## VOLUME 32, NUMBER 5

## Angular relation and energy dependence of Andreev reflection

P. A. M. Benistant, A. P. van Gelder, H. van Kempen, and P. Wyder\* Research Institute for Materials, University of Nijmegen, Toernooiveld, 6525 ED Nijmegen, The Netherlands (Received 28 May 1985)

Andreev reflection at an interface between an Ag single crystal and a Pb film is studied by an electronfocusing experiment. Quasiparticles are observed to be reflected within a cone with an angle  $\approx 10^{-4}$  rad centered about the incident direction. The energy dependence of Andreev reflection is found to be consistent with theory. In spite of the nonideal circumstances during the Pb deposition, the probability for Andreev reflection approaches that of a perfect interface.

A conduction electron in a normal metal, reaching a normal-metal-superconductor (N-S) interface, can be reflected as a hole with reversal of the signs of all three velocity components (retroreflection) and of the charge. The occurrence of this so-called Andreev reflection (AR) has been demonstrated by different types of experiments.<sup>1-8</sup> However, generally not all electrons will be reflected in this way at a N-S interface: depending on the energy of the incoming electrons and on the presence and strength of a barrier at the interface, transmission and normal reflection of the electrons are also possible. A theoretical analysis based on the Bogoliubov equations is given, for example, in Ref. 9. In this paper we describe an experiment which allows us to study in hitherto unattainable detail the precise relation between the angles of incidence and reflection in AR. With the same experimental configuration we can also measure the energy dependence of the AR probability A and so the barrier strength of the N-S interface. We find that the retroreflection is nearly ideal, however, because of our extremely high angular resolution ( $\simeq 10^{-5}$  rad), we could observe small but significant deviations. (This high angular resolution can also be applied to other problems concerning conduction-electron trajectories.) The energy dependence of A is found to be in agreement with the theoretical expectation. A related experiment has been performed by Blonder and Tinkham,<sup>7</sup> in which the energy dependence of AR was measured. In their experiment the N-S interface, constructed by pressing a superconducting Nb point contact against a Cu plate, served as both "injector" and "reflector." In our experiment, however, the injector and reflector could be individually controlled, and the measured AR signal could be switched off by applying a very small magnetic field ( $\simeq 10^{-5}$  T).

To observe the AR process, electrons were injected into a thin silver single-crystal slab through a Sharvin point-contact junction.<sup>10</sup> For the ideal Sharvin point contact the injected electrons have an energy up to eV above the Fermi energy, where V is the applied voltage across the contact. The other side of the slab was backed by a superconducting Pb layer. As can be seen from Fig. 1, only Andreev-reflected quasiparticles can be focused back on the point contact, and because the reflected particles have opposite charge, their presence will result in an increase of the current through the point contact, known as excess current.

The Ag crystal used for this experiment was a slab spark cut from a 99.9999%-pure single-crystal rod,<sup>11</sup> for which a residual resistance ratio [R(300 K)/R(4.2 K)] of 15000 was found, yielding an electron mean free path l of about 700  $\mu$ m at low temperatures. The sample was polished by mechanical and chemical techniques to a thickness d of about 200  $\mu$ m, i.e., much thinner than the electron mean free path. Next, the sample was annealed for 16 h at 800 °C at a pressure of  $10^{-3}$  Torr of air.<sup>12</sup> Finally, after sputtercleaning the sample in a glow discharge of N<sub>2</sub> for about 20 min at a pressure of  $10^{-1}$  Torr, 1.5  $\mu$ m of Pb was evaporated at a pressure of  $10^{-6}$  Torr and a temperature of 50 °C. The point contact consisted of a 50- $\mu$ m-diameter Ag wire with a point of about 1- $\mu$ m diameter etched electrolytically in a NaCN solution.<sup>13</sup>

To obtain sufficiently low magnetic fields ( $\leq 10^{-6}$  T) the insert was placed in a set of two pairs of Helmholtz coils for horizontal and vertical field compensation, respectively. The point of the Ag wire was brought into contact with the crystal surface in the helium bath at a temperature of 1.2 K. The point contacts thus obtained had resistances ranging from 1 to 7  $\Omega$ . The dynamic resistance R(V) = dV/dI was recorded as a function of applied magnetic field B and as a function of applied injection voltage V, using conventional ac modulation and phase-sensitive detection techniques. To check the quality of the point contact the voltage derivative of the resistance  $dR/dV = (d^2V/dI^2)/(dV/dI)$ , was also recorded. For good Sharvin junctions dR/dV is a measure of the electron-phonon interaction function  $\alpha^2 F^{13}$ . To ensure that the electrons were injected ballistically, only junctions producing the correct  $\alpha^2 F$  function<sup>14</sup> were used.

Some typical results (obtained with a 0.08-mA-rms modulation) are displayed in Figs. 2 and 3 as the solid curves. Both curves were measured successively on the same junction, which had a resistance of 2.41  $\Omega$  corresponding to a



FIG. 1. Experimental setup. (a) Andreev reflection at the bottom of the Ag crystal leads to focusing of the AR particles at the point contact; (b) specular reflection gives no focusing at the point contact.

3352



FIG. 2. Point-contact resistance R relative to the resistance in the absence of AR,  $R_S$ , at zero dc voltage as a function of applied magnetic field. The solid curve shows the measured result; the dots are calculated using the distribution function  $P(\gamma/\gamma_0)$  shown in the inset and the AR probability A = 0.7.  $\gamma_0 = b/d = 6.3 \times 10^{-5}$  rad, with b the radius of the point contact and d the thickness of the Ag crystal.

contact radius b of 136 Å.<sup>12</sup> It is clear from Fig. 2 that the maximum change of the resistance R is much less than the factor 0.5 expected in the ideal case. Even if the finite mean free path ( $l \approx 700 \,\mu$ m) is taken into account, the relative resistance should have been 0.71 at zero field instead of the observed 0.9955. One could try to explain this by assuming ideal retroreflection combined with a very small A ( $\approx 0.01$ ), but as will be discussed in the following and more fully in a forthcoming paper,<sup>15</sup> this leads to serious inconsistencies in the description of the resistance as a function both of applied magnetic field and of applied voltage.



FIG. 3. Relative resistance at zero magnetic field as a function of applied voltage. The solid curve shows the measured result; the dashed curve is the result of a calculation with the same conditions as used for the dots in Fig. 2.

All results can be completely understood, however, on the basis of the following model: on AR the quasiparticles are not retroreflected perfectly, but with a velocity distribution narrowly ( $\simeq 10^{-4}$  rad) centered around the ideal direction. The applied magnetic field B moves<sup>8</sup> (and distorts) the spot of Andreev-reflected particles along the top surface of the Ag crystal. So, the R(B) can be considered as a probe which measures the intensity of reflected particles at the point-contact site. From this it follows that the AR probability A can be determined straightforwardly from the  $R/R_s$ vs *B* curve, through the integral  $\int_0^\infty B[1-R(B)/R_S] dB$ , which can be shown<sup>15</sup> to be a simple function of *A* and 2d/l(the  $E_7$  function of Ref. 16), independent of the shape of the velocity distribution. In this expression  $R_S$  denotes the measured Sharvin resistance in the absence of AR at zero dc voltage. In our experiment 2d/l = 0.57 and this leads to A = 0.7. Due to the different Fermi velocities of the normal metal and the superconductor, even for a perfect N-Sinterface, A is smaller than one.<sup>7</sup> For the Ag-Pb interface we estimate<sup>17,18</sup> that  $A_{perfect} = 0.7 \pm 0.2$ . Clearly our N-S interface is very close to perfect.

To calculate R as a function of B we first consider the case of electrons injected with zero energy  $(\epsilon = 0)$ , measured from the Fermi energy  $\epsilon_F$ , and ideal retroreflection. It can be derived by straightforward geometric considerations that at a small field the displacement  $\delta$  of Andreev-reflected particles at the upper Ag surface, in units of the contact radius b, is given by

$$\delta \simeq \frac{d^2}{bR_C \cos^3\theta} [1 + \sin^2\phi (\cos^4\theta - 1)]^{1/2},\tag{1}$$

with  $\theta$  and  $\phi$  the polar angles of the injected quasiparticle and  $R_C$  the cyclotron radius of the quasiparticle orbit in the applied magnetic field, which is in the direction  $\theta = \phi = \pi/2$ . The z axis is perpendicular to the normal-metal surface. All quasiparticles flowing through the point-contact area at the same angle are equally displaced when they reach the surface again after AR, forming an area of the same size as the contact area at a distance  $\delta(\theta, \phi)$ . So, the fraction  $\eta$  of particles flowing back through the contact for a given angle of incidence is simply given by the overlap of the contact area and the shifted area divided by the contact area:  $\eta = (2/\pi) \arccos(\delta/2) - (\delta/\pi) (1 - \delta^2/4)^{1/2}.$ Obviously,  $\eta$ =0 when  $\delta \ge 2$ . Because the charge of the quasiparticles has been reversed on AR, Andreev-reflected quasiparticles that flow back through the point contact contribute to the excess current. At low voltages (V < 0.3 mV), in the absence of AR, we assume an Ohmic behavior of the point contact:  $R = R_S$ . If Andreev-reflected particles are focused on the point contact, the relative resistance becomes

$$\frac{R}{R_s} = \left(1 + \frac{A}{2\pi} \int_0^{\pi/2} \sin\theta \, d\theta \int_0^{2\pi} d\phi \eta \exp\left(-\frac{2d}{l}\cos\theta\right)\right)^{-1},$$
(2)

with  $2d/\cos\theta$  the total path length of the charge carriers. To take into account nonideal AR we have to introduce the above-mentioned velocity distribution  $P(\gamma)$ , with  $\gamma$  the angle of deviation. This results in a larger spot of the Andreev-reflected particles at the top side of the crystal, and therefore both in a broadening and a decrease of the signal.  $P(\gamma)$  is uniquely determined by the measured resistance as a function of applied magnetic field<sup>15</sup> and is given in the in-

3353

set of Fig. 2. Notice that  $P(\gamma)$  is definitely not a Gaussian, but approximately resembles an exponential  $\exp(\gamma/\gamma_0)$  with  $\gamma_0 = b/d = 6.3 \times 10^{-5}$  rad. To verify our procedure we calculated  $R(B)/R_S$  from the  $P(\gamma)$  given in the inset and the A found above. Excellent agreement (dots in Fig. 2) has been found.

To calculate the contact resistance as a function of applied voltage R(V), we use the energy dependence of A derived by Blonder, Tinkham, and Klapwijk:<sup>9</sup>

$$A(\epsilon') = [\epsilon'^2 + (1 - \epsilon'^2)(1 + 2Z^2)^2]^{-1} \quad (\epsilon' \le 1) \quad , \quad (3a)$$

$$A(\epsilon') = [\epsilon' + (\epsilon'^2 - 1)^{1/2}(1 + 2Z^2)]^{-2} \quad (\epsilon' > 1) \quad , \quad (3b)$$

where  $\epsilon'$  is the ratio of the quasiparticle energy  $\epsilon$  to the gap energy  $\Delta$  of the superconductor, and Z is a dimensionless measure of the barrier strength at the interface. Notice that A(1)=1 for any value of Z, whereas  $A(\epsilon' < 1)=1$  only if Z=0. Because only the component of the quasiparticle momentum normal to the N-S interface changes on AR, the angle of reflection differs by a small angle  $\delta\theta$  from the angle of incidence:

$$\delta\theta \simeq (\epsilon/\epsilon_F) \tan\theta. \tag{4}$$

Furthermore, we have to take into account the electronphonon interaction. The electron mean free path is now given by  $1/l = 1/l_{imp} + 1/l_{eph}$ , where  $l_{imp}$  is the energyindependent mean free path due to impurity scattering and the mean free path  $l_{eph}$  due to phonon scattering is given at low energies by<sup>19,20</sup>  $l_{eph}(\epsilon) = C/\epsilon^3$ . We obtained the best result with  $C = 3500 \ \mu \text{m meV}^3$ , which compares reasonably well with the C deduced from double-point-contact and radio-frequency size-effect experiments.<sup>19,20</sup>

In Fig. 3 the result of the calculation (dashed line) has been displayed using the  $P(\gamma)$  and the barrier strength as

- \*Also at Max-Planck-Institut für Festkörperforschung, Hochfeld-Magnetlabor, 166X, F-38042 Grenoble Cédex, France.
- <sup>1</sup>A. F. Andreev, Zh. Eksp. Teor. Fiz. **46**, 1823 (1964) [Sov. Phys. JETP **19**, 1228 (1964)].
- <sup>2</sup>W. J. Tomasch, Phys. Rev. Lett. 15, 672 (1965); 16, 16 (1966).
- <sup>3</sup>J. M. Rowell and W. L. McMillan, Phys. Rev. Lett. 16, 453 (1966).
- <sup>4</sup>G. L. Harding, A. B. Pippard, and J. R. Tomlinson, Proc. R. Soc. London, Ser. A 340, 1 (1974).
- <sup>5</sup>I. P. Krylov, in *Soviet Scientific Review, Section A: Physics Reviews,* edited by I. M. Khalatnikov (Harwood Academic, London, 1980), Vol. 2, p. 85.
- <sup>6</sup>S. I. Bozhko, V. S. Tsoi, and S. E. Yakovlev, Pis'ma Zh. Eksp. Teor. Fiz. **36**, 123 (1982) [JETP Lett. **36**, 152 (1982)].
- <sup>7</sup>G. E. Blonder and M. Tinkham, Phys. Rev. B 27, 112 (1983).
- <sup>8</sup>P. A. M. Benistant, H. van Kempen, and P. Wyder, Phys. Rev. Lett. **51**, 817 (1983).
- <sup>9</sup>G. E. Blonder, M. Tinkham, and T. M. Klapwijk, Phys. Rev. B 25, 4515 (1982).
- <sup>10</sup>A Sharvin junction is a metallic junction in the Knudsen regime (l/b >> 1, with *l* the mean-free path and *b* the contact radius).

was used for the R(B) found. The agreement with experiment is very satisfactory.

We conclude that a straightforward deposited superconducting layer on Ag can give an almost ideal Andreevreflection probability. The energy dependence of Andreev reflection is in agreement with the theory of Ref. 9. The retroreflection law is obeyed with high precision (  $\simeq 10^{-4}$ rad). However, because of the high angular resolution of the described experiment ( $\simeq 10^{-5}$  rad) we are able to observe that the particles are not exactly reflected back in the incoming direction but in a narrow cone with an angle  $\simeq 10^{-4}$  rad about this direction. The mechanism that causes this uncertainty is not yet known. On the basis of the present experiment it cannot be decided whether this effect is caused by deviations from the ideal Andreevreflection process proper, or by deviations in the quasiparticle trajectories. The last effect could, for example, be due to very small angle scattering by the long-range strain field of dislocations. Numerical estimates show that a dislocation density of  $10^6 - 10^7$  cm<sup>-2</sup> could explain the observed effect. However, more experiments are needed to determine with certainty the cause of the deviations. For both mentioned mechanisms the study of very-small-angle phenomena is essential. Clearly, the described measuring method makes a direct quantitative study possible.

The authors are grateful to T. J. Gortenmulder and the late Dr. B. Knook of the Kamerlingh Onnes Laboratory, Leiden, for growing the silver single crystal from the melt, and L. W. M. Schreurs for orienting and spark cutting the sample. This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie and was made possible by financial support from the Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek.

- Yu. V. Sharvin, Zh. Eksp. Teor. Fiz. 48, 984 (1965) [Sov. Phys. JETP 21, 655 (1965)].
- <sup>11</sup>Cominco Ltd., Trail, British Columbia, Canada.
- <sup>12</sup>A. C. Ehrlich, J. Mater. Sci. 9, 1064 (1974).
- <sup>13</sup>A. G. M. Jansen, A. P. van Gelder, and P. Wyder, J. Phys. C 13, 6073 (1980).
- <sup>14</sup>Yu. N. Shalov and I. K. Yanson, Fiz. Nizk. Temp. 3, 99 (1977) [Sov. J. Low Temp. Phys. 3, 48 (1977)].
- <sup>15</sup>A. P. van Gelder, P. A. M. Benistant, H. van Kempen, and P. Wyder (unpublished).
- <sup>16</sup>M. Abramowitz and I. A. Stegun, Handbook of Mathematical Functions (Dover, New York, 1964), Chap. 5.
- <sup>17</sup>S. Shih, Z. G. Khim, G. B. Arnold, and W. J. Tomasch, Phys. Rev. B 24, 6440 (1981).
- <sup>18</sup>B. Lengeler, W. R. Wampler, R. R. Bourassa, K. Mika, K. Wingerath, and W. Uelhoff, Phys. Rev. B 15, 5493 (1977).
  <sup>19</sup>P. van Son (private communication).
- <sup>20</sup>V. A. Gasparov, Zh. Eksp. Teor. Fiz. 68, 2259 (1975) [Sov. Phys. JETP 41, 1129 (1975)].