## Spin-Polarized Tunneling Measurement of the Antisymmetric Fermi-Liquid Parameter G<sup>0</sup> and Renormalization of the Pauli Limiting Field in Al

P. M. Tedrow

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

and

J. T. Kucera<sup>(a)</sup>

Department of Physics, Massachuesetts Institute of Technology, Cambridge, Massachusetts 02139

and

## D. Rainer

Physikalisches Institut der Universität Bayreuth, Federal Republic of Germany

and

## T. P. Orlando<sup>(b)</sup>

Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 25 July 1983)

Spin-polarized tunneling is used to measure the energy difference  $\delta$  between spin-up and spin-down electrons in a magnetic field in thin films of superconducting Al.  $\delta$  decreases from  $2\mu_B H$  at low temperature and field to  $0.8(2\mu_B H)$  as the phase boundary  $T_c(H)$  is approached. This change in  $\delta$  provides the first measurement of the l=0 antisymmetric Fermi-liquid parameter  $G^0$  in a real metal and the first direct evidence for the renormalization of the Pauli field in high-field superconductivity.

PACS numbers: 74.60.Ec, 71.38.+i, 71.45.Gm, 75.20.En

Enhanced Pauli spin susceptibility is common to many interacting systems of fermions: for example, liquid <sup>3</sup>He, nearly itinerant ferromagnetic metals (Pd and TiBe<sub>2</sub>), and some superconductors with high transition temperatures. The Pauli spin susceptibility is renormalized by many-body effects from the electron-phonon and electron-electron interactions.<sup>1</sup> Measurements of two normal-state properties, the Pauli spin susceptibility  $\chi$  and the electronic part of the heat capacity  $\gamma$ , in principle would give directly the amount of the renormalization. Unfortunately, extraction of the two required normal-state properties in real metals is complicated by large orbital contributions to the susceptibility and lattice contributions to the heat capacity. However, as Leggett<sup>2</sup> showed, superconducting properties are directly sensitive to the needed normal-state properties and, hence, to the renormalization. We describe here a tunneling experiment which exploits this sensitivity of the superconducting state, allowing measurement of the strength of the many-body interactions in Al. The effects seen in the present experiments can be traced back to a renormalization of the ratio  $N(\gamma)/N(\chi)$  where  $N(\gamma)$ and N(x) are the densities of states obtained from the normal-state electronic specific heat and from

the Pauli susceptibility, respectively. Landau's theory of Fermi liquids relates this ratio to the antisymmetric l=0 Landau parameter  $G^{0}$ ;  $N(\gamma)/N(\chi) = 1 + G^{0}$ .<sup>3</sup>

We have used a spin-sensitive tunneling technique<sup>4</sup> to measure  $G^0$  directly for the first time in a "real" metal. Specifically, we measure the energy difference between spin-up and spin-down electrons in a magnetic field in thin films of superconducting aluminum. The measurements show that  $\delta = 2\mu_B H$ at very low temperature and field but that  $\delta$  decreases as the temperature and field increase toward the phase boundary  $T_c(H)$ . The weak-coupling<sup>2, 5</sup> for superconductivity theory predicts that  $\delta \rightarrow 2\mu_{\rm B}H(1+G^0)^{-1}$  as the phase boundary is approached. This decrease in  $\delta$  arises from a temperature-dependent internal field  $H_{int}$  due to many-body interactions.

The measurement of  $G^0$  is particularly important to the quantitative understanding of the theory of high-field superconductivity and the nature of the basic interactions in high-transition-temperature superconducting materials. The traditional theory<sup>6</sup> of high-field superconductivity describes how the upper critical field  $H_{c2}$  and the tunneling density of states are affected by two pair-breaking interactions. First, the orbital pair-breaker leads to a reduction in  $T_c$  and results in a "smearing" of the density of states. Second, the Pauli spin pair-breaker leads to a further reduction in  $T_c$ ; however, the effect on the density of states is quite distinct and results in Zeeman splitting of spin-up and spin-down densities of states. The Pauli spin pair-breaking can be reduced by increasing random spin-orbit scattering which increases the critical field and mixes the densities of states of spin-up and spin-down electrons. These interactions are parametrized by the orbital pair-breaker c, the Pauli limiting field  $H_p$ , and the spin-orbit scattering rate  $b_{so}$ .

Although the traditional theory with no renormalization qualitatively explained tunneling and critical-field data, quantitative discrepancies were found.<sup>7-11</sup> Renormalization of the Pauli limiting field  $H_p$ , namely  $H_p = H_p(\text{BCS})[N(\gamma)/N(\chi)]$ , alleviated these quantitative discrepancies.<sup>8,9</sup> Although the renormalization of  $H_p$  allows quantitative agreement between the theory and experiment and among various experiment, the ratio  $N(\gamma)/N(\chi)$  has appeared as an additional fitting parameter. However, the experiment reported here allows this ratio to be measured and quantitatively demonstrates the existence of the renormalization effects.

The spin-resolved spin-polarized tunneling technique<sup>4, 12</sup> involves measuring the tunneling conductance dI/dV as a function of V of a junction between a thin superconductor and a ferromagnet in a magnetic field. The excited states of the superconductor are split in energy by the field into spin-up and spin-down parts. The ferromagnet has unequal tunneling densities of states for the two spin directions and therefore serves to make the tunneling conductance depend on the spin of the tunneling particle. The degree of polarization of the tunnel current is found<sup>13</sup> by fitting the total conductance at low temperature and moderate field using the theory of Bruno and Schwartz.<sup>14</sup> This polarization is independent of field and temperature and varies less than 3% from sample to sample for Fe films.<sup>13</sup> Hence, the tunneling conductnace then can be separated algebraically into parts related directly to the spin-up and spin-down densities of states in the superconductor as shown in Fig. 1.

The junctions used in this experiment were formed between 4-nm-thick Al ( $T_c = 2.32$  K) films and 100-nm-thick Fe films. The tunnel barrier was aluminum oxide produced by an oxygen glow discharge. The junctions were cooled in a <sup>3</sup>He cryostat and measured in an 8-T superconducting magnet.



FIG. 1. Conductance vs voltage for spin-up (shortdashed curves) and spin-down (long-dashed curves) electrons at various fields and temperatures; the solid lines are the total conductances. The horizontal bar marks  $2\mu_B H$  and is to be compared with the observed splitting  $\delta$  between the steep portion of the curves.  $\delta$ equals  $2\mu_B H$  at low temperature and fields but becomes less than  $2\mu_B H$  as the phase boundary  $T_C(H)$  is approached. Note the change of the vertical scale in the lowest graph. The scales have been chosen such that  $\sigma_{\text{total}} = a \sigma \uparrow + (1-a)\sigma \downarrow$  where a is the fraction of electrons available to tunnel from Fe that have up spin. See Ref. 13 for details.

Figure 1 shows the spin-resolved conductances at various fields and temperatures. The shapes of the conductances for spin up and spin down are nearly identical, indicating that there is very little spin mixing  $(b_{so} \le 0.05)$ . At low temperature and field Fig. 1(a) shows that the curves are separated by  $\delta = 2\mu_B H$ , where the separation is measured between the steep portions of the conductance curves.<sup>15</sup>

The traditional pair-breaking theory with no renormalizations (i.e.,  $G^0 = 0$ ) predicts that the spin conductances would always separate by  $2\mu_{\rm B}H$  independent of T for small enough spin-orbit scattering regardless of the amount of the orbital pairbreaker. However, Figs. 1(b) and 1(c) show that at either higher fields or higher temperatures the separation  $\delta$  is less than  $2\mu_B H$ , indicating that the magnetic field is being renormalized and that the renormalization is a function of applied field and temperature. The separation  $\delta$  as a function of magnetic field for two temperatures is shown in Fig. 2. The error bars reflect uncertainty in  $\delta$  arising from using different portions of the conductance curves to measure the separation. The solid curves are theoretical calculations from the (dirty limit) weak-coupling theory with  $G^0 = 0.3.^5$  Hence the measurement of  $\delta$  as a function of H and T provides the first measurement of  $G^0$  for Al and shows that the Pauli field is indeed renormalized.

The weak-coupling theory used in Fig. 2 explicitly calculates the energy separation  $\delta = 2\mu_B H_T$ , where  $H_T = H_a + H_{int}$ . Here  $H_T$  is the total field,  $H_a$  is the



FIG. 2. The observed splitting  $\delta$  of the conductances of spin-up and spin-down electrons vs applied magnetic field for different temperatures. The symbols mark the data and the solid lines the theoretical calculations of Ref. 5. The dashed lines bracket the splitting  $\delta$  between its maximum of  $2\mu_B H$  at low field and temperature and its minimum of  $0.8(2\mu_B H)$  near the superconducting phase boundary  $T_c(H)$ . Data for intermediate temperatures (not shown) also lie between these limits.

applied field, and  $H_{int}$  is the internal field. The weak-coupling theory contains  $T_c$ ,  $b_{so}$ , c, and  $G^0$  as input parameters and self-consistently finds the superconducting order parameter and the internal field. According to the theory of Fermi-liquid interactions  $H_{\rm int} = -2G^0 M / \mu_{\rm B}^2 N(\gamma)$  where M is the magnetization (see Ref. 2, for example). It should be noted that the weak-coupling theory is a generalization to high magnetic fields of Leggett's calcula $tion^2$  for the susceptibility of a superfluid. In fact, using Leggett's value of the susceptibility  $\chi$  of a superfluid (as in Ref. 1) and  $M = \chi H_a$  at low fields, one finds that  $H_{int} = 0$  at zero temperature and  $H_{int} = -G^0(1+G^0)^{-1}H_a$  near  $T_c$ . Hence, for low magnetic fields,  $\delta = 2\mu_B H_a$  at zero temperature and  $2\mu_B H_a (1+G^0)^{-1}$  near  $T_c$ . These two values of  $\delta$  bracket the splitting for all temperatures and fields and are shown as dashed lines in Fig. 2. The slope of the lower dashed line gives  $G^0 = 0.3 \pm 0.05$ . The weak-coupling theory used in Fig. 2 will be discussed in detail elsewhere,<sup>5</sup> but let us say here that it also shows the critical field can be fit with the same  $b_{so}$  as used for fitting the total tunneling conductance.

Our experiment yields the many-body enhancement of  $N(\gamma)/N(\chi)$ . This experiment by itself cannot separate the contributions of the enhancement from the electron-phonon and electronphonon interactions<sup>16</sup>; however, the additional information needed for this separation can be provided, in principle, by tunneling experiments and inversion of the Eliashberg equations<sup>17, 18</sup> which yield the electron-phonon coupling constant  $\lambda_{ep}$ . Hence, from the relationship<sup>2</sup>

$$N(\gamma)/N(\chi) = (1+G^0) = (1+\lambda_{\rm ep})(1+G^0_{\rm el})$$

the purely electronic exchange enhancement  $(1 + G_{el}^0)^{-1}$ , could be obtained. (For example, assuming a range of  $\lambda_{ep}$  of 0.4 to 0.5 for Al, one finds that  $G_{el}^0$  ranges from -0.05 to -0.15.<sup>19</sup>) An experimental determination of the exchange enhancement  $(1 + G_{el}^0)$  also is useful in view of the recently ly renewed interest in the influence of spin fluctuations on  $T_c$ . One is tempted to use  $-G_{el}^0$  as an estitimate for the enhancement parameter I of the spin-fluctuation model in which

$$N(\gamma)/N(\chi) = 1 + G^0 = (1 + \lambda_{ep}^{SF} + \lambda_{spin})(1 - \overline{I}).$$

Here  $\lambda_{ep}^{SF}$  and  $\lambda_{spin}$  are the mass renormalization from the electron-phonon interaction and spin fluctuations, respectively, in this model. Because tunneling spectroscopy of the phonons yields only the ratio<sup>20</sup>  $\lambda_{ep}^{SF}(1 + \lambda_{spin})^{-1}$  all the needed parameters ( $\lambda_{ep}^{SF}$ ,  $\lambda_{spin}$ , and I) cannot be inferred from experiments without recourse to a model-dependent relationship between  $\lambda_{spin}$  and  $\overline{I}$  which is qualitative at best.<sup>21</sup>

In summary, we have provided the first measurement of the antiysmmetric Fermi-liquid parameter  $G^0$  in aluminum and the first direct evidence that the Pauli limiting field is renormalized in high-field superconductors. The antisymmetric Fermi-liquid parameter is found to be  $G^0 = 0.3 \pm 0.05$ . This tunneling technique and analysis in the superconducting state will serve, in our opinion, as a more powerful method than the traditional normal-state analyses for determining electronic Landau parameters.

The authors gratefully acknowledge the help of J. Moodera, R. Meservey, and R. MacNabb in this research. We also thank J. A. X. Alexander, G. Hertel, and M. J. Komichak for their assistance in the digital data analysis. This research was supported in part under the Air Force Office of Scientific Research Contract No. F49620-82-K-0028 and by the National Science Foundation.

<sup>(a)</sup>Present address: Department of Physics, California Institute of Technology, Pasadena, Cal. 91125.

<sup>(b)</sup>Also, Center for Materials Science and Engineering and the Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Mass. 02139.

<sup>1</sup>The electron-phonon interaction affects the Pauli susceptibility only to order  $h\omega_{debve}/E_F = 10^{-2}$ . However, this statement is true only in the normal state. Leggett [A. J. Leggett, Phys. Rev. 140A, 1896 (1965)] showed that the susceptibility of a superfluid is  $\chi = 2\mu_B^2 N(\gamma) f(T) [1 + G^0 f(T)]^{-1}$  where f(T) is the Yosida function. For a normal metal  $T > T_c$  and f(T) = 1, and  $\chi = 2\mu_B^2 N(\gamma) (1 + G^0)^{-1}$  which is independent of the electron-phonon interaction. However, for the superfluid  $T < T_c$  and f(T) < 1, the superfluid susceptibility does depend on the electron-phonon interaction; for example, as T approaches zero.  $\chi = 2\mu_{\rm B}^2 N(\gamma) f(T)$  which depends on the heat-capacity density of states and hence the electron-phonon interaction. Note that f(0) = 0.

<sup>2</sup>Leggett, Ref. 1. For a review see A. J. Leggett, Ann. Phys. **46**, 76 (1968), and Rev. Mod. Phys. **47**, 331 (1975).

<sup>3</sup>J. W. Wilkins, in *Electrons at the Fermi-Surface*, edited by M. Springford (Cambridge Univ. Press, London, 1980). <sup>4</sup>P. M. Tedrow, J. S. Moodera, and R. Meservey, Solid State Commun. **44**, 587 (1982).

<sup>5</sup>J. A. X. Alexander *et al.*, to be published.

<sup>6</sup>For a summary of the theory of  $H_{c2}$  see, for example, A. L. Fetter and P. C. Hohenberg, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969), Vol. 2, p. 886.

<sup>7</sup>P. M. Tedrow and R. Meservey, Phys. Rev. B **8**, 5098 (1973).

<sup>8</sup>T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, Phys. Rev. B **19**, 4545 (1979).

<sup>9</sup>P. M. Tedrow, Bull. Am. Phys. Soc. 25, 356 (1980).

 $^{10}\text{P}.$  M. Tedrow and R. Meservey, Phys. Rev. B 25, 171 (1982).

<sup>11</sup>T. P. Orlando and M. R. Beasley, Phys. Rev. Lett. **46**, 1598 (1981).

<sup>12</sup>R. Meservey, P. M. Tedrow, and R. Bruno, Phys. Rev. B 11, 4224 (1975).

 $^{13}$ R. Meservey, D. Paraskevopoulous, and P. M. Tedrow, Phys. Rev. B **22**, 1331 (1980); P. M. Tedrow and R. Meservey, Phys. Rev. B **7**, 318 (1973).

<sup>14</sup>R. C. Bruno and B. B. Schwartz, Phys. Rev. B **8**, 3161 (1973).

<sup>15</sup>For  $b_{so} = 0$  the conductance curves are identical in shape but shifted. Measuring the splitting of the steep portion of the curves provides a more sensitive measurement of the shift than attempting to accurately locate the maxima of the thermally broadened conductance curves. For nonzero  $b_{so}$ , computer simulations show that the splitting of the steep portion is still the best measure of  $\delta$ . The errors in Fig. 1 are due to the uncertainty in the difference between the steep portions of each pair of curves.

<sup>16</sup>The density of states calculated from band structure N(b) can be useful and equated to  $N(\gamma)(1+\lambda_{ep})^{-1}$  only if N(b) is calculated as the density of states of normal-state quasiparticles which have been renormalized by all many-body interactions except from phonons. See, for example, D. Rainer, Physica (Utrecht) **109–110B+C**, 1671 (1982).

<sup>17</sup>G. M. Eliashberg, Zh. Eksp. Teor. Fiz. **38**, 966 (1960) [Sov. Phys. JETP **11**, 696 (1960)].

<sup>18</sup>D. J. Scalapino, J. R. Schrieffer, and J. W. Wilkins, Phys. Rev. **148**, 263 (1966).

<sup>19</sup>Note that thin films of Al have higher  $T_c$ 's than the bulk, so that the range of  $\lambda_{ep}$  used is only a rough estimate.

 $^{20}$ J. M. Daams, B. Mitrovic, and J. P. Carbotte, Phys. Rev. Lett. **46**, 65 (1981).

<sup>21</sup>Although the critical field data for V<sub>3</sub>Ga have been explained within the spin-fluctuation model (see Ref. 11), they also can be explained without spin fluctuations but with a Stoner enhancement of I = 0.5 and with  $\lambda_{ep} = 1.0$ , i.e., with  $G^0 = 0$ .