[7]U. Daybelge, unpublished.

[8]S. Chapman and T. G. Cowling, *The Mathematical Theory of Nonuniform Gases* (Cambridge U. Press, Cambridge, England, 1952), 2nd ed.

[9]C. H. Kruger and M. Mitchner, Phys. Fluids 10, 1953 (1967).

Direct Observation of Spin-State Mixing in Superconductors*

P. M. Tedrow and R. Meservey

Francis Bitter National Magnet Laboratory,† Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 30 August 1971)

We observe a peak in the tunneling conductance between two very thin superconducting aluminum films in an intense parallel magnetic field at a voltage $V=(\Delta_1+\Delta_2-2\mu H)/e$. The magnetic field dependence of this peak identifies it as that predicted by Engler and Fulde for spin-state mixing in superconductors. The spin-orbit scattering rate $b=\hbar/3\Delta\tau_{\text{s.o.}}$ obtained from this measurement is approximately 0.1.

This experiment reports the direct detection of the effect of spin-orbit scattering on the spin density of states of a superconductor. The quasiparticle energy states in superconducting Al are split in a high magnetic field by the interaction of the field with the spin magnetic moment of the quasiparticles. This splitting was recently demonstrated[1] by conductance measurements on Al-Al_2O_3-Ag tunnel junctions (Al thickness ≈ 50 Å). The measured conductances could be analyzed surprisingly well by assuming simply that the BCS[2] density of states was split into spin-up and spin-down parts displaced in energy by $\pm\mu H$ (μ being the electron magnetic moment). The success of this analysis implied that there were no significant interactions to mix the spin states. However, measurements[3,4] of the critical magnetic field H_c of such films indicated that spin-orbit scattering, though small in Al, is not negligible. In fact, theoretical fitting[5] of values of $H_c(T)$ gave a spin-orbit scattering parameter $b=\hbar/3\Delta\tau_{\text{s.o.}}\approx 0.2$, large enough to have had a significant effect on the tunneling density of states. Here 2Δ is the energy gap of the superconductor and $\tau_{\text{s.o.}}$ is the spin-orbit scattering time.

Recently Engler and Fulde[6] succeeded in calculating the density of states of a thin superconductor in a high magnetic field for various values of b. For $b=0$ and a high magnetic field, $H=0.6(\Delta/\mu)$, Fig. 1(a) shows the calculated density of states $N(E/\Delta)$. As expected, the density of states for each spin direction is just half of the BCS density of states and is shifted in energy by $+\mu H$ for spin up and by $-\mu H$ for spin down. Figure 1(b) shows the interesting theoretical result of increasing b. Here the field is the same but $b=0.2$ and the calculated behavior is qualitatively different, the spin states being partially mixed. The states near $E/\Delta=1-\mu H/\Delta$, for example, are no longer only spin down, as there is also a small spin-up peak. As b increases, the spin mixing increases, and the peaks move closer together and become more nearly equal in magnitude. By the time $b=5.0$, spin is no longer a good quantum number, and $N(E/\Delta)$ has only a single peak, which is naturally independent of H and approaches closely the single, unsplit, BCS

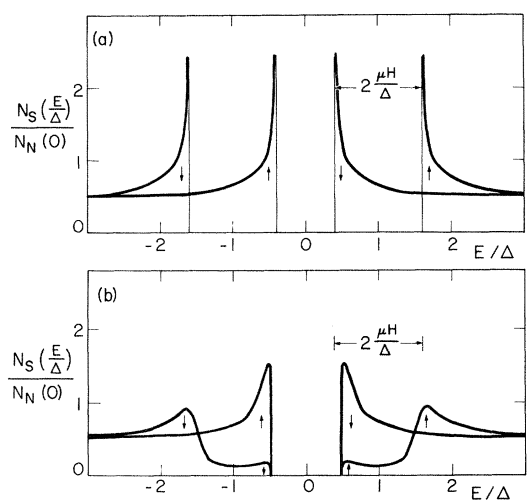

FIG. 1. Theoretical density of states for a superconcuctor in a magnetic field $H=0.6\Delta/\mu$. (a) With $b=0$, $N_s(E/\Delta)$ splits into two BCS-like curves, one for each spin direction shifted by $\pm\mu H$. (b) With some spin-orbit scattering, $b=0.2$, the spin states are mixed with some spin-up states found near the energy of the spin-down peak.

density of states.

The purpose of the present experiment was to attempt to detect directly this spin mixing of the quasiparticle states in Al by tunneling between two superconducting Al films. Junctions were formed from two crossed Al films each about 50 Å thick and separated by an Al_2O_3 insulating layer approximately 30 Å thick. The conductance $\sigma = dI/dV$ was measured as a function of applied voltage V using a lock-in amplifier operating at 5 kHz. Most measurements were made at $T=0.4$ K ($T/T_c < 0.2$) and in fields up to 55 kOe.

By referring to Fig. 1(a), we see that when $b=0$, the magnetic field has no effect on σ. Assuming that the spin of the quasiparticle does not change during the tunneling, as the voltage between the two films is increased from zero, σ has a peak only at $V=(\Delta_1+\Delta_2)/e$, independent of H. Of course, σ is symmetrical about $V=0$. When $b \neq 0$, as Fig. 1(b) shows, some spin-up states are shifted downward in energy by $2\mu H$. Therefore spin-up quasiparticles can tunnel at a voltage $2\mu H/e$ less than was the case with $b=0$, that is, when $V=(\Delta_1+\Delta_2-2\mu H)/e$. Similarly, some spin-down quasiparticles are raised in energy by $2\mu H$. Thus they, too, can tunnel at a voltage $2\mu H/e$ less than with $b=0$, again at $V=(\Delta_1+\Delta_2-2\mu H)/e$. Thus σ has a peak near $V=(\Delta_1+\Delta_2-2\mu H)/e$ which arises from the alteration of the density of states by spin-orbit scattering.

Figure 2(a) (dashed curve) shows the ratio of the conductance in the superconducting and normal states, σ_s/σ_n vs V, as calculated by Engler and Fulde[5] for $T=0$ and $H=0.6\Delta/\mu$ with $b=0.1$. They have assumed two identical Al films with $\Delta_1=\Delta_2=\Delta$ and that spin is conserved in tunneling. The large peak at $V=(\Delta_1+\Delta_2)/e$ is the usual one seen in tunneling between two superconductors.[7] The smaller peak at $V=(\Delta_1+\Delta_2-2\mu H)/e$ is produced by spin-state mixing of the density of states due to spin-orbit scattering such as was shown in Fig. 1(b). This smaller peak would not be observed if $H=0$. Its size depends on the size of b. Also shown in Fig. 2(a) (solid curve) is an experimental measurement for approximately the same conditions as the theoretical curve except that $T \neq 0$. The experimental peaks are broadened because of the finite temperature and also because of some depairing in the magnetic field. Since the films are not quite identical, the usual difference peak[7] at $V=(\Delta_1-\Delta_2)/e$ also appears in Fig. 2(a) very near $V=0$.

The broadened spin-state mixing peak in Fig. 2(a) can be convincingly identified by its magnetic field dependence. Figure 2(b) is an expanded plot of σ_s/σ_n vs V for various values of H. The arrows mark the value of $(\Delta_1+\Delta_2-2\mu H)/e$ for each curve. Figure 3 compares the measured

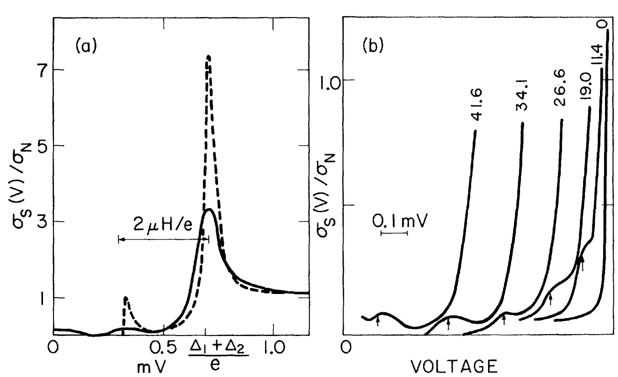

FIG. 2. (a) Normalized tunneling conductance for an Al-Al_2O_3-Al junction in a magnetic field. The dotted curve was predicted theoretically for $T=0$, $H=0.6\Delta/\mu$, and $b=0.1$. The solid curve was measured for $T=0.4$ K ($T/T_c \approx 0.16$) and $H=0.6\Delta/\mu=36.2$ kOe. (b) Measured tunneling conductance curves for various values of magnetic field with an expanded scale. The curves have been displaced horizontally for clarity. The arrows mark the voltage $V=(\Delta_1+\Delta_2-2\mu H)/e$ for each value of magnetic field, shown in kOe.

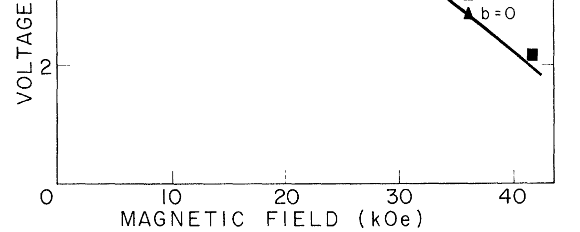

FIG. 3. Measured voltages of the spin-mixed peaks are shown as squares. The lower solid line is the theoretical prediction, $(\Delta_1+\Delta_2-2\mu H)/e$, which is obtained by subtracting $2\mu H/e$ from the measured positions of the principal conductance maxima, given by the circles. The triangles are approximate theoretical values of the voltages of the spin-mixed peak for different values of b at a field of $H=0.6\Delta/\mu$ obtained by subtracting the theoretical separation of the conductance peaks in Ref. 6 from the measured voltage of the principal peak.

voltage of the small field-dependent maxima with the theoretical value (for small values of b) $(\Delta_1+\Delta_2-2\mu H)/e$. The upper curve with circles gives the measured values of the principal maximum which is assumed to be equal to $(\Delta_1+\Delta_2)/e$. This sum peak is seen to be slightly dependent on H. Subtracting $2\mu H/e$ from this curve gives the lower curve $(\Delta_1+\Delta_2-2\mu H)/e$, which is the expected locus of the spin-mixed maxima for $b\rightarrow 0$. The squares are the measured positions of the small maxima determined from curves such as Fig. 2(b). For $H<26$ kOe the small peaks are obscured by the steep background slope of the principal maxima. In this case the positions of the small peaks were determined by subtracting an exponential curve which fitted the background curve for voltages above and below the vicinity of the small peak. The excellent quantitative agreement of the magnetic field dependence of this peak, as shown by Fig. 3, allows us to identify it as caused by spin-state mixing.

We have assumed in this analysis that there is no spin flipping in the Al_2O_3 layer.[1] Spin flipping would produce a peak at $V=(\Delta_1+\Delta_2-2\mu H)/e$, too, but a peak would also appear at $V=(\Delta_1+\Delta_2+2\mu H)/e$. Since this higher voltage peak is not observed, spin-flip tunneling apparently does not occur.

Because of the broadening of the conductance curves by thermal and depairing effects we cannot immediately determine an exact value of b from these measurements. However, it appears from the relative areas of the measured principal and spin-mixed maxima that the value of b for this sample is about equal to or perhaps slightly less than for the unbroadened theoretical curve, $b=0.1$. Another estimate for the value of b is obtained from the relative separation of the conductance peaks which according to theory should decrease continuously as b increases, for a given magnetic field. The triangles in Fig. 3 show the position for various values of b of the spin-mixed peak according to Engler and Fulde if we assume that the position of the principal peak is given by the upper solid line. For $b\rightarrow 0$ the position (for $\mu H/\Delta=0.6$) would be on the line $(\Delta_1+\Delta_2-2\mu H)/e$. For higher values of b the spin-mixed peak moves to higher voltages as shown. The experimental points lie somewhere between $b=0$ and $b=0.1$. On the other hand, measurements of $H_c(T)$ for these same films yield a calculated value of $b=0.35$, a value which seems to be outside the limit set by the present measurement. Perhaps this disagreement is to be attributed to the rather indirect determination of b from the critical-field measurements.

To summarize, we have observed extra tunneling conductance in Al-Al_2O_3-Al junctions at voltages $V=(\Delta_1+\Delta_2-2\mu H)/e$, which is caused by spin-state mixing in superconducting Al. The technique of superconductor-superconductor tunneling provides a direct and sensitive method for detecting the effects and measuring the magnitude of spin-orbit scattering. The value of the spin-orbit parameter $b\approx 0.1$ obtained from this method seems to be significantly less than the value obtained from critical-field measurements.

We are indebted to Dr. P. Fulde and Dr. H. Engler for informing us of their results before publication. We also thank R. MacNabb and M. Blaho for technical assistance.

*Work supported by the U. S. Air Force Office of Scientific Research.

†Now supported by the National Science Foundation.

[1]R. Meservey, P. M. Tedrow, and P. Fulde, Phys. Rev. Lett. 25, 1270 (1970).

[2]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

[3]P. M. Tedrow, R. Meservey, and B. B. Schwartz, Phys. Rev. Lett. 24, 1004 (1970).

[4]M. Strongin and O. F. Kammerer, Phys. Rev. Lett. 16, 456, 675(E) (1966).

[5]P. Fulde and H. Engler, private communication.

[6]H. Engler and P. Fulde, to be published.

[7]I. Giaever and K. Megerle, Phys. Rev. 122, 1101 (1961); S. Shapiro, P. H. Smith, J. Nicol, J. L. Miles, and P. F. Strong, IBM J. Res. Develop. 6, 34 (1962).