Pressure dependence of Cu, Ag, and Fe/n-GaAs Schottky barrier heights

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The Schottky barrier height of intimate Cu/GaAs (110), Ag/GaAs (110), and Fe/GaAs (100) has been measured as a function of hydrostatic pressure. The pressure dependence of the Cu and Ag/n-GaAs barrier heights of 97±4 meV/GPa fall within the uncertainty of the pressure dependence of the As_{Ga} defect and track the predicted value of As_{Ga}-rich interfaces. In contrast, the pressure dependence of the Fe/n-GaAs(100) Schottky barrier height of 109±7 meV/GPa does not fall within experimental error of these values and falls at nearly the predicted dependence of defect-free interfaces.

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I. INTRODUCTION

Control of the barrier height is critical to the successful operation of devices based on Schottky contacts, such as GaAs metal semiconductor field effect transistors (MESFET's). Unstable contacts result in barrier height changes, increased leakage current, and other undesirable effects that degrade the electrical performance of the device. However, despite years of extensive research and widespread use of Schottky contacts in device technology, the fundamental mechanism responsible for the formation of the Schottky barrier is still not fully understood. This work focuses on experimental measurements of the pressure coefficient of the Schottky barrier height to help elucidate the mechanism of Fermi level pinning in GaAs contacts.

According to Schottky's original work in 1938,[1] the barrier height is predicted to be the difference between the metal work function and the semiconductor electron affinity.[1] Experimentally determined barrier heights do not exhibit the metal work function dependence predicted by Schottky's theory.[2] In fact, Schottky contacts on many semiconductors, including GaAs and InP, form barrier heights that are relatively insensitive to the metal's work function. For example, almost all metal/n-GaAs contacts are found to have a Schottky barrier height in the range of 0.7–0.9 eV.[3–5]

Nine years after Schottky's paper was published, Bardeen proposed that the relative insensitivity of the metal work function to the Schottky barrier height is due to the presence of surface states in the semiconductor band gap.[6] This theory of Fermi level pinning is generally well accepted. However, the energy, density, and physical origin of the interfacial gap states proposed by Bardeen have not been firmly established.

There have been a large number of models proposed. We will concentrate on the two models that have gained the majority of recent support: the metal-induced gap state (MIGS) model[7–9] and the unified defect model (UDM).[10,11]

The first model is based on the intrinsic properties of the metal/semiconductor junction. Classically, the metal wave functions do not have sufficient energy to penetrate into the band gap at the semiconductor surface. However, the positional uncertainty of the metal's electrons and the electronic structure changes produced by local bonding allow them to penetrate a short distance (~5–10 Å) into the semiconductor.[7] The resulting charge extending into the semiconductor is the origin of the interfacial states, often referred to as metal-induced gap states. In 1984, Tersoff[9] suggested that the interfacial Fermi-level pinning position of the MIGS model can be directly ascribed to a universal charge neutrality level, which depends almost entirely on the host semiconductor band structure. In GaAs, the charge neutrality is predicted to fall near $E_v+0.5\,\mathrm{eV}$.[9] More recent *ab initio* calculations[12,13] have shown that the MIGS charge neutrality level is not a general property of the semiconductor and actually depends strongly on the choice of metal, contrary to the conjectures of Tersoff.

The second model proposes that semiconductor defects form near the interface during contact formation. The suggestion that the same native defect(s) form for all contacts has been used to explain the observed narrow range in Fermi-level pinning.[10,11] Spicer *et al.* and Weber *et al.* proposed the arsenic antisite (As_{Ga}) as the dominant defect responsible for pinning in metal/GaAs Schottky contacts.[14,15] This conjecture was based on the similar energies of the Fermi-level pinning position and the two As_{Ga} donor levels[14,15] and shifts in the Schottky barrier height resulting from annealing-induced changes in stoichiometry at the interface.[14] The As_{Ga} defect is known to decorate dislocations in GaAs.[16] Therefore, it is reasonable that the As_{Ga} defect may decorate the interface or free surface. Experimental evidence of As-rich regions on the surface of GaAs freshly cleaved in vacuum have been reported.[17] van Schilfgaarde and Newman demonstrated the role interfacial defects play in Fermi-level pinning using *ab initio* local density functional calculations of nonideal metal/GaAs (110) interfaces.[13] Choosing As_{Ga} as a representative defect, electronic structure calculations of the Au/GaAs (110) interface were repeated with a significant concentration of As_{Ga} defects within the first two monolayers of the interface. The results show that the Fermi level shifts from its pinning position for an ideal interface to that of the defect level. There-

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fore, defects dominate the interfacial Fermi-level pinning behavior when present at the interface in sufficient concentration.

Historically, both the defect and MIGS models have predicted pinning at similar positions in the gap.[9,11,14,15] Point defect concentrations and their respective energy levels at the interface have not been determined. Therefore, differentiation between models based on direct methods is not yet possible.

van Schilfgaarde and Newman have suggested that the pressure-induced change in the barrier height can be used to distinguish between the competing models of Schottky barrier formation. Electronic structure calculations within the local density functional approximation (LDA) were used to theoretically predict the pressure coefficient of the Schottky barrier height of ideal and defect-decorated Au/GaAs (110) and Pt/GaAs (110) interfaces.[18] The results show that $d\Phi_b/dP$ of the As_{Ga} rich interfaces exhibit distinctly different behavior than ideal defect-free interfaces. Recently, Phatak *et al.* were able to rule out MIGS's as the dominant mechanism in Fermi-level pinning for Au/n-GaAs (110) Schottky contacts based on experimental determinations of the pressure dependence of the barrier height.[19] Similar measurements on Al/GaAs (110) found that this interface tracked the predictions of ideal defect-free interfaces.[19] Bardi *et al.*[21] used theoretical calculations of $d\Phi_b/dP$ to show that the experimental results of Dobaczewski *et al.* on $Al/Ga_{1-x}Al_xAs(100)$ interfaces[20] are also consistent with those expected for ideal defect-free interfaces.[21]

The experimental measurements and theoretical calculations of $d\Phi_b/dP$ for Au/GaAs (110) provide the groundwork to analyze $d\Phi_b/dP$ of other metals on GaAs (110). The work reported here utilizes the pressure-induced change in the Schottky barrier height ($d\Phi_b/dP$) as a perturbation to examine the mechanism that determines the Schottky barrier height in Cu and Ag/GaAs(110) and Fe/GaAs (100) Schottky diodes.

II. EXPERIMENTAL PROCEDURES

A. Fabrication of atomically clean copper and silver diodes on (110)n-GaAs

To minimize interfacial contamination, diodes formed on (110) surfaces were fabricated on clean n-GaAs prepared by *in situ* cleavage and metal deposition in ultrahigh vacuum (UHV).

Ingots of n-GaAs grown using the vertical gradient freeze method were obtained from American X-tal Technology (AXT). The material was Si doped to a concentration of $5\times10^{16}\,\mathrm{cm}^{-3}$. The samples were sliced into $5\times5\times20\,\mathrm{mm}^3$ bars with the long axis along $\langle110\rangle$. Au-Ge ohmic contacts were evaporated on the sides of the GaAs bars and annealed at 450 °C for 10 min. The resistance of the Ohmic contacts was less than 10 Ω. The samples were then degreased in ultrasonic baths of acetone (10 min) and ethyl alcohol (10 min), rinsed in de-ionized water (5 min), blown dry with nitrogen, and immediately inserted into the UHV chamber. Following standard pumpdown and bakeout procedures, the UHV chamber attained a base pressure of less than 5×10^{-10} Torr. The metal evaporation sources were subsequently outgassed and the system was allowed to cool.

The GaAs bars were cleaved *in situ* to expose a clean (110) surface. Typically, 1000-Å-thick metal layers were thermally evaporated through a shadow mask to form ~500-μm-diam Schottky diodes. To minimize contamination of the metal/semiconductor interface, average pressures during the first 100 Å of deposition were maintained below 10^{-9} Torr. Transmission electron microscopic (TEM) analysis indicates that these metal films deposited on cleaved GaAs (110) at room temperature are polycrystalline with grain sizes typically in the range of ~0.1–1.0 μm.

B. Fabrication of atomically clean iron diodes on (100)n-GaAs

Samples were grown in a multichamber molecular beam epitaxy (MBE) facility on n^+-GaAs(001) wafers. They consisted of an n^+-doped buffer layer followed by a 1000–1500 Å undoped GaAs spacer layer. The growth was terminated using a flux sequence to produce a well-ordered 2×4 As-dimer terminated surface reconstruction.[23] The samples was then transferred in UHV to a second MBE chamber, where Fe was deposited from a Knudsen-cell-type source at a growth rate of 3 Å/min and a substrate temperature of 175 °C resulting in single-crystal growth of a continuous 50-Å α-Fe(001) film whose crystallographic axes aligned with those of the substrate.[23] After cooling to slightly below room temperature, a 50-Å gold film was deposited to prevent oxidation of the Fe surface. For a more complete description of the experimental procedures, see Ref. 23.

C. Barrier height measurements as a function of hydrostatic pressure

After removal from the vacuum chamber, the GaAs bars were cut into 2.5 mm×2.5 mm×1 mm samples to fit into a pressure cell manufactured by Unipress (model LOC 10). The pressure cell is constructed of a Cu-Be alloy and is designed to work to a maximum pressure of 1.0 GPa at room temperature. The sample was pressurized in a 1:1 mixture of petroleum ether and kerosene using a hydraulic press.

Electrical connection to the Schottky and Ohmic contacts were made by bonding 100-μm Cu wire to the contacts with Epo-tek brand H2OE silver epoxy. The epoxy was cured at 90 °C for 2 h. Electrical measurements of current as a function of voltage (I-V) were taken using a computer-controlled Hewlett-Packard 4140 voltage source and picoammeter.

Barrier heights were determined by fitting the I-V electrical characteristics to the standard equation of thermionic emission theory:

$$I=I_0[\exp(qV/nkT)-1], \tag{1}$$

where $I_0=SA^*T^2\exp(-q\Phi_b/kT)$, q is electronic charge, k is Boltzmann's constant, T is absolute temperature, n is the ideality factor, S is the diode area, A^* is the effective Richardson constant, and Φ_b is the Schottky barrier height. To obtain reliable values of the Schottky barrier height using thermionic emission theory, n must be less than 1.1 over a

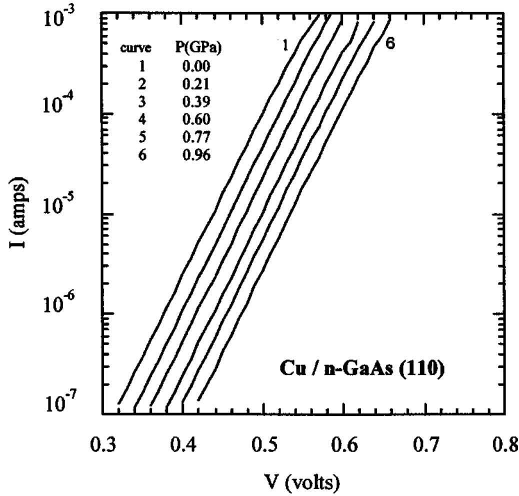

FIG. 1. Forward-biased current-voltage characteristics as a function of hydrostatic pressure for a Cu/n-GaAs(110) diode.

significant data range. The effective Richardson constant was modified to account for the pressure dependence of the effective mass using

$$\frac{m_n^\Gamma}{m_0}=0.067\frac{(1+7.4\times10^{-3}P\ \mathrm{kbar}^{-1})}{(1-3.9\times10^{-15}n^{2/3}\ \mathrm{cm}^2)},\qquad(2)$$

where m_0^Γ is the effective mass of the electron in the gamma valley, m_0 is the free electron mass, P is pressure, and n is the free electron concentration.[22]

The pressure within the cell was determined using a heavily doped n-type InSb single-crystal sensor which was manufactured and calibrated by Unipress. The measurement of the ratio of the resistance of the sensor at elevated pressure to that at 1 atm is used to infer the pressure, according to the following calibration curve:

$$\frac{R(P)}{R(0)}=0.9994+0.3518P+0.05837P^2,\qquad(3)$$

where R is the resistance of the gauge and P is the pressure in GPa. The manufacturer specified maximum error in the pressure determination to be ±0.01 GPa. The resistance of the InSb pressure gauge is highly sensitive to temperature. In order to minimize errors in the pressure determination, the temperature within the pressure cell was allowed to equilibrate with the ambient temperature. The stabilization of the temperature in the pressure cell was confirmed using a copper-Constantan thermocouple mounted on the pressure cell sensor head.

III. RESULTS

Figure 1 shows representative forward-biased current-voltage characteristics of a Cu/n-GaAs(110) Schottky diode as a function of hydrostatic pressure. The curves for this measurement, as well as for measurements on Ag/n-GaAs(110) and Fe/n-GaAs(100) diodes, exhibit exponential behavior over 4+ orders of magnitude with a corresponding ideality factor of 1.04–1.05. From the I-V measurements, the barrier height determined at ambient pressure is 0.89 eV for copper, 0.90 eV for silver, and 0.91–0.92 eV for iron. These values are consistent with earlier measurements of similarly prepared n-GaAs diodes.[3,23] It is interesting to note that the barrier height of Fe/GaAs diodes can differ significantly from this value when deposited on different surface orientations and surface stoichiometries or using different deposition conditions (e.g., temperatures, rates, etc.). This topic is discussed more extensively in Ref. 23 and references therein.

Figure 2 illustrates the corresponding change in the Schottky barrier height as a function of hydrostatic pressure as determined from I-V data (e.g., Fig. 1). A least-squares fit of the barrier heights as a function of pressure indicates that the Cu and Ag/GaAs(110) systems have a pressure coefficient ($d\Phi_B/dP$) of 97 ± 4 meV/Gpa, while Fe/GaAs(100) has $d\Phi_B/dP$ of 109 ± 7 meV/Gpa. The reported error bounds in barrier heights are estimated from the range in which the experimental data could be accurately fit with an absolute error bound of 0.1 kbar in pressure and a relative error bound of 5 meV in barrier height. This is the same procedure described in our earlier work.[19] The larger error bound reported for the $d\Phi_B/dP$ of Fe/GaAs(100) diodes arises primarily from the smaller range in pressure used than for other diodes in this study.

IV. DISCUSSION

A. Mechanism of Fermi-level pinning

The experimentally measured $d\Phi_B/dP$ of Cu and Ag on UHV cleaved $(110)n$-GaAs fall within the uncertainty of the experimentally measured pressure dependence of the As_{Ga} defect,[24–26] the experimentally measured $d\Phi_B/dP$ of Au on UHV cleaved $(110)n$-GaAs,[19] and the theoretically calculated $d\Phi_B/dP$ of both Au and Pt/GaAs (110) interfaces decorated with As_{Ga} defects.[18] The experimental uncertainty of $d\Phi_B/dP$ of Cu and Ag/n-GaAs(110) lies outside the theoretically calculated $d\Phi_B/dP$ of ideal defect-free Au and Pt/GaAs(110) interfaces.

Although electronic structure calculations of $d\Phi_B/dP$ of Cu and Ag/GaAs (110) have not been performed to date, we expect the generalizations proposed by van Schilfgaarde and Newman regarding the $d\Phi_B/dP$ of ideal and defect decorated metal/GaAs (110) interfaces to be valid for the Cu and Ag/GaAs (110) interfaces. Based on the similarity in values of $d\Phi_B/dP$ for Au and Pt/ GaAs (110), van Schilfgaarde and Newman had suggested that MIGS's are bonding in nature, and their energy position in the band gap will, in general, follow similar trends when exposed to hydrostatic pressure. Therefore, differences in the pressure dependence between ideal and defective interfaces are expected to be relatively independent of the choice of metal and will depend primarily on the presence (or absence) of defects, their energetic position within the band gap, and their concentration.

The correspondence of $d\Phi_B/dP$ for Cu and Ag/n-GaAs(110) with the theoretical calculations of $d\Phi_B/dP$ for As_{Ga}-rich metal/GaAs (110) interfaces indicates that the As_{Ga} defect is a strong candidate for Fermi-level

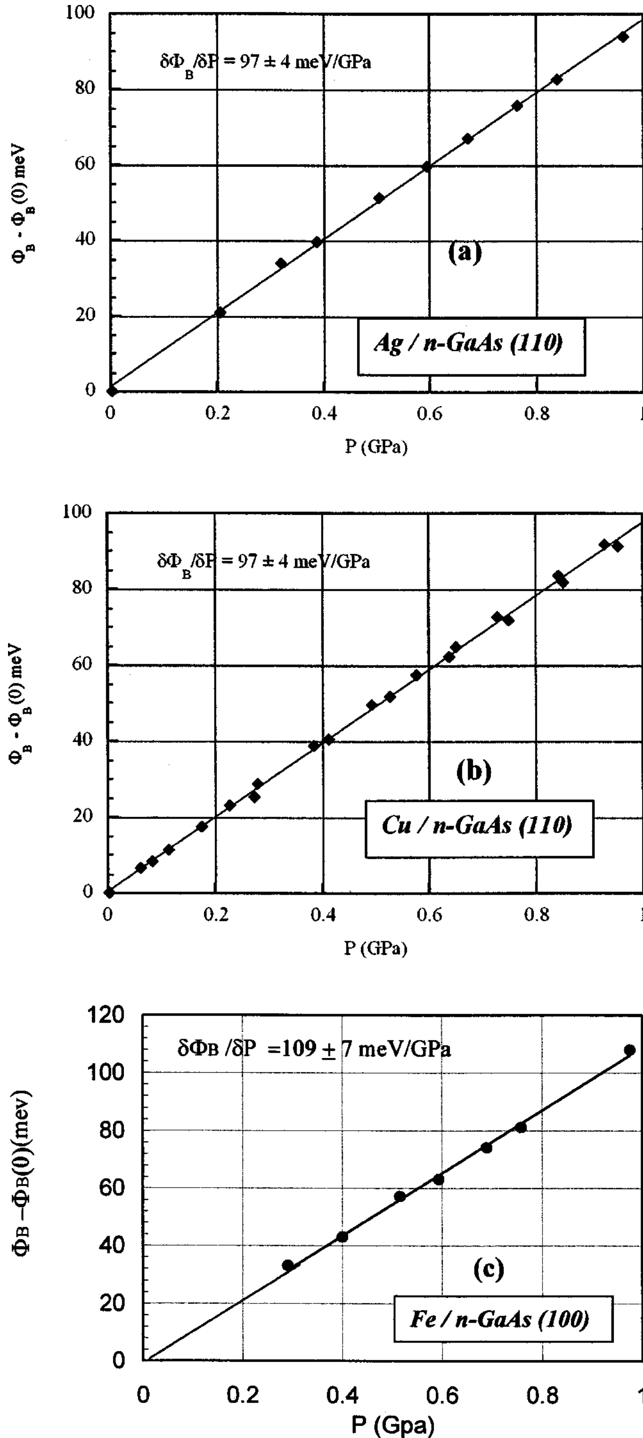

FIG. 2. Change in the Schottky barrier height determined by thermionic emission theory as a function of hydrostatic pressure for (a) Ag/n-GaAs(110), (b) Cu/n-GaAs(110), and (c) Fe/n-GaAs(100).

pinning. A comparison of previously measured hydrostatic pressure dependences of defects in GaAs in Table I (Ref. 27) allow us to rule out a number of other defects. Of the defects listed in Table I, only the As_{Ga} defect possesses both an energy level that corresponds to the Fermi-level pinning position in GaAs and a hydrostatic pressure dependence that falls within experimental error of $d\Phi_B/dP$ for Cu and Ag.

TABLE I. The energy level and its pressure coefficient for point defects in GaAs. The label for each defect corresponds to conventional notation in the literature.

Label	Energy (eV)	Pressure coefficient (meV/GPa)	Reference
As_{Ga}	$E_c-0.75$	93 ± 5	17, 18
	$E_v+0.52$		
Ga_{As}	$E_v+0.077$	119 ± 4	22
$E1$	$E_c-0.08$	0	23
$E2$	$E_c-0.14$	96 ± 10	
$E3$	$E_c-0.31$	110 ± 11	
$E4$	$E_c-0.71$	116 ± 12	
$E5$	$E_c-0.9$	116 ± 12	
$H1$	$E_v+0.29$	0	
$H0$	$E_v+0.10$	0	
$E2$	$E_c-0.14$	88	24
$E3$	$E_c-0.31$	135	
$E4$	$E_c-0.59$	105	
Cu	$E_v+0.015$	104 ± 2	25
Ag	$E_v+0.24$	102 ± 2	26

As mentioned earlier, Table I indicates that the energy of impurity levels associated with Cu and Ag in GaAs does not fall close to the measured Cu and Ag/n-GaAs(110) barrier heights. This led us to conclude that these defects do not dominate Fermi-level pinning. We do, however, note that the pressure dependences of the Cu and Ag-related defect levels are similar to that of their respective Schottky barrier heights. The significance of this observation is unclear. Whether these defects are present at the interface has not been firmly established and, if so, what role they might play in Fermi-level pinning. Within the context of the As_{Ga} defect model, an acceptor level is required to pin the Fermi level at the As_{Ga} defect donor levels for n-GaAs.[14] The Ga_{As} defect was originally proposed as the spectator acceptor.[14] Instead, the impurity levels of Cu and Ag in GaAs may be playing this acceptor role. Another alternative suggested by the calculations of van Schilfgaarde and Newman is that the MIGS's, which are amphoteric in nature, could also be performing this function.[13] There have been other models based on the movement of the metal into the semiconductor to form changes in the extrinsic and intrinsic levels at the interface,[28] although they remain highly controversial.

The fact that the uncertainty of $d\Phi_B/dP$ of Cu and Ag lies outside the theoretically calculated $d\Phi_B/dP$ of ideal metal/GaAs (110) interfaces suggests that MIGS's do not play a dominant role in Fermi-level pinning for these contacts. However, in order to fully justify these conclusions, theoretical calculations of the ideal and As_{Ga}-rich interfaces for Cu and Ag/GaAs (110) are required.

The pressure dependence of the Fe/n-GaAs(100) Schottky barrier height of 109 ± 7 meV/GPa falls at nearly the predicted dependence of ideal defect-free interfaces and is outside experimental error for that predicted for

As_{Ga}-decorated interfaces. From this observation alone, we cannot rule out the influence of defects that have similar pressure dependences to those of MIGS's. Jonker *et al.* also concluded that this interface was not pinned by large midgap point-defect concentrations, as inferred from the measured long lifetimes and small interfacial midgap defect densities obtained from photoreflectance spectroscopy measurements on similarly prepared structures.[23] Al/GaAs(100) (Ref. 21) and Al/GaAs(110) (Ref. 19) interfaces also exhibit nearly identical values (105 and 107 meV/GPa, respectively). Both Al and Fe atoms have a propensity to bond exclusively with As. When initially deposited on GaAs surfaces, they are involved in an exchange reaction in which the metal replaces Ga in the lattice and free Ga is released.[23,29] The reaction process and resulting surface stoichiometry are not expected to favor As_{Ga} formation. The observation that the Fe/GaAs pressure dependence tracks that of an interface that is not decorated by As_{Ga} defects, as is the case for Al/GaAs(110) (Ref. 19) and $Al/Ga_{1-x}Al_xAs(100)$ (Ref. 21) is extremely strong evidence that, under these circumstances, Fermi-level pinning by the As_{Ga} defect can be suppressed.

B. Comparison of the interfacial chemistry of noble metals

The noble metals Au, Cu, and Ag show similar barrier heights (0.89–0.92 eV) and pressure dependences (97 meV/GPa), even though the chemistry of these interfaces is known to vary significantly. Photoemission studies of the initial stages of Schottky barrier formation have shown that Au/GaAs (110) and Cu/GaAs (110) both have extended interface regions with Ga and As outdiffusing and metal diffusing into GaAs.[30,31] On the other hand, studies of Ag/GaAs (110) reveal an interface characterized by Ag island growth with little intermixing between the film and substrate.[32] The studies conclude that the intermixing between the metal and GaAs was highest for Cu, followed by Au, and the least intermixing for Ag.

Studies of the near interfacial stoichiometry of the Au/GaAs (110) by energy dispersive x-ray spectroscopy[33] (EDX) and surface analysis by laser ionization[34] (SALI) reveal the presence of excess As at Au/GaAs (110) interfaces. A possible manifestation of excess As is an As_{Ga} defect, although other As-dominated defects are possible, such as As interstitials, complexes thereof, or As clusters. However, the results of the interface studies and the correlation of Φ_B and $d\Phi_B/dP$ for Au/GaAs (110) with the properties of the As_{Ga} defect indicate that the As_{Ga} defect is the most likely manifestation of excess As.

In the most extensive study of the Cu/GaAs (110) interface by Joyce and Weaver,[31] the authors concluded that the interface region consists of a solid solution of Ga in Cu with As as the dominant interfacial species. The conclusions were based on the results of high-resolution synchrotron photoelectron spectroscopy and thermodynamic parameters, in conjunction to the comparison of surface studies of Cu/GaAs (110) to the surface studies of other metal/GaAs (110), especially Au/GaAs (110).

Silver shows the least interaction of the noble metals with GaAs. However, Φ_B and $d\Phi_B/dP$ are the same for all three noble metals. Therefore, it appears that the Fermi-level pinning mechanism is the same for Cu, Ag, and Au. It has been proposed that the heat released due to the Ag-Ag clustering is responsible for the formation of defects that pin the Fermi level.[32] Observation of Ag cluster formation and the delay in the Fermi-level pinning position for Ag relative to the other noble metals qualitatively supports this conjecture.[32]

V. CONCLUSIONS

The pressure dependence of the Cu, Ag, and Au/n-GaAs(110) barrier heights falls within the uncertainty of the pressure dependence of the As_{Ga} defect and tracks the predicted value of As_{Ga}-rich interfaces. This is strong evidence that these interfaces are decorated with large As_{Ga} defect concentrations. In contrast, the measured pressure dependences of Al/n-GaAs(110),[19] Al/n-GaAs(100),[20] and Fe/n-GaAs(100) Schottky barrier heights are not within experimental error of these values and fall at nearly the predicted dependence of defect-free interfaces. This is strong evidence that metals which react selectively with As and encounter an exchange reaction during Schottky barrier formation (such as Al and Fe) can suppress As_{Ga} formation and the corresponding Fermi-level pinning by interfacial defects.

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