In situ conductance characterization of Fe/Ag multilayer contacts on GaAs

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Electrical transport characteristics for epitaxially grown Ag/Fe/Ag multilayers on GaAs(100) and GaAs(110) have been studied under various growth conditions. The surfaces and structure of the multilayer were characterized by low energy electron diffraction and angle-resolved Auger electron spectroscopy/diffraction at all stages of the growth. *In situ* conductance spectroscopy performed between growth steps indicates a strong dependence of the transport characteristics on the morphology at the metal/semiconductor interface. A nanoclustered silver overlayer exhibits a reverse leakage current due to defect states in the discontinuous film; however, annealed continuous Ag overlayers on GaAs behave as a uniform rectifying diode. After completion of the magnetic multilayer growth, current–voltage characteristics indicate a sharp turn on in the reverse current at ~0.6 V with nonlinear behavior. This is characteristic of electron tunneling through the Schottky barrier from the multilayer contact into the semiconductor. The implementation of these magnetic multilayer contacts for electrical spin injection is discussed. [DOI: 10.1063/1.1577398]

I. INTRODUCTION

An active area of research in the pursuit of spin-based electronic devices aims to achieve room-temperature electrical spin injection from a ferromagnetic metallic contact into a semiconductor-based device.[1,2] There are, however, several aspects intrinsic to these hybrid material systems that present interesting challenges. These include morphological issues (reactive epitaxy, interfacial roughness) and electronic mismatch issues (conductivity, band structure), problems inherent to the metal/semiconductor interface. Despite these obstacles, promising results have been reported recently. In addressing the conductivity mismatch problem, it has been shown, both theoretically[3] and experimentally,[4] that injection by way of tunneling may be an important process for preserving spin polarization across the metal/semiconductor interface. As these problems continue to be solved, injection efficiencies will continue to improve.

In this article, we report on studies aimed at addressing the above-mentioned problems, by using silver buffer layers to prevent the reactive epitaxy associated with the Fe/GaAs system, to improve the morphological roughness using ultrasmooth films, and to overcome electronic mismatch by Schottky tunneling from quantum-confined Ag subbands into the GaAs conduction bands. The use of silver buffer layers to reduce the reactive epitaxy between Fe and GaAs is not a new idea;[5] however, in this work, the silver films were grown in a unique two-step process whereby atomically flat overlayers are obtained through a collective annealing action.[6]

This material system was chosen for investigation due to the lattice commensurability between Fe, Ag, and GaAs, in addition to the well-developed technology of III–V semiconductor heterostructures. Even though metal/semiconductor systems may be lattice matched, the dominant growth mode is a three-dimensional, clustered morphology, due for the most part to the differences in bonding and surface diffusivities for metals as opposed to semiconductors. For this reason, it may seem that predeposition of silver overlayers on GaAs would not improve the morphological roughness, which is indeed the case for room-temperature growth. However, it has been shown that atomically flat silver overlayers on various III–V substrates can be achieved through a two-step growth process.[6,7] It is this approach that has been taken in this study. Low-temperature deposition of Ag on GaAs results in the formation of a nanoclustered film; however, when followed by a gentle annealing at moderate temperatures, the film redistributes to form atomically flat epitaxial overlayers. This is then the template on which the Fe is grown, followed by a cap layer of Ag. Each film is deposited at low temperature and annealed at moderate temperatures, thereby forming a high-quality epitaxial magnetic multilayer on GaAs substrates.

II. EXPERIMENTAL DETAILS

In this study, *n*-doped (Si: $n \approx 5\times10^{24}$ m^{-3}) GaAs single crystal wafers with (100) and (110) surface orientations were used to investigate the growth and conductance characteristics under various conditions of deposition temperature and coverage. Because the metal–semiconductor interface is a crucial component in maintaining spin-polarized transport, it is imperative to obtain clean and well-ordered surfaces on the substrate. Auger electron spectroscopy (AES), low energy electron diffraction (LEED), and Auger electron diffraction (AED) were used to determine the cleanliness and crystallographic order of the substrate and of each film of the multilayer.

The GaAs wafers were cleaved into 5 mm × 5 mm samples and introduced into the ultra-high-vacuum (UHV) chamber (1×10^{-8} Pa base pressure) after a brief acetone rinse. Before the substrates were cleaned, AES indicated the presence of large quantities of carbon and oxygen contami-

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nants. Simply annealing the GaAs wafers at 850 K desorbs most of the oxygen, but it does not remove the majority of the carbon. In order to obtain clean GaAs surfaces with contaminant levels below detection by AES, two methods were employed: ion sputtering (Ne^+, 500 V, 10 mA) with the sample maintained at 850 K, and reactive desorption using an atomic hydrogen source. Both methods work well to clean the surface, but cleaning by atomic hydrogen is superior, requiring lower substrate temperatures (~525–575 K) and less cleaning time (~20 min), and producing surfaces with much larger terraces.[8] The GaAs(100) surfaces were characterized by the Ga-rich (4×2) reconstruction. Because a (1×1) LEED pattern was observed, the GaAs(110) surfaces were assumed to have the usual relaxed surface.

The initial silver buffer layers were deposited with the substrate maintained at 110 K. The silver was evaporated from a resistively heated tungsten crucible while the background pressure remained below 6×10^{-8} Pa. Silver films of varying thicknesses between 1.5 and 3 nm were investigated. Film thickness was monitored using quartz crystal monitors and cross-checked using relative Auger electron intensities. The thickness reported here is estimated to be accurate to within 10%. As expected, the unannealed silver films exhibited a diffuse LEED pattern, indicative of the disordered, nanoclustered morphology. These films were then annealed at 300 K for 1–15 h and subsequently cooled to 110 K. Iron films, 0.7–10 nm thick, were then deposited by means of an electron-beam evaporator. These films were again annealed at 300 K for 0.5 h in UHV, providing films with increased order while minimizing the intermixing of the two metal films. Finally, a 5 nm silver capping layer was deposited at 110 K, completing the sandwich structure. This final film of silver is also grown using the two-step process because of the desired formation of atomically flat films of Ag on Fe.[9] A schematic illustration of the entire multilayer is shown in Fig. 1(a).

During each step in the growth process, conductance was measured *in situ* with an indium-coated tungsten spring making electrical contact to the sample. The *in situ* measurements employed a two-probe technique, where the differential conductance and its derivative were both measured using an ac lock-in technique. Samples were also characterized with an *ex situ* four-point conductance probe.

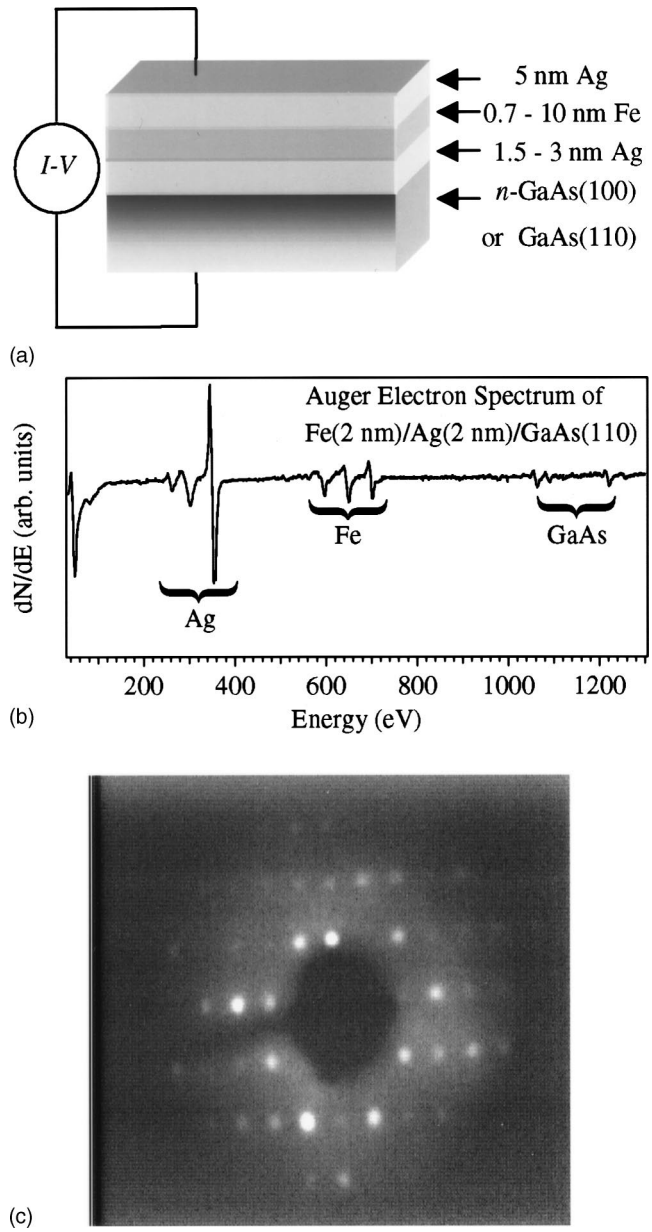

FIG. 1. (a) Schematic illustration of the entire multilayer system. GaAs(100) and (110) substrates were used. The Ag buffer layers (between 1.5 and 3 nm) were grown at low temperature (110 K) followed by a gentle anneal at 300 K [375 K for the (100) substrates]. The Fe films were also deposited at 110 K and annealed at 300 K. (b) AES and (c) LEED data (130 eV) for the Fe/Ag/GaAs(110) heteroepitaxial system qualitatively indicate clean and well-ordered growth.

III. RESULTS AND DISCUSSION

From the data in Figs. 1(b), and 1(c), it can be seen that clean, well-ordered Fe/Ag multilayers were grown on GaAs. In addition to the surface-structural data, low coercivities and a strong in-plane crystalline anisotropy measured by superconducting quantum interference device (SQUID) magnetometry indicate that the iron films have a high degree of crystallinity. Obtaining this high degree of order in these films is an important step in the formation of spin-injecting contacts because spin scattering from defects at the interfaces is most likely the dominant location of depolarization. Using epitaxial Ag buffer layers, disordered compounds at the interface, such as Fe–As complexes,[10] are avoided. Moreover, disordered and reacted species in the interfacial region are most likely the cause for the common observation of Ohmic-like I–V characteristics in Fe/GaAs grown at room temperature, seen in this study and by others,[11] meaning that for voltages around zero bias the current–voltage curves are virtually linear.

In contrast to the linear conductance characteristics for interfaces where reacted layers are present, Ag/GaAs diodes exhibit a large rectification behavior. Figure 2(a) shows typical I–V characteristics for 2 nm of Ag deposited and measured at 300 K. This diode behaves as a typical Schottky diode where, as the voltage is increased in the forward direction, the potential difference across the interface is decreased

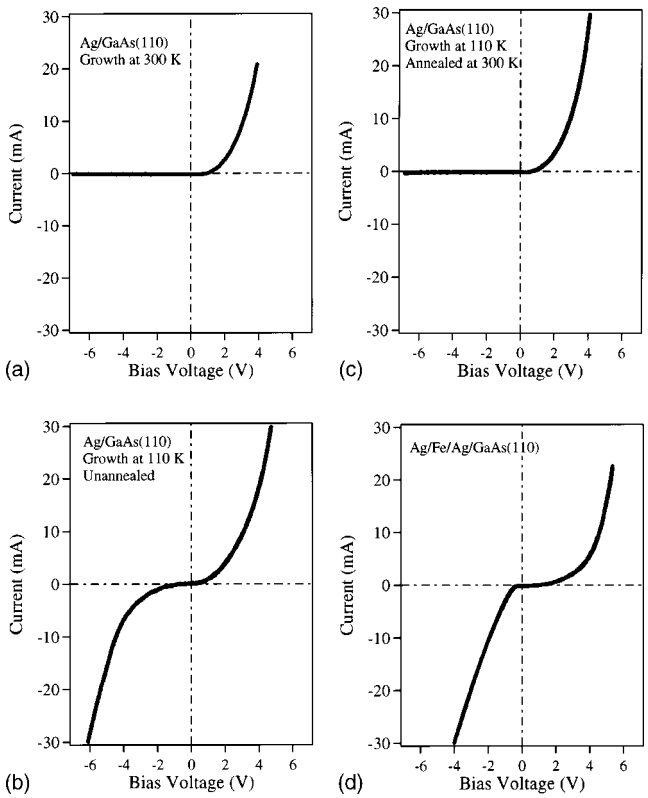

FIG. 2. *I–V* characteristics for (a) 2 nm of Ag/GaAs(110) grown and measured at 300 K, (b) 2 nm of Ag/GaAs(110) grown and measured at 110 K, (c) the same film as in (b) after annealing to 300 K, measured at 110 K, and (d) the entire multilayer system [same sample as in (b) and (c)]. The Fe film is 2 nm thick and the Ag cap layer is 5 nm thick. This sequence demonstrates the dependence of Ag/GaAs breakdown and rectification on morphology and the tunnel phenomenon occurring as a result of the entire multilayer structure.

and electrons are subsequently allowed to flow from the semiconductor into the metal. This is the case for electron *ejection*. Under reverse bias, the potential difference across the interface is increased, thereby making it harder for electrons to flow from the metal into the semiconductor. Hence, very little current flows under reverse bias for a uniform Schottky diode. This is a fundamental problem associated with electrical *injection* across Schottky barriers, notwithstanding a requirement to maintain the spin polarization. For this reason, GaAs substrates with high dopant levels have been used in order to enhance electron *tunneling* across the barrier via a thin depletion region. Moreover, as mentioned above, tunnel contacts may preserve spin orientation and ameliorate the problem of conductivity mismatch associated with metals and semiconductors.[3] In any case, Ag/GaAs diodes, grown at room temperature, show very little reverse current. This is true for both the (100)- and (110)-oriented crystals. Numerical fits to these data indicate that the ideality factors for these samples are quite large ($\sim$10); this is expected due to possible surface leakage from such large, unprocessed diodes and high dopant levels.

Figures 2(b)–2(d) show a sequence of *I–V* curves measured *in situ* for (b) 2 nm of Ag on GaAs(110) deposited and measured at 110 K, (c) the same film then annealed at 300 K and measured at 110 K, and (d) the entire Ag/Fe/Ag multilayer diode grown as described above and measured at 110 K. In Fig. 2(b), a dramatic difference is seen in the reverse-bias condition as compared to that for growth at 300 K [Fig. 2(a)] and compared to that for the same film annealed at 300 K [Fig. 2(c)]. These differences in reverse current are explained by the fact that, in the case of low-temperature deposition, due to the reduced diffusion coefficient of Ag atoms at low temperature, the morphology is that of discontinuous nanoclusters. The resultant electronic structure is characterized by defect states, which therefore give rise to breakdown at such low reverse voltages. When the film is annealed at 300 K, the Ag layers redistribute, becoming continuous across most of the GaAs surface. With a more uniform interface, the reverse bias breakdown does not occur [Fig. 2(c)].

This strong dependence of the conductance characteristics on the morphology is also seen to occur on the GaAs(100) surfaces. However, in order to obtain completely rectifying behavior, Ag/GaAs(100) samples must be annealed at 375 K. This is correlated with a smoothing of the Ag film, and is corroborated by the structure appearing in the LEED and AED images. This higher temperature for the (100) surface indicates a higher activation barrier for smoothing the Ag film. This is a direct result of the (4×2) reconstruction as opposed to the (110) unreconstructed and nonpolar surface. In addition, relative AES intensities of the Ga *LMM* and As *LMM* to the Ag *MNN* peaks have been used to determine the temperature at which the Ag film becomes more crystalline. The Ga and As peaks become relatively stronger by reduced inelastic scattering through the Ag film of higher crystalline order. Indeed, Ga and As peaks would also increase if they were floating to the top layer, but this is ruled out because Ag/GaAs is a well-known nonintermixing system[12] and the temperatures used here are very low. In addition, coverages used here are too high for this to be the result of exposing patches of bare GaAs.

In Fig. 2(d) the *I–V* curve for the entire Ag/Fe/Ag/GaAs(110) sample is shown. For this sample, the thickness of the Fe film is 2 nm and that of the Ag cap layer is 5 nm. This characteristic is the same for all films grown on the GaAs surface with smoothed Ag buffer layers. The reverse current is derived from a phenomenon completely different from that in Fig. 2(b). In Fig. 2(d) there is a sharp turn on in the reverse current at $\sim$0.6 V and the *I–V* character is nonlinear. This is indicative of electron tunneling through the Schottky barrier.[13] The origin of this behavior is not fully understood, but it is believed that quantum confinement of the Ag buffer layer gives rise to electronic quantum-well states. These states must have appropriate energy and momentum overlap with bands in the GaAs, giving rise to this substantial reverse tunnel current. Indeed, quantum-well states are known to exist in Ag layers on GaAs grown in this way.[14,15] However, this conductance characteristic does not occur until the Fe film is deposited, indicating that the necessary electronic states are a result of the collective multilayer system. A substantial difference between films grown on substrates with different symmetries [i.e., (100) vs (110)] is not observed to within the resolution of our mea-

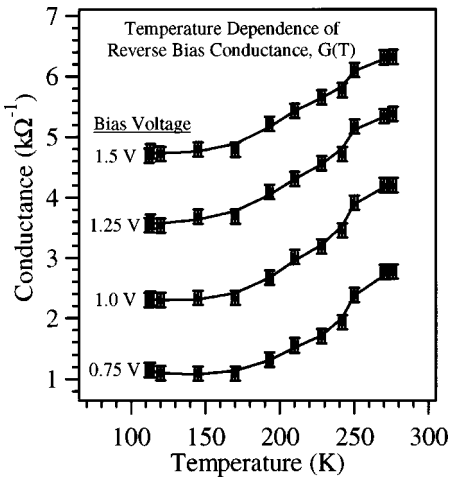

FIG. 3. Temperature dependence of the reverse-bias conductance for various applied voltages. The slight increase in conductance ($G=I/V$) with increasing temperature indicates an insulator-like behavior, indicative of Schottky tunneling. The solid lines are displayed as a guide to the eye.

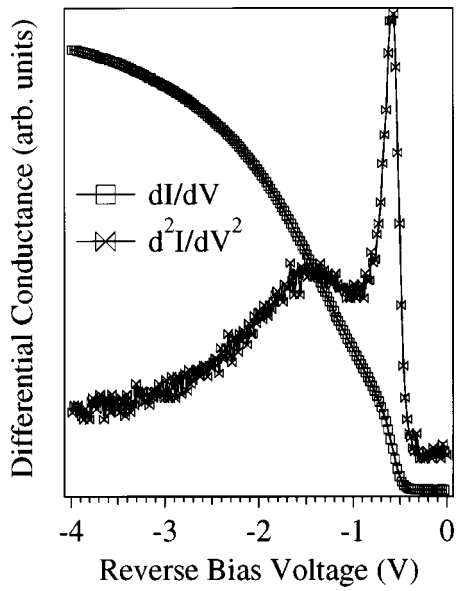

FIG. 4. Differential conductance, dI/dV, and its derivative shown for the reverse-bias condition [as in Fig. 2(d)], in arbitrary units, with an offset added for clarity. These data highlight the nonlinear behavior of this reverse tunnel current. The features seen at -0.6 and -1.36 V are coincident with previously observed quantum-well states in the Ag/GaAs (Ref. 13) and Ag/Fe (Ref. 16) systems.

surements. The turn-on voltage shifts slightly, however, the I–V characteristics are qualitatively similar.

A useful test to determine whether electron tunneling is indeed the dominant conductance mechanism is to investigate its dependence on temperature. Conduction by way of tunneling behaves with a weak insulator-like temperature dependence.[16] That is, as the temperature is decreased the conductance slightly decreases (increases the resistance). Figure 3 plots the conductance ($G=I/V$) as a function of temperature for the sample shown in Fig. 2(d). Each curve represents data for different reverse-bias conditions. Because there is only a slight increase in conductance with increasing temperature, and this behavior does not depend strongly on the applied bias after the turn-on voltage, this suggests that this transport mechanism is dominated by Schottky tunneling.

In order to investigate this reverse tunnel current in more detail, differential conductance was measured using a lock-in technique where curves for both dI/dV and d^2I/dV^2 were obtained. These data are shown in Fig. 4. The reverse-bias differential conductance and its derivative enhance the structure seen in this nonlinear behavior. This structure arises through the convoluted density of states as the voltage is swept through the spectrum. From these data, two features are observed just after the onset of current flow at -0.6 and at -1.36 V. These energies are consistent with both types of quantum-well states that are possibly playing a role in this system: those seen in Ag/GaAs (Ref. 14) and spin-polarized interface states found in the Ag/Fe system.[17] In the latter, magnetic interface states are derived from a Fe surface resonance of *s*, *p*, and *d* character hybridized with the Ag *s*–*p* band of overlapping energy. Based on this, it is highly probable that this reverse current is spin polarized on the metal side of the interface, and what needs to be determined is whether that polarization is maintained across the barrier. It is difficult to interpret the exact origin of the reverse tunnel current, but it is clear that this system provides unique conductance characteristics with rich underlying physics.

IV. SUMMARY

In conclusion, multilayer films of Ag/Fe/Ag epitaxially grown on GaAs(100)-(4×2) and GaAs(110) were studied under various growth conditions. The surface-sensitive techniques of AES, LEED, and AED were used to determine cleanliness, crystallographic order, and optimal growth conditions for this system, resulting in epitaxial, magnetic multilayer contacts of high quality. *In situ* conductance data show (a) the dependence of breakdown and rectification on morphological conditions, and (b) that the metallic multilayer as a whole participates in the manifestation of a reverse tunnel current. The reverse-bias conductance exhibits an insulator-like temperature dependence, suggesting that tunneling is the dominant transport mechanism over the range of bias and temperature investigated. Differential conductance spectroscopy enhances features in this reverse tunnel current that are coincident in energy with magnetic quantum-well states, previously seen in Ag/GaAs and Ag/Fe systems. Because of this, it is believed that these magnetic multilayer films are good candidates for spin-polarized injection into GaAs heterostructures.

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