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Spin dependent photoelectron tunnelling 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 direction. In contrast, on a (nonmagnetic) Gold film $A \approx 0\%$.

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Since the initial discovery of spin dependent tunnelling between a magnetic metal and a superconductor [1] and subsequently between two magnetic metals, [2, 3] spin dependent tunnelling has been extensively studied in fixed, all-solid junctions. This is because such studies reveal details of surface magnetism and also because tunnel junctions, in particular metallic magnetic tunnel junctions, [4] are technologically important.[5] Tunnelling from ferromagnetic and anti-ferromagnetic tips has been successfully employed to observe magnetic ordering in metals down to the atomic scale.[6] Similarly, spin polarized tunnelling 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 these cases the transient spin polarization of the post-tunnel electrons is measured via the circular polarization of the resulting luminescence. In principle the reverse process should also be possible. The tunnel current of spin polarized photoelectrons into a ferromagnetic surface should 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 tunnelling microscopy (SPSTM).[9] However, despite significant experimental work, [10, 11] the effect has never been convincingly demonstrated, with experimental difficulties attributed to parasitic optical effects yielding apparent spin dependent tunnelling, even on nonmagnetic surfaces.[11, 12]

Here we demonstrate the spin dependence of the tunnel photocurrent, $I_t^{ph}(\sigma^\pm)$, from p^+ GaAs under circularly-polarized light excitation into ultra-thin Cobalt films magnetized out-of-plane. In contrast to previous works[10–12] spin-polarized electron injection is performed from epitaxial lift-off thin GaAs films deposited using an original microfluidic method on pre-metallized quartz, with an overhanging cantilever of $65\mu\text{m}$ length (see bottom inset, Fig. 1).[13] As shown in the upper

inset of Fig. 1, the photocarriers are generated at the rear (non tunnel) surface, and then diffuse across the film before tunnelling (the film thickness, of $3\mu\text{m}$, is comparable with the charge and spin diffusion lengths for a doping level of $N_A \approx 10^{18} \text{ cm}^{-3}$ and larger than the absorption depth, $1\mu\text{m}$, for the $h\nu = 1.59 \text{ eV}$ pump light used here). The cantilevers are pressed into mechanical contact with the metal surface, as detected using the reflected part of the incident laser beam with a quadrant photodiode, so that tunnelling of photoelectrons occurs over a relatively large contact area through an interfacial oxide layer of homogeneous thickness. This simple, one-dimensional geometry i) avoids poorly controlled direct light excitation at the tip apex,[12] ii) results in a photocurrent which, unlike front surface excitation,[14] does not directly depend on tunnel bias, iii) reduces instabilities due to changes of interfacial chemistry observed for tunnelling from tips,[15] and provides a stable tunnel interface for up to 30 minutes in air at room temperature.

Tunnel injection was performed into an ultrathin Co(0001) layer (thickness ≈ 5 monolayers) epitaxially grown by electrodeposition on an atomically flat Au(111) buffer layer on Si(111).[17] The Co surface was passivated by chemisorbing CO which renders the surface resistant to oxidation in dry air and quenches empty surface states.[18] As shown (Fig. 2A) by the square magnetization loop measured with the field applied perpendicular to the surface (using the polar magneto optical Kerr effect) averaged over 1 mm^2 , these passivated Co/Au(111) ultra-thin films present a strong perpendicular anisotropy with a coercive field smaller than 200 Oe . The full zero field remanence of the magnetization after application of a magnetic field larger than the coercive field, indicates that the sample is essentially composed of a single domain whose lateral extent is larger than the contact area through which tunnelling occurs. The photoelectron polarization in the cantilevers has been analyzed using polarized luminescence (PL).

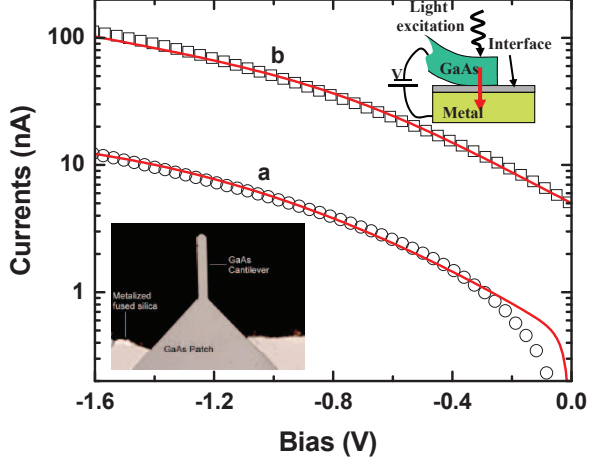


FIG. 1: The top right schematic describes the principle of the experiment in which photoelectrons injected from the rear face of a free standing GaAs layer diffuse to the front face before tunnelling. Also shown, bottom left, is an optical microscope image of the overhanging GaAs layer deposited onto a metallized quartz substrate. Curves a and b correspond to the tunnel dark and photocurrent bias dependences for tunnelling into Cobalt, respectively. The solid, red lines correspond to the calculations of the tunnel currents using a model (Ref. 16) describing tunnelling of photoelectrons into metals.

The σ^\pm polarized PL spectra $[I_{PL}(\sigma^\pm)]$ of the cantilever at a low light intensity of 50 W/cm^2 ($h\nu = 1.59 \text{ eV}$) are shown in curves a and b of Fig. 2B, respectively. As known for p^+ GaAs, the structure near 1.39 eV is due to acceptor-related recombination[19] and that the above bandgap luminescence degree of circular polarization, $[I_{PL}(\sigma^+) - I_{PL}(\sigma^-)]/[I_{PL}(\sigma^+) + I_{PL}(\sigma^-)]$ is equal to 8% as seen from curve c. This polarization corresponds to an average over all photo-electrons in the cantilever. Using this value and by numerically solving the spin diffusion equation, a spin polarization of tunnelling electrons of the order of 16% can be inferred[13] as well as a spin-lattice relaxation time for conduction electrons of 0.16 ns, in good agreement with independent measurements on doped GaAs.[20]

For the investigation of spin dependent tunnelling, the circular polarization of the pump light excitation (5 mW focussed to a spot of about $10 \mu\text{m}$ diameter) is switched by a Pockels' cell. A 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 of duration 12 ms are performed. One scan is performed in the dark and the other one under σ^+ illumination. The tunnel photocurrent $I_t^{ph}(\sigma^+)$ 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.

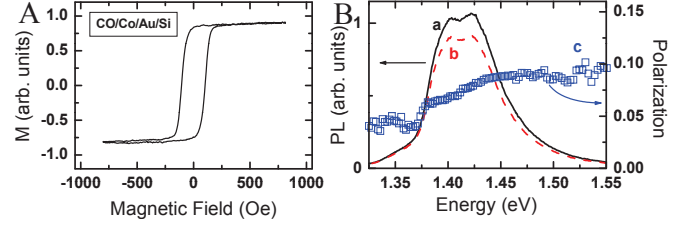


FIG. 2: (A) The magnetic field dependence of the magnetization perpendicular to the surface of the Cobalt film as measured using the polar magneto-optical Kerr effect. (B) Curves a and b show the spectra of the σ^\pm polarized components of the cantilever luminescence under circularly-polarized excitation. Curve c shows the polarization of the spectrum, about 8% for band-to-band emission.

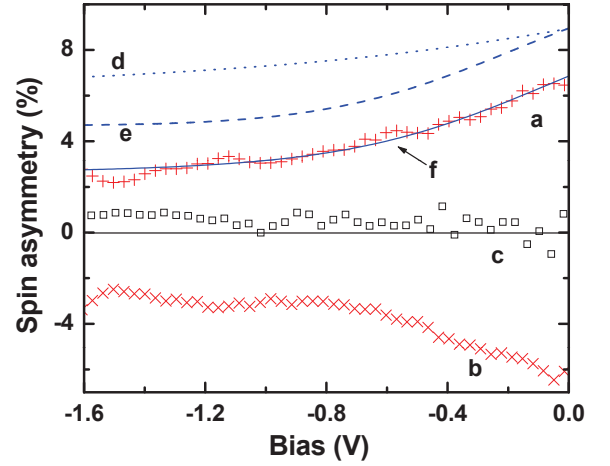


FIG. 3: Curves a and b show the measured bias dependence of the spin asymmetry of the tunnel photocurrent into magnetized Cobalt before and after reversal of the magnetization by the transient application of a magnetic field. Curve c is the asymmetry measured on a nonmagnetic Gold surface. The calculated spin dependence of the metallic density of states and of the photoelectron spin polarization, after division by factors of 10 and 1.3 respectively, are shown in curves d and e. The calculated asymmetry, shown in curve f, is in excellent agreement the measured dependence.

This procedure, lasting about 0.25 s, gives the bias dependence of the spin asymmetry factor A , defined by $A = [I_t^{ph}(\sigma^+) - I_t^{ph}(\sigma^-)]/[I_t^{ph}(\sigma^+) + I_t^{ph}(\sigma^-)]$. A may also be written [2]

$$A = \frac{\delta\rho_m}{\rho_m} \frac{\delta n_s}{n_s}, \quad (1)$$

where δX symbolizes the difference of the quantity X between + and - spins, quantized along the direction of light excitation. ρ_m and n_s are respectively the total metallic density of states at the tunnel energy and the concentration of the tunnelling electrons. Using, as shown above,

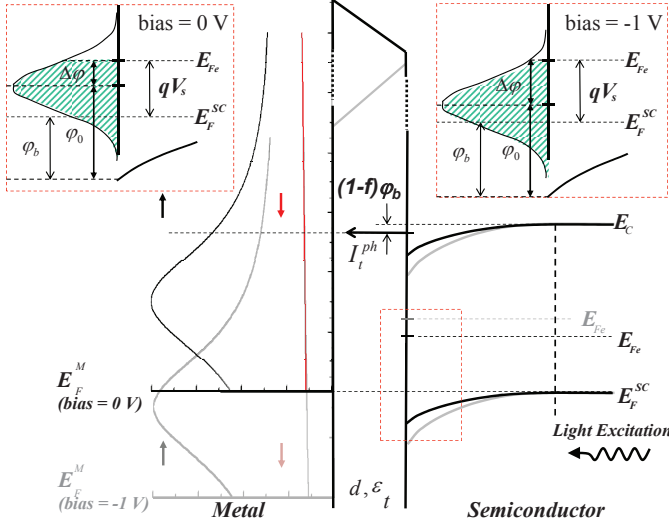


FIG. 4: Energy band structure for spin polarized tunnelling into Cobalt. The injection energy $E_g - (1-f)\phi_b$ is shown along with realistic representations of the densities of states of the majority (black) and minority (red) spins. The black (gray) lines represent the case where the applied bias is 0V (-1 V) and indicates that the spin-polarized conduction electrons are injected into the top half of the 3d minority spin band. The top left (right) inset shows the semiconductor surface density of states at an applied bias of 0V (-1 V).

$\delta n_s/n_s \approx 16\%$, and $\delta \rho_m/\rho_m \approx 70\%$ about 1 eV above the Fermi energy[18], an asymmetry of the order of 10 % is anticipated using Eq. 1.

The results averaged over 100 measurement cycles are shown in Fig. 1 and Fig. 3. Curve a of Fig. 1 shows the dependence of the dark current as a function of reverse bias applied to the GaAs cantilever and curve b shows that of the additional current, I_t^{ph} , induced by the light excitation. This current increases nonexponentially up to about 100 nA. Curve a of Fig. 3 shows that A varies from 6% at zero bias to 2% at a reverse bias of -1.6 V. The non zero value of A is due to a spin dependence of the tunnelling current since i) reversal of the magnetization of the Cobalt layer by transient application of a magnetic field larger than the coercive field induces a change of sign of the asymmetry 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 result in an asymmetry that is always smaller than 1% (curve c) and approximately 0% for zero bias. Moreover the measured asymmetry is similar to the above rough estimate.

A more quantitative interpretation of these results uses a general model recently developed for tunnel injection of photoelectrons into metals.[16] The excellent agreement between the calculated (red lines, Fig. 3) and measured bias dependences indicates that the dominant contribution to the tunnel photocurrent comes from con-

duction electrons. The injection energy is almost bias-independent and close to that of the bottom of the conduction band in the bulk since the energy loss, $(1-f)\phi_b$, in the depletion layer (see Fig. 4) is smaller than 150 meV. (The surface barrier $\phi_b \approx 0.3\text{eV}$ under light excitation and the numerical factor f is larger than about 0.5 because of surface quantization.) The energy dependence of the total Cobalt density of empty states at this injection energy cannot explain the nonexponential bias dependence of I_t^{ph} . [18] In the same way, as shown in curve d of Fig. 3, $\delta \rho_m/\rho_m$ calculated using the known spin dependent density of empty states, only decreases by 25% which, using Eq. 1, cannot explain the measured bias dependence of A . The decrease of A must therefore be dominated by $\delta n_s/n_s$.

The decrease of $\delta n_s/n_s$ and the nonexponential increase of the tunnel photocurrent are caused by the same effect, namely unpinning of the surface Fermi level.[16] As seen in Fig. 4, the application of a bias changes the semiconductor surface charge and shifts the electron quasi-Fermi level away from midgap by a quantity $\Delta\phi$ which is obtained by charge neutrality.[16] The surface recombination velocity is $S = S_0 \exp(-\Delta\phi/k_B T)/D(\Delta\phi)$ where S_0 is the value of S for $\Delta\phi=0$ and $D(\Delta\phi)$ is the relative decrease of the density of surface states.[21] The resulting bias-induced decrease of S results in an increase of the effective lifetime of the tunnelling electrons which increases their concentration and increases the spin polarization losses by spin-lattice relaxation.

The tunnel photocurrent is proportional to n_s and to the tunnel probability, for which the expressions are found in Ref. 16. Calculation of $\delta n_s/n_s$ is performed by solving the equations for spin and charge diffusion[22] from the rear surface to the plane of injection. For a cantilever of thickness l , in the limit of large recombination at the rear surface and of small absorption length, one finds

$$\frac{\delta n_s}{n_s} = \pm 0.5 \sqrt{\frac{\tau_s}{\tau}} \frac{\sinh(l/L)}{\sinh(l/L_s)} \frac{1 + S/v_d}{1 + aS/v_d} \quad (2)$$

for σ^\mp polarized light excitation, respectively. Here τ and τ_s are the bulk electron lifetime and spin lifetime, L and L_s are the charge and spin diffusion lengths, $v_d = (D/L) \coth(l/L)$ is the effective charge diffusion velocity, D is the diffusion constant and $a = (L_s/L) \coth(l/L) / \coth(l/L_s)$ is the ratio of v_d to the equivalent spin diffusion velocity (here $a < 1$ since $L_s < L$).

The bias dependences of the tunnel photocurrent, of the dark current and of A are calculated using the model of Ref. 16. The work function for passivated Cobalt is 6 eV,[23] and the dielectric constant of the tunnel gap is equal to 10, close to that of both Gallium Oxide[24] and Cobalt Oxide.[25] The spin diffusion length is $0.6\mu\text{m}$, i.e. close to independent estimates.[26] As in Ref. 16,

other parameters for non polarized tunnelling have values taken from the literature. Good agreement with the data is obtained for $0.6 < f < 1$ when the tunnel distance is adjusted between 0.6 nm and 0.75 nm. The calculated curves in Fig. 1 and Fig. 3 correspond to $f \approx 0.9$ and $d = 0.74$ nm. As seen in Fig. 1, the calculation correctly predicts the bias dependence of the tunnel dark current and photocurrent. Note that these dependences appear to be quite similar since both are determined by the degree of unpinning of the semiconductor surface Fermi level. Curve e of Fig.3 shows the calculated decrease of the polarization of injected electrons. The bias dependence of A calculated using Eq. 2 is shown in curve f 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 $\Delta\varphi$ is non negligible for the high excitation intensities used here.

We have neglected here the spin dependence of the photovoltage and therefore of $\Delta\varphi$, [10] caused by spin injection into the subsurface depletion layer. This should induce a spin dependence of the surface recombination velocity which, as for bulk spin dependent recombination, [27] increases $\delta n_s/n_s$. Conservation of spin currents shows that the relative change of $\delta n_s/n_s$ depends on the balance between the spin lattice relaxation (time T_{1s}) and the lifetime of electrons trapped at surface centers. An upper limit for this effect, found by taking for T_{1s} equal to the spin relaxation time of conduction electrons (0.16 ns), and a hole capture cross section $\sigma_p = 2 \times 10^{-18} \text{ m}^2$ equal to the maximum value obtained for a large variety of midgap centers, [28] indicates that the relative modification of the spin asymmetry is less than 10^{-3} . Finally, a possible spin dependence of the tunnel matrix element has also been neglected. While such a dependence is unknown, the 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 has been clearly demonstrated. In mechanical contact, the bias dependence of A is caused by the decrease of the electron spin polarization due to the decrease of the surface recombination velocity resulting from the unpinning of the quasi electron Fermi level. Spin injection concerns electrons of well-defined energy (comparable with $k_B T$) and this observation may finally, for larger tunnel distances where the surface recombination velocity is nearly constant, [16] open the way to spin-dependent tunnelling spectroscopy (SPSTS) and SPSTM of magnetic metals as proposed by Pierce more than 20 years ago. [9]

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