Direct Measurement of the Spin Polarization of the Magnetic Semiconductor (Ga,Mn)As

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We have carried out a direct measurement of the degree of spin polarization (***P***) of the magnetic semiconductor $Ga_{1-x}Mn_xAs$ using Andreev reflection spectroscopy. Analyses of the conductance spectra of high transparency $Ga_{0.95}Mn_{0.05}As/Ga$ junctions consistently yield an intrinsic value for ***P*** greater than 85%. Our experiments also revealed an extreme sensitivity of the measured spin polarization to the nature and quality of the interface for this material.

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Following the success of metal-based spintronics in fundamental physics as well as device applications, contemporary interest in semiconductor-based spintronics is motivated by the desire to produce three-terminal spintronic devices with potential applications in nonvolatile programmable logic, spin-based optoelectronics, and quantum computation [1]. The plausibility of such a semiconductor spintronics technology has been bolstered by recent advances such as the demonstration of coherent spin transport in homogeneous and heterogeneous semiconductors [2], the observation of spin injection from magnetic semiconductor contacts into conventional semiconductors [3], the electric field control of ferromagnetism in magnetic semiconductors [4], and the invention of a variety of ferromagnetic semiconductors [5].

A fundamental understanding of $Ga_{1-x}Mn_xAs$ is very relevant in this context since this is a "canonical" ferromagnetic semiconductor that remains the most thoroughly studied of all such materials [6]. Recent experiments demonstrate that the Curie temperature (T_C) of this material can be as high as 150 K [7], showing promise for possible technological relevance. An important parameter of direct fundamental and applied relevance is the carrier spin polarization (***P***) of $Ga_{1-x}Mn_xAs$. The large tunneling magnetoresistance observed in magnetic tunnel junctions derived from this material implies that ***P*** may be large even for small Mn concentrations [8,9]; this is consistent with band structure calculations that predicted $\boldsymbol{P} = 100\%$ for $x \geq 0.125$ [10]. However, there have been no direct measurements of ***P*** for this important material. Moreover, the ferromagnetic interaction between the Mn ions is mediated by the holes in the GaAs valence band. There exists a strong spin-orbit interaction for these holes, which poses a basic question of the existence of spin polarization in this material.

Here, we report a measurement of ***P*** using Andreev reflection spectroscopy in carefully prepared $Ga_{0.95}Mn_{0.05}As/Ga$ planar junctions. Andreev reflection (AR) [11] is a process that converts quasiparticle currents in the normal metal to supercurrent in the superconductor at a normal metal/superconductor (N/S) interface. A single electron in N with energy smaller than the superconducting gap can get into S only by pairing up with an electron of opposite momentum and opposite spin. AR results in an enhancement of the N/S junction conductance beyond its normal state value below the gap. When N is replaced by a ferromagnet, AR is suppressed due to the spin imbalance leading to a decrease of the subgap conductance [12], and a measurement of this suppression gives a quantitative measure of the magnitude of the spin polarization in the ferromagnet (AR cannot determine the sign of ***P***). Analysis of the conductance data from our $Ga_{0.95}Mn_{0.05}As/Ga$ samples in this fashion consistently yield $\boldsymbol{P} > 85\%$. We also find, however, that the preservation of this high spin polarization at semiconductor/superconductor interfaces is quite difficult and very sensitive to the nature and quality of the interfaces.

An extensive range of samples was fabricated for the purposes of this study. These include (a) superconductor/ferromagnetic semiconductor (S/FS) junctions made entirely *in situ* under ultrahigh vacuum (UHV) conditions by depositing the superconductor electrode (Ga, Al, or Zn) immediately after the $Ga_{1-x}Mn_xAs$ growth; (b) S/FS junctions made by transferring As-passivated $Ga_{1-x}Mn_xAs$ epilayers to an *ex situ* vacuum system for the deposition of the superconductor after desorption of the As cap layer; (c) S-insulator-FS ($S/I/FS$) tunnel junctions fabricated in a manner similar to (b), but after deposition of a thin layer of Al which was oxidized via exposure to O_2, before the deposition of an Al or Pb layer; (d) $S/I/FS$ junctions similar to (c), but where the $Ga_{1-x}Mn_xAs$ epilayer was exposed to an oxygen plasma before the deposition of the Al or Pb layer; (e) *in situ* grown $Ga_{1-x}Mn_xAs/AlAs/Al$ tunnel junctions wherein AlAs serves as a tunnel barrier; and (f) *ex situ* grown $Ga_{1-x}Mn_xAs/AlAs/Pb$, with the superconductor deposited as in (b). As discussed later, we find that spin polarization measurements for these systems are extremely sensitive to the details of the interfaces. Hence, we first focus on the *in situ* fabricated $Ga_{0.95}Mn_{0.05}As/Ga$ junctions since these high transparency junctions exhibit the clearest conductance spectra that indicate high $\boldsymbol{P}$ for GaMnAs.

The $Ga_{0.95}Mn_{0.05}As/Ga$ junctions were fabricated as follows: first, a 20 nm thick, p-doped GaAs:Mn buffer layer was grown on a heavily p-doped (001) GaAs:Zn substrate using molecular beam epitaxy (MBE) under standard conditions for high quality GaAs growth. A $Ga_{0.95}Mn_{0.05}As$ epilayer(typically around 100 nm thick) was then grown by low-temperature MBE ($T_{\text{substrate}} \sim 250\,^{\circ}\text{C}$) using growth conditions described elsewhere [13]. The as-grown GaMnAs film has a $T_C \sim 65$ K, as shown in Fig. 1. Immediately after the $Ga_{0.95}Mn_{0.05}As$ growth, the substrate temperature was lowered to 10 °C and a thick layer ($>$ 500 nm) of Ga was deposited under UHV conditions in the same MBE chamber. The conductance spectra of the samples were measured in a setup depicted schematically in Fig. 2(a), using phase-sensitive detection in a ^{3}He cryostat. A typical junction area is 1 mm $\times$ 1 mm. Two of the contacts were made on the conducting substrate, while the other two contacts were made on top of the Ga electrode. This setup, instead of the cross-stripe geometry, was used to circumvent the current crowding problem due to the relatively high sheet resistance of the $Ga_{0.95}Mn_{0.05}As$ compared to the low junction resistance. Typical normal state junction resistances are 10–100 Ω, while the serial resistance from the GaMnAs layer is at least 7 orders of magnitude smaller. The conductance spectra shown below were taken in zero magnetic field. We have observed no difference in field cooling and zero-field cooling.

Figure 2(b) shows the normalized conductance as a function of bias voltage taken at 370 mK for a $Ga_{0.95}Mn_{0.05}As/Ga$ junction. At first glance, this conductance spectrum is typical of that for a high transparency metallic contact between a superconductor and a ferromagnet with high $\boldsymbol{P}$: the conductance peaks at $\pm\Delta$ corresponding to quasiparticle tunneling are completely absent; on the other hand, the subgap conductance is suppressed, instead of enhanced, from G_N due to the large imbalance of spin populations in the ferromagnet. Blonder, Tinkham, and Klapwijk (BTK) [14] developed a comprehensive theory to evaluate the conductance spectra of superconductor/normal metal (S/N) junctions with arbitrary interfacial scattering strength, bridging the gap between metallic contacts and tunnel junctions. In this theory, the interfacial scattering strength is measured with a dimensionless parameter Z, with $Z = 0$ for a metallic contact, and $Z \gg 1$ for a tunnel junction. It is important to note that Z in this model is a phenomenological parameter that takes into account the effects of the physical barrier (potential scattering) as well as that of the band structure mismatch. The model was extended to include the effects of spin polarization in superconductor/ferromagnet (S/Fm) junctions [12], and the spin blockade of AR has been effectively utilized to measure the spin polarization of a variety of ferromagnets including half metals [15] with the point contact setup. With this modified BTK model, the extraction of $\boldsymbol{P}$ from a metallic contact is straightforward: when $Z = 0$, $\boldsymbol{P} = 1 - [G(0)/2]$. Using this calculation the data in Fig. 2(b) yield a spin polarization of 90% for this $Ga_{1-x}Mn_xAs$ sample. However, several aspects of the data warrant further discussion.

Although the conductance spectrum resembles that from a metallic S/Fm contact, the entire spectrum cannot be fit straightforwardly to the modified BTK theory. Moreover, the approximate energy gap for Ga inferred from the shoulders of the spectrum is ~1.4 meV, which corresponds to a T_C much higher than the T_C for bulk crystalline Ga (1.1 K). These discrepancies can be explained with a distribution of the energy gap and T_C in the Ga film. It is known that several phases of Ga have T_C substantially higher than 1.1 K, and amorphous thin films

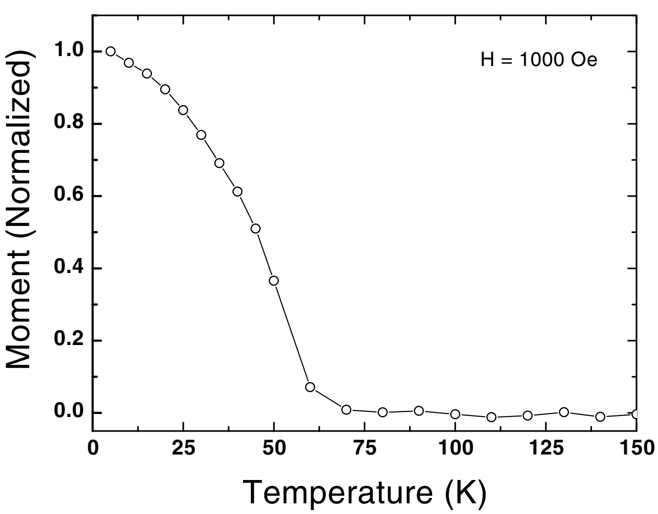

FIG. 1. Temperature dependence of the normalized magnetization of the $Ga_{0.95}Mn_{0.05}As$ layer in a junction.

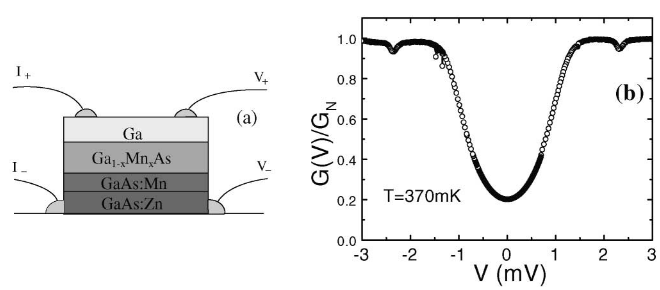

FIG. 2. (a) A schematic of the (Ga,Mn)As heterostucture and the contact scheme. (b) Normalized conductance spectrum of a $Ga_{0.95}Mn_{0.05}As/Ga$ junction exhibiting high transparency and spin polarization.

of Ga have been found to have T_C as high as 8.4 K [16]. The Ga film in our device was grown at a low temperature of 10 °C and has a granular morphology. It is probable that differences in grain size and crystallinity may result in local variations of T_C and energy gap in the film. We have attempted to fit the conductance spectra to the modified BTK theory by including a distribution of energy gaps in the superconductor. Figure 3 shows the results of such a fit for two junctions from the same growth. Clearly, excellent fits are obtained for both samples. More importantly, an identical distribution is used in both fits. The distribution was created as an *ad hoc* weighting and reflects that significant portions of the Ga film have T_C around 1.1 and 8.4 K, and less with intermediate T_C values. The fits resulted in Z values close to zero and $\boldsymbol{P}$ of 90% and 85%, respectively, consistent with values calculated from $G(0)$. Furthermore, we found that the supression of $G(0)$ persisted much above the bulk Ga T_C of 1.1 K, vanishing only when the temperature approached 8.4 K. This gives us further confidence that the broad conductance dip does not come from simple thermal broadening or inelastic effects.

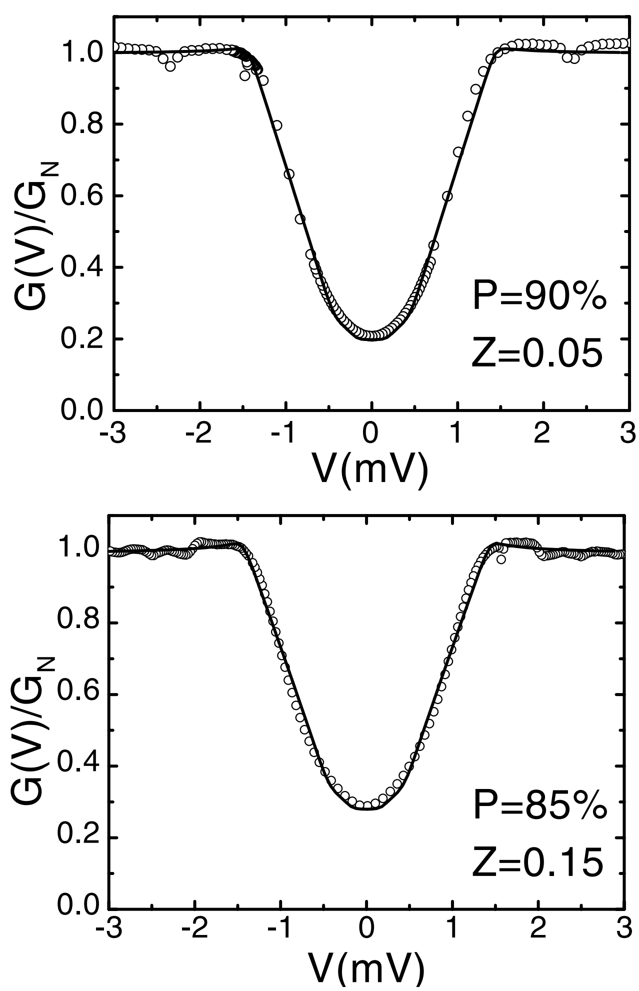

FIG. 3. Fits to modified BTK theory using a distribution of T_C and Δ for two samples of $Ga_{0.95}Mn_{0.05}As/Ga$ from the same growth. Identical distribution was used for both.

Another complication in analyzing the conductance spectra of a S/FS junction lies in the large mismatch in the Fermi velocity always present between a semiconductor and a metal. In the BTK model, the effect of the Fermi velocity mismatch can be included in the parameter Z which measures the overall interfacial scattering strength. It is therefore quite a surprise that we were able to obtain an apparent Z of zero. Under the conditions used in the MBE growth of our samples, $Ga_{1-x}Mn_xAs$ samples with $x = 0.05$ typically have a carrier (hole) density of $\sim 3 \times 10^{20}$ cm^{-3}; assuming that the (heavy) holes in $Ga_{1-x}Mn_xAs$ have the same effective mass as in GaAs ($0.45m_e$), we estimate a Fermi velocity of 4.6×10^5 m/s compared to 2.0×10^6 m/s for Ga. Such a large mismatch should result in a substantial Z even in the absence of any physical barrier at the interface. Zutic and Das Sarma [17] generalized the BTK analysis, specifically applying to superconductor/semiconductor junctions, by separating the effects of the physical barrier (potential scattering) and the mismatches in effective mass and Fermi velocity between the superconductor and the semiconductor. Indeed, they found that these mismatches lead to much decreased junction transparency for a superconductor/conventional semiconductor contact, signified by a substantial decrease of $G(0)$ from $2G_N$ and pronounced peaks at $\pm\Delta$ in the conductance spectrum even when the interfacial potential scattering is completely absent ($Z_\sigma = 0$). However, in a ferromagnetic semiconductor, the spin polarization actually enhances junction transparency. Specifically, the conductance peaks at $\pm\Delta$ from the Fermi velocity mismatch can be completely suppressed by a moderate spin polarization in the FS. In contrast, the conductance peaks at $\pm\Delta$ due to potential scattering are not affected by the spin polarization. Therefore, the complete absence of any peaks at $\pm\Delta$ in our data is consistent with high transparency of the $Ga_{0.95}Mn_{0.05}As/Ga$ interface ($Z_\sigma = 0$) and high spin polarization for the $Ga_{0.95}Mn_{0.05}As$. According to Ref. [17], the increase of $\boldsymbol{P}$ in the FS also results in a consistent decrease in $G(0)$. Hence $G(0)$ is still a good measure of the spin polarization in high transparency S/Sm junctions.

It is also somewhat surprising that we were able to obtain $Ga_{0.95}Mn_{0.05}As/Ga$ junctions with essentially no interfacial barrier considering the large differences in carrier density. On the other hand, the experience in our laboratory has shown that Ohmic contacts can be readily made on $Ga_{1-x}Mn_xAs$ with several different types of metallization. *I-V* measurements of the junctions at temperatures above T_C of Ga showed strictly linear behavior, indicating Ohmic contacts and absence of any Schottky barrier.

While the intrinsic spin polarization for $Ga_{0.95}Mn_{0.05}As$ inferred from our experiments is close

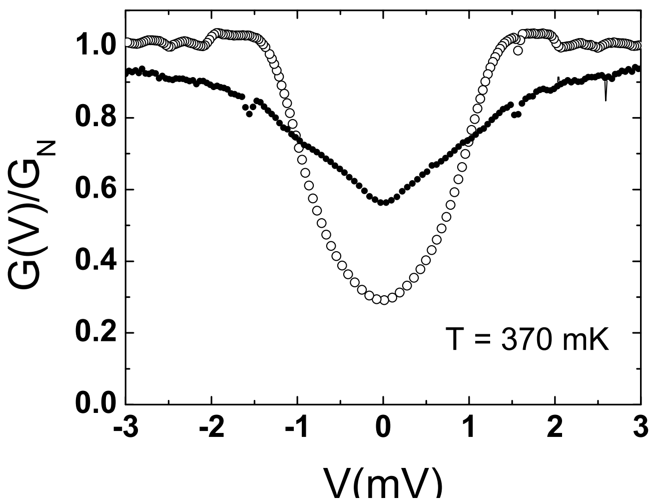

FIG. 4. Conductance spectrum of a $Ga_{0.95}Mn_{0.05}As/Ga$ junction before (open circles) and after (solid circles) annealing.

to 100%, we found that it is extremely difficult to maintain this high spin polarization at many types of GaMnAs/metal interfaces. In fact, in many cases we failed to see any signatures in the conductance spectra related to superconductivity in the counter electrode, a phenomenon also observed by others in similar setups [18]. Moreover, we have examined the effect of annealing on the $Ga_{0.95}Mn_{0.05}As/Ga$ junctions that did yield high $\boldsymbol{P}$; even a very mild vacuum annealing at 100 °C resulted in a significant deterioration of the conductance spectrum and spin polarization, as shown in Fig. 4. The spin polarization of $Ga_{1-x}Mn_xAs$ at its surface appears to be extremely sensitive to the nature and quality of the interface.

Finally, we address the issue of spin-orbit coupling since it is known that holes in the valence band are responsible for the ferromagnetic interaction in $Ga_{1-x}Mn_xAs$. It is expected that the spin-orbit interaction would greatly decrease the spin polarization of the holes, which is apparently in contradiction to the experimental results reported here and elsewhere [8]. However, resonant tunneling spectroscopy has revealed a large spontaneous spin splitting of the valence band in $Ga_{1-x}Mn_xAs$ (~ 44 meV at low temperatures for $x = 0.035$) [19]. Dietl *et al.* suggest in a mean field model that the destructive effect of the spin-orbit coupling is quickly suppressed with increasing band splitting [20]. With a band splitting of 40 meV the spin polarization is restored to above 85% even for high hole concentrations, in agreement with our observations.

In summary, we have carried out a series of experiments to directly measure the spin polarization of the ferromagnetic semiconductor $Ga_{1-x}Mn_xAs$. Andreev reflection spectroscopy from high transparency $Ga_{0.95}Mn_{0.05}As/Ga$ junctions consistently yielded a spin polarization greater than 85% for $Ga_{0.95}Mn_{0.05}As$. We believe that this may represent a lower limit of the intrinsic spin polarization for this material because of the difficulties in maintaining the high spin polarization at the interface with the superconducting metals in a planar junction device structure. The apparently high interfacial sensitivity may pose a challenge in constructing spintronics devices using $Ga_{1-x}Mn_xAs$.

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