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Spin-dependent photoelectron tunneling from GaAs into magnetic cobalt

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The spin dependence of the photoelectron tunnel current from free-standing GaAs films into out-of-plane magnetized cobalt films is demonstrated. The measured spin asymmetry (A), resulting from a change in light helicity, reaches $\pm 6\%$ around zero applied tunnel bias and drops to $\pm 2\%$ at a bias of -1.6 V applied to the GaAs. This decrease is a result of the drop in the photoelectron-spin polarization that results from a reduction in the GaAs surface-recombination velocity. The sign of A changes with that of the cobalt magnetization. In contrast, A is negligible on nonmagnetic gold films.

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Since its discovery in magnetic and superconducting junctions,[1–3] spin-polarized tunneling has been extensively studied in all-solid junctions [as opposed to a variable gap scanning tunneling microscope (STM) configuration]. This is because such studies reveal details of surface magnetism and also because magnetic tunnel junctions are technologically important.[4,5] Tunneling from ferromagnetic and antiferromagnetic tips has been successfully employed to observe magnetic ordering in metals down to the atomic scale.[6] Similarly, spin-polarized tunneling from ferromagnetic metals and ferromagnetic semiconductors into nonmagnetic semiconductors has also been reported in both all-solid junctions[7] and from a ferromagnetic tip.[8] In the latter cases, the transient spin polarization of the post-tunnel electrons is measured via the circular polarization of the resulting luminescence. Conversely, the tunnel current of spin-polarized photoelectrons into a ferromagnetic surface should also depend on the relative orientations of the photoelectron spin to the surface magnetization. This phenomenon was the basis of Pierce's proposal for GaAs tip spin-polarized scanning tunneling microscope (SPSTM).[9] While some works report this type of spin-dependent tunneling,[10] parasitic optical effects also yield apparent spin-dependent tunneling, even on nonmagnetic surfaces.[11–14] The consensus is that claims of spin-dependent tunneling must be doubted.[6,15,16]

Here, we demonstrate the spin dependence of the tunnel photocurrent $I_t^{\mathrm{ph}}(\sigma^{\pm})$ from p^+ GaAs under $\sigma^{\pm}$ circularly polarized light excitation into ultrathin perpendicularly magnetized cobalt (Co) films. In contrast to previous works,[10,12,13,17] optical pumping occurs from the rear (nontunnel) face of a 65-μm-long 3-μm-thick GaAs cantilever [see Figs. 1(a) and 1(b)].[18] Spin-polarized photoelectrons diffuse across the GaAs before tunneling, and since the cantilever thickness is larger than the absorption depth, 1 μm at $h\nu = 1.59$ eV, the light intensity (and, therefore, parasitic optical effects[12,13]) in the tunnel gap are negligible. The circular polarization of the pump light (5 mW focused to a spot of about 10-μm diameter) is controlled by a Pockels cell, and the resulting spin polarization of the tunneling electrons, $\delta n_s/n_s$, is found by analyzing the polarized luminescence (PL) from the cantilever. Here, n_s is the concentration of tunneling electrons. The $\sigma^{\pm}$-polarized PL spectra $[I_{\mathrm{PL}}(\sigma^{\pm})]$ at low light intensities (50 W/cm^2, $h\nu = 1.59$ eV) are shown in curves a and b of Fig. 1(c), respectively. The above band-gap luminescence degree of circular polarization, $[I_{\mathrm{PL}}(\sigma^+) - I_{\mathrm{PL}}(\sigma^-)]/[I_{\mathrm{PL}}(\sigma^+) + I_{\mathrm{PL}}(\sigma^-)]$, is 8% as seen from curve c. This polarization corresponds to an average over all photoelectrons in the cantilever, and by solving the spin-diffusion equation, $\delta n_s/n_s$ is found to be on the order of 16%.[18]

The cantilevers are pressed into mechanical contact with ultrathin atomically flat Co(0001) layers (thickness ≈ 5 monolayers) epitaxially grown by electrodeposition on an Au(111) buffer layer on Si(111).[19] The Co surface is passivated by chemisorbing CO, which renders the surface resistant to oxidation in dry air and quenches empty surface states.[20] Using the polar magneto-optical Kerr effect (MOKE), the CO-covered Co films are found to be magnetized out of plane, parallel (or antiparallel) with the quantization axis of the photoelectron spins [see Fig. 1(d)]. The coercive field is ≈ 200 Oe. The full zero-field remanence of the magnetization after saturation, the squareness of the hysteresis loop, as well as spatially resolved MOKE,[19] indicate that the saturated samples are composed of a single magnetic domain whose lateral extent is larger than the contact area through which tunneling via an oxide layer of homogenous thickness occurs.[21] This one-dimensional geometry reduces surface-chemistry instabilities observed for tunneling from tips in air,[22] and stable tunneling is possible for up to 30 min on a given sample at room temperature.

Tunneling takes place in zero-magnetic field with the remanently magnetized sample. The measurement cycle consists of the following phases: (i) The tunnel current is stabilized at 11 nA in the dark by the feedback loop for a GaAs bias of -1.5 V. (ii) The feedback loop is opened, and two bias scans for the duration of 12 ms are performed, one in the dark and the other one under σ^+ illumination. The tunnel photocurrent $I_t^{\mathrm{ph}}(\sigma^+)$ is obtained by difference. (iii) After a new stabilization sequence, two bias scans are again taken, one in the dark and the other one with a σ^--polarized laser. The measurement, averaged over 100 such cycles, each lasting 0.25 s, gives

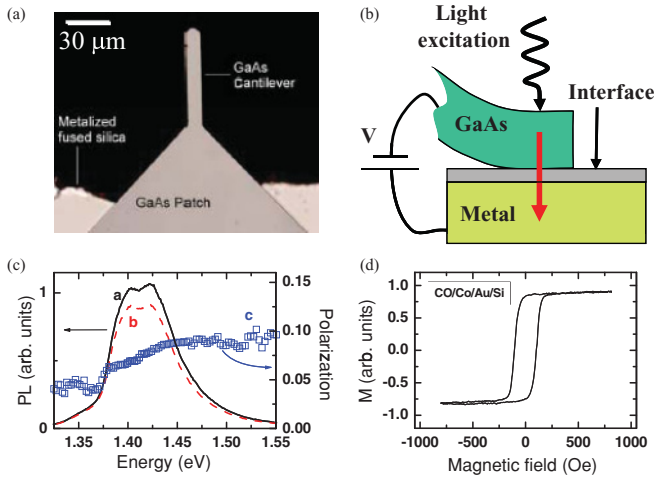

FIG. 1. (Color online) (a) A top view of the GaAs cantilever. (b) The principle of the experiment showing optical excitation at the rear face of the GaAs cantilever. (c) Curves a and b show the spectra of the $\sigma^{\pm}$-polarized components of the cantilever luminescence under circularly polarized excitation. Curve c shows the polarization of the spectrum, about 8% for band-to-band emission. (d) The magnetic-field dependence of the magnetization perpendicular to the surface of the Co film as measured using the polar magneto-optical Kerr effect.

the bias dependence of $I_t^{\rm ph} = [I_t^{\rm ph}(\sigma^+) + I_t^{\rm ph}(\sigma^-)]/2$, and of the spin asymmetry $A = [I_t^{\rm ph}(\sigma^+) - I_t^{\rm ph}(\sigma^-)]/[I_t^{\rm ph}(\sigma^+) + I_t^{\rm ph}(\sigma^-)]$. A may also be written[2]

$$A = \frac{\delta\rho_m}{\rho_m}\frac{\delta n_s}{n_s}, \tag{1}$$

where ρ_m is the total metallic density of states at the energy of the tunneling electrons and $\delta\rho_m$ is the difference in ρ_m between + and − spins, quantized along the direction of light

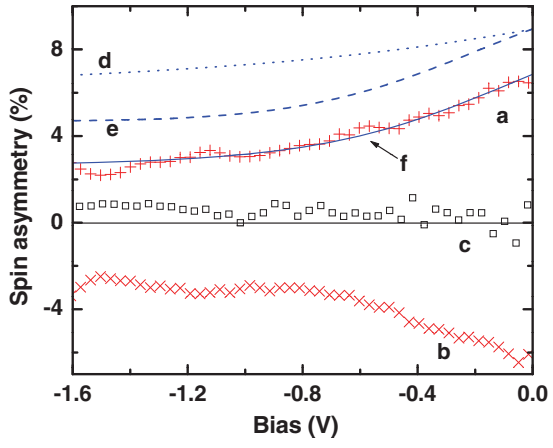

FIG. 2. (Color online) Curves a and b show the measured bias dependence of the spin asymmetry of the tunnel photocurrent into positively and negatively magnetized Co, respectively. Curve c is the asymmetry measured on a nonmagnetic gold surface. The bias dependence of $\delta\rho_m/\rho_m$ and of $\delta n_s/n_s$, normalized by factors of 10 and 1.3, respectively, are shown in curves d and e. The asymmetry, calculated using Eq. (1), is shown in curve f, and is in excellent agreement with the measured asymmetry.

excitation. As will be described below, at the tunneling energy of electrons corresponding to zero bias, $\delta\rho_m/\rho_m \approx 70\%$ so that A on the order of 10% is expected.

Figure 2 presents the main experimental result of this Rapid Communication. As seen in curve a, the measured value of A varies from 6% at zero bias to 2% at a reverse bias of −1.6 V, close to the qualitative estimation above. The nonzero value of A is due to a spin dependence of the tunneling current since (i) reversal of the magnetization of the Co induces a change in sign for A without any significant modification of either the absolute value or the bias dependence (curve b in Fig. 2), and (ii) measurements on (nonmagnetic) gold films yield A smaller than 1% (curve c) and ≈0% for small bias. Of the ten cobalt samples tested, more than half are perpendicularly magnetized as revealed by the MOKE, and all of these samples show a nonzero asymmetry that reverses sign with a change in the sample magnetization.[23]

In order to quantitatively understand the bias dependence of A, the bias dependences of the two terms of Eq. (1) are treated separately. The mechanisms that determine the concentration and tunneling energy of the photoelectrons at the GaAs surface are first analyzed using the bias dependences of the dark current (curve a) and $I_t^{\rm ph}$ (curve b) as shown in Fig. 3. These curves are in very good agreement with the predictions of a detailed model developed to describe unpolarized photoassisted tunneling[21,24] (see solid lines, Fig. 3) and lead to two distinct conclusions.

First, conduction electrons provide the dominant contribution to the tunnel photocurrent. As seen in Fig. 4, the energy of tunneling electrons is almost bias independent and close to that of the bottom of the conduction band. This is because the energy loss in the depletion layer, $(1-f)\varphi_b$, is smaller than 150 meV (the surface barrier $\varphi_b \approx 0.3$ eV under light excitation and the numerical factor $f \geqslant 0.5$ because of surface quantization). The parameters used to calculate the current curves in Fig. 3 are as follows: $f \approx 0.9$, $d = 0.74$ nm, the passivated Co work function is 6 eV,[25] and the dielectric

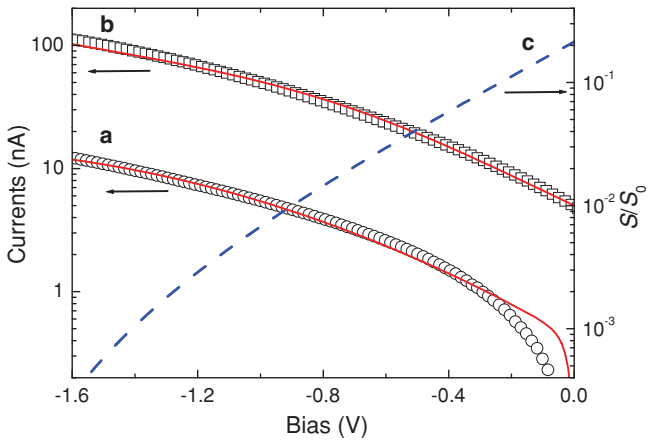

FIG. 3. (Color online) Curves a and b, respectively, correspond to the unpolarized dark and photocurrents for tunneling into Co. The solid lines correspond to the calculations of the tunnel currents using a model[21] describing unpolarized tunneling of photoelectrons into metals. Curve c shows the resulting bias variation of the normalized recombination velocity S/S_0.

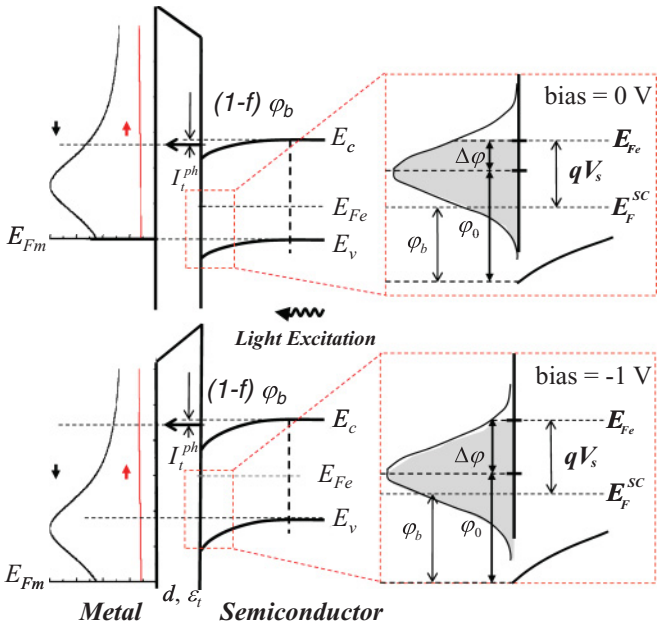

FIG. 4. (Color online) The tunnel junction and semiconductor-band structure versus position at applied biases of 0 V (top) and −1 V (bottom). The insets show the semiconductor surface density of states in both cases. To the left of the tunnel junction is the metallic density of states plotted against energy. Spin-polarized conduction electrons are injected into the top half of the 3*d* minority spin band of Co. The injection energy $E_g - (1 - f)\varphi_b$ is shown along with realistic representations of the densities of states of the majority (black) and minority (red) spins.

constant of the tunnel gap is 10, close to that of gallium oxide.[26] All other parameters are identical to those in Ref. 21. Once the energy of tunneling electrons is known, the spin-dependent metallic density of states $\delta\rho_m/\rho_m$ can be determined, as shown in Fig. 4. This value is approximated by the $k = 0$ spin-polarized inverse photoemission data for the same type of sample.[20] The use of the zone-center density of states is reasonable since the dispersion of majority and minority spin states is almost flat throughout the Brillouin zone.[27] The zero-bias value of $\delta\rho_m/\rho_m$ and its bias dependence are evaluated using Ref. 20 at the energy of tunneling electrons shown in Fig. 4. The result, shown in curve d of Fig. 2, only changes slowly with bias, and this alone cannot explain the measured bias dependence of A according to Eq. (1). Note that, while in Ref. 20, the experimental resolution is on the order of 450 meV; in the energy range considered here (1.4–2.9 eV above the Fermi energy), the energy dependence of the density of empty states is relatively flat and is only weakly affected by experimental resolution. Therefore, the decrease of A must be determined mainly by the bias dependence of $\delta n_s/n_s$.

Second, the nonexponential increase of both the dark current and the I_t^{ph} is caused by unpinning of the surface-Fermi level as shown in Fig. 4, which shifts the electron quasi-Fermi level away from the midgap by a quantity $\Delta\varphi$. This decreases the surface recombination velocity given by $S = S_0 \exp(-\Delta\varphi/k_BT)/D(\Delta\varphi)$, where S_0 is the value of S for $\Delta\varphi = 0$ and $D(\Delta\varphi)$ is the relative decrease of the density of surface states.[28] The 2 orders-of-magnitude decrease in S/S_0 with the applied bias (see curve c of Fig. 3) results in an increase of the concentration of tunneling electrons, which increases I_t^{ph}. This variation also results in a reduction of the spin polarization, which, as discussed below, is responsible for the bias-induced decrease of A.

Quantitatively, the bias variation of $\delta n_s/n_s$ is obtained by solving the equations for spin and charge diffusion.[29] For a cantilever of thickness l, in the limit of large S at the rear surface and absorption length $< l$, one finds a dependence on S of the form

$$\frac{\delta n_s}{n_s} = P_0 \frac{1 + S/v_d}{1 + S/v_{ds}}. \tag{2}$$

Here, $v_d = (D/L)\coth(l/L)$ and $v_{ds} = (D/L_s)\coth(l/L_s)$ are effective charge and spin-diffusion velocities, (L and L_s are the charge and spin-diffusion lengths, and D is the diffusion constant). For $\sigma^{\mp}$-polarized light excitation, and in the limit of large recombination velocity of the rear surface, the reduced electronic spin polarization P_0 is given by

$$P_0 = \pm 0.5 \frac{\tau_s}{\tau} \frac{h(L_s)}{h(L)}, \tag{3}$$

where the positive function,

$$h(x) = \frac{e^{\alpha l}\ \cosh^{-1}(l/x) - \alpha x\ \tanh(l/x) - 1}{(\alpha x)^2 - 1} \tag{4}$$

depends on the electron lifetime τ, the spin lifetime τ_s, and the light absorption coefficient α but not on S. As seen from Eq. (2), the bias-induced decrease of S induces a decrease of the spin polarization of tunneling electrons. Curve e of Fig. 2 shows this decrease calculated using the bias-induced decrease of S shown in Fig. 3, and $L_s = 0.6\ \mu$m. Equations (3) and (4) show that, because of surface recombination, a significant spin polarization can be created at the tunneling surface even if $L_s < l$. This is because surface recombination reduces the effective minority carrier lifetime so that spins have less time to relax and the steady-state polarization is larger than would otherwise be expected.[30] With the parameters for charge transport used for the fits in Fig. 3, the calculated bias dependence of A is shown in curve f of Fig. 2 and agrees very well with the measured dependence. The zero-bias asymmetry is also well accounted for and is smaller than the rough estimate made above because, as shown in Fig. 3, the surface recombination velocity is already reduced at zero bias for the high-excitation intensities used here.

Here, we have neglected the possible spin dependence of $\Delta\varphi$ (Ref. 17) and, hence, S, caused by spin injection into the subsurface depletion layer. A spin dependence of S should increase $\delta n_s/n_s$ as known for bulk spin-dependent recombination.[31] Conservation of spin currents shows that the relative change of $\delta n_s/n_s$ depends on the balance between the spin relaxation (time T_{1s}) and the lifetime of electrons trapped at surface centers. The upper limit for the relative modification of the spin asymmetry is less than 10^{-3}. This is found by taking the value obtained from the PL measurements for T_{1s} for conduction electrons $T_1 = 0.16$ ns.[18] The hole capture cross section $\sigma_p = 2 \times 10^{-18}$ m^2 is taken to be equal to the maximum value obtained for a large variety of midgap centers.[32] Finally, a possible spin dependence of the tunnel matrix element also has been neglected. While such a

dependence is unknown, good agreement between the model and the experimental results of Fig. 2 indicates that it does not play a crucial role.

In conclusion, the spin dependence of the tunnel current of conduction photoelectrons into a magnetic metal clearly has been demonstrated. In mechanical contact, the bias dependence of A depends on the spin-dependent metallic density of states *and* on the bias-dependent recombination velocity at the GaAs surface. For larger tunneling distances, the applied voltage principally will be dropped in the tunnel junction so that the band curvature at the GaAs surface no longer changes with bias. As such, S should be constant,[21] thus, permitting spin-resolved spectroscopy of the metallic surface. Provided tunneling still occurs from conduction-band states, the ideas developed here should also be valid for GaAs tips excited from the rear. In this case, surface chemical and electronic passivation could significantly improve the electronic-spin polarization and the chemical stability, although this has only started to be addressed recently.[18,30] Bearing these challenges in mind, the present Rapid Communication may finally open the way to spin-dependent tunneling spectroscopy and SPSTM as proposed by Pierce more than 20 years ago.[9]

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