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ABSTRACT

The amphoteric native defect model of the Schottky barrier formation is used to analyze the Fermi level pinning at metal/semiconductor interfaces for submonolayer metal coverages. It is assumed that the energy required for defect generation is released in the process of surface back-relaxation. Model calculations for metal/GaAs interfaces show a weak dependence of the Fermi level pinning on the thickness of metal deposited at room temperature. This weak dependence indicates a strong dependence of the defect formation energy on the Fermi level, a unique feature of amphoteric native defects. This result is in very good agreement with experimental data. It is shown that a very distinct asymmetry in the Fermi level pinning on p- and n-type GaAs observed at liquid nitrogen temperatures can be understood in terms of much different recombination rates for amphoteric native defects in those two types of materials. Also, it is demonstrated that the Fermi level stabilization energy, a central concept of the amphoteric defect system, plays a fundamental role in other phenomena in semiconductors such as semiconductor/semiconductor heterointerface intermixing and saturation of free carrier concentration.
I. Introduction

Despite four decades of studies there exists no commonly accepted microscopic model for Schottky barrier formation at metal semiconductor (M/S) interfaces.\textsuperscript{1,2} For metals deposited on covalent or weakly ionic semiconductors the Fermi level is pinned in a narrow range of energies. This fact has led to the realization that high densities of localized states must exist at the M/S interfaces. The microscopic nature of these defects and the mechanisms leading to their formation remain poorly understood despite considerable experimental and theoretical efforts. Among existing proposals, native defects\textsuperscript{3} and metal induced gap states\textsuperscript{4-7} (MIGS) have attracted most of the attention. A large variety of native defects including vacancies, antisites and even some more complex defects such as EL2 in GaAs were considered to play an important role in pinning of the Fermi energy.\textsuperscript{3,8} Experimentally observed general trends in the Fermi level pinning position among various semiconductors were successfully described by the theoretically calculated average energy of neutral vacancies\textsuperscript{8} as well as by the neutrality point energy,\textsuperscript{7} which has been suggested to represent the pinning energy in the MIGS model.

Recent years have brought significant progress through experimental studies of metal semiconductor interfaces. Measurements of the Fermi level pinning for extremely low metal coverages,\textsuperscript{9} as well as experiments with metals deposited on the substrates kept at low temperatures, have been performed.\textsuperscript{10,11} These experimental developments pose new challenges and provide critical tests for the applicability of various models of Schottky barrier formation.

Most recently a new mechanism of Schottky barrier formation has been proposed.\textsuperscript{12} The mechanism is based on the existence of a class of native
defects (amphoteric defects) whose electrical identity is controlled by the Fermi level position. Contrary to previous defect models, the amphoteric defect model relates the Fermi level pinning to the thermodynamic properties of the whole defect system rather than to energy levels of specific defects. The major features of this model relevant to thick metal coverages have been discussed before. In this paper the consequences of the model for Fermi level position as a function of behavior at submonolayer metal coverages at both low and room temperature are presented. We also show that the central concept of the model, intrinsic Fermi level stabilization, can be utilized to explain other phenomena such as maximum doping levels in semiconductors and doping induced heterointerface intermixing.

II. Properties of amphoteric native defects

The formation energy of a crystal lattice defect is the sum of the energy of the structural rearrangement of the lattice, $E_{\text{str.}}$, and of the electronic energy, $E_{\text{el}}$, required to transfer charge from the defect to the Fermi sea (i.e. holes to or from the valence band, electrons to or from the conduction band). It is obvious that $E_{\text{el}}$ is determined by the position of the Fermi level with respect to the energy levels introduced by the defect. $E_{\text{el}}$ can be quite large for defects which can support multiple charges. In such cases the position of the Fermi level will strongly affect the probability of defect generation. The electronic part of the defect formation energy is responsible for the well-known self-compensation effect. This effect was clearly observed in weakly bound ionic II-VI semiconductors where some type of doping cannot be realized because of the compensation by the native defects. In more strongly bound III-V semiconductors this effect is less pronounced. It will be shown here that in the case of III-V semiconductors the abundances of
already existing defects are controlled by the electronic part of the defect energy.

We have found that in III-V semiconductors the introduction of large concentrations of native defects leads to the stabilization of the Fermi energy. The stabilization energy, $E_{FS}$, does not depend on the type of doping or the doping level of the original material. In fig. 1, the stabilization energy deduced from the Fermi level position in heavily irradiated semiconductors along with the Fermi level position governing Schottky barrier heights are shown. We observe that for all semiconductors for which data are available there exists good agreement between these two Fermi energy positions in a given material. This may be interpreted to indicate that a similar mechanism is responsible for the Fermi level stabilization in both cases.

It has been demonstrated previously that amphoteric native defects are responsible for the Fermi level stabilization. When vacancy interstitial pairs on either anion or cation sites are generated in GaAs, the defects undergo transformations according to the reactions:

\begin{align*}
V_{Ga} + Ga_{I} + As_{As} & \leftrightarrow As_{Ga} + V_{As} + Ga_{I} \\
Ga_{As} + V_{Ga} + As_{I} & \leftrightarrow V_{As} + As_{I} + Ga_{Ga}
\end{align*}

where $V$ stands for vacancy and $I$ for interstitial. When the concentrations of interstitials are low, the reactions take the forms:

\begin{align*}
V_{Ga} + As_{As} & \leftrightarrow As_{Ga} + V_{As} \\
Ga_{As} + V_{Ga} & \leftrightarrow V_{As} + Ga_{Ga}
\end{align*}
Although transformations between Ga and As sites occur in these reactions, for the sake of simplicity the defects described by eqs. (1a) and (2a) (1b and 2b) will be called Ga(As) sublattice defects. The total energies of the defects participating in reactions (1) and (2) are presented in figs. 2 and 3. The diagrams were obtained using reported total defect energy calculations, in GaAs.\textsuperscript{16,17} It is seen from these figures that in n-type GaAs the reaction represented by eqs. 1 and 2 are driven to the left-hand side, i.e. acceptor-like defects are formed. The electrons are transferred from the Fermi sea to the defects. This results in a downward shift of the Fermi level upon defect generation. On the other hand, in p-type material the reactions 1 and 2 are driven to the right-hand side which means that donors are formed which will shift the Fermi energy upward. Eventually, for large enough concentrations of defects, the Fermi energy will reach the level where equilibrium conditions for the defect reactions 1 or 2 are attained. This corresponds to the Fermi level position where there is no charge transfer from or to the Fermi sea even if new defects are formed. The energy at which this happens is defined as the Fermi level stabilization energy. From fig. 2 and 3 we find that depending on the defects involved, $E_{FS}$ ranges from $E_v + 0.6$ eV to $E_v + 1.0$ eV. The actual stabilization energy in this range depends on the relative contributions of different defects to the total charge balance.

II. Mechanism of defect formation and Fermi level pinning

A major property of the amphoteric native defect system is the strong dependence of the various defect formation energies on the Fermi level position. As is seen in fig. 2, defect formation energies decrease rapidly with either p- or n-type doping. However, even in heavily doped GaAs
considerable energy is required to create a vacancy-interstitial pair in GaAs. The energy of a metal atom impinging on a GaAs surface is not sufficient to directly create defects. Two alternate sources of energy which could lead to the formation of defects were identified. It has been argued that a large energy is released during formation of metal atom clusters on the GaAs surface.\(^{18}\) The amount of energy released in such a process depends on the size of stable clusters and may be as high as 3 eV. Another process in which energy is released is back-relaxation of relaxed cleaved surfaces. It has been found that an energy of \(E_o = 0.35\) eV per surface atom is released in the process of Al induced back-relaxation of a cleaved (110) GaAs surface.\(^{19}\) In the following, it is assumed that surface back-relaxation is the major source of the energy which is required to form defects.

In order to model the process of defect formation we adopted calculated bulk values for the Fermi dependent defect formation energies as given in fig. 2. Also, we assume that the probability \(p(N)\) that \(N\) surface atoms back-relax is given by the Poisson distribution,

\[
p(N) = e^{-<N>} \frac{<N>^N}{N!} \tag{3}
\]

where \(<N>\) is the mean value of the distribution. Thus the probability of creating a defect with the formation energy \(E_{\text{def}}(E_F)\) is:

\[
G(E_F) = p(N > N_o) = \int_{N_o}^{\infty} p(N) dN \tag{4}
\]

where \(N_o = E_{\text{def}}(E_F)/E_o\)
In the presence of interface defects of the concentration $N_{\text{def}}$ the position of the Fermi energy, $E_F$, measured with respect to the bulk Fermi energy $E_{Fb}$ is,

$$E_{Fb} - E_F = \frac{2\pi}{\varepsilon} \left( \frac{N_{\text{def}}Q}{N_i} \right)^2$$  \hspace{1cm} (5)

where $\varepsilon$ is the dielectric constant, and $Q$ is the net charge transfer from or to the defect. $N_i$ is the concentration of free carriers in the bulk. From eqs. (3) and (4) one obtains the relationship between the position of the Fermi energy and the concentration of the metal atoms on the surface,

$$dN_{\text{at}} = \left( \frac{\varepsilon N_i}{2\pi} \right)^{1/2} \frac{dE_F}{2|Q|\sqrt{|E_{Fb} - E_F|} G(E_F) (1-R)}$$  \hspace{1cm} (6)

The net charge transferred from the defects to the Fermi gas is

$$Q = \frac{Q_D \exp \left[ (E_F - E_{FS})/kT \right] - Q_A}{1 + \exp \left[ (E_F - E_{FS})/kT \right]}$$  \hspace{1cm} (7)

$Q_D$ and $Q_A$ is the charge transferred for the defects which are stable for $E_F > E_{FS}$ and $E_F < E_{FS}$, respectively. The probability of recombination of a vacancy-interstitial pair $R$ depends on the mobility of the interstitials as well as on the charge located on the interstitial and the accompanying defect. At room temperatures the interstitials are mobile, they rapidly outdiffuse and do not participate in the defect reactions. The stabilization
of the Fermi energy is determined by the defect reactions 2a and 2b with $R \gg 0$. At low temperature interstitials are much less mobile. They stay longer close to the sites they originate from. Because for all values of $E_F$, both types of interstitials are positively charged donors. The probability of defect recombination depends on the charge state of the defect associated with a given interstitial. According to the defect reactions 1a and 1b $\text{As}_{\text{Ga}}^+ + \text{V}_{\text{As}}$ associated with $\text{Ga}_I$, as well as $\text{V}_{\text{As}}$ associated with $\text{As}_I$, are positively charged donors. They repel the interstitials and therefore $R \gg 0$. This situation assures fast pinning in p-type GaAs. On the contrary, in n-type material the acceptors $\text{V}_{\text{Ga}}$ and $\text{Ga}_{\text{As}}^+ + \text{V}_{\text{Ga}}$ are associated with $\text{Ga}_I$ and $\text{As}_I$, respectively. The negatively charged acceptors are attracted to the interstitial donors and recombine according to the reactions:

$$\text{Ga}_I + \text{V}_{\text{Ga}} \rightarrow \text{Ga}_{\text{Ga}} \quad (8a)$$

and

$$\text{As}_I + (\text{As}_{\text{Ga}}^+ + \text{V}_{\text{Ga}}) \rightarrow \text{As}_{\text{Ga}} + \text{Ga}_{\text{As}} \quad (8b)$$

The antistructure pair in the latter reaction is a donor with the energy level $(0/+)$ in the lower half of the bandgap. 16 Such a defect does not affect the charge balance in the n-type material. From this analysis we deduce that in n-type GaAs the defect recombination probability $R=1$ and no pinning for submonolayer coverage is expected. These qualitative considerations are in very good agreement with the data on Fermi energy at submonolayer metal coverages. It has been found that deposition of a metal on cleaved (110) GaAs surfaces at room temperatures leads to symmetrical pinning for n- and p-type material. 9 At liquid nitrogen temperature fast pinning in p-type GaAs is observed, whereas very slow pinning has been found on n-type GaAs. 10,11
Fermi energy as a function of metal concentration obtained by solving eq. (6) is shown in fig. 4. These model calculations were performed for the defects represented by reactions 1a and 2a. The most notable feature of this dependence is slow Fermi level pinning extending over two orders of magnitude of the metal coverage. Such behavior is a direct consequence of a strong dependence of the defect formation energy on the Fermi level position. As is seen in fig. 4 for a constant defect formation energy, $G(E_F) = \text{const}$, the pinning is much faster. This behavior is in obvious disagreement with existing experimental data which show a slow logarithmic dependence on the metal coverage.

Previously, an attempt has been made to explain this slow dependence by considering the effects of screening of the interface charges by metal clusters. Such a mechanism depends strongly on the cluster size. Large differences in the pinning rate should be observed for metals showing different clustering properties. Also, in its current form, such a model cannot account for the Fermi level pinning behavior at low temperatures. Here we show that the slow pinning is a straightforward manifestation of the dependence of defect formation energy on the Fermi level position. We argue therefore that it is a universal feature, independent of the deposited metal. The final pinning position is determined by the zero net-charge transfer from or to defects. Thus, as is seen in fig. 2, it depends on the relative contribution of "As" or "Ga" sublattice defects. In any case, it is limited to the energy range $E_v + 0.6 \text{ eV} \leq E_{FS} \leq E_v + 0.8 \text{ eV}$.

Low temperatures. As it has been discussed above, the major difference between room and low temperature behavior of the amphoteric native defects is an asymmetry in the defect recombination rates for n- and p-type material. In n-type $R=1$ and there is no pinning, whereas in p-type $R=0$ and fast pinning
is expected. Fig. 5 shows the calculated Fermi level pinning at low temperatures for different values of $<N>$ using eq. (6). Because of the presence of additional interstitial donors, the pinning is faster at low temperatures than at room temperatures in the case of p-type GaAs. This finding is again in general agreement with the experimental data.\textsuperscript{10,11} Although the diffusion of the interstitials is largely suppressed at low temperatures, one has to consider another factor which can affect the concentrations of Ga\textsubscript{I} and/or As\textsubscript{I}. Since at low temperatures the defect interstitial pairs are created on or in the close vicinity of the surface, the presence of a metal can, to some extent, affect out-diffusion of the interstitials which are stable in p-type material. In general, more electronegative metals such as Au would have a tendency to attract Ga\textsubscript{I}, whereas more electropositive metals, i.e. In, would react more readily with As\textsubscript{I}. Such interactions affect the abundance of different defects represented in figs. 2 and 3 and thus also the Fermi level stabilization energy. For Ga depleted interfaces, the Fermi level is controlled by the reaction 2a, and therefore it will be stabilized at $E_V + 0.6$ eV, whereas for As depleted interface Fermi level is stabilized in the energy range $E_V + 0.8$ eV to $E_V + 1.0$ eV (see fig. 2). The actual stabilization energy will depend in a complex way on the relative concentrations of the defects on the "Ga" and the "As" sublattice and on the extent of metal-induced Ga\textsubscript{I} and/or As\textsubscript{I} interstitials out-diffusion.

\section*{IV. Doping induced heterointerface intermixing}

We have shown that amphoteric properties of simple native defects well account for various characteristics of the Schottky barrier formations,\textsuperscript{12} as well as for compensation in mechanisms in irradiated semiconductors.\textsuperscript{14} In
both phenomena the defects generated at the interface or in the bulk compensate the original electrical activity of a semiconductor.

The fact that the defect formation energy depends on the Fermi level also has important consequences for other phenomena observed at elevated temperatures in semiconductors. At high temperatures the Fermi energy is close to \( E_{FS} \) in an intrinsic or lightly doped semiconductor. In this case the concentrations of acceptor and donor-like defects are very similar and therefore the defects recombine upon cooling so that a low concentration of electrically active native defects are present at room temperature. On the other hand, in heavily doped material the energy difference \( |E_F - E_{FS}| \) which measures the electronic part of the total defect energy can be large even at high temperatures. Thus the reactions (1) and (2) are driven towards high acceptor (donor) concentrations in n- (p-) type material. During the cooling of the crystal either acceptors or donors will dominate and one can expect strong compensation of donor or acceptor impurities. We propose that this one of the principal mechanisms limiting free hole or electron concentrations in semiconductors.\(^{21}\)

It is known that in GaAs it is impossible to obtain free electron concentrations higher than \( \sim 3 \) to \( 5 \times 10^{18} \) cm\(^{-3} \) using ion implantation techniques.\(^{22}\) On the other hand, doping levels exceeding \( 10^{20} \) cm\(^{-3} \) are easily accessible in p-type material.\(^{23}\) In InP the reverse behavior is observed. One can easily dope n-type InP to obtain free carrier concentrations\(^{24}\) up to \( 10^{20} \) cm\(^{-3} \), whereas p-type doping\(^{25}\) is limited to about \( 4 \times 10^{18} \) cm\(^{-3} \). This striking asymmetry has a simple explanation in terms of the Fermi level stabilization concept. As has been shown before, the formation energy of the compensating native defects is reduced by an energy
which is proportional to the energy separation $|E_F - E_{FS}|$. For a nondegenerate electron or hole gas $E_F \sim \ln(n)$, where $n$ is the carrier concentration. The equilibrium defect concentration is proportional to $\exp \left[ -E_{el}/kT \right]$, where the electronic part of the defect energy is given by $E_{el} = Q \cdot |E_F - E_{FS}|$. For the charge transfer $Q > 1$, $N_{def}(E_F)$ increases faster than $n(E_F)$ with increasing $E_F$. Thus, for $E_F$ satisfying the condition $N_{def} \cdot |Q| = n$, saturation of free carrier concentration is achieved. Further doping leads to generation of defects which compensate the impurities so that the free carrier concentration and the Fermi energy remain constant. The value of the Fermi energy at which such saturation occurs depends on the position of the Fermi level stabilization energy. In semiconductors with $E_{FS}$ located close to the conduction (valence) band, higher free electron (hole) saturation limits are expected. As is seen in fig. 6, for the case of n-type GaAs, the saturation of free electron concentration at $\sim 4 \times 10^{18} \text{cm}^{-3}$ corresponds to $E_F - E_{FS} = 0.5 \text{eV}$. For this position of the Fermi level, the formation energy of the triply ionized $V_{Ga}$ acceptor is reduced by 1.5 eV compared with the formation energy of this defect in intrinsic GaAs. The reduction in the formation energy will result in an increase of $V_{Ga}$ which, in this case, plays the role of the defect compensating intentionally-introduced impurities.

The doping induced, increased probability of native defect formation provides a straightforward explanation of another very extensively studied phenomenon, so-called doping enhanced heterointerface intermixing.\textsuperscript{26} It has been reported that high n-type doping induces very rapid intermixing of GaAs–AlAs superlattices.\textsuperscript{27,28} The intermixing does not depend on the donor species and is equally efficient for group IV as for group VI donors. The doping threshold for the intermixing\textsuperscript{29} is about $3 \times 10^{18} \text{cm}^{-3}$. This value
corresponds precisely to the maximum free electron concentration in GaAs and therefore also to the threshold for increased generation of $V_{Ga}$. The same considerations about defect generation can be expected for AlAs, although the threshold doping for the generation of $V_{Al}$ may be slightly different because of the different cohesive energy for AlAs. In any case, the increased generation of $V_{Ga}$ and $V_{Al}$ at high n-type doping leads to large defect concentration gradients, and therefore also to increased diffusion of Ga and Al at the heterointerface. Since the $V_{Ga}$ formation energy at a doping level of $3 \times 10^{18}$ is reduced by $\Delta E_{el} \approx 1.5 \text{ eV}$ compared with the intrinsic GaAs. Thus the equilibrium concentration of $V_{Ga}$ will be increased by the factor $e^{\Delta E_{el}/kT}$. At 1200 K this leads to an increase of the gallium vacancy concentration by more than 6 orders of magnitude. Similar mechanisms for the doping (or Fermi level) induced intermixing has been proposed before.\textsuperscript{28,31} The important novelty of our approach is that by introducing the concept of the Fermi level stabilization energy we can evaluate the electronic part of the defect formation energy for different types of doping in various semiconductors. In particular, we find that the small value of $E_F - E_{FS}$ for p-type GaAs indicates that this mechanism is not operational for hole concentrations lower than $10^{20}$ cm\textsuperscript{-3}. This conclusion is confirmed by the experimental data which indicate that there is no universal intermixing induced by p-type doping. It has been demonstrated that doping with Be to the level $4 \times 10^{19}$ cm\textsuperscript{-3} does not contribute to any enhancement of the intermixing.\textsuperscript{32} Extensively reported intermixing of GaAs/AlGaAs heterostructures induced by Zn diffusion finds a different explanation in terms of processes controlling uncommonly high diffusion of this acceptor in GaAs.\textsuperscript{30,33}
On the basis of our model we can predict trends in doping induced intermixing for other heterointerfaces. It is quite evident that in the case of InP/InGaAs heterostructures with $E_{FS}$ located close to the conduction band, acceptors are more efficient in causing the intermixing than donors. Using the same reasoning as in the case of GaAs, one can argue that for acceptor doping levels higher than the free hole saturation limit of $4 \times 10^{18} \text{ cm}^{-3}$ large concentrations of $V_p$ is generated. Those defects will facilitate interdiffusion of group V species. Because the free electron saturation level is high in InP, the intermixing of group III elements will not be efficient in this heterostructure.

In summary, we have shown that the recently introduced concept of amphoteric native defects well accounts for major features of Fermi level pinning at metal-semiconductor interfaces for submonolayer as well as for thick metal coverages. A slow dependence of the Fermi level pinning on the metal thickness at low coverages is a direct consequence of the Fermi level dependent defect formation energy, a unique feature of the amphoteric native defects. Also, an unusual behavior of the Fermi energy at low temperatures can be qualitatively understood in terms of the electrical and thermodynamic properties of native defects.

We also show that the Fermi energy pinning at (M/S) interfaces is not the only manifestation of the fundamentally important concept of the Fermi level stabilization energy. The stabilization energy provides the energy reference level to evaluate abundances of native defects in semiconductors. In particular, within this concept, the previously unexplained trends in maximum free carrier concentrations attainable in semiconductors and doping induced heterointerface intermixing find for the first time, a common and simple physical explanation.
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References

Figure captions

Fig. 1 Position of the Fermi level stabilization energy deduced from the Fermi energy position in a heavily irradiated semiconductor (●) and deduced from the Schottky barrier heights (○). Also the midgap or neutrality point energy7 (+) is also given. The semiconductor band lineups were adopted after ref. 15 with the GaAs affinity of 4.1 eV.

Fig. 2 Defect formation energies for simple vacancy-interstitial pairs and related defects. The numbers in the figure represent net charge transfers from the Fermi sea to the defects. The stabilization energies correspond to zero net charge transfer and are confined to the energy range $E_v + 0.6$ eV to $E_v + 1.0$ eV.

Fig. 3 The same as fig. 2 for $V_{Ga}$, $V_{As}$ and related $As_{Ga} + V_{As}$, $Ga_{As} + V_{Ga}$ defects.

Fig. 4 Room temperature Fermi level pinning in n- and p-type GaAs obtained from a model calculation for $V_{Ga}$, $As_{Ga} + V_{As}$ defect system with $<N> = 10$. The broken curves represent the pinning for defects with Fermi level independent defect formation energy.

Fig. 5 Fermi level pinning in p-type GaAs at low temperatures for different values of the parameter $<N>$. The limits of the stabilization energies for "As" and "Ga" sublattice defects indicating the range of the Fermi level pinning positions are shown.

Fig. 6 Concentration dependence of the Fermi level energy separation from the stabilization energy in n- and p-type GaAs and InP at high temperature of 1200 K typical for superlattice intermixing experiments.
Figure 1.
Figure 2.
Figure 3.
Figure 4.
Figure 5.

Concentration of metal atoms (cm$^{-2}$) vs Fermi energy (eV) for GaAs.

- $E_{FS}(As)$
- $E_{FS}(Ga)$

$\langle N \rangle = 14$, 12, 10

VBM and CBM energy levels are indicated.
Figure 6.

- GaAs
- InP

$T = 1200 \, \text{K}$