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MAGNETIC RESONANCE OF THE IONIC GROUND STATE OF ATOMS THROUGH EXCHANGE COLLISIONS WITH THE METASTABLE STATE; AND INTENSITY BEATS DUE TO COHERENT EXCITATION

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Chung-Heng Liu
July 27, 1967
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AND INTENSITY BEATS DUE TO COHERENT EXCITATION

Chung-Heng Liu
(Ph.D. Thesis)
July 27, 1967
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MAGNETIC RESONANCE OF THE IONIC GROUND STATE OF ATOMS
THROUGH EXCHANGE COLLISIONS WITH THE METASTABLE STATE;
AND INTENSITY BEATS DUE TO COHERENT EXCITATION

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ABSTRACT

Radio-frequency resonances of the metastable state $^{3}P_{2}$ of both even and odd isotopes of XeI were observed by monitoring the change of absorption of the $\lambda = 8409 \ \AA$ resonance radiation. The metastable state $^{3}P_{2}$ of Xe was produced and aligned by the impact of a unidirectional electron beam having high electron-current density and low energy. The high electron-current density beam has been achieved by using a diode-structure electron gun under the space-charge-neutralization condition. Several values of the $g$ factor of odd isotopes of Xe were measured as

\begin{align*}
g(J = 2, I = 1/2, F = 5/2) &= -1.2007 \pm 0.0040, \\
g(J = 2, I = 3/2, F = 3/2) &= -1.2007 \pm 0.0040, \\
g(J = 2, I = 1/2, F = 3/2) &= -1.7986 \pm 0.0023.
\end{align*}

By using the same experimental arrangement and technique, we have observed spin-exchange-type collisions between the singly ionized ground state $^{2}P_{3/2}$ of Xe and the metastable state $^{3}P_{2}$ of neutral Xe, both being aligned, not oriented by electron impact. So far as we know, this is the first observation of spin-exchange collisions between the aligned
ground state of ionic atoms and the metastable state of neutral atoms both having $P$ configuration. Up to the present stage of the experiment, the mechanism of this kind of collisions is still not clear; only a phenomenological explanation has been given. Calculation of the signal with the help of the solution of rate equations (in which it has been assumed that the angular momentum changes by only one unit at each collision) showed fair agreement with the experimental results. The value of the $g_J$ factor of the ionic ground state $^2P_{3/2}$ of Xe was measured as $g_J(J = 3/2, I = 0) = -1.3344\pm0.0003$.

We observed intensity beats in the resonance absorption by the coherently excited nondegenerate magnetic sublevels of the metastable state $^3P_2$ of Hg that was formed by impact excitation by a high-frequency modulated low-energy, unidirectional electron beam. Since no radio-frequency oscillating magnetic field is needed, this experiment can offer a new method for investigating the excited state of atoms having total angular momentum $J > 1/2$. 
I. INTRODUCTION

A. Radio-Frequency Resonance of the Metastable State $^{3}P_{2}$ of the Neutral Xenon Atom

To observe the radio-frequency resonance one must obtain a reasonable number of the metastable atoms that are either aligned or oriented. Though the lifetime of the metastable states of some noble gases such as helium, neon, and argon is known to be a few milliseconds at a pressure of about 1 mm of Hg,\textsuperscript{1,2} all the metastable states of the noble gases except helium have very fast relaxation times of alignment even with a buffer gas of $^{1}S_{0}$ ground state, this fast relaxation is due to the P configuration of the parent state of the metastable states of all the noble gases except helium.\textsuperscript{3} This implies that success in observing the rf resonance of the metastable states of noble gases with a P configuration of the parent state depends on a means of attaining very fast pumping speed. The difficulty in this has been overcome by Hadeishi et al. in their experiment on rf resonance of the metastable state of neon\textsuperscript{3} by using a diode-structure electron gun which can emit a high-current-density, low-energy, unidirectional electron beam under the space-charge-neutralization condition to excite a sufficient amount of the metastable states of neon; the neon metastable state becomes aligned during the electron transit of a distance corresponding approximately to the atomic diameter.

In addition to the fast pumping speed, the electron-impact excitation can put the atoms in any excited state and any ionic state with fewer selection-rule restrictions than optical excitation. But these excited atoms can be only aligned and not oriented (this is shown in
Sec. II-A). This is apparently a disadvantage of electron-impact excitation. However, we converted this disadvantage into a useful condition for the "spin-exchange" type of collisions between the metastable state of neutral xenon atoms and the ground state of singly ionized xenon.

For electron energy near the threshold of the metastable state $^3P_2$, the magnetic sublevel with $m_J = 0$ (which is excited from the ground state $^1S_0$) is preferentially populated. If the electron-exchange process is taken into account, $m_J = \pm 1$ of the $^3P_2$ state are populated, too. Now if we apply a radio-frequency oscillating magnetic field perpendicular to the external static magnetic field and at the Larmor frequency of the $^3P_2$ state, and if the intensity of the rf magnetic field is high enough, a population equalization will occur among all the magnetic sublevels of the $^3P_2$ state—or we can say that the relaxation time among the magnetic sublevels is caused to approach zero. Therefore as the absorption of a selected spectral line by the $^3P_2$ state changes, a resonance curve will be obtained, if we steadily change the frequency of the rf magnetic field or the magnitude of the external-static magnetic field near the neighborhood of the resonance frequency or field. This is the basic idea of rf resonance spectroscopy.

For the rf resonance of the metastable state $^3P_2$ of xenon, the experimental arrangement was almost identical to that used by Hadeishi in his neon experiment. But some improvements have been made, namely the optical system arrangement uses two photomultipliers instead of one to cancel out the 120-Hz fluctuation from the light source, which
was being discharged by a microwave cavity, and rf amplitude modulation is employed instead of external static magnetic-field modulation.

The reasons we chose xenon are: (a) xenon has a relatively large atomic number, so that one could achieve space-charge neutralized electron flow quite effectively; (b) among all the noble gases, only xenon has even and odd natural isotopes evenly distributed: \( ^{54}_{129}\text{Xe} \): 52.39%; \( ^{129}_{54}\text{Xe} \): 47.62% (\(^{129}\text{Xe} \) with nuclear spin \( I = 1/2 \), 26.44% (\(^{131}\text{Xe} \) with \( I = 3/2 \), 21.18%); (c) the odd isotopes of xenon have different nuclear spin states.

The relevant energy levels of a xenon atom used in our experiment are shown in Fig. 1.

B. Radio-Frequency Resonance of the Ground State of Singly Ionized Xenon Observed Through the Collision with the Metastable State of the Neutral Xenon Atom

One of the newer forms of high-precision radio-frequency spectroscopy is spin-exchange optical pumping, in which an electron with specified spin orientation is interchanged with a second electron with a different spin orientation. In this type of experiment two species, A and B (e.g., sodium atoms and quasi-free electrons) are contained in a vapor cell; A is oriented by the absorption of circularly polarized resonance radiation. The degree of orientation of A is observed by monitoring the change of absorption of the incident circularly polarized resonance radiation by the vapor cell. Through spin-exchange collisions, the orientation is transferred to the other species, B. If a radio-frequency oscillating magnetic field with suitable frequency is now applied to B to disorient it, this disorientation transfers back to
Fig. 1. Relevant energy levels of a Xe atom used in the experiment.
A via spin-exchange collisions; this results in an increase in absorption of the incident circularly polarized resonance radiation by species A in the vapor cell. This method of studying one species in terms of a second species is a very powerful experimental technique. One can thus measure the properties of atoms that cannot be observed directly.

Dehmelt, the first to perform this kind of technique, determined a very precise value of the Lande g factor for the electron by utilizing the spin exchange between optically oriented sodium atoms and quasi-free electrons. This is of considerable interest in testing for the theoretical values

$$\frac{\mu_S}{\mu_0} = 1 + \frac{a}{2\pi} + \text{higher-order terms},$$

obtained from quantum electrodynamics.

Spin exchange has been used to orient the K, Rb, and Cs atoms through collisions with other optically oriented alkali atoms, and to study the ground-state hyperfine structures of H, D, T, $^1\text{H}$, $^1\text{H}$, and $^1\text{H}$. In addition, nuclear orientation of $^3\text{He}$ has been achieved by this kind of technique.

The technique of transferring orientation from optically pumped oriented atoms to the ground state of ionized atoms was also first introduced by Dehmelt. By using a sophisticated rf quadrupole ion trap he succeeded in observing spin-exchange collisions between oriented Cs atoms and ionized $^4\text{He}^+$. The same kind of ion trap was used and a very high resolution of $^3\text{He}^+$ ion hyperfine-structure spectra was studied recently.

So far as we know, all the observations and measurements through the spin-exchange mechanism dealt only with the orbital angular momentum
L = 0 states of atoms or ions. If only those states with L = 0 are considered, then the transformation of orientation of atoms or ions through the collisions between two different species of atoms or ions can obviously be attributed to "spin exchange"; of course, the electron exchange is also to be counted in certain cases.

But in our "spin-exchange" experiments on the metastable state of neutral xenon atoms and the ground state of singly ionized xenon, instead of being oriented, both states were aligned as a result of the high-current-density electron-beam impact excitation. Furthermore, both states have an orbital angular momentum L = 1 unit. Though we names "spin exchange" the mechanism for the transformation of angular momenta, the mechanism is not so obvious as in collision between an oriented atom and an unoriented atom, both with zero orbital angular momentum. Before making a more thorough study theoretically and experimentally, to understand the mechanism involved, we tried to avoid this kind of complicated situation by assuming, in addition to the conservation of the total angular momentum of the two species, that the change of the total angular momentum component J_z of each atom can be only one unit during the collision between these two atoms Xe and Xe^+. Under these assumptions, the rate equations have been written, the relative signals then can be calculated from their solution for both rf resonance of the metastable state of neutral xenon atoms, and rf resonance of the ground state of the singly ionized xenon atoms. The calculation shows fairly good agreement with the experimental results. It also shows that, since both atoms are aligned, no rf resonance effect of the ionic ground state will be observed by monitoring the
absorption change of the metastable state of the neutral atoms caused through the "spin-exchange" collisions, unless the total angular momentum $J$ of the ionic ground state is larger than $1/2$. This can be readily seen from the aligned production of the ionic ground state and the equally probable collisions with the metastable state of neutral atoms, for which both magnetic sublevels $m_J = +1/2$ and $-1/2$ are equally populated no matter whether the ionic ground state is at rf resonance or not.

The relevant energy levels of neutral xenon and of singly ionized xenon used in this experiment are shown in Fig. 2.

Since the spectral lines terminating on the ionic ground state of $Xe^+$ lie in the vacuum ultraviolet region, the values of $g$ factors and the hyperfine-structure constant of this state have not yet been determined. The "spin-exchange" technique used here probably offers a comparatively easy way to measure these values not only for $Xe^+$ but also in other noble gases having total angular momentum $J > 1/2$.

C. Intensity Beats in Resonance Absorption Due to Coherent Excitation of the Metastable State of the Mercury Atom

Breit has shown a possibility of coherent excitation of nondegenerate levels if the exciting resonance radiation is pulsed in a time much shorter than the relaxation time of the excited states. A reason for this is that the shorter the light pulse, the broader the range of the spectral frequency. Therefore, if the width of the line $\Delta \nu$ is larger than the neutral width and the separation of the nondegenerate levels, these levels are excited in a time of order $\Delta t \approx 1/\Delta \nu < \tau$. 
Fig. 2. Relevant energy levels of a Xe atom and of a singly ionized Xe used in the experiment.
where $\tau$ is the natural lifetime of the excited states; this means that all the nondegenerate states are excited by one wave packet, provided the excitations are allowed by the optical selection rule. Under such excitation conditions the phase differences among the nondegenerate levels are perfectly definite. Such excited states are called coherently excited states. The interference of resonance radiations emitted or absorbed by coherently produced states results in beat phenomena.

In recent years, several experiments have demonstrated the existence of intensity beats in the spontaneous radiation of atoms with a nondegenerate excited state. Thus far three methods have been used to produce uniformity of phase in the beats: (1) pulse excitation, (2) harmonic modulation of the intensity of excitation, (3) modulation of frequency of the transition between the nondegenerate levels. Excited states have been produced coherently not only by modulated or pulsed light, but in some experiments also have been obtained by modulated electron-beam impact.

That coherent excitation can be achieved by electron impact is obvious. Since, in general, the electron-beam energy is not monoenergetic, for nondegenerate levels (Zeeman magnetic sublevels) the necessary condition that the spread of the electron energy $\Delta E$ be larger than the Zeeman-level separation under the weak externally applied magnetic field is well satisfied. In addition to this requirement, there must be preferential or selective excitation of certain Zeeman levels in order to have coherence among the Zeeman levels.

In our experiment, the metastable state $^3P_2$ of Hg atoms was coherently excited by modulated, unidirectional electron-beam impact; we used optical absorption to detect coherent excitation (see Fig. 3).
Fig. 3. Relevant energy levels of a Hg atom used in the experiment.
II. THEORY

A. Alignment of the Excited State of an Atom by Electron Impact

By electron-impact excitation, the excited atoms are aligned. This means that the sublevels with $M$ and $-M$ are produced with equal probability. Suppose that the energy of the incident electron is only slightly higher than the threshold of the excited state, and the angular momentum of the incident electron is zero before the collision, then the angular momentum of the scattered electron is zero, since the electron's linear momentum is near zero after the collision. From consideration of the conservation of angular momentum and neglect of the effect of electron exchange, we expect the sublevel of the excited state having the same $M$ value to be selectively excited; i.e., in this case excitation of $\Delta M = 0$ results. Electron exchange results in $\Delta M = \pm 1$.

For excitation energies higher than the threshold of the excited state, Percival and Seaton\textsuperscript{23} used the Born approximation to calculate the excitation cross section for H and He. Though Born approximation is good only for the high-energy case, the results were in fairly good agreement with the experimental results except in the region very close to the threshold, as shown in Figs. 4 and 5, where the percentage polarization is defined by $P = 100(I\parallel - I\perp)/I\parallel + I\perp$, $I\parallel$ and $I\perp$ are the radiation intensities in a direction perpendicular to the electron beam with the electric vector parallel and perpendicular respectively to the electron beam, and $V_0$ is the threshold energy of the upper excited state divided by electronic charge $e$.

Here we do a brief calculation for the transition of hydrogen from (100) state to (21 m) state by using the Golden rule instead of the Born
Fig. 1. Percentage polarization for $^1D \rightarrow ^1P$ transitions as functions of $\sqrt{V/V_0}$. Dashed curve gives calculated result for He $^3^1D \rightarrow ^1P$ (Born approximation). Full-line curve gives experimental result for Hg $^1D \rightarrow ^6^1P$ (Skinner and Appleyard 1927). Reproduced from Percival and Seaton, Ref. 23.
Fig. 5. Polarization of Hg lines as functions of $\sqrt{V/V_0}$. Full-line curves give experimental results of Skinner and Appleyard (1927). Crosses give calculated threshold polarizations (for isotopes with $I = 0$ and assuming LS coupling for the collision process). Dashed curves are extrapolations of full-line curves neglecting experimental results for energies less than those at which maximum polarization was measured. Reproduced from Percival and Seaton, Ref. 23.
approximation and neglecting the spin-orbit coupling, electron exchange, etc. This computation shows the alignment of the \((2l_m)\) states, as the incident electron energy is higher than that of the threshold of the \((2l_m)\) states.

Consider the inelastic collision of a fast electron with a hydrogen in its ground state (Fig. 6); the differential cross section \(d\sigma/d\Omega\) for the scattering of the electron accompanied by the excitation of the hydrogen atom to an excited state is calculated as follows. The unperturbed Hamiltonian is

\[
H_0 = -\frac{\hbar^2}{2m} \nabla_1^2 - \frac{\hbar^2}{2m} \nabla_2^2 - \frac{e^2}{r_2}.
\]

(1)

The perturbation is the electrostatic energy of interaction between the incident electron and the valence electron, the nucleus of the atom,

\[
H' = \frac{e^2}{r_{12}} - \frac{e^2}{r_1}.
\]

(2)

The unperturbed wavefunctions are eigenfunctions of \(H_0\), which we choose to be

\[
V^{-1/2} \exp(ik_0 \cdot r_1) u_{100}(r_2), \quad \text{initial state},
\]

\[
V^{-1/2} \exp(ik \cdot r_1) u_{2l_m 1}(r_2), \quad \text{final state},
\]

where \(V\) is the volume of the box normalization, \(k^2 = k_0^2 - (2\pi/m\hbar^2)(3e^2/\varepsilon a_o)\) by considering the energy conservation

\[
\frac{\hbar^2 k_0^2}{2m} + E_{2l_m 1} = \frac{\hbar^2 k^2}{2m} + E_{100};
\]

(3)

\(a_o\) is the radius of the first Bohr orbit.

The matrix element of \(H'\) between \((100)\) and \((2l_m 1)\) states is
Fig. 6. Collision of a fast electron and a H atom.
where \( K = k_0 - k \).

By using the Golden rule, the transition rate \( w = (2\pi/\hbar)|H_{f_1}^i|^2\rho(f) \), and knowing that

\[
\rho(f) = \frac{mv}{8\pi^3h^2}k_d\Omega ,
\]

we get

\[
d\sigma(\theta,\phi) = \frac{w}{\sqrt{v}} = \frac{w}{\hbar k_0/mv} = \frac{k}{k_0} \left( \frac{m}{2\pi\hbar^2} \right)^2 v^2 |H_{f_1}^i|^2 d\Omega ;
\]

therefore

\[
\frac{d\sigma}{d\Omega} = \frac{k}{k_0} \left( \frac{m}{2\pi\hbar^2} \right)^2 v^2 |H_{f_1}^i|^2 .
\]

Now let us calculate the matrix element \( H_{f_1}^i \). Because of the orthogonality of \( u_{2lm_1} \) and \( u_{100} \), the term \( e^2/r_1 \) in the integrand contributes nothing; this is to be expected physically, since interaction between the incident electron and nucleus cannot produce excitation of the atomic electron. Thus

\[
H_{f_1}^i = \frac{1}{v} \int \int \exp(ik \cdot r_1) u_{2lm_1}^*(r_2) \frac{e^2}{r_{12}} u_{100}(r_2) d\tau_1 d\tau_2 ,
\]

where \( R = \sqrt{R_1^2 + R_2^2 + R_3^2} \), and \( J(R_{11}^2 R_{22} R_{33}) = 1 \).

Now

\[
\int \frac{\exp(ik \cdot r_1)}{r_{12}} d\tau_R = \exp(ik \cdot r_2) \int \frac{\exp(ik \cdot R)}{R} d\tau_R
\]

\[
= \exp(ik \cdot r_2) \int \int \exp(iKR \cos \theta) R \sin \theta dR d\theta d\phi \,
\]
where we have taken the polar axis of $R$ along the direction of momentum transfer vector $K$. Therefore

\[
\int \frac{\exp(ik \cdot r_1)}{r_{12}^2} = \frac{4\pi}{K} \exp(ik \cdot r_2) \int_0^\infty \sin KRdR
\]

\[
= \frac{4\pi}{K} \exp(ik \cdot r_2) \lim_{\alpha \to 0} \int_0^\infty \sin R e^{-\alpha R}dR
\]

\[
= \frac{4\pi}{K^2} \exp(ik \cdot r_2) .
\]

Thus

\[
H'_{f1} = v^{-1} \int \frac{4\pi e^2}{K^2} u^*_{2lm_1}(r_2) \exp(ik \cdot r_2) u_{100}(r_2) d\tau_2
\]

\[
= v^{-1} \frac{4\pi e^2}{K^2} \int \int \int \int u^*_{2lm_1}(r_2) \exp(iKr_2 \cos \theta_2) u_{100}(r_2) r_2^2 \sin \theta_2 \, dr_2 \, d\theta_2 \, d\phi_2 .
\]

If we choose the three final states $m_1 = \pm 1, 0$ with their polar axis along the momentum transfer vector $K$, it is readily seen that

\[
H'_{f1}(m_1 = \pm 1) = 0 ,
\]

\[
H'_{f1}(m_1 = 0) \neq 0 .
\]

Therefore

\[
\frac{d\sigma}{d\Omega} \text{ (final state is } 2l \pm 1) = 0 ,
\]

\[
\frac{d\sigma}{d\Omega} \text{ (final state is } 2l 0) \neq 0
\]

show only the $(2l 0)$ state being excited, as shown in Fig. 7.

Since in our experiment the direction of the unidirectional electron beam is regarded as the quantization axis whenever there is no externally applied magnetic field present. Those states of the hydrogen atom which are excited can be obtained by operating on the above-defined $(2l 0)$ state with a rotation operator $J^{(1)}$, which in matrix form is
Fig. 7. If the momentum transfer vector $\mathbf{K}$ is taken as the quantization axis, only the state $(2 \ 1 \ 0)$ of H atom can be excited by a fast electron.
\[ D^{(1)}(\alpha, \beta, \gamma) = \begin{pmatrix} e^{-i\alpha \frac{1 + \cos \beta}{2} e^{-i\gamma}} & e^{-i\alpha \sin \beta} & e^{-i\alpha \frac{1 - \cos \beta}{2} e^{i\gamma}} \\ \frac{\sin \beta}{\sqrt{2}} e^{-i\gamma} & \cos \beta & -\frac{\sin \beta}{\sqrt{2}} e^{i\gamma} \\ e^{i\alpha \frac{1 - \cos \beta}{2} e^{-i\gamma}} & e^{i\alpha \sin \beta} & e^{i\alpha \frac{1 + \cos \beta}{2} e^{i\gamma}} \end{pmatrix} \] (11)

The resulting state function is now a linear combination of (211)(210) and (2 1 -1) with coefficient \( c_1 = -e^{-i\alpha}(\sin \beta/\sqrt{2}) \), \( c_0 = \cos \beta \), and \( c_{-1} = e^{i\alpha}(\sin \beta/\sqrt{2}) \). We can notice that

\[ c_1^* c_1 = \frac{\sin^2 \beta}{2} = c_{-1}^* c_{-1} \] (12)

(2 1 1) and (2 1 -1) states are equally populated. This indicates that an alignment of the excited state can be achieved by a unidirectional electron-beam impact.

B. Rate Equations of the Metastable State of the Neutral Xe Atom Colliding

With the Ionic Ground State of the Xe\(^+\) Atom

By electron-impact excitation the magnetic sublevels of Xe\(^+\)(2P\(_{3/2}\)) and Xe(3P\(_2\)) are selectively excited. The magnetic sublevels \( M_J = 0 \) and \( \pm 1 \) of Xe(3P\(_2\)) should be selectively excited at the energy of the corresponding threshold energy of excitation (an experimental proof is given at the end of this section showing that even though the energy of the incident electron is a few eV higher than the threshold energy of the excitation, this statement still holds true). Because no theoretical calculation or prediction has been given for the population distribution of the ionic ground state of atoms under electron impact, it is not reasonable to assume that the magnetic sublevels \( M_J' = \pm 1/2 \).
of Xe\(^{+}(2P_{3/2})\) would be selectively excited at the energy of the corresponding threshold energy of excitation; furthermore, since we expect no threshold-energy condition to be rigorously satisfied, we assign the production rates \(\alpha'\) and \(\beta'\) for the magnetic sublevels \(M'_{J} = \pm 3/2\) and \(\pm 1/2\) of Xe\(^{+}(2P_{3/2})\). Similarly, we assign the production rates \(\alpha, \beta,\) and \(\gamma\) for the magnetic sublevels \(M_{J} = \pm 2, \pm 1, \) and 0 of Xe\((3P_{2})\). Since the electron-impact excitation aligns the excited atoms only, the production rate for \(M_{J}\) of \(M'_{J}\) is the same as for \(-M_{J}\) or \(-M'_{J}\).

The metastable state and ionic ground-state spin-relaxation rates \(W_{ij}\) and \(W'_{ij}\) represent the processes that tend to cause the magnetic sublevel population to reach its thermal equilibrium distribution by collisions with the container wall, electrodes, and impurities, etc., excluding the process involving collision interaction with metastable ionic ground states.

Coupling between the metastable-state atoms and the ionic ground-state atoms are induced through collisions. Equilibrium between the two systems is established in the characteristic time \(t \approx \frac{1}{\nu \sigma N}\) for the ionic ground state, and \(T \approx \frac{1}{\nu \sigma n}\) for the metastable, where \(\sigma, \nu, N,\) and \(n\) are respectively the total exchange cross section ("spin exchange" associated with electron exchange), relative velocity, metastable-state atom density, and ionic ground-state atom density.

In addition to ionic ground-state — metastable-state collisions, we should consider collisions of the metastable state with \(^{1}S_{0}\) isotropic atoms whose nuclear spins are not zero, as well as collision of atoms having the same configuration. (Nuclear alignment of the \(^{131}\)Xe ground state through exchange collision with the aligned metastable state was
observed recently by us. A brief description is given in this thesis.) For simplicity, we include such processes in \( W_{ij} \) and \( w_{ij} \). We further assume that due to collisions between \( \text{Xe}^+(2\,^3P_{3/2}) \) and \( \text{Xe}(3\,^3P_2) \) the angular momentum component \( J_z \) changes at most one unit per collision. Table I shows all the possibilities of \( J_z \) change under the condition of conservation of total angular momentum of the two colliding systems.

Let \( N_2, N_1, N_0, N_{-1}, \) and \( N_{-2} \) be the population densities of the magnetic sublevels of \( \text{Xe}(3\,^3P_2) \) corresponding \( M_J = 2, 1, 0, -1, \) and \( -2 \). Similarly let \( n_{3/2}, n_{1/2}, n_{-1/2}, \) and \( n_{-3/2} \) be the population densities of the magnetic sublevels of \( \text{Xe}^+(2\,^3P_{3/2}) \) corresponding to \( M_J' = 3/2, 1/2, -1/2, \) and \( -3/2 \) (though we already know that \( N_2 = N_{-2}, N_1 = N_{-1}, \) etc., the population density notations used in the above way will simplify the calculation of the rate equations). From those assumptions, we construct the following rate equations:

\[
\frac{dN_2}{dt} = -\sum_{j \neq 2} N_2 W_{2j} + \sum_{j \neq 2} N_2 W_{j2} - \frac{1}{\tau} N_2 + \alpha + v\sigma[-N_2(n - n_{3/2}) + N_1 n_{3/2}], \tag{13}
\]

\[
\frac{dN_1}{dt} = -\sum_{j \neq 1} N_1 W_{1j} + \sum_{j \neq 1} N_1 W_{j1} - \frac{1}{\tau} N_1 + \beta + v\sigma[N_2(n - n_{3/2}) - N_1 n + N_0 n_{3/2}], \tag{14}
\]

\[
\frac{dN_0}{dt} = -\sum_{j \neq 0} N_0 W_{0j} + \sum_{j \neq 0} N_0 W_{j0} - \frac{1}{\tau} N_0 + \gamma + v\sigma[N_1 n_{3/2} - N_0 n + N_{-1} n_{3/2}], \tag{15}
\]
Table I. $M_j + M_j' = M_j + M_j'$.

<table>
<thead>
<tr>
<th></th>
<th>$2 + \frac{1}{2} = 1 + \frac{3}{2}$</th>
<th>$2 + (-\frac{1}{2}) = 1 + \frac{1}{2}$</th>
<th>$2 + (-\frac{3}{2}) = 1 + (-\frac{1}{2})$</th>
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<tbody>
<tr>
<td>$1 + \frac{3}{2} = 2 + \frac{1}{2}$</td>
<td>$1 + \frac{1}{2} = 2 + (-\frac{1}{2})$</td>
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<td>$1 + (-\frac{3}{2})$</td>
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<td>$0 + \frac{1}{2} = 1 + (-\frac{1}{2})$</td>
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<tr>
<td></td>
<td>$= (-1) + \frac{3}{2}$</td>
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<td>$= (-1) + (-\frac{1}{2})$</td>
</tr>
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<td>$-1 + \frac{1}{2} = 0 + (-\frac{1}{2})$</td>
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<td>$-2 + (-\frac{1}{2}) = -1 + (-\frac{3}{2})$</td>
<td></td>
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</tbody>
</table>
\[
\frac{dN}{dt} = - \sum_{j=-1}^{j=1} N_{j} W_{j-1} - \sum_{j=-2}^{j=2} N_{j} W_{j-2} - \frac{1}{\tau} N_{-1} + \beta
\]
\[+ \nu \sigma[N_{0} \frac{n}{2} N_{-1} + N_{-2}(n - n_{3/2})], \quad (16)\]

\[
\frac{dN}{dt} = - \sum_{j=-1}^{j=1} N_{j} W_{j-1} + \sum_{j=-2}^{j=2} N_{j} W_{j-2} - \frac{1}{\tau} N_{-2} + \alpha
\]
\[+ \nu \sigma[N_{0} \frac{n}{2} N_{-1} - N_{-2}(n - n_{3/2})], \quad (17)\]

and

\[
\frac{d\rho_{3/2}}{dt} = - \sum_{j=-3/2}^{j=3/2} \rho_{j} W_{j-3/2} - \sum_{j=-1/2}^{j=1/2} \rho_{j} W_{j-1/2} - \frac{1}{\tau'} \rho_{-1/2} + \alpha'
\]
\[+ \nu \sigma[n_{3/2}(N - N_{2}) + n_{1/2} N_{2}], \quad (18)\]

\[
\frac{d\rho_{1/2}}{dt} = - \sum_{j=-1/2}^{j=1/2} \rho_{j} W_{j-1/2} - \sum_{j=-3/2}^{j=3/2} \rho_{j} W_{j-3/2} - \frac{1}{\tau} \rho_{-1/2} + \beta'
\]
\[+ \nu \sigma[n_{1/2} \frac{N}{2} - n_{-1/2} N + n_{-3/2}(N - N_{2})], \quad (19)\]

\[
\frac{d\rho_{-1/2}}{dt} = - \sum_{j=-3/2}^{j=3/2} \rho_{j} W_{j-3/2} - \sum_{j=-1/2}^{j=1/2} \rho_{j} W_{j-1/2} - \frac{1}{\tau} \rho_{-1/2} + \beta'
\]
\[+ \nu \sigma[n_{-1/2} \frac{N}{2} - n_{-3/2} N + n_{-3/2}(N - N_{2})], \quad (20)\]

\[
\frac{d\rho_{-3/2}}{dt} = - \sum_{j=-3/2}^{j=3/2} \rho_{j} W_{j-3/2} - \sum_{j=-1/2}^{j=1/2} \rho_{j} W_{j-1/2} - \frac{1}{\tau'} \rho_{-3/2} + \alpha'
\]
\[+ \nu \sigma[n_{-1/2} \frac{N}{2} - n_{-3/2} N + n_{-3/2}(N - N_{2})], \quad (21)\]

where

\[
N = \sum_{j=-2}^{j=2} N_{j} \quad \text{and} \quad n = \sum_{j=-3/2}^{j=3/2} n_{j}
\]

\(\tau\) and \(\tau'\) are respectively the relaxation times of the \(\text{Xe}(3\,P_{2})\) and the \(\text{Xe}^{+}(3\,P_{3/2})\) to other configuration states.
For simplicity, we assume a uniform spin-relaxation rate and write

\[ W = W_{ij}, \quad w = w_{ij}; \]

then, in a steady state, \( \frac{dN_j}{dt} = 0, \frac{dn_j}{dt} = 0. \) We have

\[ - \frac{4n_2}{W} + n_1 + n_0 + n_{-1} + n_{-2} = \frac{1}{\tau W} N_2 - \frac{\alpha}{W} \]

\[ - \frac{\nu \sigma}{W} \left[ - n_2(n - n_{3/2}) + n_1 n_{1/2} \right], \quad (22) \]

\[ N_2 - \frac{4n_1}{W} + n_0 + n_{-1} + n_{-2} = \frac{1}{\tau W} N_1 - \frac{\beta}{W} \]

\[ - \frac{\nu \sigma}{W} \left[ N_2(n - n_{3/2}) - n_1 n_1 + n_0 n \right], \quad (23) \]

\[ N_2 + n_1 - \frac{4n_0}{W} + n_{-1} + n_{-2} = \frac{1}{\tau W} N_0 - \frac{\gamma}{W} - \frac{\nu \sigma}{W} \left[ n_1 n_{1/2} - n_0 n + n_{-1} n_{3/2} \right], \quad (24) \]

\[ N_2 + n_1 + n_0 - \frac{4n_{-1}}{W} + n_{-2} = \frac{1}{\tau W} N_{-1} - \frac{\beta}{W} \]

\[ - \frac{\nu \sigma}{W} \left[ n_0 n_{1/2} - n_{-1} n_{-2} n + n_{-2}(n - n_{3/2}) \right], \quad (25) \]

\[ N_2 + n_1 + n_0 + n_{-1} - \frac{4n_{-2}}{W} = \frac{1}{\tau W} n_{-2} - \frac{\alpha}{W} \]

\[ - \frac{\nu \sigma}{W} \left[ n_{-1} n_{1/2} - n_{-2}(n - n_{3/2}) \right], \quad (26) \]

and

\[ - \frac{3n_{3/2}}{W} + n_{1/2} + n_{-1/2} + n_{-3/2} = \frac{1}{\tau W} n_{3/2} - \frac{\alpha'_{1/2}}{W} \]

\[ - \frac{\nu \sigma}{W} \left[ - n_{3/2}(N - N_2) + n_{1/2} n_{1/2} \right], \quad (27) \]

\[ n_{3/2} - \frac{3n_{1/2}}{W} + n_{-1/2} + n_{-3/2} = \frac{1}{\tau W} n_{1/2} - \frac{\beta'}{W} - \frac{\nu \sigma}{W} \left[ n_{3/2}(N - N_2) - n_{1/2} n_{1/2} \right], \quad (28) \]
Adding Eqs. (22-26) and (27-30) gives us

\[ N_2 + N_1 + N_0 + N_{-1} + N_{-2} = \tau(2\alpha + 2\beta + \gamma) = T \]  

(31)

and

\[ n_{3/2} + n_{1/2} + n_{-1/2} + n_{-3/2} = \tau'(2\alpha' + 2\beta') = n \]  

(32)

satisfying the conservation of particles.

Now we solve for \( N \)'s first, subtracting each of Eqs. (22-26) from Eq. (31), and, noting that \( N_2 = N_{-2}, N_1 = N_{-1} \), we get

\[ 5N_2 = N - \frac{1}{\tau W} N_2 + \frac{\alpha}{W} + \frac{\nu \sigma}{W} [-(n - n_{3/2})N_2 + \frac{\beta}{2} N_1] \]  

(33)

\[ 5N_1 = N - \frac{1}{\tau W} N_1 + \frac{\beta}{W} + \frac{\nu \sigma}{W} [(n - n_{3/2})N_2 - nN_1 + \frac{\alpha}{2} N_0] \]  

(34)

\[ 5N_0 = N - \frac{1}{\tau W} N_0 + \frac{\gamma}{W} + \frac{\nu \sigma}{W} [nN_1 - nN_0] \]  

(35)

Solving Eqs. (33-35), we get

\[ N_2 = \frac{N + \frac{\alpha}{W}}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W} (n-n_{3/2})} + \frac{1}{2} \frac{\nu \sigma}{W} \left( \frac{nN_1 - nN_0}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W} (n-n_{3/2})} \right) N_1 \]  

(36)
Now we shall calculate the magnetic resonance signal of the $^3P_2$ metastable state first. When a rf oscillating magnetic field is applied at right angles to the externally applied magnetic field $H_0$, and if the intensity of the rf oscillating magnetic field is high enough so that its amplitude $H_1$ satisfies $\sin \gamma H_1 T/2 \approx 1$, where $\gamma$ is the gyromagnetic ratio of the state under magnetic resonance, and $T$ is its relaxation time, the magnetic-dipole interaction causes the atom to transit between the adjoining magnetic sublevels of the $^3P_2$ state. Since the rate of change of the population of each magnetic sublevel is not only dependent on the transition rate but also on its own population, therefore, the population of the originally more populated sublevel will transit toward the adjoining originally less populated sublevels faster than those immigrating into this sublevel until all magnetic sublevels are equalized, and an equilibrium state (steady state) is then reached. From Eqs. (36-38), we see that if we let the spin-relaxation time for each magnetic sublevel of the $^3P_2$ metastable state approach zero, $N_{+2}$, $N_{+1}$, and $N_0$ will all approach $N/5$. This indicates that when some state is at rf magnetic resonance, its spin-relaxation time tends to equal zero.

\[ N_1 = \frac{(N + \frac{\gamma}{W}) + \frac{\nu \sigma}{W} (n-n_3/2)(N + \frac{\gamma}{W})}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W} (n-n_3/2)} + \frac{1}{2} \frac{\nu \sigma}{W} (N + \frac{\gamma}{W}) 5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W} \]  

and

\[ N_0 = \frac{N + \frac{\gamma}{W}}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W}} + \frac{\nu \sigma}{W} \frac{N_1}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W}} \]
Table II gives the relative absorption probabilities for linearly polarized light corresponding to $^3P_2 - ^3S_1$ electric-dipole transitions. 

When the externally applied magnetic field $H_0$ is parallel to the electron-beam direction, and the rf oscillating magnetic field perpendicular to it, the change of absorption of the incident resonance radiation linearly polarized parallel to $H_0$ due to the magnetic resonance of the $^3P_2$ metastable state is

$$\text{signal} \parallel \propto (\delta)(\Delta N_o) + 2(6)(\Delta N_1) \approx - \frac{h}{5} \left( \beta + \gamma - 2\alpha \right) \frac{1}{W}, \quad (39)$$

where

$$\Delta N_o \equiv (N_o)_{rf \text{ resonance}} - (N_o)_{steady \text{ state}} \approx \frac{N}{5} - \frac{N + \frac{\gamma}{W}}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W}}$$

$$\approx \frac{1}{25} (2\alpha + 2\beta - 4\gamma) \frac{1}{W},$$

$$\Delta N_1 \equiv (N_1)_{rf \text{ resonance}} - (N_1)_{steady \text{ state}} \approx \frac{N}{5} - \frac{N + \frac{\beta}{W}}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W}}$$

$$\approx \frac{1}{25} (\gamma + 2\alpha - 3\beta) \frac{1}{W},$$

if we neglect the term that arises from collisions.

When the incident linearly polarized resonance radiation is polarized perpendicular to the $H_0$ direction, the change of the absorption is given by

$$\text{signal} \perp \propto 2(1)(\Delta N_o) + 2(3)(\Delta N_1) + 2(6)(\Delta N_2) \approx + \frac{2}{5} (\beta + \gamma - 2\alpha) \frac{1}{W}, \quad (40)$$

where

$$\Delta N_2 \equiv (N_2)_{rf \text{ resonance}} - (N_2)_{steady \text{ state}} \approx \frac{N}{5} - \frac{N + \frac{\alpha}{W}}{5 + \frac{1}{\tau W} + \frac{\nu \sigma}{W}(n-n_{3/2})}$$

$$\approx \frac{1}{25} (2\beta + \gamma - 3\alpha) \frac{1}{W}.$$
Table II. Relative absorption probabilities between magnetic sublevels of $^3P_2$ and of $^3S_1$ states.

<table>
<thead>
<tr>
<th></th>
<th>$^3P_2$</th>
<th>2</th>
<th>1</th>
<th>0</th>
<th>-1</th>
<th>-2</th>
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<tbody>
<tr>
<td>$^3S_1$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>6</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
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<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>8</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>-1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>6</td>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>

Light polarized parallel to $H_0$ direction

<table>
<thead>
<tr>
<th></th>
<th>$^3P_2$</th>
<th>2</th>
<th>1</th>
<th>0</th>
<th>-1</th>
<th>-2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^3S_1$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>6</td>
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<td>1</td>
<td>0</td>
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<td>0</td>
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<tr>
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<td>0</td>
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<td>0</td>
<td>3</td>
<td>0</td>
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<td>-1</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>0</td>
<td>6</td>
<td></td>
</tr>
</tbody>
</table>

Light polarized perpendicular to $H_0$ direction
Since the energy of the incident electron we used is only about 3 to 5 eV above the threshold energy of excitation for the $3P_2$ state, we expect $\gamma$ and $\beta > \alpha$. Therefore, we notice that from Eqs. (37) and (40) the absorption of the linearly polarized light polarized parallel to the $H_0$ direction decreases as the magnetic resonance condition for the $3P_2$ state is satisfied. On the other hand, the resonance absorption increases when the incident resonance radiation is linearly polarized perpendicular to the $H_0$ direction. Figure 8 shows a resonance absorption curve of the Xe($3P_2^+$) state, the linearly polarized incident resonance radiation being polarized parallel to $H_0$ direction, compared with a resonance curve of the Cs ground state which was optically oriented; an identical detection system was used for the two resonances. Since we are familiar with the fact that the absorption of the oriented atoms increases when it is at magnetic resonance, then Fig. 8 verifies that the resonance absorption of the $3P_2$ state of Xe decreases as the incident resonance radiation is linearly polarized parallel to $H_0$ direction.

For the magnetic resonance of Xe($^2P_{3/2}^+$), just as for the resonance of Xe($^3P_2$), we set $n_{3/2} = n/4$, if the radio-frequency oscillating magnetic field is strong enough. From Eqs. (36-38), we get

$$\Delta N_2 \equiv (N_2)_{\text{at } n_{3/2} = n/4} - (N_2)_{\text{steady state}} = \frac{N + \frac{\alpha}{W}}{2}(n_{3/2}^{\alpha/2} - n_{3/2}^{\beta/2}) + 0\left(\frac{\gamma^2 \sigma^2}{W^2}\right),$$

$$\Delta N_1 \equiv (N_1)_{\text{at } n_{3/2} = n/4} - (N_1)_{\text{steady state}} = \frac{N + \frac{\alpha}{W}}{2}(n_{3/2}^{\alpha/2} - n_{3/2}^{\beta/2}) + 0\left(\frac{\gamma^2 \sigma^2}{W^2}\right),$$

$$\Delta N_0 \equiv (N_0)_{\text{at } n_{3/2} = n/4} - (N_0)_{\text{steady state}} = 0 + 0\left(\frac{\gamma^2 \sigma^2}{W^2}\right) \approx 0.$$
Fig. 8. Left bell-shaped curve is the rf resonance absorption curve of the oriented Cs ground state. Right curve is the rf resonance absorption of Xe($^3\text{P}_2$) as incident resonance radiation polarized parallel to $H_0$ direction.
Therefore, the signal strengths of $\text{Xe}^+\left(2p_{3/2}\right)$ ionic ground-state resonance observed by monitoring the absorption change of the metastable state are given (to the first order of $v_0/W$) by

$$\text{signal}_{||} = (8)(\Delta N_0) + 2(6)(\Delta N_1)$$

$$\approx -12 \frac{N + \frac{\alpha}{W}}{(5 + \frac{1}{\tau W})^2} \frac{n_{3/2}}{W} \approx - \frac{12}{25} \frac{n_{3/2}}{W} N v_0,$$  \hspace{1cm} (41)

$$\text{signal}_\perp = 2(1)(\Delta N_0) + 2(3)(\Delta N_1) + 2(6)(\Delta N_2)$$

$$\approx 6 \frac{N + \frac{\alpha}{W}}{(5 + \frac{1}{\tau W})^2} \frac{n_{3/2}}{W} \approx \frac{6}{25} \frac{n_{3/2}}{W} N v_0.$$ \hspace{1cm} (42)

Thus the sign of these signals compared with that of the $\text{Xe}\left(3p_{2}\right)$ resonance depends on $n_{3/2} \lesssim \frac{n}{4}$.

It is interesting to compare the signal strengths of $\text{Xe}^+\left(2p_{3/2}\right)$ and $\text{Xe}\left(3p_{2}\right)$. Taking the ratio between Eqs. (39) and (41), we have

$$\frac{\text{signal}_{||}[\text{Xe}^+\left(2p_{3/2}\right)]}{\text{signal}_{||}[\text{Xe}\left(3p_{2}\right)]} = \frac{3}{5} \frac{n_{3/2}}{W} N v_0 \approx - \frac{1}{5} n v_0 \approx \text{the order of} \frac{1}{10},$$  \hspace{1cm} (43)

since we expect $\beta \approx \gamma/2$, $\alpha \approx 0$, $n_{3/2} \approx \frac{1}{2} n$, $n \approx 10^{11}/\text{cc}$, $\tau \approx 10^{-3}$ sec, $v \approx 10^5$ cm/sec, and $\sigma \approx 10^{-13}$ cm$^2$.

This estimation is in fair agreement with the experimental result as shown in Fig. 26.

C. Experiment on Absorption of Intensity Beats

In the introduction and Part A of this section, we have shown how we can produce the excited states of atoms coherently by electron-beam impact, and which magnetic sublevels of the excited state will be
selectively populated when the externally applied magnetic field $H_0$ is parallel to the unidirectional electron beam and the energy of the electron is near the threshold energy of excitation. For the Xe($^3P_2$) state, the $M_J = 0$ and $\pm 1$ (because of the presence of electron exchange and the excitation energy of electrons above the threshold) sublevels are preferentially populated (here we neglect the small population of $M_J = \pm 2$). If now the external magnetic field is applied perpendicular to the direction of the electron beam, the states with $M_J = 0$ and $M_J = \pm 1$ transform into the coherent states, which can be obtained by the following calculation:

$$D^{(j)}_{m m'} = \sum_{m'} \psi^{(j)}_{m'} D^{(j)}(\alpha, \beta, \gamma)_{m m'}$$

where $D^{(j)}(\alpha, \beta, \gamma)$ is the rotation operator, $\alpha$, $\beta$, and $\gamma$ are Eulerian angles (Fig. 9); when $D^{(j)}(\alpha, \beta, \gamma)$ operates on the functions, $\alpha$, $\beta$, $\gamma$ are positive rotations of the functions with respect to a permanently fixed coordinate system, with the rotation by $\gamma$ being performed first.

Namely,

$$D^{(j)}(\alpha, \beta, \gamma) \equiv D^{(j)}_{\text{function}}(\alpha, \beta, \gamma).$$

Therefore,

$$D^{(j)}(\alpha, \beta, \gamma) \equiv D^{(j)}_{\text{function}}(\alpha, \beta, \gamma) = D^{(j)}_{\text{axes}}(-\gamma, -\beta, -\alpha)$$

and

$$D^{(j)}(\alpha, \beta, \gamma)_{m m'} = e^{-i m' \alpha} e^{-i m \beta} \sum_{m'} (-1)^K \sqrt{(j + m)!(j - m)!(j + m')!(j - m')!} \frac{K!}{K!(j + m - K)!(j - m' - K)!(K + m' - m)!}$$

$$\times (\cos \frac{\beta}{2} 2j - 2K - m') m' - m$$
Fig. 9. Eulerian angles.
is the general expression for the representation of the transformation properties of angular-momentum eigenfunctions under rotation through the Eulerian angles.

Thus, in our experiment,

\[ \mathcal{D}^{(2)}_{\text{axes}}(0, -\frac{\pi}{2}, 0) \psi^{(2)}_0 = \sum_{m'=-2}^{+2} \psi^{(2)}_{m'} \mathcal{D}^{(2)}(0, -\frac{\pi}{2}, 0)_{mm'} \]

\[ = \frac{\sqrt{6}}{4} \psi^{(2)}_2 - \frac{1}{2} \psi^{(2)}_0 + \frac{\sqrt{6}}{4} \psi^{(2)}_{-2} \]  \hspace{1cm} (44)

and

\[ \mathcal{D}^{(2)}_{\text{axes}}(0, -\frac{\pi}{2}, 0) \psi^{(2)}_{\pm 1} = \frac{1}{2} \psi^{(2)}_2 \pm \frac{1}{2} \psi^{(2)}_{-1} - \frac{1}{2} \psi^{(2)}_{-2} \]  \hspace{1cm} (45)

The wave function of the \( ^3P_2 \) state of a xenon atom is, therefore, a superposition of eigenfunctions \( \psi^{(2)}_{\pm 2} \), \( \psi^{(2)}_{\pm 1} \), and \( \psi^{(2)}_0 \) of magnetic sublevels; it can be written at time \( t \) as

\[ \psi^{(2)}(t) = e^{-\frac{\Gamma}{2}(t-t_0)} \left[ c_2 \psi^{(2)}_2 e^{i\omega_2(t-t_0)} + c_1 \psi^{(2)}_1 e^{i\omega_1(t-t_0)} + c_0 \psi^{(2)}_0 e^{i\omega_0(t-t_0)} \right. \]

\[ \left. + c_{-1} \psi^{(2)}_{-1} e^{i\omega_{-1}(t-t_0)} + c_{-2} \psi^{(2)}_{-2} e^{i\omega_{-2}(t-t_0)} \right], \text{ for } t > t_0, \]  \hspace{1cm} (45)

where the multiplier \( e^{-\frac{\Gamma}{2}(t-t_0)} \) takes account of the decay of the \( ^3P_2 \) state, \( \omega_2 = E_2/h \), etc., and \( t_0 \) is the instant of excitation of the atom.

The probability of detection, at the instant \( t \), of absorption of a photon with a certain polarization \( \lambda \) is proportional to the square of the absolute value of the matrix element of the electric-dipole transition to the selectively allowable higher state \( ^3S_1 \),

\[ W(t, t_0) = w(t - t_0) \propto \left| \langle ^3S_1 \mid d_\lambda \mid \psi^{(2)} \rangle \right|^2 \]

\[ \propto \sum_{m=-1}^{+1} \left| \langle ^3S_1, m \mid x \mid \psi^{(2)} \rangle \right|^2 ; \]  \hspace{1cm} (46)
since only Xe\(^{(3p_2)}\) is coherently excited, we can only sum over all the magnetic sublevels of Xe\(^{(3s_1)}\) state, and we can call the polarization direction of the incident resonance radiation which propagates along the \(H_0\) direction as the \(x\) direction.

Because

\[
\langle 3s_1, m | x | \psi(2) \rangle = e^{-\frac{\Gamma}{2}(t-t_0)} \left[ c_2^* m_2 e^{i\omega_2 (t-t_0)} + c_1 m_1 e^{i\omega_1 (t-t_0)} \right. \\
+ c_0 m_0 e^{i\omega_0 (t-t_0)} + c_{-1}^* m_{-1} e^{i\omega_{-1} (t-t_0)} \\
+ c_{-2}^* m_{-2} e^{i\omega_{-2} (t-t_0)} \left. \right] ,
\]

where \(M_{m_2} = \langle 3s_1, m | x | \psi(2) \rangle \), etc., therefore

\[
w(t-t_0) \propto e^{-\Gamma(t-t_0)} \frac{1}{\omega_0} \sum_{m=-1}^{1} [ A_m + B_m e^{i\omega_0 (t-t_0)} + B_m^* e^{i\omega_0 (t-t_0)} \\
+ C_m e^{i\omega_0 (t-t_0)} + C_m^* e^{i\omega_0 (t-t_0)} + D_m e^{i\omega_0 (t-t_0)} + D_m^* e^{i\omega_0 (t-t_0)} ],
\]

where

\[
A_m = c_2 c_2^* M_2 m_2^* + c_1 c_1^* M_1 m_1^* + c_0 c_0^* M_0 m_0^* + c_{-1} c_{-1}^* M_{-1} m_{-1}^* \\
+ c_{-2} c_{-2}^* M_{-2} m_{-2}^* ,
\]

\[
B_m = c_{-2} c_{-2}^* M_{-2} m_{-2}^* ,
\]

\[
C_m = c_{-2} c_{-2}^* M_{-2} m_{-2}^* + c_{-1} c_{-1}^* M_{-1} m_{-1}^* ,
\]

\[
D_m = c_{-2} c_{-2}^* M_{-2} m_{-2}^* + c_{-1} c_{-1}^* M_{-1} m_{-1}^* + c_{-2} c_{-2}^* M_{-2} m_{-2}^* ,
\]

\[
E_m = c_{-2} c_{-2}^* M_{-2} m_{-2}^* + c_{-1} c_{-1}^* M_{-1} m_{-1}^* + c_{-2} c_{-2}^* M_{-2} m_{-2}^* .
\]
From Table III we know that
\[\sum_{m=-1}^{+1} A_m \neq 0 \equiv A, \quad \sum_{m=-1}^{+1} B_m = 0, \quad \sum_{m=-1}^{+1} C_m = 0, \quad \sum_{m=-1}^{+1} D_m \neq 0 \equiv D,\]
\[\sum_{m=-1}^{+1} E_m = 0.\]

We have, then,
\[\omega(t-t_0) = e^{-i\omega_0(t-t_0)} \cdot i\omega_0(t-t_0), \quad (47)\]
where \(\omega_0 = \omega_{10}\), which is the Larmor angular frequency of the \(\text{Xe}(^3P_2)\) state.

Now let us consider the case in which the electron beam is modulated periodically as
\[(1 + \cos \omega t) \equiv \mu(t) \ldots \quad (48)\]

Then the number of photons absorbed per unit time at the instant \(t\) is proportional to the quantity
\[I(t) = \int_{-\infty}^{t} \mu(t_0) W(t,t_0) dt_0 = \int_{-\infty}^{t} \mu(t_0) W(t-t_0) dt_0 \quad (49)\]
Substituting (48) into (49), and transforming to the variable of integration \(t' = t - t_0\) [remembering that \((1 + \cos \omega t) = 1 + \frac{1}{2} e^{i\omega t} + \frac{1}{2} e^{-i\omega t}\)], we get
\[I(t) = \int_{-\infty}^{+\infty} \mu(t-t') W(t') dt' = \hat{W}(0) + \frac{1}{2}[\hat{W}(\omega)e^{i\omega t} + \hat{W}^*(\omega)e^{-i\omega t}],\]
where \(\hat{W}(\omega) = \int_{-\infty}^{+\infty} W(t') e^{-i\omega t'} dt'.\) Substituting Eq. (47) into this equation, we get
\[\hat{W}^*(\omega) = \frac{A}{\Gamma + i\omega} + \frac{D}{\Gamma + i(\omega + 2\omega_0)} + \frac{D^*}{\Gamma + i(\omega - 2\omega_0)};\]
Table III. Matrix elements of the x-component of electric
dipole transition between $J = 2$ and $J = 1$ states (from Ref. 28).

<table>
<thead>
<tr>
<th>$M_{12}$</th>
<th>$M_{11}$</th>
<th>$M_{10}$</th>
<th>$M_{1,-1}$</th>
<th>$M_{1,-2}$</th>
<th>$M_{02}$</th>
<th>$M_{01}$</th>
<th>$M_{00}$</th>
<th>$M_{0,-1}$</th>
<th>$M_{0,-2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\langle 1,1</td>
<td>x</td>
<td>2,2 \rangle$</td>
<td>$\langle 1,1</td>
<td>x</td>
<td>2,1 \rangle$</td>
<td>$\frac{1}{\sqrt{2}} \langle 1</td>
<td>x</td>
<td>2 \rangle$</td>
<td>$\langle 1,1</td>
</tr>
<tr>
<td>$-\sqrt{3}$</td>
<td>0</td>
<td>$\frac{1}{\sqrt{2}}$</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>$-\frac{\sqrt{6}}{2}$</td>
<td>0</td>
<td>$\frac{\sqrt{6}}{2}$</td>
<td>0</td>
</tr>
</tbody>
</table>
therefore

\[ I(t) = A \left[ \frac{1}{2} + \frac{\Gamma \cos \omega t + \omega \sin \omega t}{\Gamma^2 + \omega^2} \right] + 2|D| \left[ \frac{\Gamma \cos x + 2\omega_o \sin x}{\Gamma^2 + (2\omega_o)^2} \right] \]

\[ + |D| \left[ \frac{\Gamma \cos(\omega t - x) + (\omega - 2\omega_o)\sin(\omega t - x)}{\Gamma^2 + (\omega - 2\omega_o)^2} \right] \]

\[ + \frac{\Gamma \cos(\omega t + x) + (\omega + 2\omega_o)\sin(\omega t + x)}{\Gamma^2 + (\omega + 2\omega_o)^2} \]  ,  \hspace{1cm} (50)

where \( x = \text{arg} D \).

Thus we obtain an expected result. The first term shows the uninteresting case of resonance-absorption light being modulated with the electron-beam modulation that is always expected to occur. The second term shows the depolarization effect in the magnetic field, known as the Hanle effect. The third term shows the resonance behavior in the modulation amplitude of the absorbed light when \( \omega = 2\omega_o \) and \( \omega = -2\omega_o \). The term \( \omega = -2\omega_o \) shows the effect of reversing the direction of the magnetic field.

The above derivation dealt with an atom of which magnetic sublevels were produced coherently by a modulated electron beam. Since what we observed is the ensemble average value, it is advisable to derive the phenomena once again by using density matrix formalism. Though a very elegant theory of light beats has been formulated by Nedelec, we present a more straightforward theory to describe the phenomena observed, employing intuition without involving a great deal of complex mathematical theory.

Although the rf resonance curve of the Xe(\( ^3P_2 \)) shown in Fig. 8 points out that the substates with \( m = 0 \) and \( \pm 1 \) are more selectively
produced than \( m = \pm 2 \), there is no way of knowing by just what amount these states are produced by the electron-impact method used in the experiments. All we know is that if we take the electron-beam axis as an axis of quantization, the \( ^3P_2 \) state's wave function can be represented as

\[
\psi = a_2(t)|2\rangle + a_1(t)|1\rangle + a_0(t)|0\rangle + a_{-1}(t)|-1\rangle + a_{-2}(t)|-2\rangle,
\]
such that \( \rho_{22} = \rho_{-22}, \rho_{11} = \rho_{-1,-1} \), and \( \rho_{00} \) and \( \rho_{11} > \rho_{22} \), where \( \rho_{mn} \) is the diagonal density matrix element, defined as \( \rho_{mn} = a_m^\dagger(t)a_m(t) \).

Also in this coordinate system, the phases of \( a_m's \) are completely random, because of the cylindrical symmetry about the beam axis. Therefore, \( \rho_{mm'} = 0 \), if \( m \neq m' \). However, if one would apply a magnetic field at some angle with respect to the electron beam, the density matrix \( \rho^o \) in this new coordinate system would be given by a similarity transformation as

\[
\rho^o = \mathcal{D}^{(2)} \rho^o(t=0) \mathcal{D}^{(2)\dagger},
\]

where \( \mathcal{D}^{(2)} \) is the rotation operator for \( J = 2 \). Therefore, in this coordinate system \( \rho^o_{mm} \neq 0 \), for certain \( m \) and \( m' \). Because of the observed fact that at \( t = 0 \), \( \rho_{00}, \rho_{11} > \rho_{22} \), we are certainly justified in assigning the production rate \( \alpha, \beta, \) and \( \gamma \) to indicate the rate of production of the states with \( m = \pm 2, \pm 1, \) and \( 0 \). The initial magnetic sublevel populations at the instant of excitation can be represented by

\[
\rho_o = \frac{1}{2\alpha + 2\beta + \gamma} \begin{pmatrix}
\alpha & 0 & 0 & 0 \\
0 & \beta & 0 & 0 \\
0 & 0 & \gamma & 0 \\
0 & 0 & 0 & \alpha
\end{pmatrix}.
\]

(51)
In addition, it was found experimentally that the selective excitation rate is independent of the magnitude of the externally applied magnetic field $H_0$. Suppose now the electron beam is injected perpendicular to $H_0$, then we can write $\rho^o$ as

$$\rho^o = D^{(2)}(0, -\frac{\pi}{2}, 0)\rho_o D^{(2)*}(0, -\frac{\pi}{2}, 0),$$

where

$$D^{(2)}(0, -\frac{\pi}{2}, 0) = \begin{pmatrix}
\frac{1}{4} & \frac{1}{2} & \frac{\sqrt{6}}{4} & \frac{1}{2} & \frac{1}{4} \\
\frac{1}{2} & -\frac{1}{2} & 0 & \frac{1}{2} & -\frac{1}{2} \\
\frac{\sqrt{6}}{4} & 0 & \frac{1}{2} & 0 & \frac{\sqrt{6}}{4} \\
\frac{1}{2} & \frac{1}{2} & 0 & \frac{1}{2} & \frac{1}{2} \\
\frac{1}{4} & \frac{1}{2} & \frac{\sqrt{6}}{4} & \frac{1}{2} & \frac{1}{4}
\end{pmatrix};$$

therefore, we get

$$\rho^o = \frac{1}{2\alpha+2\beta+y} \begin{pmatrix}
\frac{1}{8}(\alpha+4\beta+3y) & 0 & \frac{\sqrt{6}}{8}(\alpha-y) & 0 & \frac{1}{8}(\alpha-4\beta+3y) \\
0 & \frac{1}{2}(\alpha+\beta) & 0 & \frac{1}{2}(\alpha-\beta) & 0 \\
0 & \frac{\sqrt{6}}{8}(\alpha+y) & 0 & \frac{\sqrt{6}}{8}(\alpha-y) & 0 \\
0 & \frac{1}{2}(\alpha-\beta) & 0 & \frac{1}{2}(\alpha+\beta) & 0 \\
\frac{1}{8}(\alpha-4\beta+3y) & 0 & \frac{\sqrt{6}}{8}(\alpha-y) & 0 & \frac{1}{8}(\alpha+4\beta+3y)
\end{pmatrix}.$$ (52)

The time-rate change of the density matrix is then given by

$$\frac{d\rho}{dt} = -\frac{i}{\hbar} [H, \rho] - \Gamma \rho + \rho^o \frac{1}{T}$$, (53)
where $T_p$ is the electron pumping time (similar to that of an optical pumping time), $-\frac{i}{\hbar} [H, \rho]$ describes the process of time evolution of the excited state due to interaction of the magnetic dipole moment with the field $H_0$, and $\Gamma$ is the spin-relaxation time. Since the Hamiltonian $H$ in Eq. (53) is equal to $-\mu \cdot H_0$, Eq. (53) can be written as

$$\frac{d\rho_{mm'}}{dt} = -i(m-m')\omega_o \rho_{mm'}, -\Gamma \rho_{mm'}, + \rho_{mm'}^0 \frac{1}{T_p}, \quad (54)$$

where $\omega_o$ is the Larmor angular frequency of the $Hg(^3P_2)$ state.

The time-dependent intensity of the light absorbed was derived by Barrat, as given by

$$I(t) = B \sum_{mm'} \langle \mu | e_\lambda \cdot D | m \rangle \rho_{mm'}, \langle m' | e_\lambda \cdot D | \mu \rangle, \quad (55)$$

where $B$ is a constant multiplication factor, and $D$ the vector electric-dipole operator. Thus $e_\lambda \cdot D$ denotes the polarization of the light in the $e_\lambda$ direction.

Since the electron beam is modulated periodically as $(1 + \cos \omega t)$, Eq. (54) can be written as

$$\frac{d\rho_{mm'}}{dt} = \frac{1}{T_p} \rho_{mm'}^0 (1 + \cos \omega t) - i(m-m')\omega_o \rho_{mm'}, -\Gamma \rho_{mm'}, \quad (56)$$

The solution of this equation, noting that $1 + \cos \omega t = 1 + \frac{1}{2} e^{i\omega t} + \frac{1}{2} e^{-i\omega t}$, is given by

$$\rho_{mm'}(t) = \frac{1}{T_p} \rho_{mm'}^0 \left[ \frac{1}{\Gamma + i(m-m')\omega_o} + \frac{1}{\Gamma + i\omega + i(m-m')\omega_o} \right] \left[ \frac{1}{2} e^{i\omega t} + \frac{1}{2} e^{-i\omega t} \right]. \quad (57)$$
Substituting (52) into (57), we get

\[ \rho_{22} = c \frac{1}{8} (\alpha + 4\beta + 3\gamma) \left[ \frac{1}{\Gamma} + \frac{1}{2} e^{i\omega t} + \frac{1}{2} e^{-i\omega t} \right] = \rho_{-2,-2}, \]

\[ \rho_{20} = c \sqrt{6} (\alpha - \gamma) \left[ \frac{1}{\Gamma + i2\omega} + \frac{1}{\Gamma + i(\omega + 2\omega_o)} + \frac{1}{\Gamma - i(\omega - 2\omega_o)} \right] = \rho_{0,-2}, \]

\[ \rho_{2,-2} = c \frac{1}{8} (\alpha - 4\beta + 3\gamma) \left[ \frac{1}{\Gamma + i2\omega} + \frac{1}{\Gamma + i(\omega + 2\omega_o)} + \frac{1}{\Gamma - i(\omega - 2\omega_o)} \right], \]

\[ \rho_{11} = c \frac{1}{2} (\alpha + \beta) \left[ \frac{1}{\Gamma} + \frac{1}{2} e^{i\omega t} + \frac{1}{2} e^{-i\omega t} \right] = \rho_{-1,-1}, \]

\[ \rho_{1,-1} = c \frac{1}{2} (\alpha - \beta) \left[ \frac{1}{\Gamma + i2\omega} + \frac{1}{\Gamma + i(\omega + 2\omega_o)} + \frac{1}{\Gamma - i(\omega - 2\omega_o)} \right], \quad (58) \]

\[ \rho_{02} = c \sqrt{6} (\alpha - \gamma) \left[ \frac{1}{\Gamma + i2\omega} + \frac{1}{\Gamma + i(\omega - 2\omega_o)} + \frac{1}{\Gamma - i(\omega + 2\omega_o)} \right] = \rho_{-2,0}, \]

\[ \rho_{00} = c \frac{1}{4} (3\alpha + \gamma) \left[ \frac{1}{\Gamma} + \frac{1}{2} e^{i\omega t} + \frac{1}{2} e^{-i\omega t} \right], \]

\[ \rho_{-1,1} = c \frac{1}{2} (\alpha - \beta) \left[ \frac{1}{\Gamma - i2\omega} + \frac{1}{\Gamma + i(\omega - 2\omega_o)} + \frac{1}{\Gamma - i(\omega + 2\omega_o)} \right], \]

\[ \rho_{-2,2} = c \frac{1}{6} (\alpha - 4\beta + 3\gamma) \left[ \frac{1}{\Gamma - i2\omega} + \frac{1}{\Gamma + i(\omega - 2\omega_o)} + \frac{1}{\Gamma - i(\omega + 2\omega_o)} \right], \]

where \( c = \frac{1}{2\alpha + 2\beta + \gamma} \).

Next, we consider the observation of the resonance radiation absorption of linearly polarized light polarized parallel to the \( x \) direction and propagating along the \( H_o \) direction; i.e., \( z \) direction.

Equation (55) then becomes

\[ I(t) \propto M_{12}^0 M_{12}^* + M_{12}^0 M_{10}^* + M_{10}^0 M_{12}^* + M_{10}^0 M_{10}^* + M_{01}^0 M_{01}^* \]
\[ + M_{01}^0 M_{01}^* + M_{00}^0 M_{00}^* + M_{00}^0 M_{00}^* + M_{00}^0 M_{00}^* + M_{00}^0 M_{00}^* + M_{00}^0 M_{00}^* \]
\[ + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* \]
\[ + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* + M_{10}^0 M_{10}^* \], \quad (59)
where the $M_{ij}$'s have been defined in the first part of this section, and their values can be found from Table III.

Therefore

$$I(t) = A \left[ \frac{1}{\Gamma} + \frac{1}{\Gamma + i\omega} + \frac{1}{\Gamma - i\omega} \right] + D \left[ \frac{1}{\Gamma + i2\omega} + \frac{1}{\Gamma + i(\omega + 2\omega)} \right]$$

$$+ \frac{1}{\Gamma - i(\omega - 2\omega)} + \frac{1}{\Gamma - i2\omega} + \frac{1}{\Gamma - i(\omega - 2\omega)} + \frac{1}{\Gamma - i(\omega + 2\omega)} \right] \),

where $A$ and $D$ are constant multiplication factors containing linear combinations of $\alpha$, $\beta$, and $\gamma$ and the reduced matrix element, etc.

By rationalizing the denominators of the above equation, we get

$$I(t) = A \left[ \frac{1}{\Gamma} + \frac{\Gamma \cos \omega t + \omega \sin \omega t}{\Gamma^2 + \omega^2} \right] + D \frac{2\Gamma}{\Gamma^2 + (2\omega)^2}$$

$$+ \frac{\Gamma \cos \omega t + (\omega - 2\omega) \sin \omega t}{\Gamma^2 + (\omega - 2\omega)^2} + \frac{\Gamma \cos \omega t + (\omega + 2\omega) \sin \omega t}{\Gamma^2 + (\omega + 2\omega)^2} \right], (60)$$

which is identical to Eq. (50) derived before.

Equations (50) and (60) also satisfy the case in which we expect no observable beat phenomena if the production rate is $\alpha = \beta = \gamma$; that is, there is no selective excitation of the Zeeman sublevels. Therefore, selective excitation must be achieved in order to observe the beat phenomena.
III. EXPERIMENTAL METHOD AND APPARATUS

A. Electron Gun and Light Source

The electron gun, which is simply a diode structure, is the most important part in our experiments. For a detectable signal to be obtained, the following conditions should be met in the first place: (a) The electron beam should be unidirectional; therefore, the gas pressure should be low to avoid multiple scattering of the electron beam. (b) The electron-beam current density should be high so enough of the Xe(3P₂) and Xe⁺(2P₃/₂) states can be obtained.

The maximum electron current that can flow from a hot cathode to an anode in high vacuum is limited by the space charge of the electrons. Normally the current under such conditions is too small to produce a sufficient amount of atoms in the metastable state and ionic ground state. However, from Langmuir's theory of space-charge-neutralized electron flow, we know that if even a small amount of gas is present and the applied voltage is higher than the ionization potential, the positive ions formed tend to neutralize the electron space charge. This neutralization allows the current to increase until it is limited only by the electron emission of the cathode, which depends on the temperature of the cathode. Under the condition of space-charge neutralization, the cathode surface is covered by the positive ion sheath, and the rest of the space is filled by a nearly field-free plasma; the voltage difference between the cathode and anode is concentrated between the cathode and ion sheath, so that the electrons receive most of their acceleration between the cathode and the sheath, and enter the relatively field-free plasma perpendicular to the cathode surface.
Thus, by the space-charge-neutralization method, the requirement of unidirectional high-electron-beam current can be satisfied.

The electron gun is a simple diode structure, as shown in Fig. 10, consisting of a 3/4-in.-diameter indirectly heated Phillips cathode, which is barium oxide imbedded in molybdenum. This cathode can withstand the bombardment of positive ions produced by the electron impact, an anode made of a 35-mil-thick 1-in.-diameter Ta disk, spot-welded to a tungsten rod which extends through the Pyrex tube envelope. The separation between the anode and the cathode is 2 cm. The electron gun was carefully processed by baking at 375°C for about 8 hours before and after cathode activation under a vacuum pressure of about 10^{-8} mm of Hg. All electrodes were rf-bombarded at about 500°C for about 2 hours. After activation the heat-shielded cathode was operated at 1050°C with a heater input power of 50 watts. The bifilar-wound indirect-heating element was constructed of a 20-mil tungsten wire coated with alundum \((\text{Al}_2\text{O}_3)\) by the cataphoresis process. Reagent-type Xe gas was introduced into the gun, which was then operated by applying a dc voltage of about Xe ionization potential across the anode and the cathode for a few hours. The structure was flushed several times, saturating the electrodes with Xe gas to minimize the ion-pumping effect. After several flushings, Xe gas at the desired pressure \((10^{-14} \text{ to } 10^{-3} \text{ mm of Hg})\) was introduced into the gun and the gun was sealed off. Ordinarily, the gun with a glass bulb of 1 liter attached to it as a gas reservoir can be operated almost continuously for several months without any noticeable sign of deterioration. The electron gun used in the intensity-beats experiment is a little different from the electron gun filled with Xe.
Fig. 10. Electron gun.
gas; it has a side tube containing a large drop of Hg, and the side tube was kept at room temperature, giving a vapor pressure of Hg about $10^{-3}$ torr. Using the Phillips cathode, we found it possible to expose the gun to the air for several short periods without any noticeable sign of a drop in electron emission. Although a reprocess cycle of the gun is necessary after each exposure to the air, the same sealed-off electron gun can be reopened to be cleaned up and to change the gas pressure. Characteristic curves for the electron gun are shown in Fig. 11.

An electrodeless discharge of Xe gas, or of Hg mixed with Ne gas, was used as a resonance-radiation light source. A quartz tube (8 mm i.d. and 10 cm long) filled with Xe at a pressure of about 0.9 torr or filled with Ne at a pressure of a few torr and a few little drops of mercury, was placed in a coaxial resonance cavity driven by a Diathermy microwave generator at about 2450 MHz (Diathermy MW/200). Maximum power output of the microwave generator used is about 100 watts. Although light intensity was high at maximum power output of the microwave generator, it was necessary to operate at lower power in order to prevent the quartz tubing from melting from heat generated by the discharge. Because of the relatively hard driving of the discharge, the lamp usually lasted only about a few hundred hours; frequent replacement of the lamp was required, although considerable care was taken in making the lamp by baking the quartz tubing at 375°C for about 5 to 8 hours, and then heating it to about 1000°C as well as rf-discharging it while it was connected to the pump station during several flushings with Xe or Ne gas. The reason for its short life may be that the temperature of the sealed-off lamp under the microwave discharge in
Fig. 11. Characteristic curves for the electron gun when filled with Xe gas at a pressure of about $10^{-4}$ torr.
the cavity was higher than the temperature it reached during its processing in the pump station, so that further outgassing of the quartz tubing took place, while He gas diffused into the tube. Since the gas-filled tube is very simple to construct, we made no further improvement on the lamps.

Since, in our experiments, the atoms were aligned by electron impact, not oriented by circularly polarized pumping light, we found that it was possible to use the resonance radiation emitted from the nearly field-free plasma inside the gun as a light source to monitor the change in the resonance absorption by the metastable state of Xe or Hg existing in the same region. A very rough calculation shows (Fig. 12) that the absorption is at the same order of magnitude if the plasma in the gun has the same intensity as that of the light source.

Suppose that the shape of the plasma column is rectangular, and that the atoms which emit the light of frequency ν are uniformly distributed in this column, then we can assign an $A_ν$ such that the light intensity emitted from the layer of thickness $dx$, at a distance $x$ from the left edge, is $A_ν dx$. This intensity drops to $dI_ν = (A_ν dx)e^{-k_ν(x-L)}$ as it propagates to the right through the plasma column, where $k_ν$ is the absorption coefficient. Therefore, the total intensity after absorption becomes

$$I_ν = \int dI_ν = A_ν \int_0^L e^{-k_ν(x-L)} dx \approx I_{ov}(1 - \frac{1}{2}kL), \quad (61)$$

where $I_{ov} = A_ν L$ is the total intensity of frequency $ν$ emitted from the plasma in the gun; and the light intensity being absorbed, $I_{ov}(\frac{1}{2}kL)$, has the same order of magnitude as shown by the ordinary absorption formula $I_ν = I_{ov}e^{-kL} \approx I_{ov}(1 - kL)$. 
Fig. 12. Calculation of the absorption of the light emitted from the plasma by the metastable states of atoms existing in the same region.
Although the light emitted from the Xe or Hg lamp is much more intense than that emitted from the gun, we did not gain any advantage by using the lamp as a light source. In addition to the small size of the radiant portion (the front end of the lamp which was facing the gun) and the limitation of the subtended solid angle of the incident light by the lens, there always existed a 120-Hz light fluctuation of the lamp which was introduced from the microwave generator. These fluctuations were so large they saturated the preamplifier unless we reduced the voltage of the photomultiplier tube. The result of the reduction of the photomultiplier tube's voltage was obviously a decrease in signal level.

For the experiments reported in this thesis, we did not use the electron gun as a light source; the 120-Hz light fluctuation of the lamp had been overcome by either using a higher maximum input voltage preamplifier such as Tektronix 122 (in the intensity-beat experiment), or using two photomultiplier tubes and a simple bridge, which gave an output as the difference of the outputs from the two photomultiplier tubes [in the magnetic resonances of the Xe($3P_2$) and Xe$^+$(2P$3/2$)-state experiments]. This is given in detail in the next section.

Two different optical arrangements have been used in the experiments, as will be seen in Figs. 17 and 18.

B. Arrangement of Two Photomultiplier Tubes for Reducing the Fluctuation of the Light of the Lamp

In the experiment on the magnetic resonance of the Xe($3P_2$) and the Xe$^+$(2P$3/2$) states, resonance-absorption light is split into two
beams by a beam splitter. One beam is polarized parallel to the electron beam and to the direction of the externally applied magnetic field $H_0$, while the other beam is polarized perpendicular to it. The two beams are detected by two identical RCA 7102 photomultiplier tubes. Since we amplitude-modulated the rf oscillating magnetic field at about 700 Hz, only the ac part of the photomultiplier tube output needed to be detected. A capacitor of 0.1 $\mu$F and a resistor of 1 M$\Omega$ is arranged so as to block off the dc part. The modulated signal and the 120-Hz fluctuation were then sent into a bridge construction, as shown in Fig. 13, the output from which was connected to the differential input of a PAR low-noise amplifier powered by rechargeable batteries. The difference between the two outputs of the 120-Hz fluctuation can be eliminated completely if the bridge and the applied voltage of the two photomultiplier tubes have been suitably adjusted. Contrarily, because of their opposite signs, the two modulated signal outputs will add to each other, increasing the signal-to-noise ratio. That the signs of the two signals—of which one corresponds to the light beam polarized parallel to $H_0$, the other corresponds to the other light beam polarized perpendicular to $H_0$—are opposite to each other has been shown in Sec. II.B, when we calculated the absorption signals from the $3P_2$ to the $3S_{1/2}$ states of Xe. A more general calculation is given in the following.

For simplicity, we consider only the cases in which both $J$ and $J'$ are integers, and $J = J' + 1$ and $J = J'$ (Fig. 14). From consideration of the transition matrix element, we know readily that the signs of signal$_\parallel$ and signal$_\perp$ should, respectively, be the same for the $J = J' - 1$ case as for the $J = J' + 1$ case.
Fig. 13. Balance of two identical photomultipliers.
Fig. 14. Absorption of the linearly polarized light by that state of the atom which has a total angular momentum $J$. 
We have

\[ \text{signal}_\parallel \propto \sum_{MM'} W_{MM'} \delta_{M'M} \left( \frac{N}{2J+1} - N_M \right) , \]

\[ \text{signal}_\perp \propto \sum_{MM'} W_{MM'} \delta_{M',M\pm 1} \left( \frac{N}{2J+1} - N_M \right) , \]

where \( W_{MM'} \) denotes the optical transition rate between the \(|JM\rangle\) and \(|J'M'\rangle\) levels; \( N/2J+1 \) is the population of each \( M \) sublevel of the \( J \) state at rf resonance; \( N_M \) is the population of the \( M \) sublevel of the \( J \) state at steady state; and \( \text{signal}_\parallel \) and \( \text{signal}_\perp \) are defined as in Sec. II.B.

We furthermore assume that at steady state only the \( M = 0 \) sublevel of the \( J \) state is populated; i.e., \( N_0 = N \) and all other \( N_M \)'s are equal to zero.

Therefore, in the \( J = J' + 1 \) case, we have

\[ \text{signal}_\parallel \propto W_{00} \left( \frac{N}{2J+1} - N \right) + \sum_{M=J', \ldots, J+1} W_{MM} \left( \frac{N}{2J+1} \right) \]

\[ \propto J^2 \frac{N}{2J+1} - J^2 N + \frac{2N}{2J+1} \sum_{J-1}^{J+1} (J^2 - M^2) \quad \text{(Ref. 28)} \]

\[ = J^2 \frac{N}{2J+1} - J^2 N + \frac{2N}{2J+1} \sum_{M=1}^{J-1} (J - 1)J^2 - \frac{2N}{2J+1} \sum_{M=1}^{J-1} M^2 \]

\[ = - \frac{N}{3} J(J + 1) < 0 , \]

\[ \text{signal}_\perp \propto 2 \sum_{MM'} W_{MM'} \delta_{M',M-1} \left( \frac{N}{2J+1} - N_M \right) \]

\[ \propto 2 \sum_{M=0}^{J} \frac{1}{4} (J + M - 1)(J + M) \left( \frac{N}{2J+1} - N_M \right) \]

\[ = \frac{1}{2} \left[ - (J - 1)J(\frac{N}{2J+1})2J + \sum_{M=1}^{J} (J + M - 1)(J + M) \frac{N}{2J+1} \right] \]

\[ = \frac{1}{6} \frac{N}{2J+1} J(J^2 + 6J - 1) > 0 . \]
In the $J = J'$ case, we have

$$\text{signal}_\| \propto W_{00}(\frac{N}{2J+1} - N) + \sum_{M=\pm J, \ldots, \pm 1} W_{MM}(\frac{N}{2J+1})$$

$$= \frac{2N}{2J+1} \sum_{M=1}^{J} M^2$$

$$\text{signal}_\perp \propto 2 \sum_{M=0}^{J} \frac{1}{4} (J - M + 1) (J + M)(\frac{N}{2J+1} - N_M)$$

$$= -\frac{1}{2} \frac{N}{2J+1} [2J^2(J+1) - \sum_{M=1}^{J} (J - M + 1)(J + M)] < 0 ,$$

by inspection.

C. Externally Applied Magnetic Field $H_0$

Three Helmholtz coils have been used in the experiments. One coil provided the cancellation of the unneeded terrestrial magnetic component, and was powered by a current-regulated dc power supply (North Hills precision current source model CS-11). The other two coils provided the field $H_0$. A dc current was sent flowing into one of them by a Trygon T50-750 current-regulated power supply to produce a steady magnetic field of the desired magnitude. In order to avoid an overload of the power supply, this was connected to the third coil to produce a saw-tooth linearly changed magnetic field, which was driven and triggered by an Exact-250 function generator. The current from this power supply was regulated to better than one part in $10^4$. The circuit diagram is shown in Fig. 15.
Fig. 15a. Circuit diagram of magnetic field sweep power supply.
Fig. 15b. Circuit diagram of magnetic field sweep power supply.

NOTES:
1. SHEETS TO BE ENCLOSED IN SEALED ENVELOPE.
2. SHEET HEADING: 6 x RIGHT & 4 x LEFT.
3. SCALE: 10:1 on FA.
4. Elevation 0.01% X 0.01% line change.
5. COILS IN SHELL 1 in. due to sheet tear cause a single coil at FA.

BLOCK DIAGRAM SHOWN ON 3X4333
FIG. 15C. Circuit diagram of magnetic field sweep power supply.
Fig. 15a. Circuit diagram of magnetic field sweep power supply.

NOTES:
1. MOUNT ON 304TL PLATE RATING AND AIR COOL
2. DIAGRAM SHOWS MAXIMUM SUGGESTED WIRE SIZES AND AIR COOL
3. BLACK DIAMETER SHOW ON 3X-151

SPEC:
1. POWER SUPPLY (FOH @ PA)
   1 4A 4 PAKE BATT 412

XBL 678-4448
D. Mechanical Switch

To eliminate the stray magnetic field of about 0.5 gauss produced by the indirect cathode heating element of the electron gun, in which usually 5-A current was maintained, the heater current was turned on and off at about 50 Hz, and the signal from the preamplifier was detected only when the heater circuit was off. This process was accomplished by a mechanical switch driven by a Hewlett-Packard 2114A square-wave generator; the circuit diagram of this mechanical switch is shown in Fig. 16. Because of the massive cathode used, the electron-beam current changed by a negligible amount during switching. Since the signal was not received by the phase-sensitive amplifier (Princeton Applied Research JB-5) at all times, and some noise arose from the action of this switch, the signal accumulation time of the time-averaging system must be extended. Some improvements in making a perfectly non-inductive heating element are under investigation.

E. Experiments on rf Resonances of the Xe\(^{3}P_{2}\) and Xe\(^{+}S_{3/2}\) States

The experimental apparatus is almost identical to that used in the experiment on the magnetic resonance of the Ne metastable state,\(^3\) with slight improvements in the optical arrangement and the electronic detection system. Figures 17 and 18 are block diagrams of the experimental arrangements.

The resonance radiation at 8409 Å, corresponding to the \(3S_1 - 3P_2\) transition, was linearly polarized and filtered by a narrow-band filter made by Spectrolab, Inc. at about 35% transmission with a bandwidth of 15 Å. One or two RCA 7102 photomultiplier tubes were used for light
Fig. 16. Circuit diagram of mechanical switch designed by M. Hartley of LRL—Berkeley.
Fig. 17. Block diagram of the experimental arrangement for the experiments on rf resonances of the Xe($2p_2$) and Xe($2p_3/2$) states.

XBL 678-4438
Fig. 18. Block diagram of the slightly improved experimental arrangement for the experiments on rf resonances of the Xe($^3P_2$) and Xe$^+(^2P_{3/2})$ states.
detection. No liquid nitrogen cooling was used, since the time-averaging system we used can overcome the tube-noise problem.

The rf oscillating magnetic field was created and amplitude-modulated at about 700 Hz by means of a General Radio 805-C high-frequency generator whose output was amplitude-modulated by the audio-frequency output of the PAR JB-5 lock-in amplifier.

The signal-to-noise ratio of the Xe\(^{+}\left(\frac{2}{3}P_{3/2}\right)\) resonance was quite small, although that of the Xe\(^{3}P_{2}\) resonance was quite large. This required long-time averaging by assigning a linearly changed field value to the channels of the multichannel pulse-height analyzer. The signal strength was converted to pulses of varying height corresponding to the channel number (using the arrangement as shown in Fig. 17). Alternatively, these signals were converted to data pulses in a single channel at constant height with the PHA operating in time mode (using the arrangement as shown in Figs. 18 and 19). Typically, an integration time of about 3 hours was required for the Xe\(^{+}\left(\frac{2}{3}P_{3/2}\right)\) resonance. The signal-to-noise ratio of the Xe\(^{3}P_{2}\) resonance was high enough so that one could observe it from an Esterline-Angus recorder connected to the monitor of the lock-in amplifier. However, this output was also fed into the PHA in order to compare the relative amplitudes and signs of the two resonance curves, and to calibrate the magnetic-field sweep region.

F. Experiment on Intensity Beats in Resonance Absorption

1. Principle of the Experiment

If the externally applied magnetic field \(H_0\) is perpendicular to the electron beam, the system of atoms in the metastable state is not
Fig. 19. Circuit diagram of time mode for PHA.
cylindrically symmetrical about the field $H_0$. Therefore, the system commences a Larmor precession about the field $H_0$. Since the system is aligned, not oriented, precession by 180 degrees is the same as 360-deg precession about $H_0$. Now consider the case of pulsed electron-beam excitation; the duration of the pulse is much shorter than the Larmor period; the first pulse excites and aligns the system of metastable atoms, which then executes a Larmor precession about $H_0$. The second pulse, coming in $\frac{1}{2}$ Larmor period later, creates another group of aligned metastable atoms, which precess about $H_0$ in phase with the metastable atoms already aligned by the first pulse. Thus from this classical point of view, an aligned population will precess about the external magnetic field $H_0$ synchronously (coherently) when it is produced by short electron pulses repeated at half the Larmor period; i.e., at twice the Larmor frequency. The decay of the aligned population due to spin relaxation will be compensated by the successive excitation by the pulsed electron beam.

If one monitors such a system with linearly polarized resonance radiation connecting the metastable state to a higher state, one finds that the light absorption is modulated at twice the Larmor frequency. This can also be visualized by a classical picture, as shown in Fig. 20. A virtual electric dipole in the atom is created by the linearly polarized light. Because of Larmor precession of the atom due to a magnetic-dipole interaction with $H_0$, the virtual electric-dipole oscillator will also precess about $H_0$; then the amount of light absorbed by the atom will reach two maxima in one precession, whenever the virtual linear oscillator is parallel to the direction of the polarization of the light. Thus the light absorption is modulated at twice the Larmor frequency.
Fig. 20. Classical picture for the intensity beats experiment.
2. Modulation of the Electron Beam

As shown in Fig. 21, for achieving high-frequency modulation of a space-charge-neutralized high-current electron beam, we interrupted the electron flow from the cathode to the ground, using as a switch a fast-response transistor 2N2893 capable of drawing a high current. We found that we were able to achieve 100% modulation of the electron beam with a peak-to-peak current of 400 mA at about 3 MHz.

Although the lifetime of the metastable state of Hg is typically a few milliseconds, the spin-relaxation time is much shorter; spin-relaxation time can be readily measured either by the bandwidth associated with paramagnetic resonance of the metastable state or by the magnetic bandwidth associated with the Hanle effect of resonance radiation absorption. Since the experimental setup for the observation of the Hanle effect needed only very little modification to satisfy the experimental conditions for the beat experiment, we determined the spin-relaxation time of the metastable state of Hg by observing the Hanle effect. The signal is shown in Figs. 22 and 23. By matching this curve to the second term of Eq. (50) or Eq. (60), we obtained the spin-relaxation time $\tau$ of the $^{3}P_{2}$ state of Hg; under our experimental conditions it was $2 \times 10^{-4}$ sec. As we have already stated in the introduction, the Larmor frequency $\Delta \nu = \omega_{0}/2\pi \gg \frac{1}{\tau}$; in order to observe the beat phenomena, we decided to use 2.89 MHz for electron-beam modulation (because we had a narrow-band amplifier tuned at this frequency).

Because the externally applied magnetic field $H_{0}$, which was at right angle to the modulated electron beam, was only about 1 gauss, the deflection of the electron beam was so small that no correction needed to be considered.
Fig. 21. Method used to modulate the space-charge-neutralized electron beam at high frequency (≈ 3 MHz).
Fig. 22. Hanle-effect signal. The externally applied magnetic field was modulated at about 700 Hz.
Fig. 23. Hanle-effect signal. The integrated curve of Fig. 22.
3. **Experimental Arrangement**

Figure 24 shows the block diagram of the experimental setup. For observing the Hanle effect, the only modifications are (a) connecting the output of photomultiplier directly to the preamplifier, and (b) changing the magnetic field sweep to the neighborhood of zero value to include zero field. The highest frequency that can be locked by the PAR JB-5 lock-in amplifier is 150 kHz, much smaller than the modulation frequency of the light absorption. In order to use the lock-in amplifier, we modulated the external magnetic field $H_0$ at about 700 Hz; therefore, the shape of the signal appeared as a dispersion curve instead of a bell shape, as was the case when we modulated the rf oscillating magnetic field in the magnetic resonances of the $\text{Xe}(^3\text{P}_2)$ and $\text{Xe}^+(^2\text{P}_{3/2})$ states.
Fig. 24. Block diagram of the experimental apparatus of the resonance absorption beat experiment.
IV. EXPERIMENTAL RESULTS AND ANALYSIS

A. Experiments on the Magnetic Resonances of Xe(3P₂) and Xe⁺(2P₃/₂)

Figures 25 and 26 show the optical absorption curves of Xe(3P₂) and Xe⁺(2P₃/₂) as a function of the applied magnetic field $H_0$ with the frequency of the rf oscillating magnetic field fixed. The magnetic field value was calibrated from the magnetic resonance of a metastable state of the even isotope of Xe whose $g_J$ value is known from the atomic beam magnetic resonance to be $-1.50089 \pm 0.00006$. The Xe⁺(2P₃/₂) magnetic resonance was identified by noting its resonance positions corresponding to the calculated value of $g_J$ with respect to the $g_J$ and $g_F$ values of the metastable state of even and odd isotopes of Xe.

The signals shown in Fig. 25 are correlated with resonances in the Xe(3P₂) and Xe⁺(2P₃/₂) states of even isotopes. Figure 26 shows the magnetic resonances, at a higher rf power level than in Fig. 25, of (a) odd isotopes of $^{129}$Xe(3P₂) and $^{131}$Xe(3P₂), and (b) of even isotopes of Xe⁺(2P₃/₂) which have a power dip, in addition to (c) a resonance, corresponding to $g = -1.2530 \pm 0.0040$, that is due to cascading from the long-lived excited state that decays to the short-lived intermediate state by emitting infrared light, and then decays to the metastable state while maintaining its initial alignment because of unidirectional electron-impact excitation (a detailed description of the method for identifying the long-lived excited state will be published elsewhere).

Measured $g$ values of $^{129}$Xe(3P₂) with $I = 1/2$, $F = 5/2$, and $^{131}$Xe(3P₂) with $I = 3/2$ and $F = 3/2$ are both $-1.2007 \pm 0.0040$. Figure 27 shows a magnetic resonance corresponding to $g_F = -1.7986 \pm 0.0023$ which belongs to $^{129}$Xe(3P₂) with $F = 3/2$. 
Fig. 25. Magnetic resonances of \( \text{Xe}(^3\!P_2) \) and \( \text{Xe}^+(^2\!P_{3/2}) \) states when a single photomultiplier tube is used.
Fig. 26. Magnetic resonances of $^{129}\text{Xe}(^3P_2)$, $^{131}\text{Xe}(^3P_2)$ corresponding to $F = 5/2, J = 2, I = 1/2$ and $F = 3/2, J = 2, I = 3/2$ respectively, and of even $\text{Xe}^+(^2P_{3/2})$ state.
Fig. 27. Magnetic resonance of $^{129}\text{Xe}(^3P_2)$ with $F = 3/2, J = 2, I = 1/2$. 
It is interesting that the relative signs of the resonance signals of Xe\(^{3}\,P_2\) and Xe\(^{+}\,^{2}\,P_{3/2}\) are opposite to each other. From Eqs. (39) through (42), we conclude that \(n_{3/2}\) is greater than \(n_{1/2}\) when Xe\(^{+}\,^{2}\,P_{3/2}\) reaches its steady state, namely \(M' = \pm \frac{3}{2}\) magnetic sublevels are more populated than \(M' = \pm \frac{1}{2}\) sublevels by unidirectional electron-beam excitation of the Xe ground state or \(^3\,P_2\) state.

At first glance, this surprising conclusion about the population distribution, drawn from the results of the rate equations we calculated, may fail to convince those who are acquainted with the Dehmelt type "spin exchange" experiments. In those experiments, two species of atoms were contained in a cell. The No. 1 species of atoms was oriented by circularly polarized resonance radiation; through spin exchange, this orientation transferred to the No. 2 species of atoms. If then a rf oscillating magnetic field was applied which was capable of equalizing the population of the No. 2 species among their magnetic sublevels, this equalization transferred back to the No. 1 species through its spin exchange in collisions with a depolarizing species No. 2.

With this clear physical picture in mind, one notices that \(n_{3/2} > n_{1/2}\) at the steady state of Xe\(^{+}\,^{2}\,P_{3/2}\), as shown in Fig. 28. A thicker line expresses more population in that magnetic sublevel. We already know the population distribution of Xe\(^{3}\,P_2\) states in the steady state. An equalization of the population of Xe\(^{+}\,^{2}\,P_{3/2}\) caused by a rf magnetic field will transfer to the Xe\(^{3}\,P_2\) state through the "spin-exchange" collisions, giving the same result (though with different degree) as applying a rf magnetic field to directly resonate the Xe\(^{3}\,P_2\) state. Therefore, the signs of the two magnetic resonance signals would seem to be the same!
Fig. 28. Magnetic sublevels and population distributions of $\text{Xe}^+\left(\frac{3}{2}\right)$ state and $\text{Xe}\left(\frac{3}{2}\right)$ state in our experiment.
The contradiction can be resolved when one notes that the initial conditions for the Dehmelt-type spin experiments and our "spin-exchange" experiment are different. In the former case, an orientation of the No. 2 species was produced by spin-exchange collisions; and in our case, the alignment of No. 2 species was predominantly achieved by unidirectional electron-beam impact. Two collisions between the two species are needed for observing the rf magnetic resonance of the No. 2 species in the former case, but only one collision between the two species is needed in this experiment. For simplicity, suppose that each species has only two magnetic sublevels, and that the population distribution is just reversed at steady state, as shown in Fig. 29. Under the condition of conservation of total angular momentum, only when the $m' = \pm 1/2$ sublevel in species No. 2 interacts with the $m = \pm 1/2$ sublevel in species No. 1, there is a net population-distribution change of both species. If we consider the spin-exchange interaction only, the rate of population change of any sublevel is proportional to the occupations of the two interacting sublevels. But when we consider also the production rate to each sublevel of each species by electron impact, the decay rate, and the spin-relaxation rate, etc., the situation is not so simple. At steady state, though level C ($m' = 1/2$) of No. 2 species and level B ($m = -1/2$) of No. 1 species are more populated, there is no net population change of any sublevel, unless one of the sublevels such as level D of species No. 2 increases its population because of some other interaction. Then the population-enhanced level D starts to interact with the level A, which is in the other species and has opposite sign, to reduce the increased population in level D. Because of the conservation
Fig. 29. Illustration of the "antidepolarization through spin-exchange collisions" argument.
of the particles, as the population of level D of species No. 2 is increased, that of level C of the same species is decreased. But level C does not interact with the other species, since if it did so its population would be further reduced. From the above argument, we know immediately that when a rf magnetic field is used to equalize the population of the levels C and D of species No. 2, the population-increased sublevel, level D, will interact with level A of species No. 1. This results in an "antidepolarization" of species No. 1. Therefore, the arguments on "the depolarization transformation through the spin-exchange collision" cannot be generally used in all the "spin-exchange" experiments.

Comparing this simplified example with our experiment, we notice that if \( n_{3/2} > n_{1/2} \) at the steady state of \( \text{Xe}^+ (2P_{3/2}) \), the rf magnetic resonance of \( \text{Xe}^+ (2P_{3/2}) \) will cause \( m = 0 \) of \( \text{Xe} (3P_2) \) state to be more populated; as a result the resonance signal of \( \text{Xe}^+ (2P_{3/2}) \) will have opposite sign from that of magnetic resonance of \( \text{Xe} (3P_2) \) state.

Although we still assume \( n_{3/2} > n_{1/2} \) at the steady state of \( \text{Xe} (2P_{3/2}) \) in order to make the calculation consistent with experimental result, we believe that some detailed theoretical derivation and experimental proof are necessary to find the right answer, or to give a possible reason for the relative populations. We can still examine our rate equations and the assumptions we made by calculating some other known experimental results.

Firstly, we check with the Dehmelt-type spin-exchange experiment, as shown in Fig. 30, using the identical notations as used in Sec. II.B. We write the rate equations of the No. 1 species as
Fig. 30. Calculations of the depolarization and antidepolarization.
\[ \frac{d N_{1/2}}{dt} = W(- N_{1/2} + N_{-1/2}) - \frac{1}{\tau} N_{1/2} + \alpha + \nu \sigma (N_{1/2}^2 N_{-1/2} + N_{-1/2}^2 N_{1/2}) \]

and

\[ \frac{d N_{-1/2}}{dt} = W(N_{1/2} - N_{-1/2}) - \frac{1}{\tau} N_{-1/2} + \beta + \nu \sigma (N_{1/2}^2 N_{-1/2} - N_{-1/2}^2 N_{1/2}) . \]

At steady state, \( \frac{d N_{1/2}}{dt} = \frac{d N_{-1/2}}{dt} = 0 \), we get

\[ (- W - \frac{1}{\tau} - \nu \sigma_{-1/2}) N_{1/2} + (W + \nu \sigma_{1/2}) N_{-1/2} + \alpha = 0 \]

and

\[ (W + \nu \sigma_{-1/2}) N_{1/2} + (- W - \frac{1}{\tau} - \nu \sigma_{1/2}) N_{-1/2} + \beta = 0 . \]

Adding them together, we obtain \( N_{1/2} + N_{-1/2} = \tau (\alpha + \beta) = N \), the conservation of particles. Solving for \( N_{1/2} \), we get

\[ N_{1/2} = \frac{(1 + \frac{\nu \sigma_{1/2}}{W}) N + \frac{\alpha}{W}}{2 + \frac{1}{\tau W} + \frac{\nu \sigma}{W}} . \]

(I) Suppose that the initial conditions are \( \alpha \neq 0, \beta = 0 \), \( n_{1/2} \approx n, n_{-1/2} \approx 0 \). Then

\[ \Delta N_{1/2} = (N_{1/2})_{n_{1/2}=n/2} - (N_{1/2})_{\text{at steady state}} \]

\[ = - \frac{\nu \sigma N}{2W} \frac{\nu \sigma N}{2 + \frac{1}{\tau W} + \frac{\nu \sigma}{W}} < 0 , \]

which means that when we depolarize the No. 2 species by a rf magnetic field, the No. 1 species is also depolarized through the spin-exchange collisions with No. 2 species, as shown in Fig. 30.

(II) Suppose that the initial conditions are \( \alpha = 0, \beta \neq 0 \), \( n_{1/2} \approx n, n_{-1/2} \approx 0 \), as in Fig. 29. Then
\[ \Delta n_{1/2} = (n_{1/2})' - (n_{1/2}) \text{ at steady state} \]

\[ \frac{v_{\text{on}} N}{2W} < 0 \]

We get the same results as the explanation given on page 82.

Now let us consider an experiment on the nuclear alignment of the \(^{131}\text{Xe}\) ground state by metastability exchange.\(^{25}\) The \(^{131}\text{Xe}\) ground state \(^{1S_0}\) with \(I = 3/2\) was aligned through exchange collisions with the aligned metastable state of Xe excited by unidirectional electron-beam impact. Since \(^{131}\text{Xe}(1S_0)\) has \(I = 3/2\), the rate equations are more or less as those we have used in Sec. II.B with the production rates \(\alpha'\) and \(\beta'\) equaling zero, and the decay time \(\tau'\) approaching \(\infty\). Considering the hfs of \(^{131}\text{Xe}(3P_2)\), we get

\[ n_{3/2} = \frac{(n + \frac{\alpha'}{W}) + \frac{v_{\sigma}}{4W} Nn}{4[1 + \frac{1}{4\tau' W} + \frac{v_{\sigma}}{4W} (\frac{3N}{2} - N_{7/2})]} \]

and

\[ n_{1/2} = \frac{(n + \frac{\beta'}{W}) + \frac{v_{\text{on}}}{2W} (N - N_2)}{4[1 + \frac{1}{4\tau' W} + \frac{v_{\sigma}}{4W} (\frac{3N}{2} - N_{7/2})]} \]

Setting \(\alpha' = \beta' = 0\), \(\tau' \to \infty\), we have, at steady state,

\[ n_{3/2} \text{ of } ^{131}\text{Xe}(1S_0) \text{ with } I = 3/2 = \frac{n + \frac{v_{\sigma}}{4W} Nn}{4[1 + \frac{v_{\sigma}}{4W} (\frac{3N}{2} - N_{7/2})]} \]

\[ n_{1/2} \text{ of } ^{131}\text{Xe}(1S_0) \text{ with } I = 3/2 = \frac{n + \frac{v_{\sigma}}{2W} (N - N_2)n}{4[1 + \frac{v_{\sigma}}{4W} (\frac{3N}{2} - N_{7/2})]} \]
Therefore \( n_{3/2} < n_{1/2} \), since \( N_{7/2} < N/8 \) at steady state (see Fig. 31).

From this result, the observed magnetic resonance signal of the \( ^{131}\text{Xe}(1S_{0}) \) with \( I = 3/2 \) will have the same sign as the magnetic resonance of \( \text{Xe}(3P_{2}) \). Figure 32 shows the two signals; it gives the evidence for the above statement.

When there is no magnetic field gradient in the space between the anode and cathode of the electron gun, the half-bandwidth of the magnetic resonance curve is the natural half-bandwidth \( \Delta \nu \). From \( \Delta \nu = W/\pi \), the spin-relaxation time \( 1/W \) can be obtained. This has been derived by Colegrove,\(^{31}\) using the time-dependent transition probability \( P(J,m,m',t) \)\(^{35}\) for the transitions between the magnetic sublevels of the He metastable state induced by a rf magnetic field (instead of \( 1/W \)) in the signal calculation based on the solution of the rate equations.

**B. Experiments on Intensity Beats**

Figure 33 shows the 2.89-MHz component of the optical absorption as a function of the externally applied magnetic field; the peak absorption corresponds to a magnetic field of \( H = 688\pm10 \text{ mG} \) at \( \nu_L = 1.445 \text{ MHz} \). To make certain that the signals were real, we shifted the electron-beam modulation frequency to 2.72 MHz and observed the proper shift of the beat signal, as shown in Fig. 34.
Fig. 31. Schematic diagram of the hyperfine-structure levels of $^{131}$Xe in a magnetic field.
Fig. 32. The first trace (a) shows the magnetic resonance of Xe\(^{3}\)P\(_2\) even isotopes. The second trace (b) shows the nuclear magnetic resonance of \(^{131}\)Xe in the \(^{4}\)S\(_0\) ground state with a linearly oscillating magnetic field of \(\nu = 2.5680\) kHz. The third trace (c) shows a typical NMR signal by a digitalized signal-averaging system for four sweeps over the resonance at different \(H_0\) and \(J\) values from those of trace (b).
Fig. 33. Resonance-radiation-absorption beat signal with electron beam modulated at 2.89 MHz.
Fig. 34. Resonance-radiation beat signal with electron beam modulated at 2.72 MHz.
V. CONCLUSION

Using a simple diode-structure electron gun which can emit a high-current-density, low-energy and unidirectional electron beam under the space-charge-neutralization condition, and employing relatively simple apparatus, we have readily observed the ionic ground state $^{2}\text{P}_{3/2}$ of the xenon atom (which is very difficult to observe by more conventional methods). We also observed the beats in the resonance absorption of the metastable state $^{3}\text{P}_{2}$ of the mercury atom; the absorption is, in principle, quite similar to that observed by Bell and Bloom$^{36}$ using the optical method. In addition to the advantage of the fast pumping speed of the space-charge-neutralized high-current-density electron beam which has been exhibited in our experiments, we also have the advantage of electron-impact excitation, which can put the atoms into any excited state, and into the ionic ground state with less selection-rule restriction than can optical excitation. Since electron-impact excitation can only align the atoms, our method should work with a variety of easily vaporized atoms having a total angular momentum larger than 1/2 for the ionic ground state. Recently, we have made some efforts to observe the nuclear orientation of $^{129}\text{Xe}$ with nuclear spin $I = 1/2$ by "electron pumping," "optical pumping," and possible "metastability exchange," using nearly the same experimental arrangement, as we used in the rf resonance of $\text{Xe}^{+}(^{2}\text{P}_{3/2})$ state through "spin exchange" with the $\text{Xe}(^{3}\text{P}_{2})$ state except that we changed the polarization of the incident resonance radiation and the direction of the externally applied magnetic field; the first few tries have indicated that our method worked. The advantages of our method, besides the
above mentioned, are: (a) only one collision is needed to observe the rf resonance, while the conventional method demands at least two, thus requiring relatively long spin-relaxation times of the colliding species or higher pressure; (b) our electron gun operated under lower gas pressure (about $10^{-3}$ to $10^{-4}$ torr), therefore the collision of the aligned or oriented atoms with other atoms could be reduced; (c) the high-flux electron beam in the electron gun was very stable, and therefore the fluctuation of the electron temperature was very little.

Since no rf magnetic field, which perturbs the excited state, is needed in the beats experiment, this experiment can offer a new method for investigating the excited state of atoms that has a total angular momentum $J > 0$, and for giving information about the hyperfine structure.
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REFERENCES


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