Observation of Spin-Polarized-Electron Tunneling from a Ferromagnet into GaAs

Santos F. Alvarado and Philippe Renaud

IBM Research Division, Zurich Research Laboratory, Säumerstrasse 4, CH-8803 Rüschlikon, Switzerland (Received 25 November 1991)

(Received 25 November 1991)

Experimental evidence is presented for the tunneling of polarized electrons from the apex of a ferromagnetic Ni tip into GaAs(110). The polarization is found to be negative and of highest magnitude at very low injection energies, which shows that highly polarized *minority* 3d electrons are preferentially extracted from the Fermi level of the tip.

PACS numbers: 75.30.Pd, 73.40.Gk, 75.70.Cn, 78.55.Cr

Ten years after its invention by Binnig and Rohrer the scanning tunneling microscope (STM) [1] has proven to be an extremely useful instrument for research in a vast range of areas and topics [2]. It also marked the beginning of a new era in the study of nanometer-sized structures. Recently, the detection of electron-spin-polarization effects in STM has received some attention. Manassen et al. [3] reported on beautiful experiments on the observation of individual paramagnetic spins in oxidized silicon surfaces. More recently, Wiesendanger et al. [4] reported on STM experiments using a CrO_2 tip and a Cr(001) single crystal. Here we report on STM experiments demonstrating tunneling of spin-polarized electrons from a ferromagnetic tip into a semiconductor. The experiment is based on the measurement of the circular polarization of the recombination luminescence excited by electrons tunneling from a ferromagnetic Ni tip into GaAs(110). In fact, GaAs acts as a spin detector [5] by employing the reverse of the optical orientation of spins [6], so-called optical pumping. This effect is used, for example, for photoemission of spin-polarized electrons from negative-electron-affinity GaAs by using circularly polarized light for excitation [7]. The advantage of the present approach is that it allows the observation of electron-spin-polarization effects without the interference of topographic features, e.g., roughness, steps, etc., of the sample under study. By definition the polarization of light is a normalized quantity. Thus the detection of the spin-polarization effects through it does not depend critically on the tunneling current or on its fluctuations. Furthermore the spin-polarization effects can be observed even in an external magnetic field. We note that Brechet et al. [8] were the first to report preliminary results on the injection of spin-polarized electrons from vacuum into GaAs with reduced electron affinity. Similar experiments were reported by Fromme et al. [9].

The process of injection and recombination of charge carriers from atomically clean Ni tips into III-V compounds has been studied in detail [10]. The GaAs samples were doped with Zn at a concentration of $p=10^{19}$ cm⁻³. We also used GaAs grown by molecular-beam epitaxy, Be doped at $p=10^{18}$, 3×10^{18} , and 10^{19} cm⁻³. Clean atomically flat GaAs(110) surfaces were produced by cleaving *in situ*. The high doping level of the GaAs

was chosen in order to position the Fermi level close to the Γ_8 point, the uppermost of the spin-orbit-split valence bands. Thus a sizable concentration of holes is available, making possible the radiative recombination of minority charge carriers injected at low energies. Ni tips were made by electrochemically etching polycrystalline wire with diluted HCl. The shape of their shaft and apex was characterized before and after the experiments by inspection with a scanning electron microscope. The tips exhibit typical cone angles of 9° to 15° and apex radii of 40 to 70 nm. Cleaning in the ultrahigh vacuum (UHV) chamber was done by Ne-ion bombardment at 1 keV and subsequent heating to 890 K for about 15 min. The crystal orientation of the apex of the tip was not determined. The tip was magnetized in situ by means of an electromagnet (see Fig. 1). The measurements were taken with the tips remanently magnetized. The orientation of the magnetization of the apex side of the tips was determined by measuring the stray magnetic field with a Hall



FIG. 1. Experimental setup. See text.

probe gaussmeter. It is known that the magnetization of the micrometer-sized region at the apex of the tip is preferentially oriented along the tip's axis [11]. The results below show that we are now able to flip this magnetic domain by 180°. Additional experimental details are given in Refs. [10,12].

The experimental setup is illustrated in Fig. 1. In the present work no attempt was made to observe whether there is any difference between tunneling into Ga or As sites. The results thus represent an average for electron injection at different lattice sites. Light passes through a Pockels cell, and then through the linear polarizer. The intensities for $\pm \lambda/4$ retardation, I_+ and I_- , are measured by a photomultiplier in the counting mode. The degree of circular polarization is

$$\rho = (I_{+} - I_{-})/(I_{+} + I_{-}). \tag{1}$$

The circular polarization of the emitted radiation is proportional to the degree of spin polarization of the recombining minority carriers at the fundamental band gap. Because of the relatively large spin orbit splitting at the valence band, $\Delta_{s.o.} = 0.34$ eV, the recombination takes place predominantly at the uppermost valence band Γ_8 . Hence, assuming integration over all electron impulse directions in the semiconductor in the steady-state regime, we measure [7]

$$\rho = \rho_r P_i \cos\theta \,, \tag{2}$$

where $\rho_r = 0.5(1 + \tau/\tau_s)^{-1}$ is the spin-polarization detection sensitivity, τ is the electron lifetime at the bottom of the conduction band, τ_s is the spin relaxation time [13], and $P_i(E_k) = (n_1 - n_1)/(n_1 + n_1)$ is the initial polarization of the electrons injected into the conduction band, with n_1 and n_1 the number of majority and minority electrons of the ferromagnet, respectively. The maximum electron injection energy above the bottom of the conduction band E_C is $E_k = eV_T - E_C$, where V_T is the tunneling potential applied to the tip. θ is the angle between the electron-spin-polarization vector \mathbf{P}_i and the direction of light propagation towards the optical detector. In order to determine the spin polarization P_M associated with the magnetic state of the tip, the circular polarization of the luminescence, ρ^+ and ρ^- , is measured for both polarities of the magnetizing electromagnet. Thus, using Eq. (2), we obtain

$$P_M = \frac{1}{\rho_r \cos\theta} \frac{\rho^+ - \rho^-}{2}, \qquad (3)$$

where $P_M = (P_i^+ - P_i^-)/2$.

Figure 2 shows data collected for two different tips giving clear evidence of the injection of polarized electrons from ferromagnetic Ni into GaAs. Several measurements were performed with different Ni tips and GaAs samples. The inset in Fig. 2 shows a typical luminescence intensity (I) vs E_k curve. Near threshold, $E_k \approx 100$ meV, we measure I = 30 to 100 counts/snA. The interpretation





FIG. 2. Degree of spin polarization vs maximum injection energy of electrons tunneling from a Ni tip into p-doped GaAs(110). The error bars are the statistical error. Inset: The luminescence intensity (*I*, in arbitrary units).

of the I vs E_k curves is discussed elsewhere [10]. The polarization spectra show an increasing polarization magnitude with decreasing E_k . A local minimum of the magnitude of the polarization appears at $E_k \approx 0.7$ eV. For its definitive confirmation, however, further work is necessary. The magnitude of P_M reaches the background level at $V_t = -2.7 \pm 0.2$ eV, i.e., at $E_k = 1.3 \pm 0.1$ eV. This strong decrease of the polarization is not accounted for by the creation of electron-hole pairs, since the threshold for such processes lies at a higher energy [10]. We find that insufficient cleaning of the tip or contamination of the apex caused by prolonged exposure to the residual gases in the UHV chamber, or after tunneling for several hours, would result in a loss of the spin polarization associated with the tip magnetization. This means that a low number of contaminant atoms, maybe even a single one, can drastically influence the spin polarization of the charge carriers tunneling into GaAs. We also observe a polarization contribution that does not show measurable changes upon reversal of the magnetization. This residual polarization is nearly zero at low injection energies and increases with increasing energy. It remains even if the magnetic effect is quenched by tip contamination. We believe that this is a manifestation of the spontaneous polarization P_{Ω} of conduction electrons in GaAs observed in photoemission experiments [14]. This effect is associated with the energy-dependent spin splitting of the conduction band [13]. Preliminary results show that the background can be minimized by orienting the GaAs crystal such that \mathbf{P}_{Ω} lies perpendicular to the optical detection axis. This is the case depicted in Fig. 1 for which \boldsymbol{P}_{Ω} is along the $[\bar{1}10]$ direction [14]. We also expect a precession of \mathbf{P}_M by an angle θ_p about the [110] direction [15]. For small injection energies this effect may be neglected, but for $E_k \gtrsim 1$ eV we might expect precession angles of up to 30°. Finally we note that the luminescence radiation also exhibits a small linear polarization component lying in a direction parallel to the axis of the tip.

To determine P_M from the measured circular polarized luminescence we need to know the quantity ρ_r . Likewise if $P_i \cos\theta$ is known one can, using Eq. (2), determine the ratio τ/τ_s within a region the size of a micrometer or smaller, depending on the diffusion length and dimensions of the semiconductor structure where depolarization takes place. Measurements of the depolarization of conduction electrons in highly Zn-doped GaAs at room temperature give sample-dependent results in the range $\rho_r = 8\%$ to 26% [13,16]. Because of this uncertainty, and lacking a calibration of the GaAs samples used in the present measurements, we can only attempt to estimate a lower limit of $|P_i|$ by taking a reasonable upper limit [17], $\rho_r = 20\%$, and noting that no significant difference in the depolarization is expected between Be- or Zn-doped samples [18]. Hence we determine from Eq. (2) with $\cos\theta = 0.45$ the polarization scale for P_M shown in Fig. 2. The negative sign indicates dominant minority electron injection. From this data we get $P_M = (-31 \pm 5.6)\%$ for $E_k = 300$ meV. The highest circular polarization of magnetic origin we report is $\rho_M = (4.35 \pm 0.56)\%$ which gives P_M $=(-48\pm5)\%$. High spin polarization, P=-30%, of minority electrons at the Fermi level was observed in photoemission on Ni(001) by Eib and Alvarado [19]. Those measurements also reveal an abrupt crossover to positive polarization at $h\omega \simeq \Phi + 0.05$ eV, and that the positive polarization peaks at 34% for $h\omega + \Phi \simeq 1$ eV, where Φ is the work function. High negative polarization at the Fermi level was subsequently reported for other low-indexed Ni(hkl) [20,21] surfaces, the (110) being of particular interest because in that case P = -100% was found [21]. The negative spin polarization observed at the Fermi level is predicted by the Stoner-Wohlfahrt-Slater theory of ferromagnetism [22]. A negative spin polarization of magnitude < 5% at the Fermi level for various Ni(*hkl*) surfaces [23] has been found in spin-polarized field emission (SPFEM) experiments. Calculations are consistent with the observed spin polarization of the field-emitted electrons although in the case of Ni(111) a higher magnitude of the polarization is predicted [24]. Rau and Eichner [25] have reported an electron-capture spectroscopy study of Ni(hkl) surfaces that indicate very high negative polarization of electrons from the Fermi level, up to $P_{\rm Ni} = -95\%$, one exception being the Ni(120) surface which yields 15%.

It should be mentioned in this context that Thedrow and Messervey [26] reported positive polarization in spin-dependent tunneling between thin films of superconducting Al and polycrystalline Ni thin films. Those findings support theoretical considerations of the spindependent tunneling from transition-metal ferromagnets which conclude that the tunneling current is predominantly due to s-d hybridized bands exhibiting positive polarization [27], despite the much greater density of dstates of minority type at the Fermi level. In the present STM results, however, the magnitude and sign of the measured polarization seem to indicate that, for our particular experimental conditions, tunneling from the minority spin 3d states at the Fermi level is dominant.

We have presented experimental evidence for the tunneling of spin-polarized electrons in STM experiments. The degree of negative spin polarization of the electrons extracted from the Ni tip is large, $P_M = (-31 \pm 5.6)\%$, where the magnitude is an estimated lower limit. The sign indicates that minority spin electrons are a dominant contribution of the tunneling current. In light of the present results the low magnitude of the negative polarization observed in the SPFEM experiments [23] with Ni tips becomes a puzzling yet interesting finding. The difference in magnitude and/or sign of the polarization determined using different techniques could have many different explanations. Sample quality and experimental conditions can surely affect the outcome. More fundamentally one may consider the decay of the spin polarization of the electron cloud P(z) at ferromagnetic surfaces, which was predicted for the case of increasing the distance z from the surface into vacuum [28]. Interestingly, SPFEM calculations show that the spin polarization of the emitted electrons depends critically on the shape of the surface-barrier potential [24]. In STM experiments the distance between the tip and the sample can be exactly adjusted within the range of interest, e.g., by changing the tunneling current, to probe the polarized electron cloud and test theoretical predictions. We propose that the tunneling effect reported here may be useful to image magnetic domains and to realize spin-polarized spectroscopy with nanometer resolution as well as to study small magnetic particles. Furthermore it opens new possibilities for the study of the transport of polarized electrons in semiconductors.

Many thanks to F. Meier for help in cross-checking the calibration of our circular polarizer and magnetization arrangement. Thanks to M. Lutz, F. Meier, and H. C. Siegmann for fruitful conversations. Thanks to H. Meier for providing GaAs samples. We are indebted to B. Weiss for technical help. We are grateful to U. Dürig and D. Pohl for support. Thanks to H. Rohrer for making this project possible and for fruitful discussions.

- G. Binnig, Ch. Gerber, E. Weibel, and H. Rohrer, Phys. Rev. Lett. 50, 120 (1983).
- [2] Proceedings of the International Conference on STM, Interlaken, Switzerland, 1991 [Ultramicroscopy (to be published)].
- [3] Y. Manassen, R. H. Hamers, J. E. Demuth, and A. J. Castellano, Jr., Phys. Rev. Lett. 62, 2531 (1989).
- [4] R. Wiesendanger, H.-J. Güntherodt, G. Güntherodt, R. J. Gambino, and R. Ruf, Phys. Rev. Lett. 65, 247 (1990).
- [5] H. C. Siegmann, Europhys. News 14, 9 (1983).
- [6] M. I. Dyakonov and V. I. Perel, in *Optical Orientation*, edited by F. Meier and B. P. Zacharchenya, Modern Problems in Condensed Matter Sciences Vol. 8 (North-

Holland, Amsterdam, 1984), p. 11.

- [7] D. T. Pierce and F. Meier, Phys. Rev. B 13, 5484 (1976).
- [8] Ph. Brechet, M. Campbell, G. Lampel, and D. Paget, in Proceedings of the Nineteenth International Conference on Semiconductors, 1988, edited by W. Zawadski (Institute of Physics, Polish Academy of Science, Warsaw, 1988), Vol. 2, p. 1369.
- [9] B. Fromme, G. Baum, D. Glöckel, and W. Raith, Phys. Rev. B 40, 12312 (1989).
- [10] S. F. Alvarado, Ph. Renaud, D. L. Abraham, Ch. Schönenberger, D. J. Arent, and H. P. Meier, J. Vac. Sci. Technol. B 9, 409 (1991); Ph. Renaud and S. F. Alvarado, Phys. Rev. B 44, 6340 (1991).
- [11] Ch. Schönenberger and S. F. Alvarado, Z. Phys. B 80, 373 (1990).
- [12] D. L. Abraham, A. Veider, Ch. Schönenberger, H. P. Meier, D. J. Arent, and S. F. Alvarado, Appl. Phys. Lett. 56, 1564 (1990).
- [13] G. E. Pikus and A. N. Titkov (Ref. [6]), p. 133.
- [14] S. F. Alvarado, H. Riechert, and N. E. Christensen, Phys. Rev. Lett. 55, 2716 (1985).
- [15] H. Riechert, S. F. Alvarado, A. N. Titkov, and V. I. Safarov, Phys. Rev. Lett. 52, 2297 (1984); in *Proceedings* of the Seventeenth International Conference on the Physics of Semiconductors, edited by J. D. Chadi and W. H. Harrison (Springer, New York, 1985), p. 1361.
- [16] G. Lampel and M. Eminyan, J. Phys. Soc. Jpn. Suppl. A 49, 627 (1980); H.-J. Drouhin, C. Hermann, and G. Lampel, Phys. Rev. B 31, 3872 (1985); K. Zerrouati, F. Fabre, G. Bacquet, J. Bandet, J. Frandon, G. Lampel,

and D. Paget, Phys. Rev. B **37**, 1334 (1988); R. C. Miller, D. A. Kleinman, W. A. Nordland, Jr., and R. A. Logan, Phys. Rev. B **23**, 4399 (1981).

- [17] A lower limit $\rho_r \approx 7\%$ is estimated from the maximum polarization measured at $E_k \approx 1.5$ eV by taking $P_i = 1$.
- [18] S. F. Alvarado, F. Ciccacci, S. Valeri, M. Campagna, R. Feder, and H. Pleyer, Z. Phys. B 44, 259 (1981).
- [19] W. Eib and S. F. Alvarado, Phys. Rev. Lett. 37, 444 (1976).
- [20] E. Kisker, W. Gudat, M. Campagna, E. Kuhlmann, E. Hopster, and E. Moore, Phys. Rev. Lett. 43, 966 (1979).
- [21] E. Kisker, W. Gudat, E. Kuhlmann, R. Clauberg, and M. Campagna, Phys. Rev. Lett. 45, 2053 (1980).
- [22] E. P. Wohlfahrt, Phys. Lett. 36A, 131 (1971).
- [23] M. Landolt and M. Campagna, Phys. Rev. Lett. 38, 663 (1977).
- [24] A. Modinos, Field, Thermionic and Secondary Electron Emission Spectroscopy (Plenum, New York, 1984), p. 158.
- [25] C. Rau and S. Eichner, Phys. Rev. Lett. 47, 939 (1981);
 C. Rau, J. Magn. Magn. Mater. 30, 141 (1982).
- [26] P. M. Thedrow and R. Messervey, Phys. Rev. Lett. 26, 192 (1971); Phys. Rev. B 7, 318 (1973); J. S. Moodera and R. Messervey, Phys. Rev. B 29, 2943 (1984).
- [27] J. A. Hertz and Koya Aoi, Phys. Rev. B 8, 3252 (1973);
 M. B. Stearns, J. Magn. Magn. Mater. 5, 167 (1977); R. Messervey, J. Appl. Phys. 61, 3709 (1987).
- [28] J. S. Helman and W. Baltensperger, Mod. Phys. Lett. B (to be published).