

ANOMALOUS GAPLESS SUPERCONDUCTIVITY DUE TO SCATTERING  
FROM LOCALIZED NONINTERACTING SPINS

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Electron tunneling measurements made on lanthanum-cerium alloy films show large deviations from the Abrikosov-Gor'kov theory of gapless superconductivity. It has been suggested that such deviations should occur for an alloy system such as cerium in lanthanum which exhibits resonant scattering.

Abrikosov and Gor'kov<sup>1,2</sup> (AG) predicted that magnetic impurities would cause a drastic alteration in the density of states of superconductors, leading to gapless superconductivity. Reif and Woolf<sup>3</sup> performed tunneling measurements on quenched films of Pb-Gd, Pb-Mn, and In-Fe. The measured conductance curves for the Pd-Gd films agreed reasonably well with the predictions of AG theory. For the In-Fe and Pb-Mn films the agreement was very poor. From their data on the latter samples one would conclude that the density of states at low energies was greater than that predicted by the AG theory. Reif and Woolf suggested that this discrepancy may have been due to the nonlocalization of the Mn or Fe moment.

This Letter reports tunneling measurements on La-Ce films which show deviations from the AG theory similar to those observed by Reif and Woolf. The present measurements represent an improvement in that they were made on a system which is stable (unlike the quenched films of Reif and Woolf) and hence easier to characterize. It is a simple system in that cerium only has one  $4f$  electron and the magnetic moment is localized. Further, La-Ce alloys exhibit a Kondo effect<sup>4</sup> and probably resonant scattering.<sup>5</sup> Maki<sup>6</sup> has suggested that deviations from the usual AG theory may be expected in the case of resonant scattering. Matthias, Suhl, and Corenzwit<sup>7</sup> measured the depression of the  $T_c$  of lanthanum caused by the addition of 1% of the various rare earths. Except for cerium, they found a smooth variation throughout the series which was consistent with a nearly constant  $s$ - $f$  exchange integral  $J$ . The depression of  $T_c$  caused by Ce is anomalously large. The magnitudes of  $J$  from the resistivity measurements and from the depression of  $T_c$  agree and are equal to about 0.3 eV under the assumption of strong spin-orbit coupling.<sup>4</sup> Coqblin and Blandin<sup>8</sup> state that the anomalously large  $J$  is due to resonant scattering.

The diodes were of the form Al-Al<sub>2</sub>O<sub>3</sub>-La<sub>1-x</sub>Ce<sub>x</sub>. Details of sample preparation are the same as employed on a different system.<sup>9</sup> A film of Ag followed by a wider film of Al doped with Mn was evaporated. The Al film was oxidized for  $\frac{1}{2}$  h and painted with Formvar paint so that there remained only a small exposed area near the center. The La-Ce alloy was then evaporated rapidly (600 Å/sec) via Ohmic heating. It was found that the diodes deteriorate unless stored under vacuum at 77°K. The resistive transitions had a width of about 0.1°K or less. Films prepared under the same conditions nearly always had the same transition temperature. The composition of the La-Ce films was determined by x-ray fluorescence to be as much as 50% richer in Ce than the starting material.

The alloy films switched from the fcc structure of the pure material to the dhcp phase with the addition of less than 1% Ce. The fact that the crystal structure changed in the concentration range of interest is probably not too serious a difficulty since the ratio of the energy gap to the temperature is nearly the same for the two phases<sup>9,11</sup> and the  $T_c$  of the pure fcc film is approximately equal to the  $T_c$  of bulk dhcp La.<sup>9</sup> Consistent with this is the fact that a plot of the  $T_c$ 's of the alloys versus composition forms a curve essentially parallel to the one measured by Matthias, Suhl, and Corenzwit<sup>7</sup> but displaced downward by about 1°K.

The differential resistance was measured by conventional means. The conductance curves for three different compositions are shown in Fig. 1 along with data for an undoped sample. The conductance has been normalized by the large voltage-limiting conductance. To compare these curves with theory the measured  $T_c$ 's shown in Fig. 1 were used to determine a value for the pair-breaking parameter  $\Gamma$  from the formulas given in Ref. 1. According to the AG theory the value of  $\Gamma$  uniquely determines the value of the order parameter  $\Delta$ ,

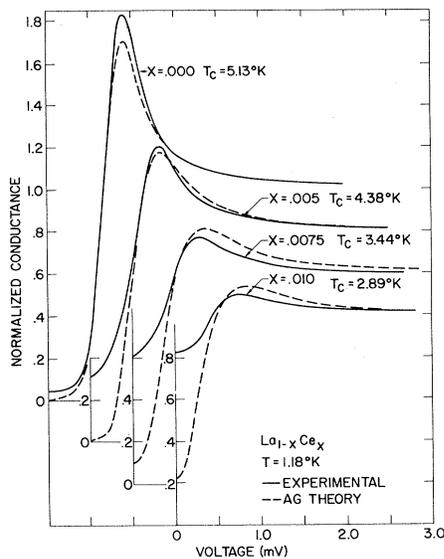


FIG. 1. Conductance curves for several diodes taken at 1.18°K. The composition of the starting material employed in the evaporation is used as the nominal composition  $X$ . The dashed curves are the predictions of AG theory. Successive sets of curves are displaced by 0.5 mV in voltage and 0.2 in normalized conductance.

the energy gap  $\Omega$ , and the density of states at each temperature. Hence the density of states and the conductance for each sample at  $T = 1.18^\circ\text{K}$  were computed. Values of  $T_c = 5.2^\circ\text{K}$  and  $\Delta = 0.778$  meV were used for the pure material. The resulting curves are shown dashed in Fig. 1. The agreement between the theoretical and experimental curves is satisfactory for the undoped sample but becomes increasingly poor with increased doping. The conductance maximum, which was greater than the theoretical prediction for the doped sample, becomes smaller than theory predicts as the concentration of impurities is increased. The most striking feature of the data is that with increasing doping the experimental conductance becomes much larger than the theoretical prediction at low voltages.

Figure 2 shows a plot of the zero-voltage conductance  $g(0)$  for one of the diodes versus reduced temperature  $t = T/T_c$ . Data for an undoped sample and the BCS prediction in this case are also included. The inset shows a plot of  $\Delta g = 1 - g(0)$  near  $T_c$ . It is seen that as  $t \rightarrow 1$ ,  $\Delta g \rightarrow 0$  with zero slope. The values for the doped sample are more than an order of magnitude smaller than for the undoped sample near  $T_c$ . These properties are consistent with the expected gaplessness near  $T_c$ .

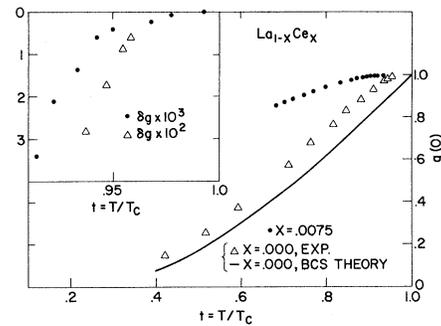


FIG. 2. Plot of the zero-voltage conductance  $g(0)$  versus reduced temperature  $t = T/T_c$  for one doped sample and one undoped sample. The curve represents the BCS prediction. The inset shows the deviation from the normal-state conductance  $\delta g = 1 - g(0)$  near  $T_c$ .

The large deviations at low energies are not due to the impurity spins becoming correlated since susceptibility measurements<sup>12</sup> indicate these correlations are small for concentrations less than 8%. The deviations are consistent with Maki's suggestion<sup>6</sup> that there may be states in the gap for systems which exhibit resonant scattering. These states result from the energy dependence of the pair lifetime  $\tau$ . Work is in process on checking Maki's prediction<sup>6</sup> that near  $T_c$  AG theory should be corrected by the insertion of the correct energy-dependent pair lifetime  $\tau(E)$ .

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Note added in proof.—Sugawara and Eguchi have recently revised their estimate of  $J$  for Ce in La to  $-0.053$  eV.<sup>13</sup>

<sup>1</sup>A. A. Abrikosov and L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **39**, 1781 (1960) [translation: Soviet Phys. -JETP **12**, 1243 (1961)].

<sup>2</sup>S. Skalski, O. Betbeder-Matibet, and P. R. Weiss, Phys. Rev. **136**, A1500 (1964).

<sup>3</sup>F. Reif and Michael A. Woolf, Phys. Rev. Letters **9**, 315 (1962); Rev. Mod. Phys. **36**, 238 (1964). Michael A. Woolf and F. Reif, Phys. Rev. **137**, A557 (1965).

<sup>4</sup>Tadashi Sugawara and Hiroko Eguchi, J. Phys. Soc. Japan **21**, 725 (1966).

<sup>5</sup>One can treat the interactions of conduction electrons with a magnetic impurity as a scattering problem. The scattering is large for electrons near the Fermi surface if the ratio of the energy separation of the impurity level from the Fermi surface to the im-

purity energy level width is small (10 or less). This situation is described as resonant scattering.

<sup>6</sup>Kazumi Maki, Phys. Rev. 153, 428 (1967).

<sup>7</sup>B. T. Matthias, H. Suhl, and E. Corenzwit, Phys. Rev. Letters 1, 92 (1958).

<sup>8</sup>B. Coqblin and A. Blandin, to be published.

<sup>9</sup>A. S. Edelstein, Phys. Rev. (to be published).

<sup>10</sup>D. L. Johnson and D. K. Finnemore, Phys. Rev. 158, 376 (1967).

<sup>11</sup>H. J. Levinstein, V. G. Chirba, and J. E. Kunzler, Phys. Letters 24A, 362 (1967).

<sup>12</sup>Magnetic measurements have been made on bulk fcc samples. Our interpretation of these measurements differs from that of Ref. 4 and will be published subsequently.

<sup>13</sup>Tadashi Sugawara and Hiroko Eguchi, Institute for Solid State Physics, University of Tokyo, Technical Report Ser. A, No. 271, 1967 (unpublished).

## EVIDENCE FOR THE EXISTENCE OF THE HYPERNUCLEUS $\Lambda \text{Li}^6$

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An event has been observed in nuclear emulsion which is attributed to the decay of a  $\Lambda \text{Li}^6$  hypernucleus,  $\Lambda \text{Li}^6 \rightarrow \pi^- + p + p + \text{He}^4$ , with a total binding energy of  $B = 3.92 \pm 0.37$  MeV relative to the free-particle system  $\Lambda + p + \text{He}^4$ .

During a systematic investigation of mesonic decays of hypernuclei into four charged particles, an event was observed which is best interpreted as the decay

$$\Lambda \text{Li}^6 \rightarrow \pi^- + p + p + \text{He}^4, \quad (1)$$

with a binding energy  $B = 3.92 \pm 0.37$  MeV relative to the free-particle system  $\Lambda + p + \text{He}^4$ .

A stack of Ilford K5 nuclear emulsion was exposed to about  $10^6$  stopping  $K^-$  mesons from a separated  $K^-$  beam at the Brookhaven alternating-gradient synchrotron. The density of the emulsion pellicles at the time of exposure was determined by the measurement of the range of 38 proton tracks from the decay of  $\Sigma^+$  hyperons. The protons have a mean range of  $R = 1707 \pm 4 \mu$ , compared with  $R = 1677 \pm 2 \mu$  for standard nuclear emulsion (density  $\rho = 3.815 \text{ g/cm}^3$ ).

The hypernucleus is emitted from a  $K^-$ -meson capture star together with three other nucleonic charged particles. The hypernucleus track, which has a dip angle of  $7^\circ$  relative to the emulsion plane, exhibits a thinning down of its profile towards the end of its range which is typical of stopping fragments of charge  $Z \geq 3$ .

It has a range of  $38.1 \mu$  and decays mesonically into four charged particles. The pion from the decay star comes to rest in the emulsion giving rise to a three-prong star, and its track does not exhibit detectable inelastic collisions. The measurement data for the production and the decay star are given in Table I. A photomicrograph of the event is shown in Fig. 1.

For the analysis of the decay star a  $\chi^2$ -min-

imizing computer program<sup>1</sup> has been used. This enables a certain probability to be attributed to each of the solutions fitting the kinematics of the decay. Fits were tried under the assumptions that the hypernucleus decayed at rest and that, in turn, (a) only particles giving visible tracks were emitted from the decay; (b) one neutron could be emitted in the decay; and (c) one charged particle making no visible track could be emitted in addition to the visible ones. Also tried were fits for a hypernucleus decay in flight.

Only one fit under assumption (a) was found, i.e., the decay  $\Lambda \text{Li}^6 \rightarrow \pi^- + p + p + \text{He}^4$ , yielding a momentum unbalance before the fit of  $\Delta P = 14.7 \text{ MeV}/c$ , and a value of the binding energy, relative to the free-particle system ( $\Lambda + p + \text{He}^4$ ) after the fit,  $B(\Lambda \text{Li}^6) = 3.92 \pm 0.37 \text{ MeV}$ . The  $\chi^2$  value for this hypothesis, correspond-

Table I. Measurement data of the event 5K-70-21.

Track identity	Range ( $\mu\text{m}$ )	Dip angle $\lambda$ (deg)	Azimuth $\phi$ (deg)
Production star			
1	1580	-34.99	347.52
2	151.8	-39.39	351.53
3	63.0	0.00	273.59
Hyperfragment	38.1	7.06	238.93
Decay star			
$\pi^-$	3434	-7.37	242.95
$p$	1551	11.24	0.00
$p$	25.7	-7.48	192.21
$\text{He}^4$	7.5	-13.94	153.53