Spin polarization of (Ga,Mn)As measured by Andreev Spectroscopy: The role of spin-active scattering

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We investigate the spin-polarization of the ferromagnetic semiconductor (Ga,Mn)As by Andreev point contact spectroscopy. We analyze the conductance spectra with the theoretical model of [R. Grein *et al.*, Phys. Rev. B **81**, 094508 (2010)], which accounts for momentum- and spin-dependent scattering at the interface. We show that this allows for fitting the data without resorting to an effective temperature or statistical distribution of superconducting gaps, as it is the case for the spin-dependent version of the standard Blonder-Tinkham-Klapwijk (BTK) model. We find a transport polarization $P_C \approx 56\%$ at the Fermi level and comparing it to the $\vec{k}\cdot\vec{p}$ kinetic-exchange model of (Ga,Mn)As, we achieve a considerably better agreement than estimates inferred from the spin-dependent BTK model which are significantly higher. The temperature dependence of the conductance spectra is fully analyzed.

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Introduction.— The quickly evolving field of spintronics, which broadly concerns the manipulation and exploitation of the quantum mechanical spin of an object, has led to an intense search for spin-polarized materials that are promising candidates for applications. Current metal spintronic devices and most proposed semiconductor spintronic devices aim to exploit the net spin-polarization of charge carriers (holes in the case of ferromagnetic semiconductors) to encode and/or process information. The advantage of using ferromagnetic (FM) semiconductors is their potential to serve as spin-polarized carrier sources and the possibility to easily integrate them into semiconductor devices [1]. Recently, the family of (III,Mn)V FM semiconductor has attracted much attention for its potential applications in non-volatile memory, spin-based optoelectronics and quantum computation [2]. In particular, (Ga,Mn)As, with a Curie temperature as high as 185 K [3, 4] is one of the most promising candidates for such applications. Hence, understanding and controlling the electrical and magnetic properties of these materials is an important step towards spintronics devices.

The degree of spin-polarization is the key parameter for most spintronic functionalities. The injection of spin polarized currents from FM semiconductors into nonmagnetic semiconductor devices has been demonstrated by measurements of the optical polarization of light emitted after recombination of spin polarized holes with electrons in nonmagnetic semiconductors [5]. However, the resulting polarization is strongly dependent on the experimental set-up and the spin relaxation rate in the nonmagnetic part of the device, so that it is very difficult to infer reliable values of spin-polarization from such measurements [6]. Specialized techniques based on superconductor/ferromagnet junctions have been employed frequently in recent years to obtain information on the spin-polarization in metals [7, 8]. In particular, the Point Contact Andreev Reflection (PCAR) technique has become a popular tool to measure the transport spin-polarization P_C of carriers at the Fermi level in FM materials [9]. However, the reliability of the P_C-values obtained with this technique - which is based on fitting the spectra to an extension of the Blonder-Tinkham-Klapwijk (BTK) model [10, 11] - is currently under debate [8, 12–14].

The PCAR data contains information regarding the carrier spin-polarization because the Andreev reflection process is spin-sensitive. Andreev reflection is the only allowed process of charge transfer across a contact between a superconductor (SC) and a normal metal at energies below the SC gap. In this process, a Cooper pair is created in the SC when a quasiparticle tunnels from the normal metal and a hole is reflected back coherently. Effectively, two particles with opposite spin are transferred, and hence the probability of this process to occur is reduced when the density of states for spin up/down carriers is different. This results in a suppression of the conductance for voltages below the superconducting gap and thus the degree of spin-polarization can be estimated from such conductance spectra [11, 15].

In this Letter, we present a detailed PCAR study of spin-polarization of (Ga,Mn)As. The measurements have been carried out at different temperatures starting from about 2K up to the point where the SC gap in the tip closes. The shape of the experimental conductance spectra shows a suppression of the conductance for voltages smaller than the SC gap and a complete absence of co-

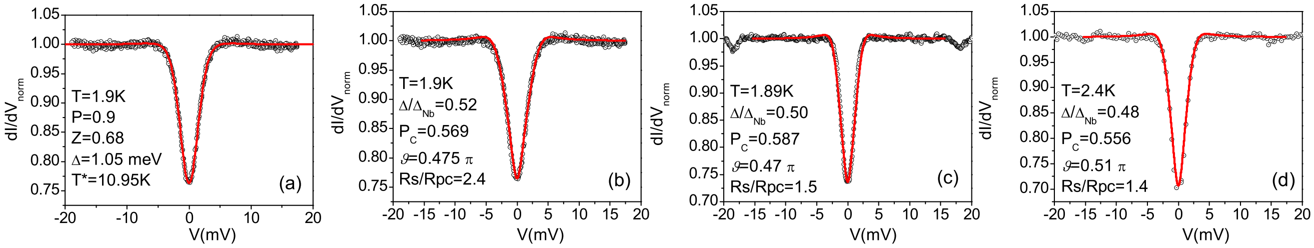

FIG. 1: (Color online) Experimental PCAR data (black dots) with the theoretical fit (red line), (a) in agreement with the modified BTK model; and [(b), (c) and (d)] according to the theoretical model in Ref. [12].

herence peaks at the gap edge. However, in agreement with earlier PCAR measurements on this material, the conductance suppression below the gap energy is rather small, with a minimum of about 0.7 G_N, where G_N is the conductance in the normal state. This feature would usually hint at an intermediate spin-polarization of the FM-material. Yet, if one tries to fit these spectra with the BTK theory, it turns out that the two key features, the absence of coherence peaks and the rather high zero-bias conductance, cannot be reconciled within this model. To remedy this situation, one needs to appeal to an "effective" temperature, T^*, which we find to be almost 6 times as high as the real temperature of the sample in our case. With this additional fit parameter, T^*, excellent fits can be achieved and usually a high value of the spin-polarization is inferred.

Alternatively, we propose an interpretation of the data using a model of interface scattering that goes beyond the BTK theory and shows that good agreement can be achieved *without* an effective temperature. From this analysis we infer a value of the spin-polarization of about 57%. We also notice a substantial reduction of the superconducting energy gap and critical temperature of the Nb tip, which is probably related to an inverse magnetic proximity effect. From measurements of the conductance spectra at different temperatures, we extrapolate the temperature dependence of the superconducting energy gap.

Experiment and BTK fitting.— We have analyzed samples which are 7% Mn doped and 25 nm thick. They are grown on a 200nm thick, highly Carbon p-doped ($\sim 10^{19}$ cm^{-3}) buffer layer to minimise series resistance. As grown the Curie temperature, T_C, is $\sim$ 70 K, and the resistivity (ρ) at $T = 4.2$ K is $\sim$ 4 mΩcmm. After low temperature annealing (which removes compensating Mn interstitials) T_C increases to $\sim$ 140 K, and $\rho(T = 4.2)$ decreases to $\sim$ 2 mΩcm. The details of the sample growth and preparation are described elsewhere [18]. The experiments were carried out by means of a variable temperature (1.5–300 K) cryostat. To realize our experiments we used a chemically etched Nb tip, . Sample and tip were introduced into the PCAR probe, in which a piezo motor and scan tube can vary the distance between tip and sample. The PCAR junctions were formed by pushing the Nb tip on the(Ga,Mn)As surface with the probe thermalized in ^{4}He gas. The current-voltage I vs V characteristics were measured by using a conventional four-probe method and, by using a small ac modulation of the current, a lock-in technique was used to measure the differential conductance dI/dV vs V spectra as function of the applied voltage directly.

In Fig. 1 we show conductance spectra at low temperature ($T \sim 2$ K). The data of Fig.1(a) and (b) is identical, but a is fitted with the extended BTK model and b-d with the theory of [12]. The data has been normalized using the background conductance estimated at large voltage ($V \gg \Delta_{\rm Nb}/e$) regions, where $\Delta_{\rm Nb}$ is the superconducting gap of Nb ($\Delta_{\rm Nb} \sim 1.5$ meV). All conductance spectra show a moderate dip and completely suppressed coherence peaks at the gap edge. No significant difference in the spectra has been noticed before and after annealing.

To fit the experimental data in Fig.1(a), we have used as free parameters: P_C; the strength of the barrier, Z; the superconducting energy gap, Δ; and T^*. From the theoretical model we infer a value of the spin-polarization of about 90%, consistent with the other values reported in literature and a reduction of the superconducting energy gap. We underline that using the BTK model requires a very high effective temperature, $T^* = 10.95$ K, which is more than 5 times higher than the measured temperature of 1.9 K. According to Ref. [17], this effective temperature accounts for inelastic scattering in the (Ga,Mn)As sample, but in any case it is a parameter introduced "ad hoc", and whether such a high value of T^* can be justified on this basis is not clear.

Spin-active Scattering.— Recently, a theoretical model was introduced which allows for a more realistic description of interface scattering in the calculation of charge and spin transport across such point contacts [12, 19]. When a contact with a magnetic material is created, one would expect that the scattering properties of quasiparticles depend on their spin. When no tunneling potential is present, the transparency of the interface is controlled by wave vector mismatches. Since wave vectors of $\uparrow$- and $\downarrow$-spin quasiparticles are different in the FM material, their transmission probabilities should differ accordingly. Moreover, it was shown that scattering phases can play an important role in this case [20]. While a global phase will never affect any physical properties, the relative phase difference, that quasiparticles incident from the SC may acquire upon being reflected at the magnetic interface, induces a triplet proximity effect and leads to substantial modifications of conductance spectra [12]. This relative scattering phase is called *spin-mixing angle* or spin-dependent interface phase shift. In the case of point-contacts, it suppresses the coherence peaks at the gap-edge and shifts their spectral weight to energies below the gap, where interface Andreev bound states are induced. This mechanism allows for an alternative interpretation of the PCAR spectra analyzed here. Using a minimal model of spin-active scattering, we show that good fits can be achieved without resorting to an effective temperature.

The transport theory proposed in Ref. [12] relies on the normal-state scattering matrix of the interface as a phenomenological parameter. This S-matrix generally depends on the impact angle of the incident quasiparticles. We assume that spin-flip scattering due to spin-orbit coupling can be neglected (this approximation is appropriate for sufficiently strong spin polarization), hence the S-matrix is diagonal in spin-space, yet we allow for different transmission probabilities $t_\uparrow \neq t_\downarrow$ and a spin-mixing angle ϑ. Since there is no insulating interlayer, we assume that the transmission probability is controlled by wave vector mismatches. We use the averaged Fermi wave vectors of the (Ga,Mn)As spin bands $k_{\uparrow,\downarrow}/k_{SC}$ as fit parameters. Here, k_{SC} is the Fermi wave vector in the SC. $t_\uparrow(\theta)$ and $t_\downarrow(\theta)$ are then calculated for any impact angle θ by wave function matching. The density of of states $N_{\uparrow,\downarrow}$ (at Fermi energy E_F) of the respective spin-band is assumed to be proportional to $k_{\uparrow,\downarrow}$ and independent of energy on the relevant scale of the SC gap. The third parameter describing the interface is the spin-mixing angle ϑ which also depends on the impact angle. If the conduction bands of the materials are assumed to be unperturbed at the interface, this relative scattering phase will not occur. However, if the transition from one material to the other is smoothed on the scale of the Fermi-wavelength in Nb [12], it will. We assume that this mixing phase is maximal for perpendicular impact and goes to zero for grazing trajectories. For definiteness, this is modeled by $\vartheta(\theta) = \vartheta \cdot \cos(\theta)$, but even if $\vartheta(\theta) = const$ is assumed, it does not change anything about our conclusions, since grazing trajectories contribute little to the total conductance. Additionally, the value of the SC gap and a spread resistance are fitted, while the temperature is taken from experiment. For details of the calculations involved we refer the reader to Ref. [12, 19].

Based on the assumptions made above, the (transport) spin-polarization can be directly inferred from the fitted ratio $r = k_\downarrow/k_\uparrow$ of the individual spin-band wave vectors. Adhering to the notation of Ref. [9], we define the spin-

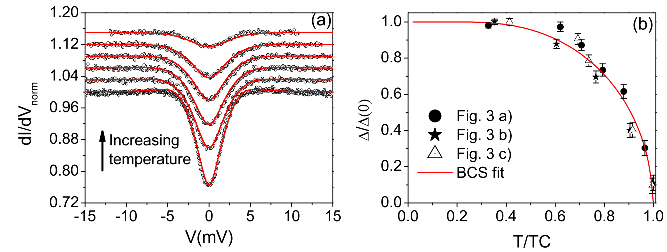

FIG. 2: (Color online) (a) Spectra as a function of temperature (black dots) with the theoretical fittings obtained by considering the spin-mixing effect with the energy gap as only free parameter. Spectra are shifted and shown for temperatures between $1.9K$ and $5.6K$. (b) Temperature dependence of the superconducting energy gap as inferred from the theoretical fittings rescaled to the BCS relation.

polarization, P, and transport spin polarization (at the Fermi surface), P_C, as

$$P = 2\frac{\langle s_z\rangle_\uparrow + \langle s_z\rangle_\downarrow}{\langle 1\rangle_\uparrow + \langle 1\rangle_\downarrow}, \qquad P_C = 2\frac{\langle s_z v\rangle_\uparrow + \langle s_z v\rangle_\downarrow}{\langle v\rangle_\uparrow + \langle v\rangle_\downarrow}. \quad (1)$$

where $\langle f\rangle_i \equiv \int d^3k f(\vec{k})\delta(E_F - \varepsilon_{\vec{k},i})$ denote the Fermi surface averages of the function f over the $i \in \{\uparrow,\downarrow\}$ spin-bands and $\varepsilon_{\vec{k},i}$ the band dispersions (i.e. $\langle 1\rangle_i = 8\pi^3 N_i$); v and s_z are the group velocities $(1/\hbar)|\nabla_k \varepsilon_{\vec{k},i}|$ and the spin expectation value projected to the direction of magnetization (taken to be z here). If the effective masses in both spin-bands are approximately the same, we get $P \approx (1-r)/(1+r)$ and $P_C \approx (1-r^2)/(1+r^2)$. Various normalized conductance spectra shown in Fig. 1 (b) (c) and (d), obtained by establishing different contacts on different areas of the (Ga,Mn)As sample, lead to a nearly constant transport spin-polarization $57 \pm 2\%$ (or equivalently $P \approx 0.31$) at low temperatures when P_C is used as the fit parameter together with ϑ, R_s/R_{pc}, and Δ within our BTK theory. Wavevectors inferred vary little around $k_\uparrow/k_{SC} = 0.447$ and $k_\downarrow/k_{SC} = 0.237$ and we find $0.47 \lesssim \vartheta/\pi \lesssim 0.51$. The spread resistance R_s arises from the resistance of the sample between the junction and one of the measuring contacts [8] renormalizing both the voltage that drops across the contact and the normalized conductance. The value of R_s/R_{pc} found by fitting (between 2.4 and 1.4 in Fig. 1 (b)–(d)) is rather high as the conductance of (Ga,Mn)As is low compared to metallic samples and it is likely that multiple shunted contacts are established when the tip is pressed into the sample.

Using the spin-active scattering model for fitting, we also find a reduction of the superconducting energy gap. We estimate the reduction of the gap to be about 50% with respect to the zero temperature bulk value, reported to be $1.5meV$ in Nb for the lowest temperature spectra we measured. T_c is reduced to $5.4 - 5.8K$ which we infer from the disappearance of all SC features at this temperature. This implies a deviation from the theoretical strong-coupling BCS ratio [23] for Nb ($2\Delta/K_BT_C \approx$

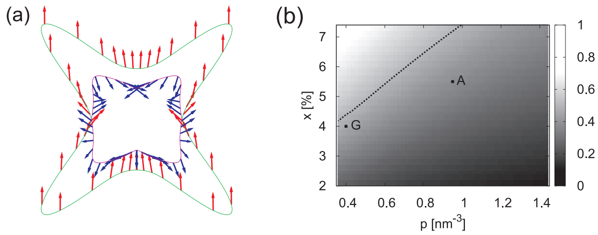

FIG. 3: (Ga,Mn)As $\vec{k}\cdot\vec{p}$ model [21] used to evaluate (a) spin-textures in the heavy-hole bands at the Fermi surface (k_y, k_z section is shown) and (b) total P_C calculated by generalizing Eq. (1) to all six bands involved (the isoline corresponds to $P_C = 0.5$). Total hole concentration p and $x = x_S - x_i$ (as explained in text and in Ref. [22]) are taken as independent parameters in (b) while $x = 5.5\%$ and $p = 0.8$ nm^{-3} applies for (a).

3.95), instead we find $2\Delta/K_BT_C \sim (3.3 \pm 0.3)$. The fitting for different temperatures at the same measurement location was done by only varying Δ, all other parameters are kept constant. Remarkably, the quality of the fits for all temperatures is still very good (see Fig.2(a)). Rescaling the obtained gap values to the BCS relation also yields reasonable agreement (Fig.2(b)). The reduction of the gap found here is stronger than in other experiments, indicating that an inverse proximity effect may be important.

In order to study the influence of Fermi surface anisotropy and spin orbit coupling we compare our results with a theoretical warped six-band $\vec{k}\cdot\vec{p}$ model with mean field kinetic-exchange due to Mn [21]. Focusing on the heavy-hole bands only, non-trivial spin-textures of the majority and minority bands (Fig. 3(a)), witnessing the appreciable spin-orbit interaction, can nevertheless, at least on average, be interpreted as belonging to spin $\uparrow$ and $\downarrow$ bands. Evaluating Eq. (1) for the full warped six-band model [21], we find a satisfactory agreement with the inferred values of r, P, and P_C from the spin active model. The warping and non-parabolicity effects, although sizable in Fig. 3(a), therefore seem to play only a minor role (e.g. the majority heavy hole band can still be reasonably described by a single averaged $k_\uparrow$). Specifically, taking $x_S = 5.5\%$ and $x_I = 1.5\%$ as a typical content of substitutional and interstitial Mn in a nominally 7%-doped as-grown sample [22], we evaluate $P_C \approx 0.46$ as indicated by the point marked 'G' in Fig. 3(b) while after annealing (interstitial Mn removed) we find a rather close value of ≈ 0.38 corresponding to 'A' in the same plot (small additional compensation corresponding to -0.25 nm^{-3} holes was added, due to e.g. As-antisites). The precise role of impurity scattering in the strongly doped materials has not been elucidated so far, however as our results show, the spin polarization obtained from the mean field kinetic-exchange model is in good agreement with the value we obtain from our fits assuming isotropic bands.

Conclusions.—In summary we have studied the spin-polarisation at the Fermi level in (Ga,Mn)As with the PCAR technique, using a recently developed theory that accounts for spin-active scattering at the interface to model the experimental results. Compared to previous work on PCAR with this material, this allowed us to drop the assumption of an effective temperature. The value of the spin-polarisation we obtain from this analysis is sizeable but significantly smaller than that inferred by earlier studies and it now agrees better with predictions of the $\vec{k}\cdot\vec{p}$ kinetic-exchange model of (Ga,Mn)As. We also investigated the full temperature dependence of the spectra and find a strong suppression of the SC gap. The temperature dependence of the fitted gap values is in qualitative agreement with the BCS relation.

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