Experimental Test of the Theory of High-Field Superconductivity

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The theory of high-field superconductivity contains two parameters, one of which, the spin-orbit scattering rate, previously has been treated as a variable in fitting the theory to the experimental data. We have made critical-field measurements on thin-film superconductors and have simultaneously determined both parameters from an independent spin-polarized tunneling measurement, allowing for the first time a test of the theory with no adjustable parameters. A significant lack of agreement was found.

When the critical magnetic field H_c of a superconductor is very high, the interaction of a magnetic field with the magnet moment of the electrons can have an important effect on the magnitude and temperature dependence of H_c.[1] This effect can be reduced, however, by spin-orbit scattering.[2] The theory of high-field superconductivity including the effect of electron spin was formulated by Maki,[3] by Werthamer, Helfand, and Hohenberg,[4] and by Fulde and co-workers[5] using a generalization of the Ginzberg-Landau theory. The resulting theory contains two parameters; the first describes the importance of the spin paramagnetism compared with the usual orbital diamagnetism while the second,[6] $b = \hbar/3\Delta\tau_{s.o.}$, is the rate of spin-orbit scattering $\tau_{s.o.}^{-1}$ normalized by the zero-field, zero-temperature superconducting order parameter Δ. In the early work to test the theory by comparing its predictions with measured critical fields,[7] the first parameter [$a = \mu/eD$ in the case of type-II superconductors and $c = De^2d^2\Delta/6\hbar\mu^2$ in the case of thin films] was assumed to be known from resistivity measurements or from measurements of the slope of H_c versus temperature for temperatures near the transition temperature T_c. Here, $D = V_f l/3$ is the electron diffusion constant, e and μ are the electron charge and magnetic moment, respectively, and d is the film thickness. The spin-orbit scattering rate, on the other hand, could not be determined separately and so was taken as a free parameter to be varied to give the best fit of theory to experiment. Quite reasonable fits could be obtained, although as time went on, it became apparent that the values of $\tau_{s.o.}^{-1}$ obtained from the theory were too large.[8]

For example, in recent work by Orlando *et al.*,[9] the value of $\tau_{s.o.}^{-1}$ needed to fit the H_c data for some $A15$ superconducting thin films was larger than the transport momentum scattering rate τ^{-1} and, in fact, approached infinity. Orlando *et al.* were able to ameliorate the problem somewhat by noticing that the theory neglected to include renormalization of the density of states in the superconductor, thereby introducing a factor of $(1+\lambda_{eph})^{1/2}$ into the spin paramagnetic limiting field[1] H_p. Here λ_{eph} is the usual electron-phonon interaction parameter. Even with this improvement, the value of $\tau_{s.o.}^{-1}$ for Nb_3Sn was still greater than τ^{-1} in some cases. These results appeared to indicate a serious flaw in the theory; however, the value of $\tau_{s.o.}^{-1}$ determined in these experiments is still only a fitting parameter.

In recent years, the technique of spin-polarized tunneling has been developed,[10] and, when applied to thin films, furnishes a method of independent

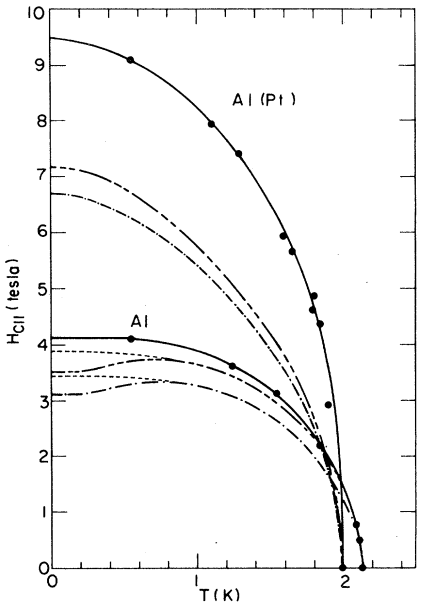

FIG. 1. Parallel critical magnetic field of Al and Al(Pt) films as a function of temperature. Dash-square-dash curve indicates measured values; dash-dot-dash curve indicates calculated using Refs. 3 and 5; and long-dash–short-dash curve indicates calculated with the help of Ref. 9 and with use of $\lambda_{eph} = 0.4$. The dashed curves show the approximate location of the theoretical first-order transition expected for the bare Al film.

determination of $\tau_{s.o.}^{-1}$. The method is based on the fact[5] that the density of states of the superconductor in a magnetic field depends on $\tau_{s.o.}^{-1}$. Densities of states as a function of the normalized spin-orbit scattering rate b were calculated by Engler and Fulde[11] and by Bruno and Schwartz.[12] Since a measurement of the tunneling conductance dI/dV versus voltage V of a normal-metal–superconductor junction allows a determination of the superconductor density of states, such a measurement in an applied magnetic field provides a way of determining the value of b for the superconductor.[10] We have carried out such measurements on a number of junctions and have measured the critical fields of the same films. Since the density-of-states calculations include the orbital-depairing effects, we can then independently obtain both the parameters b and c from the tunneling measurements to insert into the theory and calculate the theoretical $H_c(T)$ curves with no adjustable parameters.

In one experiment ultrathin Al films were used because the effect of electron spins in such films is well understood.[5] To increase the spin-orbit scattering rate, the Al films were coated with a layer of Pt about one monolayer thick. For a metal sample with at least one dimension very small, it has been shown[13] recently that $\tau_{s.o.}^{-1}$ is dominated by surface scattering and varies approximately as Z^4 (Z is the atomic number) as predicted by Abrikosov and Gor'kov.[14] According to this relation one expects that coating one side of the Al with Pt should greatly increase $\tau_{s.o.}^{-1}$.

The Al films were made by vapor deposition onto liquid-nitrogen–cooled substrates. Usually, four junctions were made at one time with two being immediately overcoated with Pt. The counter electrode was also Al but was thick enough (15 nm) to have a low critical field (less than about 2 T). The tunneling measurements were carried out at a temperature of 0.5 K and the magnetic field was oriented parallel to the film plane. H_c was determined either by four-terminal dc resistance measurement or by measuring the junction resistance dV/dI at $V=0$.

Figure 1 shows the measured parallel critical field as a function of temperature for two 4-nm-thick Al films formed simultaneously on the same substrate with one film subsequently covered by 0.2 nm of Pt. The critical field $H_{c\parallel}$ of the Al is dramatically increased by the addition of the Pt layer. This increase is consistant with an increase in the spin-orbit scattering rate b. Note that T_c is hardly affected by the Pt. Figure 2 shows the tunneling conductance dI/dV vs V of junctions between each of junctions between each of these films and the normal-metal film for various values of magnetic field. The conductance of the bare Al junction in Fig. 2(a) shows the emerging Zeeman splitting of the density of states characteristic of a thin superconductor with small b. The conductance of the junction containing the Pt-coated Al film in Fig. 2(b)

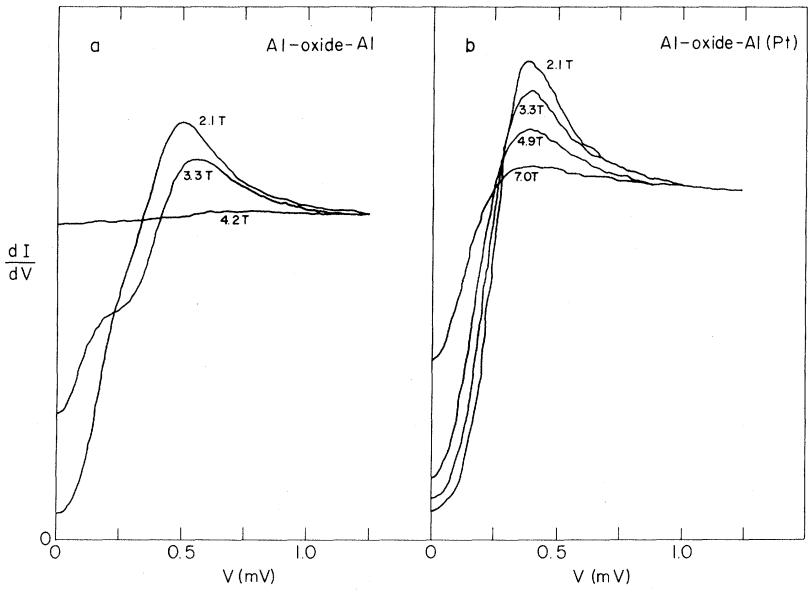

FIG. 2. Tunneling conductance dI/dV vs voltage V for various applied parallel magnetic fields (a) for a bare Al film and (b) for an Al(Pt) film.

shows no Zeeman splitting but instead shows the gradual broadening typical of orbital depairing as the field is increased. This behavior is expected if the value of b for the superconductor is large. Thus the Pt has at least qualitatively produced the expected effect.

We have fitted theoretical tunneling conductances[12] to data such as are shown in Fig. 2 and, thereby, have obtained values of the parameters b and c for the films. We find that $b = 0.05$ and $c = 0.29$ for the bare Al and that $b = 3.5$ and $c = 0.18$ for the Pt-coated Al. Thus, the Pt has increased b by a factor of about 70. From the work of Ref. 13, we expect the maximum allowable increase in b to be about $(Z_{\mathrm{Pt}}/Z_{\mathrm{Al}})^4/2 = 648$ if the Al and Pt films were uniform and each electron encountered a Pt atom every time it was scattered from the top surface of the film. Although the topography of the Al film and the distribution of the Pt are not known, the observed enhancement of b does not seem unreasonable.

Given the values of b and c from the tunneling data, we can now calculate the theoretical critical field with no adjustable parameters. The results are shown by the broken curves in Fig. 1. We have done the calculations both neglecting and and including the renormalization factor $(1+\lambda_{e\,\mathrm{ph}})^{1/2}$ brought to light by Orlando *et al.*[9] In Fig. 1, the lower (dash-dotted) curve in each case is obtained neglecting the renormalization (or taking $\lambda_{e\,\mathrm{ph}} = 0$) and the upper (long-short-dashed) curve is found using $\lambda_{e\,\mathrm{ph}} = 0.4$ ($\lambda_{e\,\mathrm{ph}}$ was obtained by solving the McMillan equation[15] relating $\lambda_{e\,\mathrm{ph}}$, the transition temperature T_c, the Debye temperature, and the Coulomb pseudopotential μ^*). Although the effect of the renormalization correction for Al is not very large, it does change the theoretical curves in the direction of better agreement with the data. The theoretical curve with $\lambda_{e\,\mathrm{ph}} = 0.4$ for the bare Al film fits the data above 1 K fairly well. Below this temperature the transition to the normal state is of first order; since the theory depends on the transition being of second order, for $T < 1$ K the theoretical curve does not represent the true transition field but rather the supercooling field. The dashed line shows the approximate first-order–transition curve. For the Pt-coated film the disparity between theory and experiment is quite large at all temperatures.

From our data it appears that the theory always gives too low a value for H_c and works best when paramagnetic limiting is the dominant factor in determining H_c. Thus, for the above data the theory fails badly in the case where b is large while it does fairly well when b is small.

A similar conclusion is implied by our work on V-Ti alloy films. We have carried out experiments similar to those described here for bare Al with the difference that since these alloys are extreme type-II superconductors the experiment can be done with $\vec{H}$ perpendicular as well as parallel to the films. In this case, the theory predicts $H_{c\parallel}$ quite closely, but the calculated $H_{c\perp}$ falls too low. The perpendicular orientation corresponds most closely to the situation with practical bulk high-field superconducting alloys. These alloys often also are expected to have large values of $\tau_{s.o.}^{-1}$. Thus, the theory as presently formulated appears to fare worst in cases which most closely approximate practical high-field superconductors.

To summarize, in the high-field region where electron spin effects can be important, there is a discrepancy between the measured values of $H_c(T)$ and those calculated from theory with use of values of the spin-orbit and orbital-depairing parameters as determined by spin-polarized tunneling. This discrepancy is only somewhat reduced by including electron-phonon renormalization in the calculation. The origin of this disagreement could be an inadequacy in the Maki theory as used to calculate $H_c(T)$ or in the theoretical tunneling conductance and is manifest as a smaller observed orbital depairing than theoretically predicted in the high-field regime.

The combined use of critical-field measurements and spin-polarized tunneling gives a method of comparing the behavior of high-field superconductors with theory without using any free parameters.

The method of coating a superconductor of low atomic number with a layer of a metal of high atomic number has potential as a method of measuring $\tau_{s.o.}$ of the latter metal.

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Many-Particle Effects in the Optical Excitations of a Semiconductor

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We present (a) a general formulation of the electron-hole interaction which takes into account both screened electron-hole attraction and its exchange counterpart, giving rise to the excitonic and local-field effects, respectively; (b) a quantitative calculation of absorption and modulation spectrum in Si, which demonstrates the dominant role played by the continuum-exciton effect on the main optical absorption of a covalent semiconductor; and (c) a model analysis of the continuum-excitonic effect on the optical excitations of semiconductors in general.

It is well established by now[1-5] that the optical absorption spectrum of solids derived from non-interacting electron-hole pairs is modified by interaction effects. They consist primarily of the attraction between the electron and the hole and its exchange counterpart, which gives rise to the local-field effect. Ample evidence for the importance of these many-particle effects can be found among the optical data of a wide class of insulators[1,3] and semiconductors.[1-6] A typical example is silicon, where the single-particle treatment gives the oscillator strength for the E_1 peak about one-half of the observed value and that of the E_2 peak somewhat too large.[7]

Recently, much effort has been made to clarify the role of the local-field effect on the electronic excitations in semiconductors and insulators.[7-11] Usually, only the local-field effect in the random-phase approximation (RPA) is considered which takes the electron and its hole as noninteracting. In a recent Letter[7] this RPA local-field effect has been shown in Si to shift absorption strength to higher energies as compared to the one-particle calculation. In the main absorption region this behavior increases the discrepancy between the calculated absorption and experiment. It has also been found in recent work on insulators, independent of whether a pseudopotential representation[7,9] or the local-orbital scheme, which we have developed,[8] was used.[8,10,11] On the other hand, following an empirical treatment by Phillips,[3] saddle excitons have been considered as the cause of the discrepancy in the effective-mass approximation (EMA),[12,13] or in a short-range Slater-Koster model[6,14,15] for the electron-hole attraction.

On the basis of a very general formulation of the electron-hole interaction, employing the local-orbital method, also in vogue recently for the impurity problem,[16,17] we present here (a) a numerical investigation for Si which demonstrates that the combined effect of exciton interaction and its exchange increases the intensity of the E_1 peak by about a factor of 2 and shifts the peak position by 0.2 eV to lower energies compared to the single-particle interband transi-

