystematic uncertainties in $G_F^\#$. Furthermore, there are two obvious systematic effects which would tend to make $d_m > d_e$. These are: (1) a surface oxide layer, and (2) a film which is rough on a scale smaller than the profiler stylus tip radius (typically \approx 1 \mu m). Analysis of our films with Secondary Ion Mass Spectroscopy (SIMS) confirms the presence of oxygen at the sample surface, and electron micrographs of the films do show roughness on the scale of a crystallite dimension.
For consistency, all further analysis of the ANL samples will assume the sample dimensions to be the measured values of \( l \) and \( w \), and an "effective" sample thickness \( d^* \equiv (l/w)d_F^* \). Table 3.1 lists relevant sample parameters for all samples used at ANL. We see there that, typically, \( (d^*/d_m) = 0.88 \pm 10\% \), so the different methods of determining \( d \) (and \( G_F \)) agree with each other reasonably well. We take \( \pm 15\% \) as an estimate of the absolute uncertainty in \( d \), \( G_F \), and \( \Delta \rho \), though the relative uncertainty in comparing different samples is probably better than this.

### III.E. Noise Measurement

The apparatus used to measure low frequency resistance fluctuations is shown in Fig. 3.2. The five-terminal bridge circuit is based on a suggestion by Voss and Clarke.\(^{60}\) Scofield\(^{61}\) had independently developed similar apparatus. A sample (held inside the TEM) with five contacts and total resistance \( R_s = 10 \Omega \) formed two arms of a Wheatstone Bridge, each arm with approximately equal resistance (\( R_1 = R_2 = R_s/2 \)). The bridge was driven by a bias voltage \( V_B(t) = V_0 \cos(2\pi f_0 t) \), where \( f_0 \sim 2010 \text{ Hz} \). Large, low-noise wire-wound resistors (\( R_{B1} = R_{B2} = 1000 \Omega \gg R_s \)) were used in the other two bridge arms in order to assure a nearly constant-amplitude current \( i_0 = V_0/R_B \) through each sample arm. A 50 \( \Omega \) ten-turn variable resistor \( R_v \) and a trim air-capacitor \( C \) were used to precisely balance the bridge. As the resistance of \( R_1 \) and \( R_2 \) fluctuate an imbalance voltage \( V \) appears across the sample:

\[
V = i_0 \cos(2\pi f_0 t) \cdot \delta R(t) \tag{3.2}
\]

where
Table 3.1. Thin Cu film samples used at Argonne National Laboratory. RRR is the room-temperature-to-helium-temperature residual resistance ratio. Sample width $w$ measured by an optical microscope; the physical thickness $d_m$ measured with a stylus profiler or a calibrated quartz-crystal monitor during deposition. All samples had nominal length $L = 480 \mu m$. The effective geometry factor $G_F^*$, the effective thickness $d^*$, and the initial trapping term $Q_o$ are defined in the text.

<table>
<thead>
<tr>
<th>sample</th>
<th>date measured</th>
<th>RRR</th>
<th>$G_F^*$ (nm)</th>
<th>$w$ (µm)</th>
<th>$d^*$ (nm)</th>
<th>$d_m$ (nm)</th>
<th>$d^*/d_m$</th>
<th>$Q_o$ (nncm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu01</td>
<td>8/7/86</td>
<td>5.72</td>
<td>0.732</td>
<td>4.6</td>
<td>87</td>
<td>100</td>
<td>0.87</td>
<td>--</td>
</tr>
<tr>
<td>Cu02</td>
<td>11/20/86</td>
<td>4.80</td>
<td>0.842</td>
<td>4.8</td>
<td>85</td>
<td>100</td>
<td>0.85</td>
<td>6.7</td>
</tr>
<tr>
<td>Cu03</td>
<td>5/13/86</td>
<td>6.41</td>
<td>0.732</td>
<td>4.0</td>
<td>88</td>
<td>100</td>
<td>0.88</td>
<td>3.9</td>
</tr>
<tr>
<td>Cu04</td>
<td>4/22/87</td>
<td>7.40</td>
<td>0.821</td>
<td>3.8</td>
<td>105</td>
<td>120</td>
<td>0.88</td>
<td>6.5</td>
</tr>
<tr>
<td>Cu05</td>
<td>4/24/87</td>
<td>6.95</td>
<td>0.727</td>
<td>3.3</td>
<td>106</td>
<td>120</td>
<td>0.88</td>
<td>--</td>
</tr>
<tr>
<td>Cu06</td>
<td>4/27/87</td>
<td>7.40</td>
<td>0.803</td>
<td>3.8</td>
<td>103</td>
<td>120</td>
<td>0.86</td>
<td>4.8</td>
</tr>
<tr>
<td>CuIn1a</td>
<td>8/9/86</td>
<td>5.89</td>
<td>1.081</td>
<td>4.7</td>
<td>112</td>
<td>140</td>
<td>0.80</td>
<td>14.0</td>
</tr>
<tr>
<td>CuIn2b</td>
<td>2/20/87</td>
<td>4.40</td>
<td>0.883</td>
<td>4.2</td>
<td>95</td>
<td>104</td>
<td>0.91</td>
<td>38.</td>
</tr>
<tr>
<td>CuBe1c</td>
<td>11/18/86</td>
<td>5.73</td>
<td>0.978</td>
<td>3.4</td>
<td>139</td>
<td>145</td>
<td>0.96</td>
<td>7.3</td>
</tr>
<tr>
<td>CuBe2d</td>
<td>2/27/87</td>
<td>5.93</td>
<td>0.857</td>
<td>4.0</td>
<td>104</td>
<td>120</td>
<td>0.87</td>
<td>10.6</td>
</tr>
</tbody>
</table>

*a* pellet-drop thermal evaporation (see text). Indium content ~ 270 at.ppm, determined by X-ray florescence spectroscopy.

*b* electron-beam evaporation. Indium content ~ 0.45 at. %, determined by Rutherford Back Scattering.

*c* thermal-evaporation. Source material had ~ 680 at.ppm Be. Low trapping efficiency thought to be due to oxidation of Be during thermal evaporation (A. Baily, private communication). Sample 1/f noise not measured.

*d* electron-beam evaporation. Source material ~ 680 at.ppm Be.
Fig. 3.2 Bridge circuit and apparatus used for low-frequency noise measurements.
\[ \delta R(t) = [\delta R_1(t) - \delta R_2(t)]. \] (3.3)

This signal voltage was amplified by a liquid-nitrogen cooled transformer connected to a low-noise Brookdeal 5004 pre-amplifier, then filtered through a 1 kHz - 3 kHz passband and demodulated with a lock-in detector. When the lock-in phase is set to sense resistive (as opposed to reactive) sample fluctuations, its low-frequency output voltage is proportional to \( i_0 \delta R(t) \). This signal was filtered over a 0.1 Hz - 25 Hz passband and analyzed with a digital spectrum analyzer in conjunction with a desk-top computer. The resulting spectral density obtained from the difference fluctuations \( \delta R = (\delta R_1 - \delta R_2) \) should be the same as that obtained from the sum \( \delta R' = (\delta R_1 + \delta R_2) \) since the spatial correlation length of the resistance fluctuations is small compared with sample length.\(^{15}\) This latter spectral density is that measured by conventional four-probe schemes.\(^1\) Test measurements on our Cu films with the five-probe ac scheme and the four-probe dc scheme produced roughly equal spectral densities, provided the root-mean-square voltage across each sample arm in the ac-scheme is set equal to one-half the dc voltage across the sample in the dc-scheme. The ac method for measuring sample noise has a distinct advantage over the dc method since signal amplification is done in a small frequency band near \( f_0 \), thus avoiding low-frequency roll-offs in the transformer and pre-amplifier, as well as pre-amplifier 1/f noise. It also helped minimize effects of low-frequency pick-up transients present inside the TEM.

The samples were each checked for contact noise, which was found to be insignificant in all noise measurements presented here.

An unshielded Triad G-4 transformer configured with a turns ratio \( N_T \)
and cooled in a small dewar of liquid nitrogen was used to match the low sample impedance to the optimum input impedance of the pre-amp. The cooling reduced the Nyquist noise in the transformer windings by about 12 dB, to a level lower than the Nyquist noise of the sample at 90 K.

III.F. Noise Analysis

To analyze the excess low-frequency noise, a background noise spectral density without current modulation (predominantly the Nyquist noise of the sample) was subtracted from the spectral density with current. At least 50 scans were averaged for each spectral density. For the LBL measurements, least-squares linear fits were made to log-log plots of the data to obtain the noise magnitude and slope to better than ±5%. For the ANL measurements, a routine which minimized the goodness-of-fit parameter \( \chi^2 \) was used to fit the noise data to a function of the form

\[
S_N(f) = \frac{A}{f^m} + \frac{B}{f^2},
\]

where \( A, B, \text{ and } m \) are free fitting parameters. The first term is the sample 1/f noise, the second is the system 1/f^2 noise introduced by the small temperature fluctuations of the cold stage. These fits accurately yield the 1/f noise magnitude at higher frequencies (the \( B/f^2 \) term was always less than 10% of the \( A/f^m \) term for \( f > 5 \text{ Hz} \)), but a rather large "trough" of roughly equal \( \chi^2 \) often existed in the free-parameter space, which produced large uncertainties in the parameter \( m \). Consequently, for the ANL measurements we focus primarily on questions related to the 1/f noise magnitude.
IV. EXPERIMENTAL RESULTS

Since vacancies and interstitials migrate freely and recombine at room temperature, it was necessary to cool the samples during irradiation in order to build up a substantial excess defect population. All irradiations described here were performed with a sample at 90 K or colder. Test irradiations performed at room temperature resulted in no measurable increase in sample noise or resistivity.

IV. A. LBL Results

IV. A.1. Procedure

In a first set of experiments performed at LBL, a sample (Fig. 3.1(a)) was cooled to 90 K in the HU-650 TEM, and the average sample resistance and noise were measured. We then irradiated the sample with a beam of 500 keV electrons with a typical intensity of $10^{17}$ cm$^{-2}$s$^{-1}$ for a specified time. By use of the sample resistance as a thermometer during irradiation, we established that the sample temperature rose by at most 1 K. After each irradiation process, which could last longer than 1 h for the largest doses, we measured the resistance and noise. Typically 30 min elapsed between the irradiation and the noise measurements. At the end of this sequence, we annealed the film for 5 min at each of a series of progressively higher temperatures, remeasuring the resistance and noise at 90 K after each annealing step. The noise magnitude remained constant (within ±5%) for up to 2 h provided that the sample was maintained at 90 K. All the data reported in this section are from a single Cu sample. Similar results were
IV.A.2. 90 K Damage Rate

Figure 4.1 shows the change in sample resistivity $\Delta \rho$ vs. the total electron dose $\phi$, for three irradiation sequences on the same sample, each separated by a room-temperature annealing process of at least 12 h. We see that $\Delta \rho$ scales approximately as $\phi^{1/2}$, consistent with the "unsaturable trap model" described earlier. This scaling is consistent with the behavior observed in bulk materials at similar sample temperatures. The observed initial damage rate $d\Delta \rho/d\phi$ is, however, significantly larger (~factor of 5) than that observed in bulk materials under similar irradiation conditions. Even though the absolute dose calibration could be off by as much as a factor of 2, this damage rate still appears large compared with previous measurements. Tests performed on thicker (~300 nm) Cu films indicated a lower 500 keV damage rate, while a 100 nm film which had not been annealed at 400° C following evaporation was found to have a damage rate more than a factor of three larger than the well annealed films of the same thickness. A significant amount of "subthreshold damage" for electron energies < 400 keV was also observed. These results will be discussed later in conjunction with the work done at ANL, where the electron dosimetry and resistivity measurements were much more accurate.

IV.A.3. Radiation-Induced 1/f Noise

Figure 4.2 shows several voltage power spectra, $S_V(f)$ (with background noise subtracted) for the sample biased with a current $I_{\text{rms}}$ = 20 mA, but with progressively higher electron doses. The spectral
Fig. 4.1 Change in sample resistivity $\Delta \rho$ vs. electron dose $\phi$, for three different irradiation sequences. The dashed line $\Delta \rho \propto \phi^{1/2}$ is drawn for comparison.
Fig. 4.2  Typical excess-voltage-noise power spectra and least-squares fits for different electron doses $\phi$. Data taken at LBL.
density of the 1/f noise measured over this frequency range increases by more than an order of magnitude as radiation-induced defects are added to the film. This directly shows that crystalline defects are a source of 1/f noise in metal films. This large increase in noise magnitude is accompanied by only about a 10% increase in sample resistivity. In Fig. 4.2, the slope of the spectral density on the log-log plot steepens by about 10%, with most of the increase occurring after the first irradiation. The noise remains "1/f-like" over the full range of irradiations.

Here we characterize the noise in terms of the parameters $m$ and $\alpha$, defined by Eqs. (2.3) and (2.4). In Eq. (2.4), $V$ is twice the rms voltage across half the sample, $N = 2.9 \times 10^{12} (\pm 30\%)$ is the estimated number of atoms in the sample, and $f_0$ is taken to be 1 Hz. Before each irradiation sequence, the initial value of $\alpha$ at 90 K was within 10% of $5.5 \times 10^{-4}$. In Fig. 4.3 we plot $\Delta \alpha$ vs. $\Delta \rho$ for the three data runs illustrated in Fig. 4.1; $\Delta \alpha$ and $\Delta \rho$ are the changes in $\alpha$ and $\rho$ relative to the values before a particular run. The values of $\Delta \alpha$ obtained after successive irradiations fall approximately on the dashed line $\Delta \alpha \propto \Delta \rho^{0.6}$. Since $\Delta \rho$ is roughly proportional to the added fractional Frenkel Pair concentration $C_d$ ($C_d \equiv C_V = C_I$), these data indicate that the noise magnitude scales as $C_d^{0.6}$. At first sight, this dependence is surprising. If all the added defects act as equivalent, independent, local noise sources, then one would expect a linear dependence of the noise magnitude on $C_d$. However, we note that $C_d$ is a measure of the total number of added defects, but it is not necessary that all the added defects contribute equally to the noise. For example, existing defect-noise models relate the noise magnitude...
Fig. 4.3 Change in 1/f noise magnitude $\Delta \alpha$ vs. change in sample resistivity $\Delta \rho$. The dashed line $\Delta \alpha \propto \Delta \rho^{0.6}$ is drawn as a guide to the eye. Points along this line correspond to increasing electron dose $\phi$, while points along the dotted line correspond to annealing at successively higher temperatures. The datum point for $T_A = 239$ K (not shown) is $\Delta \rho = 11.6 \, \text{n}\Omega\text{cm}$, $\Delta \alpha = 7 \times 10^{-5}$. 
only to the concentration $C_m$ of mobile defects which change position or orientation in the same frequency range as the observed noise. Since the total added defect concentration $C_d$ serves as an upper limit on the added mobile defect concentration $C_m$, one can use these noise measurements to check whether a particular noise model is able to produce sufficient noise levels.\cite{29,49} The observed scaling $\Delta \alpha \propto C_d^{0.6}$, however, may be primarily an indication of how $C_m$ scales with $C_d$, rather than how the noise magnitude depends on $C_m$.

We emphasize here that we describe as mobile only those defects which move with characteristic frequencies which are within the experimental noise bandwidth (0.1 Hz - 25 Hz). In a broader sense, essentially all the added defects are mobile since they are metastable. If one heats the sample and/or waits a sufficiently long time, they will eventually migrate and recombine. However, those defects which move at frequencies less than ~0.1 Hz at a particular temperature do not add noise within the experimental bandwidth, and thus are considered to be "immobile" and "quiet."

IV.A.4. Annealing Behavior

The dependence of $\Delta \alpha$ on $\Delta \rho$ after the samples were annealed is very different, as is shown by the dotted line in Fig. 4.3. The annealing process reduces the noise much more rapidly than the resistivity, producing hysteresis in the plot of $\Delta \alpha$ vs. $\Delta \rho$. This behavior is also illustrated in Fig. 4.4(a), which shows the annealing data plotted as recovery curves, namely $\Delta \rho/\Delta \rho_{\text{max}}$ and $\Delta \alpha/\Delta \alpha_{\text{max}}$ vs. the annealing temperature $T_A$. The three different symbols in the figure correspond to three separate measurement sequences, taken on different days. Most
Fig. 4.4  Annealing behavior of an irradiated Cu film with $\Delta \rho_{\text{max}} = 90 \, \text{n\Omega cm}$ and $\Delta \alpha_{\text{max}} = 6 \times 10^{-3}$ prior to annealing. (a) Recovery of the $1/f$ noise magnitude ($\Delta \alpha/\Delta \alpha_{\text{max}}$) and resistivity ($\Delta \rho/\Delta \rho_{\text{max}}$) vs. annealing temperature $T_A$. (b) the frequency exponent $m$ vs. annealing temperature $T_A$. 
of the resistivity recovery occurs in the range $200 \, K < T_A < 300 \, K$, and is similar to the "stage III recovery" well documented for irradiated Cu.\textsuperscript{31} This recovery step is generally associated with the onset of the free migration of monovacancies.\textsuperscript{37} The noise magnitude, $\Delta \alpha$, recovers partially over this same temperature range, but also exhibits substantial recovery below $150 \, K$ that is not readily apparent in the resistivity curves. These data support the idea that certain subpopulations of the added defects contribute disproportionately to the noise. The observed difference in the recovery of $\Delta \rho$ and $\Delta \alpha$ is readily explained if one assumes that a population of "noisy" defects is deactivated (via thermally activated trap-release, recombination, or clustering, for example) at lower temperatures than the bulk of the added defects.

This interpretation is consistent with a model which ascribes the noise to defect motion, since thermally activated defect motion is also known to be responsible for defect annealing observed in irradiated metals.\textsuperscript{31} The type of defect motions which lead to annealing may well be related to motions which produce noise. It is quite plausible that defects which anneal easily at a particular temperature are also particularly noisy. We note here that Fleetwood and Giordano\textsuperscript{21} have interpreted noise data taken after annealing sputtered AuPd films by making a related assumption: that defects with low noise activation energies also have low annealing activation energies. In both cases, the assumed relation between defect motions which produce noise and those that lead to annealing are physically plausible, but nevertheless only hypothetical. Such a relation does not as yet have a firm experimental or theoretical justification.
In terms of the ideas of populations of "quiet" and "noisy" defects, one can speculate on several possible origins of the less-than-linear dependence of $\Delta \alpha$ on $\Delta \rho$ observed during sample irradiation. Interactions between defects might lead to suppression of the activity of noisy defects when the added defect concentration $C_d$ becomes large. There could also exist limited numbers of noisy sites for added defects. For example, a particular kind of trap might allow interstitials trapped at 90 K to move or reorient locally, but not to migrate freely. The occupancy of such sites could saturate at larger $C_d$, leading to a sub-linear increase of $\Delta \alpha$ with $\Delta \rho$. Another possibility is suggested by the fact that noisy defects anneal more easily than the bulk of the added defects. If noisy defects introduced during early irradiations are partially annealed during subsequent irradiations, then the sub-linear dependence of $\Delta \alpha$ on $\Delta \rho$ follows naturally. We noted earlier that the irradiation-induced noise appeared to be at least quasi-stationary at 90 K, in that the noise magnitude did not decrease significantly over several hours after the electron beam had been turned off and the sample had been allowed to sit for 30 min. Thus annealing simply due to the extra time taken to perform subsequent irradiations cannot have produced enough reduction in $\Delta \alpha$ to account for the observed $C_d^{0.6}$ dependence. One can, however, speculate that effects of the electron beam itself and the excess population of freely-migrating interstitials present during the irradiation might stimulate annealing of the noisy defects. For example, "subthreshold" recoil events and "Radiation Annealing" are thought to cause enhanced recombination of existing defects in metals. These possibilities are (for the moment) quite
speculative, but can serve as guides for further experiments and analysis.

In addition to the changes seen in the noise magnitude and sample resistivity, the frequency exponent $m$ of the noise measured at 90 K also changed during the irradiation and annealing experiments. Figure 4.2 shows that $m$ increased from about 1.0 to 1.1 during the irradiations. The annealing behavior of $m$ shown in Fig. 4.4(b) exhibits a striking dip to a value $m = 0.9$ at $T_A = 240$ K, that is reproducible in all the samples studied at LBL. We see from the figure that this behavior is extremely repeatable on a single sample, provided it was annealed at room temperature for an extended period between runs. We note that this dip occurs within the range of annealing temperatures where the "Stage III" recovery of $\Delta \rho$ and $\Delta \alpha$ is observed; temperatures at which large changes in the added defect population occur. We will return to this behavior of $m$ when the temperature dependence of the added noise is discussed.

IV.A.5. Temperature Dependence of the Noise

All the noise measurements described so far were taken with the sample held at ~90 K, and indicate that the added noise is due to defects created by the electron irradiation. The added noise can be reduced by annealing the Cu films at temperatures in the range 90 K - 300 K, a range where thermally activated trap release, thermally activated defect migration, and defect recombination are known to take place. Additional information about the processes responsible for the noise can be gained by measuring the temperature dependence of the added noise. In particular, one can check whether the added noise is
thermally activated in a manner consistent with a model proposed several years ago by Dutta, Dimon, and Horn.\textsuperscript{16} The model is briefly reviewed in Appendix A.

In the next series of measurements, a sample maintained at 90 K was first irradiated with 500 keV electrons to a total resistivity increment $\Delta \rho = 85 \, \text{n\mcm}$, and then annealed at a temperature $T_A > 90$ K. The 1/f noise was then measured as a function of sample temperature for $T < T_A$. By first annealing at $T_A$, one puts the sample into a state such that any remaining noise measured at $T < T_A$ is quasi-stationary, i.e., the noise magnitude and spectral shape do not change significantly during the noise measurement.

In Fig. 4.5, the noise magnitude, expressed in terms of the parameter $\rho^2$ defined by Eq. (2.5), is plotted as a function of $T$, for $T_A = 201$ K, $T_A = 239$ K, and in the "unirradiated" (i.e. fully annealed) state. Here we express the noise magnitude using the parameter $\rho^2$ instead of the parameter $\alpha$ since $\rho$ does change by a large amount as the temperature is increased. We see from Fig. 4.5 that the noise magnitude is a very strong function of the sample temperature $T$ and the sample state (as determined by $T_A$). The noise level on a single sample can be made to vary by well over two-orders-of magnitude by varying $T$ and $T_A$. The three curves in Fig. 4.5 are quadratic fits to the three sets of data points. In Fig. 4.6, the frequency exponent $m$ is plotted as a function of $T$ for the same data series shown in Fig. 4.5. The parameter $m$ also has a strong dependence on both $T$ and $T_A$. The curves shown in Fig. 4.6 are discussed below.

If the 1/f noise measured in a system results from an ensemble of thermally activated processes with a distribution in activation rates,
Fig. 4.5 Dependence of the noise magnitude $\rho^2_*$ on temperature $T$ for the Cu film in the unirradiated state, and for $\Delta \rho_{\text{max}} = 85$ n$\Omega$cm followed by a 5 min anneal at $T_A$. Curves are log-log quadratic fits to the data points.
Fig. 4.6  Dependence of the frequency exponent $m$ on $T$ for the same data series as shown in Fig. 4.5. Curves are prediction of Eq. 4.3, using fits in Fig. 4.5.
then its spectral density should, in general, be described by a superposition of thermally activated Lorentzians\textsuperscript{17,18} (see Appendix A). Du Pre,\textsuperscript{17} and subsequently Dutta et al.\textsuperscript{16} considered such a model in which: (1) the distribution of activation rates results primarily from a distribution $D(E)$ in activation energies $E$, and (2) the coupling of the process to the measured noisy quantity (e.g. sample resistance) is temperature independent. With these assumptions, these authors showed that noise of the form $1/f^m$ (with $m = 1$) follows in a natural way provided $D(E)$ is broad with respect to $k_B T$ ($k_B$ is Boltzmann's constant), but otherwise of arbitrary form. According to this model, the shape of the distribution function $D(E)$ can be determined from the measured quantity $S_p(f,T)$, via the relation

$$D(\bar{E}) = \frac{(f/T)}{S_p(f,T)}.$$  \hfill(4.1)

Here

$$\bar{E} = -k_B T \ln(2\pi f \tau_0),$$ \hfill (4.2)

where $\tau_0^{-1}$ is the attempt rate of the activated process, assumed constant for all $E$. The attempt rate $\tau_0$ is taken to be $= 10^{14}$ s$^{-1}$, a typical phonon frequency. Dutta et al.\textsuperscript{16} extended the analysis to show that the temperature dependence of $S_p(f,T)$ and of the frequency exponent $m$ in such a model are related by

$$m(f,T) = 1 - \frac{1}{\ln(2\pi f \tau_0)} \left[ \frac{\partial \ln S_p}{\partial \ln T} - 1 \right].$$ \hfill (4.3)

The noise measured in a physical system is not consistent with this model if it does not obey this relation.\textsuperscript{68}

The curves shown in Fig. 4.6 are the expected temperature dependences of $m$ for the three annealing states, as determined by Eq. (4.3) and the fits of the noise magnitude data shown in Fig. 4.5. The
curves reproduce the general trends in the measured data points rather well, indicating that the noise observed with the sample in all three annealing states is thermally activated in a manner consistent with the type of model proposed by Dutta et al.\textsuperscript{16}

Weissman,\textsuperscript{69} and Scofield et al.\textsuperscript{70} have pointed out that qualitative agreement of Eq. (4.3) with experiment is not a sufficient condition to prove that the observed noise results from an activated process with a broad spread of activation energies. An alternate model, with a single activation energy and an appropriate distribution in the attempt rate $\tau_0^{-1}$, could also result in noise of the form $1/f^m$, and would obey a consistency relation which is qualitatively very similar to Eq. (4.3). One expects that $1/f$ noise which has been observed to agree qualitatively with Eq. (4.3) could in general result from a distribution in $E$ and in $\tau_0$. In certain small MOSFET\textsuperscript{7-9} and tunnel junction devices\textsuperscript{10-12} the low-frequency excess noise can be decomposed into a discrete set of Lorentzians, with $\tau_0$ and $E$ determined for each Lorentzian. In these systems, substantial distributions in both $\tau_0$ and $E$ are in fact found.

With this said, one should not forget that one of the most attractive features of the model of Dutta et al.\textsuperscript{16} is that a "$1/f$-like" spectrum follows in a natural way from a broad (but otherwise arbitrary) distribution $D(E)$. On the other hand, a model which assumes a fixed $E_0$ and a distribution $H(\tau_0)$ in $\tau_0$, requires that

$$H(\tau_0) = \tau_0^{(m-2)}$$  \hspace{1cm} (4.4)

in order to produce noise which scales as $1/f^m$. In this case, one must provide the physical motivation for such a distribution in $\tau_0$. With respect to the work described here, we note that a number of studies
indicate the existence of a variety of defect motions in metals, with typical activation energies in the range \(31 \, 0.1 \, eV - 3 \, eV\), and with attempt rates \(33, 45, 71\) within a few orders of magnitude of \(10^{12} \, s^{-1}\). The model of Dutta et al., which assumes that the range in characteristic times is dominated by a range in activation energy, is likely the most appropriate to the results discussed here.

With the assumption that the results reported here can be described by the Dutta-Dimon-Horn model, we can use the data in Fig. 4.5 and Eqs. (4.1) and (4.2) to infer the form of \(D(E)\), over an energy range determined by the range of measurement temperatures. This is shown in Fig. 4.7 for the three sample states. For \(T_A = 200 \, K\) and \(T_A = 240 \, K\), we see a pronounced increase in \(D(E)\) as \(E\) nears \(0.6 \, eV\). This is close to measured activation energies for the migration of monovacancies, \(72\) suggesting that there does exist a relation between the activation energies of those defect motions which cause noise, and those which lead to migration. This same general conclusion can of course be drawn directly from Fig. 4.4 and Fig. 4.5, by noting that the irradiated films become extremely noisy close to \(200 \, K\), a temperature at which defect migration causes a large scale reduction in the defect concentration. \(31\)

We see from Fig. 4.7 that, at a given value of \(E\), \(D(E)\) decreases monotonically with \(T_A\) as one might expect if noisy defects are removed during annealing. The shape of \(D(E)\) does, however, change in a subtle way. For \(T_A = 240 \, K\), we note that \(D(E)\) has a mild local minimum near \(E = 0.4 \, eV\), a minimum not present for \(T_A = 200 \, K\), nor when the film is fully annealed. Since \(D(E)\) and \(m(f,T)\) are linked via Eqs. (4.1) and (4.3), this behavior implies that the value of \(m\) measured at \(= 100 \, K\),
Fig. 4.7 Distribution of noise activation energies in irradiated film. Curves are calculated using Eqs. 4.1 and 4.2, and fits to noise data shown in Fig. 4.5.
should, as a function of $T_A$, start off larger than 1 for $T_A < 200$ K, drop below 1 for $T_A = 240$ K, then increase for larger $T_A$. This is precisely the "dip" observed in Fig. 4.4(b). In this analysis, the dip observed in $m$ (measured at 90 K) that occurs near $T_A = 240$ K is a consequence of the evolution of the shape of $D(E)$ as the sample is annealed. The shape of the curves in Fig. 4.7 at higher energy suggest that for noise measured at higher temperatures, the "dip" should become less pronounced. In fact, Fig. 4.6 does show that $m$ becomes a monotonically decreasing function of $T_A$ for $T > 120$ K. A plot of $m$ vs. $T_A$ with the noise measured at a temperature $T > 120$ K would not exhibit a "dip".

At this point we can offer no physical insight into the reason why the local minimum in $D(E)$ near $E - 0.4$ eV should appear at $T_A = 240$ K. We can only note that in samples held at 240 K, vacancy migration has commenced, and large scale reduction and redistribution of defects occur.

Finally, we would like to note the qualitative similarities between the results presented here in Figs. 4.5 - 4.7, and those described by Fleetwood and Giordano. These authors measured noise changes due to thermal annealing of defects present in AuPd films, freshly-sputtered onto room-temperature substrates. As in our study, Fleetwood and Giordano found that the noise magnitude increased strongly with temperature, especially near temperatures where annealing caused large changes in sample noise and resistivity. Annealing the films at high temperatures also caused a larger fractional reduction in the noise level measured at moderate temperatures than at relatively lower temperatures. This supports the hypothesis that defects with low noise
activation energy tend to have low annealing activation energy. They also found that changes in $S_p(f,T)$ caused by annealing were systematically reflected in the behavior of $m(T)$, consistent with Eq. (4.3). Thus even though the type and annealing activation energies of the pertinent defects in their study were quite different from those in ours, the overall behavior of the noise during defect annealing was quite similar.

IV.B. ANL Results

The following experiments were performed at the Argonne National Laboratory, in collaboration with Wayne King. The facilities at ANL are much better suited than those at LBL for quantitative measurements of the effects of defects produced in metal films by high energy electron and ion irradiation.

IV.B.1 Low Temperature Electron Damage Rates

The damage rate studies at LBL presented a few puzzles. The 500 keV electron damage rate measured at 90 K appeared at least a factor of four larger than that expected for bulk Cu, and significant subthreshold damage was found for 300 keV electron irradiation. This anomalous behavior was studied further at ANL.

The Kratos 1200 keV TEM at ANL has been extensively modified and automated to permit accurate low-temperature radiation damage studies.58,64 Low sample temperatures (~8 K) are necessary to suppress defect migration and to permit accurate ($\pm0.03 \text{ n}\Omega\text{cm}$) residual resistivity measurements. Analysis of irradiations at elevated temperatures is complicated by defect migration, recombination,
trapping, and clustering. The Kratos TEM also allowed incident electron energies up to 1.1 MeV. Damage rate measurements made at lower energies (i.e. 500 keV) may be complicated by the anisotropic threshold energy surface in Cu,$^4$ and the unknown distribution of Cu crystallites in our polycrystalline films.

The filled squares in Fig. 4.8 show the measured low-temperature damage rate $d\Delta \rho/d\phi$ vs. $\Delta \rho$, for sample Cu01 irradiated with 1.1 MeV electrons. The geometry of this sample is similar to that shown in Fig. 3.1(b), except in that the SiO$_2$ window was broken to allow an unobstructed path for electron dose calibration. The dashed line in Fig. 4.8 is the expected bulk damage rate, which for Cu is a linear function intersecting the vertical axis$^{64,73}$ at $-6 \times 10^{-27}$ $\Omega$cm$^3$, and the horizontal axis$^{64,66}$ at $\Delta \rho = \rho_m = 750$ n$\Omega$cm. The linear decrease in the bulk damage rate with increasing added defect concentration results from the spontaneous recombination of interstitials (vacancies) created close to existing vacancies (interstitials), and "sub-threshold" annealing effects.$^{66}$

The measured damage rate in sample Cu01 is fairly close to the bulk rate, and appears to decrease linearly at high $\Delta \rho$, but also exhibits enhanced damage below $\Delta \rho = 20$ n$\Omega$cm, which introduces some curvature in the plot. This curvature appears, at first glance, qualitatively similar to that measured in several other radiation damage experiments made on thin films.$^{58,64,74}$ In those studies, this effect is thought to result from Fuchs-Sondheimer$^{76,77}$ finite size corrections of the measured sample resistivity $\rho$, as compared with the intrinsic (material) resistivity $\rho_1$. These corrections are due to conduction electron scattering by the sample surfaces. They can be important when
Fig. 4.8 Low-temperature damage rate $\frac{d\Delta\rho}{d\phi}$ vs. $\Delta\rho$ for ANL sample Cu01, following 1.1 MeV electron irradiation. Filled squares are measured damage rates; dashed line is expected bulk Cu damage rate. Open symbols are corrected for conduction-electron surface scattering (see text), with assumed values $\kappa = 6 \times 10^{-12}$ $\Omega\text{cm}^2$, and $P = 0.4$ (open diamonds) or $P = 0.0$ (open circles). Solid curve is fit of Eq. (4.5) to open diamond data points.
the intrinsic electron mean free path $l_1$ is comparable to the sample thickness, and result in an enhancement of the measured damage rate as compared with the actual, intrinsic rate. The application of the Fuchs-Sondheimer theory to radiation damage measurements is discussed briefly in Appendix B.

The Fuchs-Sondheimer theory was applied to the data in Fig. 4.8 by using extrapolation formulae and numerical tables calculated by Groeger. Application of this theory requires knowledge of the sample thickness $d$, the measured resistivity, and two ill-defined materials parameters: $P$ and $\kappa$. The parameter $P$ is the fraction of electrons which are scattered specularly at the surfaces (the remainder are assumed to scatter diffusely), and $\kappa$ is the constant in the free-electron expression $\rho_1 = \kappa/l_1$.

The open diamonds and circles in Fig. 4.8 are the adjusted data with $P$ chosen to be 0.4 and 0.0, respectively, assuming a value $\kappa = 6 \times 10^{-12} \text{Ωcm}^2$. The overall magnitude of the damage rate can be modified substantially by such corrections, but the curvature in the data seen at low $\Delta\rho$ cannot be eliminated for any reasonable choice of $\kappa$ and $P$, and thus does not result simply from a Fuchs-Sondheimer effect.

The adjusted damage rate curves where each fit with a phenomenological expression

$$d\Delta\rho/d\phi = A(1 - \Delta\rho/B) + C\exp(-\Delta\rho/D). \quad (4.5)$$

The first term represents the expected bulk damage rate, which decreases linearly with $\Delta\rho$. The second term approximated the "anomalous" damage seen at small $\Delta\rho$. The value of $P$ was then varied to obtain the bulk value $B = \rho_m = 750 \text{ nΩcm}$. This was achieved with the very reasonable values of $\kappa = 6 \times 10^{-12} \text{ Ωcm}^2$ and $P = 0.4$, shown by the
open diamond symbols in Fig. 4.8. The solid line in the figure in the fit of Eq. (4.5) to the adjusted data. This adjustment actually amounts to a rather small correction to the raw data. The adjusted initial damage rate \( A = 5.2 \times 10^{-27} \, \text{Ocm}^3 \) is within experimental uncertainties of the known bulk value \( 6 \times 10^{-27} \, \text{Ocm}^3 \). The other adjusted parameter values are \( C = 0.8 \times 10^{-27} \, \text{Ocm}^3 \) and \( D = 14 \, \text{nOcm} \). Thus the "anomalous" part \( C \cdot \exp(\Delta p/D) \) represents only a small fraction of the total damage rate.

We conclude that the low temperature 1.1 MeV electron damage rate in this film is within 15% of the known bulk damage rate, but does exhibit a small anomalous enhancement visible for small \( \Delta p \).

In Fig. 4.9 we plot the low-temperature damage rate for sample Cu05, which also had its SiO\textsubscript{2} window broken to allow an unobstructed electron path. The damage rate was measured in three data segments. The first and third segments (open diamond symbols) are due to 500 keV electron irradiation, while the middle segment (open circles) is due to 1.1 MeV irradiation. The damage rate due to 1.1 MeV electrons is very similar in form and magnitude to the bulk rate, and to that measured in sample Cu01. The 500 keV damage rate is lower than the 1.1 MeV rate, but is nevertheless a factor of 2 - 3 larger than the expected bulk value\textsuperscript{46,73} for polycrystalline Cu.

We noted in section IV.A.2 that the LBL experiments indicated that thicker films had lower 500 keV damage rates than thinner ones, and that annealing the films (at 400° C) prior to irradiation significantly reduced the damage rate. Both observations are consistent with the proposal that fine crystal structure is responsible for the enhancement. King and co-workers have done preliminary radiation
Fig. 4.9 Low-temperature damage rate $d\Delta \rho / d\phi$ vs. $\Delta \rho$ for sample Cu05. Open diamonds were measured following 500 keV electron irradiation; open circles measured following 1.1 Mev electron irradiation.
damage experiments on freely-suspended Cu films specifically made to have extremely small crystal structure.\textsuperscript{59} These films were grown on a substrates at liquid nitrogen temperatures as a succession of thin (-10 nm) layers. The films were removed from the substrates, ion-milled into a rectangular geometry with four contacts, and mounted at room-temperature. Fig. 4.10 shows the low-temperature damage rate measured on such a film. The initial damage rate is over an order of magnitude larger than the bulk Cu value (shown by the dashed line in the figure), and decreases with increasing $\Delta p$ in a very uncharacteristic way. The films also exhibit nearly the same damage rate at 200 keV as at 500 keV, indicating a lack of a bulk-like energy threshold.

It has been suggested that this anomalous damage might result from interrupted replacement collision sequences.\textsuperscript{59} Figure 2.1(b) shows a typical bulk Cu replacement sequence. Following an incoming electron collision, the primary knock-on atom is displaced from its lattice position (leaving a vacancy), and replaces an adjacent atom, which replaces another, etc., until the replacement sequence ends with an atom in an interstitial position, located at some distance from the vacancy. This separation in general increases with increasing primary recoil energy, and determines the stability of the Frenkel Pair. The Pair will spontaneously recombine if the separation is less than a critical distance of about three atomic distances.\textsuperscript{45} For electron irradiation, most collision events are "subthreshold", and end with spontaneous recombination. In the presence of fine grain structure, a subthreshold replacement sequence might be interrupted by an extended defect structure which effectively traps the interstitial and stabilizes the defect. This tentative hypothesis is consistent with the high
Fig. 4.10 Low-temperature damage rate $d\Delta \rho / d\phi$ vs. $\Delta \rho$ for a fine-grained Cu film, following irradiation by electrons with three different kinetic energies. Dashed line is expected 500 keV bulk Cu damage rate. Data taken by King and co-worker at ANL; results are preliminary and unpublished.
initial damage rate followed by rapid saturation, and significant sub-threshold damage observed Fig. 4.10.

Most thin-film Cu samples used by King and co-workers\textsuperscript{46,58,64} had much larger-scale grain structure and exhibited bulk-value damage rates for 500 keV to 1.1 MeV electron irradiation.\textsuperscript{46,64} These films were thicker (typically 200 nm - 400 nm) than the samples used here, were deposited in clean conditions on substrates held at room temperature or above, and were irradiated while freely-suspended. We are not certain why the films in the current study, which were deposited in a similar manner, appear to show enhanced damage rates at 500 keV. We suspect that the presence of a substrate may be responsible. The grain structure in the films is likely affected by the substrate during evaporation, sample fabrication, and annealing. As noted above, small grain structure can lead to enhanced damage. The Si substrate and the Cu film also have significantly different thermal expansion coefficients, which result in a very large internal strain (- 0.15\%) in the Cu as it is cooled from room temperature to low temperatures.

We conclude from the low-temperature measurements that the 1.1 MeV damage rate in the Cu films is within the experimental uncertainty of the bulk damage rate, but does exhibit a small anomalous enhancement visible for small $\Delta p$. The 500 keV damage rate does however appear to be a factor of 2-3 times higher than the expected bulk rate. Small-scale grain structure and large internal strains may be connected to this enhanced damage rate.

\textbf{IV.B.2 Interstitial Traps}

The low-temperature measurements indicate that the 1.1 MeV damage
rate in the Cu films is close to the bulk rate, suggesting that most of the defects created are bulk-like vacancy-interstitial pairs. The defect population resulting from 1.1 MeV electron irradiation at 90 K is then expected to be, as in bulk Cu, primarily in the form of trapped interstitials and isolated, non-migrating vacancies (section II.B.1). It is natural at this point to consider each of these defects as a possible source of the induced 1/f noise.

On the one hand, the possibility that trapped interstitials are the primary noise source in the irradiated films is attractive from a fundamental point of view. An interstitial-impurity complex has a highly anisotropic structure, which (via conduction electron scattering) has a corresponding anisotropic contribution to the film resistivity.\textsuperscript{79,80} In chapter V it is shown that random activated reorientation of such defects can result in resistivity fluctuations large enough to account for the added 1/f noise measured in these irradiated films, provided one assumes that a sufficient fraction of the trapped interstitials reorient.\textsuperscript{29}

In contrast to interstitials, isolated bulk-like vacancies produce an isotropic resistivity in a metal,\textsuperscript{79,80} and would not be expected to cause resistivity fluctuations simply by moving from one bulk lattice site to another. Nevertheless, the LBL experiments do give strong indications that vacancies are connected to the noise. Figure 4.5 shows that the noise increases dramatically in the irradiated films when they are warmed near 200 K, just as vacancies begin to migrate.

We note here that Fig. 4.4(a) shows partial recovery of the noise when the sample was annealed in the temperature range 90 K - 150 K where interstitials are known to be released from traps,\textsuperscript{31,32} as well
as strong recovery in the range 200 K - 300 K where vacancies migrate.\textsuperscript{31} However, one should only with caution infer information about the identity of the noisy defects simply from this recovery of the added noise, since defect recombination simultaneously eliminates both a vacancy and an interstitial. The noise recovery observed for annealing temperatures in the 90 K - 150 K range could result, for example, from release of noisy, trapped interstitials. However, this recovery could also result when (quiet) interstitials are released from traps, then recombine with (and eliminate) noisy vacancies.

In order to study further the question of whether trapped interstitials in particular are primarily responsible for the radiation-induced noise, we have measured the noise at 90 K in nominally "pure" (undoped) Cu films, and in Cu films intentionally doped with In and Be. In bulk Cu, these impurities are known to act as interstitial traps. An isolated indium atom is an oversized impurity which traps interstitials with a moderate trapping energy\textsuperscript{32,33} of about - 0.3 eV. Interstitials trapped at isolated In impurities are mostly released at temperatures below 150 K. Furthermore, internal-friction measurements\textsuperscript{33,71} indicate that interstitials trapped at In impurities can undergo low-frequency, thermally activated \textit{rotations} around the In atoms in the temperature range 30 K - 150 K. Such trapped interstitials are a prime suspect as a low-frequency noise source. Beryllium, on the other hand, is an undersized impurity with a much stronger trapping energy.\textsuperscript{32,33} Ultrasonic attenuation measurements indicate that interstitials trapped at Be impurities undergo a very rapid "hopping" motion around the Be atom, with MHz hopping rates at liquid-helium temperatures.\textsuperscript{81} One therefore does not expect such defects to contribute to low-frequency noise. No
\textit{a priori} statements can be made concerning the expected noise behavior of residual traps present in all the films.

IV.B.2.a Trap Concentrations

In order to estimate the effective trap concentrations in the films, we made measurements of the "reciprocal damage rate" $d\phi/d\Delta\rho$ at 90 K, due to 1.1 MeV electron irradiation. In Fig. 4.11, $d\phi/d\Delta\rho$ is plotted vs. $\Delta\rho$ for several of the films listed in Table 3.1. After each irradiation, the sample was allowed to remain at 90 K for 10 min before it was cooled to a low temperature (-8 K) and its resistivity measured. As discussed in section II.B, the intercept with the vertical axis on such a plot is indicative of the intrinsic defect production rate, which should be approximately independent of trap concentration, while the initial slope of the plot is related to the trap concentration in each film. We see from Fig. 4.11 that the initial value of $d\Delta\phi/d\Delta\rho$ is roughly the same for all the samples. There does however exist a large range of initial slopes. In order to estimate these initial slopes, linear fits were made to the measured data points for which $\Delta\rho \lesssim 10$ n\Omega\,cm. These fits are shown by the dashed lines in Fig. 4.11. The corresponding initial values of the "trapping term" $Q_0$ (defined in Eq. (2.12)) are listed in table 3.1. If one assumes that the interstitial trapping radius $r_t$ and the vacancy capture radius $r_v$ are approximately equal for all traps observed here, these values of $Q_0$ indicate effective trap concentrations which range from about 15 - 25 atppm for the undoped films, 40-50 atppm for the CuBe and lightly doped CuIn films, to 140 atppm for heavily doped CuIn.

We note from Table 3.1 that the measured impurity contents of the
Fig. 4.11 90 K reciprocal damage rate $d\phi/d\Delta \rho$ vs. $\Delta \rho$ measured on seven samples following 1.1 MeV electron irradiation. (a) Undoped samples: CuO3 (filled triangles), CuO6 (filled circles), CuO4 (filled squares), CuO2 (open diamonds). Doped samples: CuBe2 (open circles), CuIn1 (open triangles), CuIn2 (open squares). Sample properties summarized in Table 3.1. Dashed lines are linear fits to initial data points (see text).
In-doped films are significantly larger than the above concentrations, and thus many of the traps are likely clustered rather than isolated In atoms. The main point remains that there does exist a large range of trap concentrations in the films.

**IV.B.2.b Effect of traps on 1/f noise**

In Fig. 4.12 we plot the change in the noise magnitude $\Delta(\rho^*P^2)$ measured at 80 K as a function of $\Delta\rho$ for six different samples, doped and undoped. Here we normalize the noise magnitude using $\rho^*P^2$ as defined by Eq. (2.5), with $f_0$ chosen to be 5 Hz. We see in this figure that $\Delta(\rho^*P^2)$ increases with $\Delta\rho$ by roughly the same amount in all the films measured, even though the films were fabricated with different types of traps, and with a factor of 10 range in effective trap concentration. Here we define the "specific noise" $\gamma_\rho$ of a sample as

$$
\gamma_\rho = \frac{\partial(\rho^*P^2)}{\partial(\Delta\rho)}.
$$

(4.6)

This parameter is indicative of the induced noise per added defect. In Fig. 4.12, a single line can be drawn which falls within ~30% of all but the most uncertain (lower left) data point, indicating that the specific noise $\gamma_\rho$ is quite insensitive to the type and quantity of interstitial traps present in the film.

If one assumes that trapped interstitials are responsible for the added noise, then one must produce a microscopic mechanism for the noise (e.g. an interstitial rotation, rearrangement, etc.) which is not sensitive to the type and average occupancy of the traps. This might prove difficult, as there are known differences in trapping strength between different impurities, leading one to expect differences in any noise-producing motions. Another difficulty for such a trapped-
Fig. 4.12 Change in 1/f noise magnitude $\Delta (\rho_\star^2)$ vs. $\Delta \rho$ for six Cu films following 1.1 MeV electron irradiation. Filled symbols are undoped Cu; open symbols are doped Cu. Symbols defined in Fig. 4.11.
interstitial noise mechanism is the absence of a strong saturation or change in the noise as the average trap occupancy surpasses 1. With the assumption that \( r_t - r_v \), such a saturation should occur when \( \Delta \rho > p_r c_t = Q_0 \). In Table 3.1, one sees that \( Q_0 = 4 \) nncm - 7 nncm for the undoped films, and 11 nncm - 38 nncm for the doped films. Fig. 4.12 shows that the noise in undoped films does not saturate in a particularly strong way as compared with the noise in the doped films.

If on the other hand one assumes that vacancies are responsible for the noise, then the "specific noise" should have little to do with the trapped interstitials. The observed insensitivity of the added noise to trap type and quantity would follow as a natural consequence. This possibility will be discussed further in chapter VI.

All the noise data shown in Fig. 4.12 were taken from samples irradiated with 1.1 MeV electrons. One can also compare, on a single sample, the noise induced by defects created with electron irradiation of different energies. In Fig. 4.13 the filled squares are the change in noise magnitude \( \Delta (\rho_s^2) \) plotted vs. \( \Delta \rho \) for sample Cu06, due to 1.1 MeV electron irradiation, while the open squares are due to 500 keV electron irradiation. We see that the "specific noise" is - 40\% higher for the 500 keV irradiation than for the 1.1 MeV irradiation, indicating that an "average" defect created by 500 keV electrons is somewhat noisier than that created by 1.1 MeV electrons. These results will be discussed further in chapter VI.

IV.B.3. Noise Induced by 1 MeV Kr⁺ Irradiation

As discussed in section II.B.2, high-energy ion irradiation of Cu creates large numbers of spatially correlated defects in the form of
Fig. 4.13 Change in noise magnitude $\Delta(\rho^2)$ vs. $\Delta\rho$ for sample Cu04 following 500 keV electron irradiation (open squares) and 1.1 MeV electron irradiation (filled squares).
"displacement cascades." If the irradiations are performed at elevated temperatures, most of the Frenkel Pairs will recombine or cluster in a correlated way. A fraction of the defects will, however, escape the cascades and will be able to migrate and react as isolated vacancies and interstitials. For 1 MeV Kr⁺ irradiation of Cu, the average primary recoil energy is ~60 keV, and one therefore expects most of the defects which survive intra-cascade recombination will be in the form of large clusters. Of the order of 15% of the surviving defects are, however, expected to escape the large clusters. One intuitively expects that the "specific noise", resulting from such a defect configuration will be quite different from that due to electron irradiations.

We have compared 1/f noise in Cu films following 1.1 MeV electron and 1 MeV Kr⁺ irradiation. The open circles in Fig. 4.14 show the change in the noise magnitude $\Delta(\rho^2)$ measured at 80 K as a function of $\Delta\rho$ for sample Cu02 following 1.1 MeV electron irradiation, while the open squares show the change in the noise following 1 MeV Kr⁺ irradiation. The electron irradiations preceded the ion irradiations, and the two data sets were separated by an extended room-temperature annealing process. Both irradiation sequences were performed with the sample held at 90 K. We see from Fig. 4.14 that the specific noise $\gamma_\rho$ is significantly larger for defects created by electron irradiation as compared with those created by Kr⁺ ions. Comparison of the two sets of data shows that one requires roughly a factor of 8 times the value of $\Delta\rho$ for the Kr⁺ irradiations in order to achieve the same noise level found in the film following electron irradiation. Similar results were also obtained from sample CuIn1. When compared with the estimated
Fig. 4.14 Change in noise magnitude $\Delta(\rho^2)$ vs. $\Delta\rho$ for sample CuO2 following 1.1 MeV electron irradiation (open circles) and 1 MeV Kr+ irradiation (open squares). Dashed lines are guides to the eye.
fractions of isolated and clustered defects in the ion-irradiated films, these data suggest that isolated defects are primarily responsible for the added 1/f noise, and that most (if not all) the added noise observed following Kr⁺ irradiation is due to the fraction of the defects which remain isolated. This interpretation is consistent with the known fact that large defect clusters are physically more stable than individual vacancies and interstitials.
V. A "LOCAL-INTERFERENCE" MODEL FOR 1/F NOISE IN METAL FILMS

The work presented here demonstrates that crystalline defects are a source of 1/f noise in Cu films. This agrees with the results of a number of other studies\textsuperscript{15,19,20-26} (discussed in chapter I) which indicate that defects and/or defect motions are responsible for the 1/f noise observed in a variety of metal films. The 1/f form of the spectral density and the noise kinetics are successfully described by the Dutta-Dimon-Horn thermal activation model,\textsuperscript{16} as discussed in sections IV.A.5 and Appendix A. However, the microscopic mechanism by which a defect motion gives rise to a resistivity fluctuation, and the expected magnitude of such a fluctuation remain very much open questions.

Over the past decade, it has been suggested that interference of the wave functions of conduction electrons scattered by defects might result in resistivity fluctuations as defects move.\textsuperscript{25,27-30,69,82-83} Kogan and Nagaev\textsuperscript{27} and Black, Restle, and Weissman\textsuperscript{25} have discussed changes in the scattering cross-section of asymmetric defects as they move, but did not carry out detailed calculations of the magnitude of these changes. We shall call this type of model a "Local Interference" (LI) model for reasons which will become apparent. More recently, Feng et al.\textsuperscript{28} proposed an alternative model based on "Universal Conductance Fluctuations" (UCF) in order to relate resistivity fluctuations to defect motion.

In this section, we first use the defect-scattering calculations of Martin\textsuperscript{79,80} to estimate the magnitude of the resistivity fluctuations produced in the LI model. We then apply this model to the measurements
of the 1/f noise in thin Cu films in which defects have been induced by electron-irradiation, discussed in chapter IV.A. Finally, we compare the predictions of the LI and UCF models in order to determine the conditions under which each is expected to be the predominant mechanism for 1/f noise.

The most complete LI model to date is that of Kogan and Nagaev, who in particular considered rotations of point defects with symmetry lower than the crystal. To estimate the change \( \delta \sigma \) in the average cross section \( \sigma \) of a defect when it rotates, the authors considered electron scattering from anisotropic local defect potentials and set up a Boltzmann equation to determine the effect on the conductivity. However, they did not actually solve the equation, but rather used dimensional arguments to obtain the order-of-magnitude estimate \( \delta \sigma = a_0^2 \sigma \), where \( a_0 \) is a lattice constant. One clearly requires a quantitative calculation of \( \delta \sigma \) to justify this estimate and to make a realistic comparison with both experiment and the UCF model.

In fact, J. W. Martin performed such calculations some 10 years prior to the work of Kogan and Nagaev. Martin considered scattering of conduction electrons from structural defects consisting of a number of vacant lattice sites and interstitial atoms, each of which acts as an isotropic scattering center. He showed that interference of electrons scattered by the centers causes the resistivity tensor \( \rho_{ab} \) to be anisotropic, and to change with varying separation of the centers. This interference can be included in the calculations in the form of a structure factor. The total interference term for an arbitrary configuration can be expressed as a sum of dipole interference terms, one for each pair of scattering centers. Furthermore, for a given pair...
the dependence of $\rho_{\alpha\beta}$ on the dipole orientation and on the separation $R$ of the centers is large only for $R < \lambda_F$, where $\lambda_F$ is the Fermi wavelength. For $R \gg \lambda_F$, the resistivity tensor becomes isotropic and independent of $R$. We accordingly designate a $1/f$ noise model based on this effect a "local interference" model, since the principal effect arises only from interference of electrons scattered by centers within a few lattice constants of each other. In the remainder of this chapter, we focus on the anisotropy introduced by the interference and the resulting effect on the resistivity when defects reorient. In considering more general defect rearrangements, one should also include effects of changing the separation of the scattering centers. We note here that Martin assumed the Fermi surface to be spherical, which is approximately correct for a nearly-free electron metal, and single-scattering matrix elements which are independent of position of the center relative to the lattice. Only the positions of the scattering centers relative to each other enter these resistivity calculations.

Martin calculated the tensor components $\rho_{xx}$, $\rho_{yy}$, and $\rho_{zz}$ for several defect configurations likely to occur in fcc metals after high-energy electron, neutron or ion irradiation. Here the $x$, $y$, and $z$ directions correspond to the fcc $(100)$ axes. For some of these defects one can use the defect symmetry and the properties of tensor rotations to determine the principle moments $\rho_1$, $\rho_2$, and $\rho_3$ of the resistivity tensor $\rho_{\alpha\beta}$. If one measures the resistivity in a particular direction (say the $z$ direction) and assumes random defect orientation, one can show that
\[ \langle p_{zz} \rangle \equiv \langle \rho \rangle = (p_1 + p_2 + p_3)/3, \quad (5.1) \]

\[ \langle (p_{zz} - \langle \rho \rangle)^2 \rangle \equiv \langle (\delta \rho)^2 \rangle = \]

\[ (4/45)(p_1^2 + p_2^2 + p_3^2 - p_1p_2 - p_1p_3 - p_2p_3). \quad (5.2) \]

We define a root mean square anisotropy parameter \( \beta \) by

\[ \beta^2 \equiv \langle (\delta \rho)^2 \rangle / \langle \rho \rangle^2 = \langle (\delta \rho)^2 \rangle / \langle \rho \rangle^2. \quad (5.3) \]

This parameter expresses the average fractional change of the scattering cross section of a defect when it moves.

In Table 5.1 we list several defects considered by Martin, along with \( \beta \) and the relevant tensor components. We see that values of \( \beta \) are typically a factor of 5 to 10 smaller than the estimate \( \delta \sigma - \sigma \) given by Kogan and Nagaev. Comparison of the first- and second-nearest-neighbor divacancy cases in Table 5.1 demonstrates that \( \beta \) decreases rapidly with increasing separation of the scattering centers.

We now express the resistivity noise magnitude in terms of the parameter \( \beta \) and the concentration of mobile defects. Similar expressions have been derived previously.\(^{15,27,49}\) We define:

\[ \rho = \kappa \ell^{-1}, \quad (5.4) \]

\[ \ell^{-1} = \ell_{\text{in}}^{-1} + \ell_{\text{e}}^{-1} \quad (5.5) \]

and
Table 5.1. Resistivity tensor components and anisotropy parameter $\beta$ for six types of defects. The components $\rho_{xx}$, $\rho_{yy}$ and $\rho_{zz}$ are from Ref. 80; $\rho_1$, $\rho_2$ and $\rho_3$ are the corresponding principal moments. For each defect type, the upper row lists values for the bare defect, while the lower row includes effects from surrounding lattice relaxation. All resistivities are normalized to that of the bare monovacancy.

<table>
<thead>
<tr>
<th>Defect type</th>
<th>$\rho_{xx}$</th>
<th>$\rho_{yy}$</th>
<th>$\rho_{zz}$</th>
<th>$\rho_1$</th>
<th>$\rho_2$</th>
<th>$\rho_3$</th>
<th>$\beta$</th>
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<tr>
<td>Monovacancy</td>
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<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
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<tr>
<td></td>
<td>0.92</td>
<td>0.92</td>
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<td>0.92</td>
<td>0.92</td>
<td>0.92</td>
<td>0.00</td>
</tr>
<tr>
<td>1st neighbor</td>
<td>1.80</td>
<td>1.80</td>
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<td>2.27</td>
<td>2.27</td>
<td>1.33</td>
<td>0.14</td>
</tr>
<tr>
<td>divacancy</td>
<td>1.65</td>
<td>1.65</td>
<td>2.13</td>
<td>2.13</td>
<td>2.13</td>
<td>1.17</td>
<td>0.16</td>
</tr>
<tr>
<td>2nd neighbor</td>
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<td>1.85</td>
<td>2.04</td>
<td>2.04</td>
<td>2.04</td>
<td>1.85</td>
<td>0.03</td>
</tr>
<tr>
<td>divacancy</td>
<td>1.73</td>
<td>1.70</td>
<td>1.73</td>
<td>1.73</td>
<td>1.73</td>
<td>1.70</td>
<td>0.01</td>
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<tr>
<td>$180^\circ$</td>
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<td>2.58</td>
<td>2.58</td>
<td>3.54</td>
<td>3.54</td>
<td>1.62</td>
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<td>trivacency</td>
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<td>2.45</td>
<td>3.35</td>
<td>3.35</td>
<td>1.58</td>
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<td>1.08</td>
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<td>1.08</td>
<td>1.08</td>
<td>2.33</td>
<td>0.25</td>
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<tr>
<td>interstitial</td>
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<td>2.17</td>
<td>3.63</td>
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<td>2.33</td>
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<tr>
<td>interstitial</td>
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<td>2.44</td>
<td>2.44</td>
<td>2.44</td>
<td>4.51</td>
<td>0.20</td>
</tr>
</tbody>
</table>
Here Eq. (5.4) is the free-electron expression for the resistivity $\rho$ in terms of the electron scattering length $l$, and $\kappa$ is a material-dependent parameter. Equation (5.5) is Matthiessen's law, which expresses $l$ in terms of the inelastic length $l_{in}$ and the elastic (defect) length $l_e$. In Eq. (5.6) the sum extends over all defect cross-sections $\sigma_i$ in the sample volume $V$.

If a particular defect moves and its cross section changes by an average amount $<\delta \sigma_i> = \beta \sigma$, we have

$$\delta \rho_i = K(\beta \sigma/V). \quad (5.7)$$

If we now assume a concentration $n_m$ of mobile defects with uncorrelated motion, then the total normalized fluctuation in the sample is

$$N<(\delta \rho)^2>/\rho^2 = (n\delta \sigma)^2(n_m/n), \quad (5.8)$$

where $N$ is the number of atoms in the sample and $n = N/V$.

To test this model rigorously, one should measure the noise and the concentration of mobile defects in a particular sample. To date this has not been done. However, we have measured the radiation-induced 1/f noise in Cu films maintained at 90 K as a function of the total added defect concentration. In this system, we can thus put an upper limit on the added mobile defect concentration, and can check the consistency of the LI model with these measurements.
Here, the 1/f noise magnitude will be characterized in terms of the parameter $\alpha$ defined in Eq. (2.4);

$$NS_{\rho}(f)/\rho^2 = \alpha/f.$$ 

Integrating this expression over the experimental bandwidth (0.1 Hz to 25 Hz) we obtain $N<(\delta\rho)^2/\rho^2 = \alpha \ln(25/0.1) = 5.5\alpha$. For an added resistivity $\Delta \rho = 10 \ \text{n\Omega cm}$, Fig. 4.3 shows that the parameter $\alpha$ changed by $\Delta \alpha = 1.6 \times 10^{-3}$. Using Eq. (5.8) and $n = 8.5 \times 10^{22} \ \text{cm}^{-3}$, $l = 800 \ \AA$, $\beta = 0.15$, and $e = 4\pi/k_F^2 = 6.5 \times 10^{-16} \ \text{cm}^2$, one can account for the added noise with $(n_m/n) = 2 \times 10^{-5}$. An added resistivity $\Delta \rho = 10 \ \text{n\Omega cm}$ corresponds to a fractional concentration of Frenkel Pairs $n_{FP}/n = 4 \times 10^{-5}$, and we thus require about 5% of the added defects to be mobile within the experimental bandwidth to account for the added noise. If we assume the 1/f spectrum extends over a larger bandwidth, we require larger numbers of moving defects: For example, a bandwidth of 10 decades would require 20% of the added defects to be mobile.

These numbers are quite reasonable, especially when one considers that all the added defects are metastable and hence inherently mobile. Most of the defects anneal at temperatures below 300 K, via thermally activated free migration and defect trap release. Precisely how the type of motion which produces noise relates to the motion involved in annealing has yet to be determined.

When the temperature of the films is raised above 90 K, the induced 1/f noise increases. This behavior is shown in Fig. 4.5. In particular, the films become extremely noisy at temperatures near 200 K, just below the temperature at which large scale annealing of the defects occurs via vacancy migration. To account for the high level of noise that we observe at 185 K after annealing the film at 200 K for
5 min, the LI model would require approximately 50% of the remaining defects to be mobile within the experimental bandwidth, although uncertainties in the values of $\beta$ and $\sigma$ make this estimate somewhat rough. In fact, such a large fraction may not be unreasonable since most of the defects are on the verge of annealing and a high degree of thermal motion is likely. Furthermore, in this particular case, a measureable decrease in the average resistivity of the sample occurred during the noise measurement, confirming that there was indeed substantial defect motion. This also suggests that non-stationary processes not specifically included in the LI model (e.g. defect dissociation, free migration and annihilation) may have added substantial amounts of noise.

Measurements of 1/f noise have been made on a wide variety of metal films at room temperature, with $\alpha$ in the range $10^{-5} < \alpha < 10^{-1}$. In relatively clean metals ($\lambda = \lambda_{in} = 400$ A) the LI model would require mobile defect concentrations in the range $2 \times 10^{-7} < n_m/n < 2 \times 10^{-3}$ to account for the range of 1/f noise if one assumes a 10-decade bandwidth. These concentrations of mobile defects would seem reasonable, especially if we include atoms at grain boundaries and surfaces as "lattice defects."

In concluding this discussion of the LI model we note that it requires defects which are each composed of multiple scattering centers within one or two lattice constants of each other, and not a system composed of randomly placed point scattering centers with no spatial correlation. In such a random system with average scattering center separation $R >> \lambda_F$ one expects less noise than outlined above. However, in a (mostly) crystalline real metal, anisotropic agglomerates
of near-neighbor scattering centers are in fact the norm,\textsuperscript{31} in the form of "split" interstitials, defect clusters, dislocations, grain boundaries, surfaces, etc., with the monovacancy and substitutional impurities notable exceptions. One therefore expects large interference effects in these structures, with corresponding resistivity fluctuations when scattering centers move.

We now turn to a discussion of the $1/f$ noise produced by the LI and UCF models, both of which are based on scattered electron interference effects, but which differ significantly in several ways. The LI model considers \textit{single} scattering events by defects in relatively clean systems. Only interference from nearby scattering centers produces a resistivity change when a defect moves. The UCF model,\textsuperscript{28} on the other hand, considers interference effects from \textit{multiple} elastic scattering events in disordered systems, and is non-local in the sense that the motion of any scattering center within an inelastic diffusion length $L_{1n}$ can affect the interference. The theory was originally formulated for the limit $k_F a_e \sim 1$, where $k_F$ is the Fermi wavevector, but should remain applicable when $k_F a_e > 1$ provided $l_{1n} > l_e$. To obtain a quantitative estimate of the $1/f$ noise predicted by the UCF model, we consider the three dimensional case in which all sample dimensions are larger than $L_{1n}$ and assume that the conductance fluctuations from all mobile defects within a cube of side $L_{1n}$ add incoherently. We take $\rho = m_e v_F / n e^2 l$ ($v_F$ is the Fermi velocity), assume one conduction electron per atom, and assume sufficient disorder to make $l = l_e$. From Eqs. (3) and (6) of ref. 28, assuming $\alpha(k_F \delta r) = f(\kappa / k_B T_{1n}) \sim 1$, we obtain
\[
N\langle(\delta\rho)^2\rangle/p^2 = (m_e \nu_F/\hbar)^2 (L_{in}/\ell)^3 (n_m/n) \sigma
= (m_e/\hbar)^2 (\nu_F^2 \tau_{in}^3/27\ell^3)^{1/2} (n_m/n) \sigma,
\]
(5.9)

where we have set \( L_{in} = (\nu_F \tau_{in}/3)^{1/2} \) with \( \tau_{in} \) the inelastic scattering time. Equation (5.9) is valid provided \((n_m/n) \leq (\ell/\sigma L_{in}^2)\). The noise saturates for larger \((n_m/n)\) since the conductance fluctuation per cubic volume \( L_{in}^3 \) cannot exceed \( 28 \cdot e^2/\hbar \). We see that at a given temperature and mobile defect concentration the UCF model predicts that \( N\langle(\delta\rho)^2\rangle/p^2 \) scales as \( \ell^{-3/2} \) while the LI model predicts a scaling with \( \ell^2 \). Thus, in sufficiently dirty metals the UCF mechanism is expected to dominate while in sufficiently clean metals the LI mechanism takes over.

Taking the ratio of Eqs. (5.8) and (5.9) we find

\[
\frac{\langle(\delta\rho)^2\rangle_{LI}}{\langle(\delta\rho)^2\rangle_{UCF}} = \left(\frac{nB\hbar}{m_e}\right)^2 \sigma \left(\frac{27\ell^7}{\nu_F^2 \tau_{in}^3}\right)^{1/2}.
\]
(5.10)

Using the values of \( n, B, \) and \( \sigma \) listed above with \( \nu_F = 1.6 \times 10^8 \) cms\(^{-1}\) and \( \tau_{in} = \hbar/KB_T \) at \( T = 300 \) K, we find that the two mechanisms produce equal levels of \( 1/f \) noise when \( \ell = \ell_e = 25 \) \( \text{A} \ \leq \ell_0 \). In metals with \( \ell_e > \ell_0 \) we thus expect the LI mechanism to dominate, while in clean metal films with \( \ell_e > \ell_{in} \) the UCF theory does not apply, leaving only the LI mechanism. Equation (5.10) also demonstrates that for a given mean free path the UCF mechanism becomes progressively more important as the temperature is lowered to produce an increase in \( \tau_{in} \).

In summary, we have used Martin's calculations of defect resistivity to make quantitative predictions of \( 1/f \) noise generated by moving defects. Martin's work emphasizes the importance of local interference effects, which can produce \text{rms_fractional} asymmetries in the cross-section of defects ranging from 0 to 0.25. If one assumes a
value $\beta = 0.15$, these calculations can account for the 1/f noise generated by radiation-induced defects in Cu films at 90 K provided one assumes that approximately 5% of the added defects move at frequencies within the experimental bandwidth, though a larger percentage is required when the irradiated films are at a higher temperature. In the case of metal films at room temperature, the measured range in the level of 1/f noise over a similar bandwidth requires a fractional concentration of mobile defects ($n_m/n$) between $10^{-7}$ and $10^{-3}$. At room temperature, the 1/f noise in metal films with mean free paths greater than about 25 Å should be dominated by the LI mechanism rather than by the UCF mechanism; at lower temperatures and/or shorter mean free paths the UCF mechanism is expected to dominate.

Finally, we note that Zimmerman and Webb at Cornell University are currently studying the magnitude of the resistivity fluctuations due to H$^+$ ions diffusing in Palladium films. Their measurements indicate a value for the quantity $(\delta \rho/\rho)$ to be in the range 0.2 - 0.5, in good agreement with Martin's estimates.
VI. DISCUSSION

The experimental results discussed in chapter IV give direct evidence that radiation-induced defects are a source of $1/f$ noise in Cu films, and the "Local Interference Model" discussed in chapter V can plausibly account for the observed noise magnitude. However, we also wish to determine the identity of the particular defect types responsible for the induced noise, since a variety of defect configurations can in general result from electron and ion-irradiation.

In this section, we consider several possible sources of the radiation-induced $1/f$ noise observed in these experiments. In particular, we consider several models which each assume a particular type of defect is primarily responsible for the induced noise, and compare each model with the results discussed in chapters IV and V.

Model (A): We first consider a model in which one assumes that the noise does not result from crystalline defects within the Cu at all, but rather from some interaction of the film resistivity with defects created in the underlying substrate. This model is not consistent with a number of experimental results. We note first that the added resistivity must primarily be due to defects within the Cu. This can be seen from the low-temperature 1.1 MeV damage rates (Figs. 4.8 and 4.9), which are close to bulk Cu damage rates. We have also seen that when electron irradiations are performed at 90 K, the added resistivity $\Delta \rho$ increases with electron dose $\phi$ in a way which is consistent with the "Unsaturable Trap Model" (section II.B.1) known to apply to bulk Cu. Next we note from Fig. 4.4(a) that both the added resistivity and
induced noise show strong recovery at annealing temperatures in the range 200 K - 300 K. Thus there exists a definite correlation between the induced noise and the added resistivity, which, as noted above, is primarily due to defects within the Cu film. This temperature range in which strong recovery in the noise and resistivity occurs corresponds to the range where free migration of monovacancies is known to occur in bulk Cu.

The above Model (A) is also inconsistent with the results shown in Fig. 4.12. If the induced noise results only from defects within the substrate, then it should depend only on the total electron dose $\phi$, and not depend on the trap concentration within the Cu film. Undoped Cu films maintained at 90 K must be irradiated to a larger values of $\phi$ than doped films in order to build up a given value of $\Delta \rho$, since there are fewer impurities present to trap interstitials. One would therefore expect that a plot of $\Delta(\rho^2)$ vs. $\Delta \rho$ to behave roughly as sketched in fig. 6.1(a), with the undoped films becoming increasingly noisier as $\Delta \rho$ increases. This expected behavior is not consistent with the experimental results shown in Fig. 4.12.

For these reasons, we conclude that the induced noise is connected directly to defects created within the Cu films.

One can also consider the possibility that the induced noise results simply from the small amount of vacancy-interstitial recombination which occurs during the noise measurements. Such recombination causes the sample resistivity to decrease monotonically in time, in small random steps as individual Frenkel Pairs recombine. In Appendix C we show that such "recombination noise" can neither account for the magnitude nor the spectral shape of the induced noise.
Fig. 6.1 Behavior of $\Delta(p_*^2)$ vs. $\Delta \rho$ predicted by various simple models which each attribute the induced $1/f$ noise to a particular type of defect (see text).
We now consider several models which attribute the induced noise primarily to trapped interstitials. Before considering specific cases, we make the general point that an isolated interstitial and an interstitial-impurity complex are both anisotropic defect structures which, in accordance with the "Local Interference Model" discussed in chapter V, should produce substantial resistivity fluctuations if the defects reorient. From a theoretical point of view, trapped interstitials are quite appealing as a possible noise source.

Model (B): In this model, it is assumed that interstitials trapped at dopant atoms are significantly noisier than those at residual traps, and are the dominant noise source in the Cu films. By "residual" we mean here traps which are present in all the films in roughly equal concentrations. From such a model, one would expect that the doped films would have significantly larger specific noise than the undoped films, for all values of $\Delta p$. This expected behavior is sketched in Fig. 6.1(b). It is not consistent with the measured behavior shown in Fig. 4.12, which shows the specific noise to be roughly independent of trap concentration.

Model (C): Interstitials at residual traps are assumed noisier than those at dopant atoms, and are assumed to be the dominant noise source. In the doped films, only a fraction of the interstitials will be trapped at the residual sites, and thus one would expect the undoped films to be noisier. This is sketched in Fig. 6.1(c), and is not consistent with the data.

Model (D): All trapped interstitials produce approximately equal noise levels, independent of trap type and occupancy. This proposal is consistent with the behavior shown in Fig. 4.12, and cannot be ruled
out. However, it is not physically intuitive that interstitials trapped at different impurities should exhibit similar noise behavior. This is especially true if one compares oversized impurities (e.g. In) with undersized impurities (Be), which are known to have quite different trapping energies and interstitial-impurity defect structures. Here we must qualify our experimental conclusions somewhat since the active-trap concentration in our Be-doped samples is only a factor of two larger that in the undoped films. Further experiments with higher Be trap concentrations are desireable in order to determine whether or not there might yet exist subtle differences between the noise behavior of the undoped films, and those doped with undersized and oversized impurity traps.

Before considering other possible noise sources, we note that any model which attributes the bulk of the noise to trapped interstitials is not particularly consistent with the measurements summarized in Fig. 4.5, which shows that the irradiated films become extremely noisy when heated near 200 K. Near this temperature, vacancies are known to migrate in bulk Cu, but nothing in particular is known to happen to trapped interstitials, other than that they recombine with the migrating vacancies.

Model (E): Isolated vacancies within the bulk produce the noise. In this case, the added noise should be independent of trap type, concentration, and average occupancy, consistent with the results in Fig. 4.12. One might also expect intuitively that the noise should increase as the irradiated films are heated close to temperatures at which vacancies migrate, consistent with the behavior shown in Fig. 4.5. Of the five models so far considered, this model is the most
consistent with the experimental results.

From a physical basis, however, it is hard to understand how any kind of motion of a bulk-like, symmetric, isolated vacancy could produce a resistance fluctuation. A rotation of a monovacancy has no physical significance, and a simple translation from one lattice site to another will simply reproduce the local environment around the vacancy. Below we consider a variation of the above model which avoids this translational invariance.

Model (F): Vacancies close to a surface, grain boundary, dislocation or some other extended defect structure are assumed primarily responsible for the noise. A translation of such a vacancy will change the local environment of the defect, and produce a resistivity fluctuation. By "close" here we mean a distance of the order of a Fermi wavelength $\lambda_F \sim 0.5$ nm, in accordance with the Local-Interference model discussed in chapter V.

Figure 4.13 offers additional experimental information which should also be considered. This figure indicates that the specific noise $\gamma_p$ resulting from 500 keV electron irradiation is larger than that from 1.1 MeV electrons. This is somewhat counter-intuitive, since the structure and distribution of stable defects in bulk Cu resulting from electron irradiation is not significantly different at these two energies.

One major difference is, however, that any "subthreshold" effects are expected to be fractionally more important at the lower energy, since the intrinsic (bulk) damage rate is roughly an order of magnitude smaller at 500 keV than at 1.1 MeV. In section IV.B.2 it was suggested that the anomalously high damage rates measured for 500 keV electron
irradiation was due to a subthreshold damage mechanism related to small-scale crystallite structure within the films. If in fact a subthreshold mechanism enhances defect production near surfaces and grain boundaries, then Model (F) would predict that the specific noise $Y_p$ resulting from 500 keV irradiation should be larger than that resulting from 1.1 MeV electrons. Further experiments involving a range of film thickness and grain size and a wider range of electron energies would help determine if in fact there does exist a definite relation between enhanced damage production and enhanced $Y_p$ observed at lower electron energies, as well as a specific dependence on film structure.

Model (F) is consistent with the experimental results, and with the theoretical requirement that a defect motion change its local environment. However, one now expects that only those vacancies located close to a surface, grain boundary, etc., which we shall refer to a boundary vacancies, will be available to contribute to the noise. The motion of vacancies located deep within a crystallite will not result in resistance fluctuations. In chapter V, estimates were made of the fraction of the total number of added defects which must move in order for local-interference effects to account for the induced noise. If this required fraction is instead expressed relative only to the boundary vacancies, then it must be multiplied by a factor of $F^{-1}$, where $F$ is the fraction of vacancies located within $\lambda_F$ of a surface, grain boundary, etc.

One can estimate $F$ by considering a typical Cu grain to be a cube of dimension $D$. Very roughly then,

$$F = 6\lambda_F/D.$$  \hspace{1cm} (6.1)
For $\lambda_F = 0.5$ nm and $D = 100$ nm, one has $F = 0.03$. If we accept Model (F) above, we require that the fraction of boundary vacancies which must be mobile is $1 - 2$ orders of magnitude larger than the estimates of the mobile-defect fraction made in chapter V. Those estimated fractions were of the order of $0.05 - 0.50$. Consequently, in order to apply local-interference effects to Model (F) above, one must require that nearly all the boundary vacancies be mobile in order to account for the induced $1/f$ noise. This requirement is of borderline physical plausibility.

However, one cannot rule out Model (F) simply on this basis, since several parameters in the above analysis are rather uncertain. For example, the noise magnitude predicted by Eq. (5.8) scales as $\beta^2$, where $\beta$ is the "anisotropy" parameter defined by Eq. (5.3). A value of $\beta = 0.15$ was assumed. The actual value of $\beta$ could conceivably differ from this number by as much as a factor of 3, with a corresponding change in the predicted noise level of up to an order of magnitude. Also, the fraction $F$ defined by Eq. (6.1) is a very rough estimate. The actual fraction $F$ of "boundary vacancies" could be considerably larger if the films contain large numbers of dislocations or other extended defects within the Cu grains.

Finally, we note that Model (F) is similar to a proposal made by Koch et al.\textsuperscript{24} that the $1/f$ noise measured in evaporated Al films was due to the motion of vacancies along grain boundaries.

In summary, none of the models presented here provide an entirely satisfactory explanation of the induced $1/f$ noise in terms of local-interference effects. Model (F) above appears to be most consistent with experimental results and theoretical requirements of
local-interference effects, but has some trouble accounting for the magnitude of the induced noise. Models (D) and (E) are both consistent with Fig. 4.12, but each has problems. Model (D) would require that all trapped interstitials produce roughly equal noise levels, and does not offer an obvious reason why the noise should increase dramatically at temperatures where vacancies migrate. With Model (E), it is not clear how the motion of a symmetric monovacancy can result in a resistance fluctuation. Models (A), (B), and (C) are all inconsistent with the experimental results discussed in chapter IV.

Finally, it should be noted that none of these models has considered the effects on the damage rate or induced noise of the very large internal stress present in the Cu films. By taking the difference in the thermal expansion coefficients of Cu and Si, one finds that the Cu films will experience a tensile strain of about 0.15% upon cooling from room temperature to 90 K. This strain is close to the elastic limit for bulk Cu. Further experiments in which the films are subject to variable internal stress may prove useful in determining whether internal stress is connected to the radiation-induced noise.
VII. CONCLUSIONS

VII.A. Summary

The work presented here demonstrates that high-energy electron and ion irradiation of metal films is a useful technique to study the relationship between 1/f noise and defects in metals. The radiation-induced 1/f noise in polycrystalline Cu films increases in a systematic way with the number of defects introduced, indicating a direct connection between 1/f noise and defects in metals. The difference in the recovery of the induced noise and the added resistivity after successive annealing steps suggests that a large fraction of the induced noise is generated by a subpopulation of "noisy" defects, presumed to be mobile, that are more readily annealed than the majority of the added defects. The temperature dependences of the noise magnitude and the frequency exponent m after irradiation and partial annealing are consistent with the Dutta-Dimon-Horn\textsuperscript{16} model, indicating that thermally activated kinetics govern the induced 1/f noise.

The low-temperature damage rates measured in the Cu films due to 1.1 MeV electron irradiation are found to be close to bulk Cu damage rates. In contrast, the 500 keV damage rates are found to be significantly higher than the corresponding bulk damage rates. It is suggested that a subthreshold effect related to small-scale structure within the films may be responsible for the enhanced 500 keV damage.

The measured reciprocal damage rates due to 1.1 MeV electron irradiations performed at 90 K were used to estimate the effective interstitial trap concentrations in undoped Cu films, and films doped...
with In and Be. It is found that the specific noise $\gamma_p$, which is proportional to the induced noise per added defect, is not sensitive to the type and quantity of interstitial traps present in the films. Isolated defects introduced by electron irradiation appear to generate much more noise than the mainly clustered defects introduced by 1 MeV Kr⁺ irradiation.

A simple "Local Interference" model was presented, which uses calculations of Martin⁷⁸,⁸⁰ to make quantitative estimates of the 1/f noise magnitude generated by moving defects. This model can account for the magnitude of the observed radiation-induced 1/f noise, provided one assumes a sufficient fraction of the added defects move at frequencies within the experimental bandwidth. This model was compared with an alternated model, based on "Universal Conductance Fluctuations"²⁸ (UCF's). At room temperature, the Local-Interference mechanism should dominate the 1/f noise magnitude in metal films with mean free paths greater than about 2.5 nm, while at lower temperatures and/or shorter mean free paths the alternate mechanism based on UCF's is expected to dominate.

Several simple models were examined concerning the identity of the defect responsible for the radiation-induced 1/f noise. A model which attributes the noise to the motion of vacancy-type defects close to surfaces, grain boundaries, or dislocations is most consistent with the experimental results and theoretical considerations, though none of the models considered provides an entirely satisfactory explanation of the induced 1/f noise.
VII.B. Future Work

These experiments have contributed important information concerning the identity, structure, and dynamics of the defects responsible for the induced 1/f noise in Cu films, and local-interference effects provide a highly plausible mechanism by which defect motions produce noise. Yet several puzzles remain. Further studies or radiation-induced noise could help resolve some of these puzzles.

One could investigate the effects of the crystal grain structure on the induced noise, either by systematically varying the metal film thickness (to vary the average grain size), or by using epitaxially-grown single crystal films. However, radiation-induced noise studies on epitaxial films will not be entirely straightforward. In our films, the noise measuring current creates a large amount of internal heat (typically 100 W/cm²) which must be dissipated by a sample substrate. One cannot therefore use freely-suspended films, as has been done in several previous radiation-damage studies.46,58,64 One also requires that at least part of the substrate located close to the small metal sample be transparent to electrons, in order to focus (to about 100 µm) and position the electron beam. This may prove difficult on substrates suitable for epitaxial growth, such as mica, sapphire, and rock-salt.

A promising alternative would be to create defects in epitaxial films using high energy proton irradiation. For example, the 1 MeV proton damage rate in Cu is sufficiently large31 that only a modestly focussed (- 1 mm) beam in a typical accelerator would be necessary to obtain a reasonable damage rate. Yet the average primary recoil energy from these protons is sufficiently small (- 1 keV) that most of the defects produced would be in the form of isolated Frenkel Pairs.55 The
sample-stage space requirements on most ion accelerators are much less severe than that imposed by the pole-pieces inside a TEM, so a larger, more thermally stable cold stage could be constructed.

One could also investigate the effect on the induced noise of internal stress in the metal films. This could be achieved either by using various substrate materials with different coefficients of thermal expansion, or by constructing a sample cold-stage which can apply external stress to a sample.

Finally, other metals besides Cu could be studied. Aluminum would be a particularly interesting case. Small interstitial clusters in Aluminum are known to reorient in the temperature range 20 K - 60 K, producing peaks in the low-frequency internal friction observable at several temperatures. Such interstitial reorientations could also show up as peaks in the low-frequency noise.
APPENDIX A.

NOISE AS A SUPERPOSITION OF LORENTZIANS

A random process with a characteristic time $\tau_1$ connected to a physical variable $x$ generally has a Lorentzian spectral density

$$S_x(\omega) = \frac{\tau_1}{1 + \omega^2 \tau_1^2} \quad (A1)$$

where $\omega = 2\pi f$. The spectral density from an ensemble of such processes with a range of characteristic times is then a superposition of Lorentzians

$$S_x(\omega) = \int d\tau \ D(\tau) \frac{\tau}{1 + \omega^2 \tau^2} \quad (A2)$$

where $D(\tau)$ described the distribution of characteristic times $\tau$. By a suitable choice of $D(\tau)$, one could construct any spectral density such that $0 \geq \Im S_x/\Im \omega \geq -2$ for all $\omega$. In particular, the choice $D(\tau) = \tau^{-1}$ for $\tau_1 < \tau < \tau_2$ (and zero elsewhere) yields

$$S_x(\omega) = \int_{\tau_2}^{\tau_1} \frac{d\tau}{\tau_2 (1 + \omega^2 \tau^2)} = 1/\omega \quad (A3)$$

for $\tau_2^{-1} < \omega < \tau_1^{-1}$. Some physical motivation for this form of $D(\tau)$ came from Du Pre$^{17}$ and Van der Ziel$^{18}$ who pointed out that if the processes are thermally activated,

$$\tau = \tau_0 \exp(E/k_BT), \quad (A4)$$

with a distribution $D(E)$ of activation energies $E$, then

$$D(\tau) = D(E) \cdot (d\tau/dE)^{-1} = D(E) \cdot (k_BT/\tau), \quad (A5)$$

and a constant $D(E)$ leads directly to the desired dependence $D(\tau) \sim \tau^{-1}$. Here $T$ is temperature, $k_B$ is Boltzmann's constant, and $\tau_0^{-1}$ is an attempt rate (e.g. a phonon frequency) assumed constant for all energies.
Du Pre,17 and subsequently Dutta et al.16 generalized these ideas, and showed that a non-constant distribution \( D(E) \) which is assumed only to be broad with respect to \( k_B T \) in fact results in a spectral density which is approximately of the \( 1/f \) form, i.e.

\[
S_x(\omega) = \omega^{-m}
\]

(A6)

where \( m \) is a number close to 1. These authors started by rewriting Eq. (A2) in terms of activation energies

\[
S_x(\omega) \sim \int dE \, D(E) \left[ \frac{\tau_0 \exp(E/k_B T)}{1 + \omega^2 \tau_0^2 \exp(2E/k_B T)} \right].
\]

(A7)

The quantity in the square brackets is a strongly peaked function of \( E \) with a width of the order \( k_B T \), centered at an energy

\[
\bar{E} \equiv -k_B T \ln(\omega \tau_0).
\]

(A8)

Thus, at a given temperature, the only processes which contribute significantly to the spectral density at a frequency \( \omega \) are those with activation energies within \( k_B T \) of \( \bar{E} \). Since \( D(E) \) is assumed broad with respect to \( k_B T \), one can, to first order, take the quantity \( D(\bar{E}) \) outside the integral in Eq. (A7)

\[
S_x(\omega) \sim D(\bar{E}) \int dE \left[ \frac{\tau_0 \exp(E/k_B T)}{1 + \omega^2 \tau_0^2 \exp(2E/k_B T)} \right].
\]

(A9)

With Eqs. (A2) and (A4) this reduces to

\[
S_x(\omega, T) \sim \frac{k_B T}{\omega} \ D(\bar{E}).
\]

(A10)

\( S_x(\omega, T) \) contains an explicit \( 1/\omega \) dependence, but also a weak implicit dependence through the term \( D(\bar{E}) \) and Eq. (A8), which causes \( S_x(\omega) \) to differ somewhat from an exact \( 1/\omega \) form.

Du Pre17 did not carry the analysis further. However Dutta et al.16 noted that equation (A10) implies a relation between the frequency and
temperature dependences of $S_x(\omega, T)$. For noise which scales roughly as $1/\omega$, one may express the frequency exponent $m$ as

$$m(\omega, T) = -\frac{\partial \ln S_x}{\partial \ln \omega}$$

$$= 1 + (k_B T)(\frac{\partial \ln D(E)}{\partial E}) \cdot (A11)$$

Physically, one has the following situation: In order to account for the measured temperature dependence of the noise magnitude (at a given frequency), one postulates a $D(E)$ to satisfy Eqs. (A8) and (A10). This same function $D(E)$ must also account for the temperature dependence of the parameter $m$ according to Eq. (A11). Noise measured in a given system is inconsistent with this model if one cannot find a distribution $D(E)$ which simultaneously satisfies Eqs. (A10) and (A11).

One can eliminate from Eq. (A11) any explicit reference to $D(E)$ by noting that Eq. (A10) implies

$$\frac{\partial \ln S_x}{\partial \ln T} = 1 - [k_B T \ln(\omega \tau_0)] [\frac{\partial \ln D(E)}{\partial E}] \cdot (A12)$$

One can thus rewrite Eq. (A11) as

$$m(\omega, T) = 1 - \frac{1}{\ln(\omega \tau_0)} \left[\frac{\partial \ln S_x}{\partial \ln T} - 1\right] \cdot (A13)$$

In a given physical system, the measured noise magnitude and frequency exponent $m$ must satisfy Eq. (A13) in order to be consistent with the model proposed by Dutta et al.\textsuperscript{16}

This model has proven quite successful when applied to the $1/f$ noise measured in metal films, as well as a number of other physical systems (see Weissman\textsuperscript{2} for a recent review). With a measurement frequency $\omega/2\pi - 1$ Hz and an assumed attempt rate $\tau_0^{-1} = 10^{14}$ s$^{-1}$, the consistency relation expressed by Eq. (A13) has been found to be in qualitative agreement with experimental results in a number of metal-film systems. We note here that as long as $\ln(\omega \tau_0) \ll -1$, one can vary the particular
value chosen for $\tau_0$, a free parameter in this model, by several orders of magnitude without significantly affecting the quality of agreement of the Eq. (A13), or the shape of the hypothetical distribution $D(E)$. The $1/f$ noise measured in these films over a temperature range $100 \, K < T < 500 \, K$ was used (with Eq. (A10)) to infer a finite value of $D(E)$ over an activation energy range $0.25 \, \text{ev} - 1.2 \, \text{ev}$. This range of energies is typical of processes involving atomic motion in metals.\textsuperscript{1,31}

Further support for such a model comes from selected systems such as Bi films,\textsuperscript{25} silicon-on-sapphire films,\textsuperscript{4} and Si grain boundaries\textsuperscript{87} in which the noise contains broad spectral features in addition to a strict power-law behavior. These spectral features change position in a manner consistent with a superposition of thermally activated Lorentzians, where one now assumes a relatively narrow distribution function $D(E)$.

In recent years, measurements on small area MOSFET's\textsuperscript{7-9} and tunnel junctions\textsuperscript{10-12} have shown conclusively that most of the $1/f$ noise observed in these systems indeed results from a superposition of Lorentzians. In extremely small devices at low temperatures, most of the excess noise can be fitted by a few Lorentzians, with thermally activated characteristic relaxation times. With increasing device size and/or temperature, the number of randomly-distributed Lorentzians increases in such a manner that the spectral density evolves continuously into a nearly featureless "$1/f$ - like" form.

Two features of the Dutta-Dimon-Horn model make it quite generally applicable to a number of systems. The first is simply that, as formulated, it makes no explicit reference to the identity of the microscopic random process responsible for the noise, nor to its
coupling to the measured fluctuating quantity (e.g. sample resistance). The second is that the model demonstrates in a simple way that a 1/f-like spectral density is an expected consequence of a process with a reasonably broad distribution of activation energies, independent of most details of the shape of the distribution, or its origin in a particular system. The extremely general nature of the model may well go a long way towards explaining why "1/f-like" noise is observed in such a variety of systems in which the origin and coupling of the noisy microscopic processes are quite diverse.

In recent years, there has been some discussion on various assumptions made by Dutta et al. in formulating their model. Weissman and Scofield et al. have noted that one could obtain noise of an approximate 1/f^m form by assuming a fixed activation energy E_0 and an appropriate distribution in the pre-factor τ_0. From such a model, Scofield et al. derived a relation analogous to Eq. (A13),

\[ m(\omega, T) = 1 + \left[ \frac{k_B T}{E_0} \right] \left[ \alpha \ln S_x \right] \left[ \alpha \ln T \right] \]  (A14)

With appropriate choice of the free parameter E_0, Eq. (A14) agrees as well with experimental data as Eq. (A13). The point of this should be well taken: qualitative experimental agreement with Eq. (A13) does not, by itself, assure that the 1/f noise seen in a given system results from a process with the distribution of activation energies expressed by Eq. (A10). One can be reasonably confident that the noise is thermally activated and results from a broad spread of relaxation times, but it remains ambiguous whether the spread is due to a distribution in E, in τ_0, or in both. In systems where the noise can be decomposed into discrete Lorentzians with E and τ_0 independently
determined,\textsuperscript{7-12} it is not surprising that a substantial distribution in both quantities is found.

As noted in the main text, a number of studies indicate a variety of point defect motions in metals, with typical activation energies in the range\textsuperscript{31} 0.1 eV - 3 eV, and with attempt times\textsuperscript{33,45,71} within a few orders of magnitude of $10^{-12}$ s. A noise model which assumes that the range in characteristic times is dominated by a range in activation energy is probably most appropriate to the results discussed in this thesis.

In their model, Dutta et al.\textsuperscript{16} assume that the contribution to the total variance in the noise,

$$\langle \delta x^2 \rangle = \int S_x(f) df,$$  \hspace{1cm} (A15)

by each microscopic process is temperature independent. A more general form of Eq. (A7) which includes an explicit temperature dependence should read\textsuperscript{2,70}

$$S_x(\omega) = \int dE \psi(E,T) \left[ \frac{\tau_0 \exp(E/k_B T)}{1 + \omega^2 \tau_0^2 \exp(2E/k_B T)} \right].$$  \hspace{1cm} (A16)

This introduces modifications to the consistency relations expressed by Eqs. (A13) and (A14). Scofield et al.\textsuperscript{70} and Weissman\textsuperscript{2} discuss in detail consequences of such an explicit temperature dependence. In the specific case where \(\psi(E,T)\) can be factored as

$$\psi(E,T) = D(E) \Theta(T),$$  \hspace{1cm} (A17)

the coupling function \(\Theta(T)\) can be taken out of the above integral. One should now replace Eq. (A13) with

$$m(\omega,T) = \frac{1}{\ln(\omega_0)} \left[ \frac{\Delta \ln S_x}{\Delta \ln T} - 1 - \frac{\Delta \ln \Theta}{\Delta \ln T} \right].$$  \hspace{1cm} (A18)

The value of \(m(\omega,T)\) expressed here differs from that in Eq. (A19) by an
amount

\[ \Delta m = [\partial \ln \Theta / \partial \ln T] [1/\ln(\omega T_0)]. \] \hspace{1cm} (A19)

Assuming typical values: \( \omega/2\pi = 10 \text{ Hz} \) and \( \tau_0 = 10^{-14} \text{ s} \), we find \( \ln(\omega T_0) = -30 \). Experimental uncertainties in \( m \) are typically \( \pm(0.05 - 0.10)\), so only the existence of a coupling coefficient \( \Theta(T) \) which is strongly temperature dependent will seriously affect the quality of agreement of current experimental results and the unmodified expression Eq. (A13). Fleetwood et al.\textsuperscript{88}, and Scofield et al.\textsuperscript{70} have considered the specific case

\[ \Theta(T) = T^\delta. \] \hspace{1cm} (A20)

In this case, \( \Delta m = \delta/\ln(\omega T_0) \), so that an assumed value of \( \delta \) anywhere in the range \(-3 < \delta < 3\) only changes the value of \( m \) by \( \pm10\% \) from that expressed in the unmodified expression Eq. (A13).
APPENDIX B.

FINITE-SIZE CORRECTIONS TO DAMAGE RATES

Fuchs76 and Sondheimer77 first developed the theory of the resistivity of conducting films and wires in which surface scattering plays an important role. This theory relates the measured resistivity \( \rho \) to the bulk or "intrinsic" resistivity \( \rho_i = \kappa / l_1 \), where \( l_1 \) is the electron mean free path of the material in the absence of surfaces and \( \kappa \) is a parameter of the material. The ratio \( \rho / \rho_i \) becomes large when \( l_1 > d \), where \( d \) is the film thickness. A parameter in the theory is the fraction \( P \) of electrons which reflect specularly from a surface (i.e. conserving in-plane momentum). The remainder of the electrons are assumed to scatter diffusely. As originally formulated, the theory predicts that

\[
\frac{\rho}{\rho_i} = \frac{\phi(d/l_1, P)}{l_1},
\]

where \( \phi(x, P) \) can be evaluated numerically. With the definition \( \gamma = d/k \), one can eliminate explicit reference to \( l_1 \), and write

\[
\frac{\rho}{\rho_i} = \phi(\gamma_{\rho_i}, P),
\]

where \( \phi(x, P) \) is the same function as above. Thus with tables of \( \phi(x, P) \), a suitable extrapolation formula, and a choice of \( \gamma \) and \( P \), one can determine \( \rho_i \) directly from \( \rho \).

This theory has been applied to radiation damage experiments in thin metal films, where one wants to determine the intrinsic damage function \( d\Delta \rho_i / d\phi \) vs. \( \Delta \rho_i \) from the measured function \( d\rho / d\phi \) vs. \( \rho \). However, the parameters \( \gamma \) and (especially) \( P \) are not accurately known, and are likely sample dependent. The damage function for bulk Cu is known to be a straight line (section IV.B.1), so the following
procedure has been adopted for correcting data taken on thin Cu films: Fix the parameter $P$ (usually at zero) and vary $Y$ until the corrected damage function approximates a straight line. The parameters $\kappa$ and $P$ affect $\phi(Y_0, P)$ in a very similar way, so it is generally only necessary to vary one or the other to obtain an optimal straight line. Figure B.1, taken from Ref. 46, shows an example of this procedure applied to data taken from a 400 nm $<100>$ single crystal Cu film. The corrections to the data are quite large at low $\Delta \rho$ (where $l_1 > d$), but become smaller as increasing defect concentration reduces $l_1$. Most (but not all) thin-film damage rate data on Cu can be transformed into reasonably straight lines with approximately the correct bulk behavior by assuming reasonable values of $Y$. However, certain damage function measurements, especially those done with ion irradiation, show enhanced damage at low $\Delta \rho$ which cannot be explained simply by finite-size effects.
Fig. B.1 Damage rate $d\rho/d\phi$ vs. $\Delta \rho$, showing the experimental and size-effect corrected data for a 400 nm <100> Cu film irradiated at 1.1 MeV. Figure from Ref. 46.
APPENDIX C.

FRENKEL PAIR RECOMBINATION NOISE

It is clear from the annealing behavior shown in Fig. 4.4 that both the added resistivity and the added noise measured at 90 K are metastable phenomena. Both quantities return nearly to their initial values when the Cu films are annealed at elevated temperatures. If the irradiated films are maintained at the irradiation temperature of 90 K, small but continuous reductions in both the added noise and resistivity must occur. One might therefore question whether the observed added 1/f noise might be a simple consequence of fluctuations in the sample resistance due to the small amount of recombination of Frenkel Pairs which occurs during the noise measurement.

In this section we show that neither the magnitude nor the 1/f shape of the added noise measured in these experiments can result as a simple consequence of the uncorrelated recombination of Frenkel Pairs. We consider a rectangular sample of total length 2L, width w, and thickness d, configured with five contact leads as in Fig. 3.1. We further assume a fractional concentration $C_F$ of Frenkel Pairs, which produce an added resistivity in the films

$$\Delta \rho = \rho_F C_F .$$

Here, $\rho_F = 2.75 \times 10^{-4} \ \Omega \text{cm}$ is the average Frenkel resistivity.\textsuperscript{46} In contrast to the discussion in chapter V, the present analysis does not consider details of the specific structure and orientation of defects within the film.

We now assume that the Frenkel Pairs are randomly recombining at an average rate
If \( \bar{v} \ll 1/T \), where \( T \) is the length of the time record analyzed by the FFT Spectrum Analyzer, then this recombination is quasi-stationary, i.e. it is roughly constant during the time \( T \).

We now consider the two arms of the film as separate volumes, and define \( \Delta \rho_L \) and \( \Delta \rho_R \) as the average added resistivity of the left and right arms of the sample, respectively. We define

\[
\rho_d \equiv (\Delta \rho_L - \Delta \rho_R) .
\]

It is fluctuations in the quantity \( \rho_d \) that are actually measured experimentally (see section III.E). If a single Frenkel Pair in the left arm of the sample recombines, for example, then \( \rho_d \) will change by an an amount

\[
\delta \rho_d = \delta (\Delta \rho_L) = -\rho_p/\eta Lw d = -2\rho_p/N \equiv -\delta \rho_S ,
\]

where \( n \) is the concentration of atoms in the metal, and \( N = 2nLwd \) is the total number of atoms in the sample. Thus, as Frenkel Pairs recombine randomly in the two arms of the sample, the quantity \( \rho_d \) will follow a random walk with step size \( \delta \rho_S \), and a mean time \( \tau \) between steps of

\[
\tau = \frac{1}{2\bar{v} C_p N} .
\]

The spectral density \( S_\rho(f) \) of the fluctuations in \( \rho_S \) may be calculated by making use of the mathematical relation\(^{90}\) between the spectral densities of a quantity \( x \) and of its time-derivative \( \dot{x} \);

\[
S_x(f) = \frac{1}{(2\pi)^2} S_{\dot{x}}(f) .
\]
occurring with average frequency $t^{-1}$. The spectral density of $\dot{\rho}_d$ is of the well known shot-noise form $^{91}$,

$$S_\ld(f) = \frac{2f_2}{\tau} = \frac{8\rho_F^2}{N} \nu C_F .$$

(C7)

With reference to Eq. (C6), this implies that

$$S_\ld(f) = \left(\frac{2\rho_F^2}{\pi^2 N}\right)^{1/2} = \frac{A}{f^2} .$$

(C8)

We see immediately that this "recombination noise" does not have the correct spectral shape to account for the radiation-induced $1/f$ noise. We next show that this recombination-noise is also not of sufficient magnitude to account for the induced noise. The experimental bandwidth of the noise measurements was roughly $f_1 < f < f_2$, where $f_1 = 0.1$ Hz, and $f_2 = 25$ Hz. The low-frequency limit is simply $f_1 = T^{-1}$, where $T = 10$ s is the time-record length of the digital spectrum analyzer. By integrating Eq. (C8) over the experimental bandwidth, one obtains the recombination-noise variance $<\langle(\delta \rho)^2\rangle_{RN}$:

$$<\langle(\delta \rho)^2\rangle_{RN} = \frac{A}{f^2} \int_{f_1}^{f_2} f_2 = A T .$$

(C9)

We wish now to compare this with the radiation-induced noise measured over the same bandwidth. As an example, we refer to Fig. 4.3 and note that $\Delta \alpha = 1.6 \times 10^{-3}$ when $\Delta \rho = 10$ n$\Omega$cm. The induced-noise variance $<\langle(\delta \rho)^2\rangle_{IN}$ measured over the experimental bandwidth is then roughly

$$<\langle(\delta \rho)^2\rangle_{IN} = \frac{\Delta \alpha \rho_0^2}{N} \int_{f_1}^{f_2} \frac{df}{f} = \frac{\Delta \alpha \rho_0^2}{N} \ln(f_2/f_1) ,$$

(C10)

where $\rho_0 = 10^{-6}$ $\Omega$cm is the resistivity of the Cu film. In order that $<\langle(\delta \rho)^2\rangle_{RN} = <\langle(\delta \rho)^2\rangle_{IN}$, Eqs. (C8) - (C10) require that
With reference to Eqs. (C1) and (C2), this implies that during a single time record of length \( T = 10 \) s, the average sample resistivity must decrease by an amount

\[
\delta(\Delta \rho) = \sqrt{C_F T \rho_F} = 0.2 \text{ nÅcm}.
\]  

This is, however, some 2 to 3 orders of magnitude larger than the upper limit of the measured decrease in sample resistivity during a 10 s interval. We therefore conclude that simple "recombination-noise" magnitude actually present in the sample is far too small to account for the measured level of radiation-induced 1/f noise.
REFERENCES

3. It is essentially a matter of taste how close m must be to one for the noise to be considered "1/f". For example Ref. 1 defines "generic" 1/f noise by the requirement that 0.8 < m < 1.4.


56. We greatly acknowledge R. Hammond and P. Rosenthal for assistance with the electron-beam evaporations.


59. Wayne E. King, private communication.
65. In comparing relative noise levels measured at 90 K, it makes little difference here whether one normalizes the noise using Eq. (2.4) or Eq. (2.5) since the sample resistivity changes by less than 10% as defects are added.
68. Appendix A discusses implicit assumptions made by Dutta et al. in deriving Eq. (4.3). Relaxing these assumptions can cause the two sides of Eq. (4.3) to be offset by a quantity which varies with temperature. Under most conditions, one expects this offset to be fairly small (-1/ln(2πτ_0) - .03) and to vary slowly with temperature, allowing one to detect common structure in both sides of Eq. (4.3).


78. V. Groger, Cryogenics 19, 56 (1979).


84. S. Hershfield (Ref. 30) has independently done similar calculations.

85. As an example we consider a defect with axial symmetry ($\rho_1 = \rho_2 = \rho_3$). For a $<100>$ defect orientation one has $\rho_1 = \rho_{xx}$, and $\rho_3 = \rho_{zz}$. For a $<110>$ orientation one has $\rho_1 = \rho_{zz}$, and $\rho_3 = (2\rho_{xx} - \rho_{zz})$. One cannot uniquely determine the principal moments for a $<111>$ orientation.

86. N. Zimmerman, private communication.


89. Karl Merkle, private communication.

