

Direct measurement of the spin diffusion length by Andreev spectroscopy

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The application of point contact Andreev spectroscopy for the measurement of the spin diffusion length is demonstrated by studying the spin relaxation in platinum thin films grown on the top of a ferromagnetic cobalt layer. Using this local probe technique, the temperature dependence of the spin diffusion length was determined, and various sources of the spin relaxation in platinum were identified. At low temperatures the spin lifetime is found to be three orders of magnitude larger than the momentum lifetime. The applied method is selectively sensitive to spin-flip processes even in the presence of a huge background of momentum scattering. © 2011 American Institute of Physics. [doi:10.1063/1.3593959]

The spin transport through nonmagnetic metals has attracted special attention since the discovery of the giant magnetoresistance (GMR) (Refs. 1 and 2) in multilayer structures. Nonequilibrium spin polarization exists in a paramagnetic metal on the length scale of the spin diffusion length, l_s , which is controlled by the different contributions to the spin relaxation, including electron-phonon interactions and scattering on magnetic impurities.³ In metallic systems the spin transport greatly influences the amplitude of the GMR and determines the efficiency of the spin injection and detection.⁴

In this letter we address the spin transport in Pt thin layers using point contact Andreev spectroscopy. Measuring the decay of the local spin polarization on the top of a Pt thin film evaporated on a ferromagnetic Co layer, we determine the spin diffusion length and the relaxation time in the temperature range of 1.5–8.0 K. The measurement geometry is the same as that of the current perpendicular to the plane (CPP) measurements in multilayer structures, in which Pt is often used as nonmagnetic spacer.^{5,6}

In superconductor-normal (SN) metal junctions, two electrons with opposite spin are transferred from the normal to the superconducting electrode by the Andreev reflection. This process, however, is suppressed by the spin imbalance in the normal metal. Based on this property, point contact Andreev spectroscopy has become an important tool to detect local spin polarization of the charge carriers, P_c .⁷ It has been shown that for small enough ballistic junctions the bulk spin polarization of the charge carriers in a ferromagnetic system can be reliably determined from the bias dependence of the differential conductance, $G(V)$.^{8–10}

The spin relaxation in a paramagnetic metal is studied on the model system of thin film Co/Pt samples prepared using molecular beam epitaxy with a base pressure of 1.7×10^{-10} mbar. First a Co layer of 100 nm was evaporated on the Si substrate, then the Pt layer was deposited with a varying thickness d , for different samples. Both Co and Pt were evaporated from an electron gun at a rate of 0.022 nm/s. In this letter, we investigate $d=20, 40, 80$ nm layer thickness

expected to be around the spin diffusion length in the temperature range of our experiments.

Point contact Andreev reflection experiments were carried out using our custom built system. Mechanically sharpened Nb tips were used as the superconducting counterelectrode gently touching the top of the Pt layer. High stability point contacts were created using a screw mechanism for coarse and a piezo actuator for fine tuning. In order to preserve the well-defined Pt layer thickness and to prevent indentation of the Nb tip to the thin layer, the junction diameter was continuously controlled during the measurement and was always kept below 10 nm, as calculated from the junction resistance using the Wexler equation.^{11,12} The upper temperature limit of our measurements is defined by the gradual decrease in the $\Delta(T)$ BCS gap of Nb while the thermal smearing $k_B T$ increases. With the bulk $T_c=9.2$ K of Nb, we were able to make reliable measurements up to 8.0 K.

Typical $G(V)$ curves are shown in Fig. 1 fitted using the

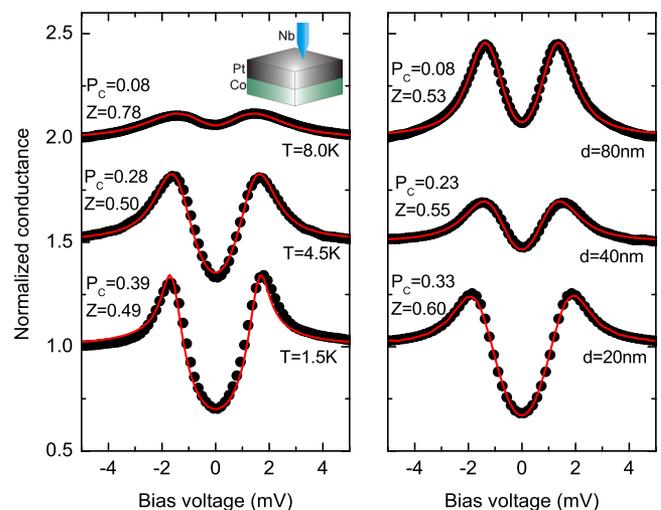


FIG. 1. (Color online) Measured normalized differential conductance curves ($G_{\text{normal}}^{-1} \times dI/dV$). The curves are shifted for clarity. Solid lines show fitted curves using the modified BTK theory. The fitted P_c and Z values are provided for each curve. Left panel: different temperatures with a fixed $d=20$ nm layer thickness. Right panel: different layer thicknesses at a fixed intermediate temperature of $T=3.5$ K. Inset shows the measurement geometry.

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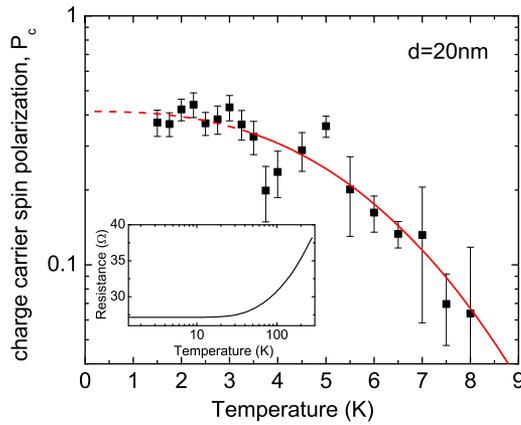


FIG. 2. (Color online) Charge carrier spin polarization as a function of the sample temperature in the range of 1.5–8.0 K for $d=20$ nm Pt layer thickness. The line plot shows fitting with a power law expression of the spin diffusion length. Error bars represent the scattering of multiple-independent junctions. Inset shows the four point probe resistance of a 20 nm thick Pt film between 1.5 K and room temperature.

modified Blonder-Tinkham-Klawijk (BTK) theory^{7,8,13} including the charge carrier spin polarization, P_c and the interface barrier strength Z , the latter measuring the backscattering of the electrons on the SN interface due to inhomogeneities of the metallic surface, or the Fermi velocity mismatch.⁸ We note that the typical value of $Z \approx 0.5$ corresponds to an average transmission of $\tau = (1 + Z^2)^{-1} \approx 0.8$. Such a high transmission is essential for the accurate determination of P_c , as for $\tau \ll 1$ the second order Andreev reflection process is always suppressed regardless of the spin polarization. It was also shown that for high transparency junctions the fitted value of P_c is not sensitive to the model^{8,9,13} describing the transport on the SN interface.¹⁰ In the left panel of Fig. 1 the temperature dependence of the $G(V)$ curves is plotted at a fixed layer thickness of $d = 20$ nm. In the right panel, measurement data taken at a fixed temperature of $T = 3.5$ K on Pt layers with thicknesses of $d = 20, 40, 80$ nm are shown. The change in P_c significantly exceeds the uncertainty of the fitting procedure estimated to be $\Delta P_c \approx 0.05$. This shows that the spin diffusion length, l_s , in Pt is indeed in the range of 10 nm in good agreement with earlier CPP spin valve measurements in the same geometry.⁵

In order to gain good statistics, several junctions at different lateral positions on the sample were created and evaluated for P_c . Through this letter we present the mean value of the spin polarization for different contacts, averaging out the fluctuations due to the lateral inhomogeneities of the sample.

First we investigate the temperature evolution of P_c at a fixed layer thickness of $d = 20$ nm. Figure 2 shows that an order of magnitude decay of P_c is found over the temperature range of $T = 1.5$ –8.0 K. In contrast, the four probe resistivity of a single Pt layer of 20 nm thickness, measured as a reference sample, already saturates at much higher temperatures (inset in Fig. 2). The residual resistivity is $\rho_0 = 200$ n Ω m with a relative change in less than 10^{-4} below $T = 10$ K, i.e., the transport lifetime is dominated by elastic scatterings with a temperature-independent contribution to the resistivity. The Andreev reflection measurements, however, are able to selectively resolve the temperature dependence of the spin relaxation.

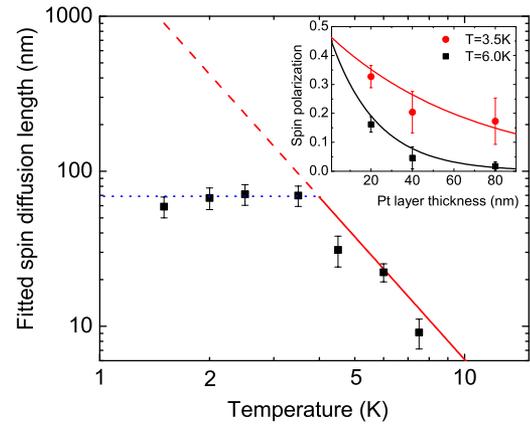


FIG. 3. (Color online) Temperature evolution of the fitted spin diffusion length, l_s . Continuous and dashed line is equivalent to the one in Fig. 2. Individual points are based on measurements with varied Pt layer thickness at a fixed temperature. Dotted line shows the saturation of the spin diffusion length below $T = 3.5$ K. Inset shows measurement data at $T = 3.5$ and 6.0 K with the fitted exponential decay as the function of the Pt layer thickness.

The data are evaluated in Fig. 2 assuming an exponential decay of the spin polarization over the characteristic length of l_s ,

$$P_c(d, T) = P_0 e^{-d/l_s(T)}. \quad (1)$$

Well below the Curie temperature P_0 can be regarded as a constant spin polarization of the Co layer, thus the temperature dependence is solely related to the spin relaxation in the Pt layer. The extrapolated value of $P_0 = 0.41 \pm 0.04$ is in good agreement with earlier Andreev spectroscopy measurements performed on bare Co layers providing P_c in the range of 0.4–0.45.^{8,9}

The data are well fitted with a power law behavior of $l_s \sim T^{-\alpha}$ with an exponent of $\alpha = 2.6 \pm 0.8$ (line graph in Fig. 2). Our results define $l_s = 60_{-19}^{+26}$ nm at $T = 4.2$ K with a gradual decrease to $l_s = 11 \pm 2$ nm at $T = 8.0$ K.

It is important to note that for $d \ll l_s$ the decay of P_c in the thin Pt layer becomes exponentially small thus it cannot be used to determine the value l_s . This is the case at low temperatures for the $d = 20$ nm thick sample, where P_c is close to the saturation value, P_0 . In order to address this problem we made further experiments on thicker Pt layers and independently demonstrated the exponential decay of the spin polarization at fixed temperature values with varying sample thickness.

The results are shown in Fig. 3, where $l_s(T)$, deduced from fitting the data to Eq. (1), is plotted. P_0 was set as a common fitting parameter for all temperatures. In the inset of Fig. 3, the decay of P_c is shown at 3.5 and 6.0 K together with the fitted exponential decay providing the value of l_s . In the main panel, the such determined spin diffusion length is plotted together with the $l_s \sim T^{-\alpha}$ curve obtained in Fig. 2. At $T > 3.5$ K we find a good agreement between the two measurements. At lower temperatures, however, we find a saturation of the spin diffusion length at $l_s = 67 \pm 5$ nm, which could not be recovered in Fig. 2, as in the $d = 20$ nm sample the spin polarization is already close to its maximum value, P_0 , in this temperature range (dashed part of the line plot in Fig. 2).

Using the material parameters of bulk Pt (Refs. 14 and 15) the elastic and spin-flip scattering rate of the electrons can be compared. Based on the residual resistivity, ρ_0

$=200$ n Ω m of the Pt layer, the elastic mean free path is estimated to be $l_m=4$ nm, yielding a transport lifetime of $t_m=l_m/v_F=1.4\times 10^{-14}$ s. In comparison, assuming diffusive transport ($t_m\ll t_s$) the spin lifetime can be expressed by $t_s(T)=l_s^2(T)/D$, where $D=(1/3)v_F l_m$ is the diffusion constant.

Substituting $l_s=67$ nm in the low temperature limit we get $t_s=1.2\times 10^{-11}$ s, thus the ratio of the spin lifetime to the transport lifetime is $\sim 10^3$. At $T=8.0$ K the spin diffusion length decreases to $l_s=11$ nm yielding a spin lifetime of $t_s=3.0\times 10^{-13}$ s, which is 20 times larger than t_m justifying the assumption of diffusive spin transport.

As D can be considered as a constant in the temperature range of our experiments, we can directly connect the measured power law behavior of $l_s\sim T^{-\alpha}$ to earlier calculations¹⁶ of the temperature dependence of the spin lifetime $t_s\sim T^{-2\alpha}$. The observed decay of the spin lifetime above $T=3.5$ K with $2\alpha=5.2$ is in good agreement with $2\alpha=5$ expected for spin relaxation caused by electron–phonon scattering via the spin–orbit coupling.¹⁷ The saturation observed in the low temperature limit is attributed to the contribution of the elastic scattering on the defects mediated by the spin–orbit interaction.

We note that the saturation of the spin lifetime occurs at a considerably lower temperature ($T\approx 3.5$ K) than that of the transport lifetime ($T\approx 20$ K) meaning that t_s/t_m is not a temperature-independent constant. However the ratio of t_s and t_m is not expected to be the same for the scattering on lattice defects as for the electron–phonon interaction. Accordingly, the saturation temperature, where the temperature-independent contribution of the scattering on defects becomes dominant, is different for the spin relaxation and the charge transport. The presence of spin hot spots¹⁶ on the Fermi surface of Pt (Ref. 18) introduces a strong anisotropy of the spin relaxation that can further enhance this difference.

In conclusion, we investigated the decay of the spin polarization in Pt thin films evaporated on the top of a ferromagnetic Co layer. Using point contact Andreev spectroscopy we probed the spin polarization locally on the surface of the Pt film and investigated it both as a function of the temperature and the Pt layer thickness. These measurements enable a direct determination of the spin diffusion length in the nonmagnetic Pt layer. The temperature dependence of the spin lifetime is attributed to the elastic and electron–phonon

scattering mediated by the spin–orbit coupling.

Our results demonstrate that a simple transport measurement based on Andreev spectroscopy is a sensitive probe to study spin relaxation, even if the spin lifetime is orders of magnitude larger than the transport lifetime. This method carries a unique possibility to be implemented as a local probe technique, and to study spin relaxation in nanostructures locally, on the nanometer scale.

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¹M. N. Baibich, J. M. Broto, A. Fert, F. N. Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, *Phys. Rev. Lett.* **61**, 2472 (1988).

²G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, *Phys. Rev. B* **39**, 4828 (1989).

³J. Bass and W. P. Pratt, *J. Phys.: Condens. Matter* **19**, 183201 (2007).

⁴T. Kimura, J. Hamrle, Y. Otani, K. Tsukagoshi, and Y. Aoyagi, *Appl. Phys. Lett.* **85**, 3501 (2004).

⁵H. Kurt, R. Loloee, K. Eid, J. W. P. Pratt, and J. Bass, *Appl. Phys. Lett.* **81**, 4787 (2002).

⁶A. Rajanikanth, S. Kasai, N. Ohshima, and K. Hono, *Appl. Phys. Lett.* **97**, 022505 (2010).

⁷R. J. Soulen, Jr., J. M. Byers, M. S. Osofsky, B. Nadgorny, T. Ambrose, S. F. Cheng, P. R. Broussard, C. T. Tanaka, J. Nowak, J. S. Moodera, A. Barry, and J. M. Coey, *Science* **282**, 85 (1998).

⁸G. J. Strijkers, Y. Ji, F. Y. Yang, C. L. Chien, and J. M. Byers, *Phys. Rev. B* **63**, 104510 (2001).

⁹F. Pérez-Willard, J. C. Cuevas, C. Stürgers, P. Pfundstein, J. Kopu, M. Eschrig, and H. v. Löhneysen, *Phys. Rev. B* **69**, 140502 (2004).

¹⁰A. Geresdi, A. Halbritter, M. Csontos, S. Csonka, G. Mihály, T. Wojtowicz, X. Liu, B. Jankó, and J. K. Furdyna, *Phys. Rev. B* **77**, 233304 (2008).

¹¹G. Wexler, *Proc. Phys. Soc.* **89**, 927 (1966).

¹²B. Nikolić and P. B. Allen, *Phys. Rev. B* **60**, 3963 (1999).

¹³I. I. Mazin, A. A. Golubov, and B. Nadgorny, *J. Appl. Phys.* **89**, 7576 (2001).

¹⁴T. Dosdale and D. Livesey, *J. Phys. F: Met. Phys.* **4**, 68 (1974).

¹⁵W. W. Schulz, P. B. Allen, and N. Trivedi, *Phys. Rev. B* **45**, 10886 (1992).

¹⁶I. Žutić, J. Fabian, and S. Das Sarma, *Rev. Mod. Phys.* **76**, 323 (2004).

¹⁷R. J. Elliott, *Phys. Rev.* **96**, 266 (1954); Y. Yafet, in *Advances in Research and Applications*, Solid State Physics Vol. 14, edited by F. Seitz and D. Turnbull (Academic, New York, 1963), pp. 1–98.

¹⁸M. Gradhand, M. Czerner, D. V. Fedorov, P. Zahn, B. Y. Yavorsky, L. Szunyogh, and I. Mertig, *Phys. Rev. B* **80**, 224413 (2009).