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Electron Tunneling into Dilute CuCr and CuFe Alloy Films

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ABSTRACT

Tunneling experiments on dilute homogeneous magnetic alloys of Cu by means of Al/oxide/CuCr and Al/oxide/CuFe junctions have shown that there is no concentration dependent structure in the tunneling density of states to $< 10^{-3}$ of the background conductance over a wide range of temperature, concentration and magnetic fields, in agreement with recent theoretical calculations. Some concentration independent structure was observed which appears to be due to the Cu itself, and is not presently understood.

In a search for structure due to the Kondo effect in the tunneling density of states, we have performed two independent sets of experiments on dilute magnetic alloys of Cu by means of thin film tunnel junctions of the form Al/oxide/CuX, where X=Cr for the Berkeley experiments and X=Fe for the Orsay experiments. The junction characteristics were studied as a function of temperature, impurity concentration, and magnetic field; in no case have we observed any effect attributable to the presence of magnetic impurities in the Cu.

The experiments may be summarized briefly. The temperature dependence of the resistivity of the CuX films was studied from 0.9°K up. The results were consistent with temperature dependences reported for bulk alloys[1,2] and were used to determine the impurity concentration. Low temperature tunneling measurements performed on the same films by means of Al/oxide/CuX junctions did not exhibit any concentration or magnetic field dependent structure over the measured bias range of $|eV| \lesssim 100$ meV. The normalized differential conductance, G(V) was, to the precision of our measurements, the same for pure Cu as for the CuX alloys up to the solubility limit ($\lesssim 10^3$ ppm) of X. This contradicts previous experiments[3] in which large zero-bias conductance peaks were observed. The junctions reported here all showed excellent quantitative agreement with calculations[4] based on the BCS theory at 0.9°K where the Al was superconducting and exhibited no zero bias anomalies.[5]

The CuX films were evaporated from specimens of bulk alloy by means of electron-beam bombardment in an ion-pumped ultra-high vacuum system.[6] Films were generally several microns thick and were deposited

onto a substrate containing previously oxidized Al cross strips in a geometry which allowed resistance probes to be attached directly to the CuX film. Both groups verified that their pure Cu alone exhibited no resistance minimum to $\lesssim 10^{-4}$, and that the operation of the Ti getter pump introduced no structure. The properties of the films studied are given in Table I. Consistent determinations of impurity concentration were made by comparing the temperature dependence of the resistivity with known data for bulk alloys.[1,2] The impurity concentration was of the order of 1/2 (X=Fe) to 1/10 (X=Cr) that of the initial ingot, which appears to be due to fractional distillation during the evaporation. However, only a small portion of the evaporation was used for our data, and one of us (GR) several times verified that three consecutive films produced during a single evaporation had almost identical resistivity curves. These facts and the resistive behavior indicate that the impurities are homogeneously distributed throughout the film.

In the Orsay experiments on CuFe, the shape of the conductance is shown in Fig. 1. For the alloys listed in Table I, no concentration dependent structure is observed. There is, however, a definite symmetrical, concentration independent structure centered at V=0. This structure was found to be quantitatively the same in all junctions studied (even in very pure Cu with < 1 ppm nominal Fe concentration) and with very different conditions of evaporation. It was absent in Al/oxide/normal Pb junctions prepared under the same conditions.

In the Berkeley experiments on CuCr typical conductance curves for a given concentration are shown in Fig. 2. The small peaks shown

are residual superconductivity at the edges of the Al film. The conductance is clearly temperatnre and magnetic field independent to better than .05%. The data were also concentration independent over this bias range. The unusual concentration-independent structure mentioned in the Orsay experiments was also observed at Berkeley; it was qualitatively the same, but its magnitude varied markedly from sample to sample (completely uncorrelated with the Cr concentration).

Summarizing the two sets of experiments, we have observed that: a) the presence of Fe or Cr impurities in Kondo alloys of Cu produces no visible structure in the tunneling characteristics of Al/oxide/CuX diodes over a wide range of concentrations and magnetic fields, and at temperatures both above and below the Kondo temperature, T_k; and b) there is some symmetrical structure about the origin, apparently due to the Cu alone, the magnitude of which varies between our laboratories. Qualitatively we agree that a careful fit of the background conductance to a parabolic shape for 35 meV $<|eV|<$ 70 meV shows this structure to consist of a symmetric pair of resistance peaks at $|eV| \sim 20$ meV with a resistance dip below the parabolic fit near $|eV| = 0$. Of course, this analysis depends strongly on how one chooses the background conductance.

In order to compare the experiments with theory, it is necessary to separate the two possible contributions of magnetic impurities to the conductance. The usual expression for the tunneling current in terms of one-electron Green's functions $G_{\underset{\sim}{k}\alpha}(\varepsilon)$ is[7]

$$I(V) = \frac{2\pi e}{\hbar} \sum_{kk'\alpha\alpha'} |T_{\underset{\sim}{k}\underset{\sim}{k}'\alpha\alpha'}|^2 \int_{-\infty}^{\infty} \frac{d\varepsilon}{\pi} \int_{-\infty}^{\infty} \frac{d\varepsilon'}{\pi} \times$$

$$\times \, \mathrm{Im}G_{\underset{\sim}{k}\alpha}(\varepsilon) \, \mathrm{Im}G'_{\underset{\sim}{k}'\alpha'}(\varepsilon)[f(\varepsilon)-f(\varepsilon')]\delta(\varepsilon'-\varepsilon-eV), \qquad (1)$$

where the primed terms refer to one metal and the unprimed to the other, $\underset{\sim}{k}$ and α are the momentum and spin of an electron, $|T|^2$ is the transition matrix element, $f(\varepsilon)$ is the Fermi function and $\mathrm{Im}G_{\underset{\sim}{k}\alpha}(\varepsilon)$ is the many-body spectral weight function. Solyom and Zawadowski have recently shown[8] that a <u>spatial</u> variation of impurities in the immediate vicinity of the barrier leads to local variations of the self energy and modifies the spectral weight function.[9] This local modification is responsible for the "zero-bias anomalies" observed in many systems. Therefore the absence of such anomalies in our data self-consistently verifies the homogeneity of our films and the validity of the theoretical model.[10]

For homogeneous alloys, their calculation shows that one regains the usual expression for the spectral weight function,

$$\mathrm{Im}G_{\underset{\sim}{k}\alpha}(\omega) = \frac{\Sigma_2(\underset{\sim}{k},\alpha,\omega)}{[\omega-\varepsilon_{\underset{\sim}{k}\alpha}-\Sigma_1(\underset{\sim}{k},\alpha,\omega)]^2 + [\Sigma_2(\underset{\sim}{k},\alpha,\omega)]^2}, \qquad (2)$$

where $\varepsilon_{\underset{\sim}{k}\alpha}$ is the bare particle energy and $\Sigma(\underset{\sim}{k},\alpha,\omega) = \Sigma_1 + i\Sigma_2$ is the self energy contribution. Assuming the quasi-particle approximation to be valid, we decompose Σ_2 into Coulomb, electron-phonon, and exchange[11] contributions,

$$\Sigma_2(\underset{\sim}{k},\alpha,\omega) = \Sigma_2^{\mathrm{coul}} + \Sigma_2^{\mathrm{el-ph}} + \Sigma_2^{\mathrm{Kondo}}. \qquad (3)$$

Making the usual approximations,[7] $\Sigma_2^{\mathrm{el-ph}}$ contributes at most

$\sim (\theta_D/T_F) \lesssim 10^{-2}$ to the conductance of a normal junction. By comparison, neglecting the momentum dependence of Σ_2^{Kondo}, it will contribute at most a term of order $(T_K/T_F) < 10^{-3}$. To the extent that the quasiparticle approximation is valid for our CuX systems this is consistent with our experimental results.

To summarize, there is good agreement between present theoretical models and our observations of the absence of any concentration-dependent symmetric structure when tunneling into homogeneous CuX alloy films. The unusual symmetric structure appears to be a property of the Cu film, and further investigations are being made.

FOOTNOTES

* Laboratoire associe' au CNRS.

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1. DAYBELL, M. D. and STEYERT, W. A., Phys. Rev. Lett. 20, 195 (1968).
2. MONOD, P., Phys. Rev. Lett. 19, 1113 (1967) and private communication. L. D. and E. G. wish to thank P. Monod for many discussions on the Kondo problem.
3. HEDGCOCK, F. T. and RIZZUTO, C., Phys. Lett. 24A, 17 (1967).
4. BERMON, S., Technical Report No. 1, Grant #NSF-GP1100, University of Illinois, 1964 (unpublished).
5. G. R. has observed some anomalous structure for concentrations exceeding the solid solubility of Cr in Cu.
6. Previous experiments by L. D. and E. G. using joule heated crucibles at $\sim 10^{-7}$ torr frequently displayed non-reproducible spurious structures.
7. See, for example, SCHRIEFFER, J. R., Tunneling in Solids, E. Burstein and S. Lundquist, eds., p. 287, Plenum Press, New York (1969).
8. SOLYOM, J. and ZAWADOWSKI, A., Phys. Kondens. Materie 7, 325 (1968).
9. APPELBAUM, J. A. and BRINKMAN, W. F., Phys. Rev. 186, 464 (1969), and private communications have derived the same result by a somewhat different method.
10. The zero-bias anomaly formally resembles the Kondo problem in that both effects are due to the sharpness of the Fermi surface when scattering is computed in the second Born approximation, leading to anomalously long lifetimes near the Fermi level. This does not

imply that they must be simultaneously observable in a given physical system.

11. BOWEN, S. P., Phys. Rev. (to be published) and private communication has calculated the imaginary self energy term due to the Kondo effect in the quasi-particle approximation and has shown that its magnitude is of the order of $k_B T_k$ for these alloys. G. R. wishes to thank S. Bowen for communicating his results prior to publication and for helpful discussions.

TABLE I

Physical parameters of the CuX films used in the Al/oxide/CuX tunneling experiments and the deviation of the tunneling conductance from the results on pure Cu.

Sample	X	CuX thickness (Angstroms)	Nominal Impurity Concentration* (ppm)	Deviation from Pure Cu behavior
Orsay D_1†	Fe	3,500	70	$< 10^{-3}$
Orsay B_4†	Fe	3,600	250	$< 10^{-3}$
Orsay C_5†	Fe	13,000	800	$< 10^{-3}$
Berkeley 8‡	Cr	3,500	8	$< 2\times10^{-4}$
Berkeley 7‡	Cr	16,500	12	$< 2\times10^{-4}$

* Derived from film resistivity data.

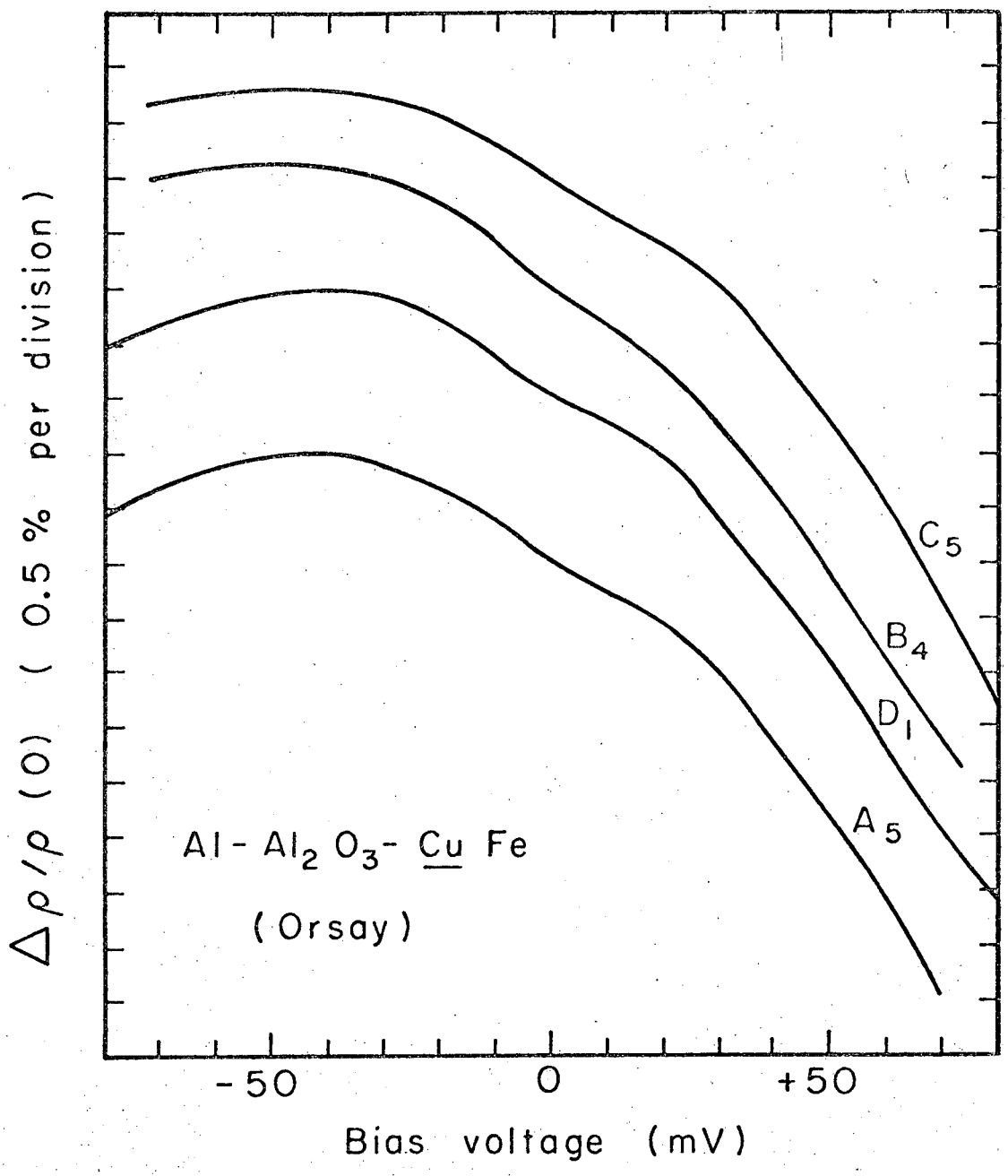
† Conductance independent of magnetic fields up to 35,000 Oe at 1°K.

‡ Conductance independent of magnetic fields up to 25,000 Oe at 1°K.

FIGURE CAPTIONS

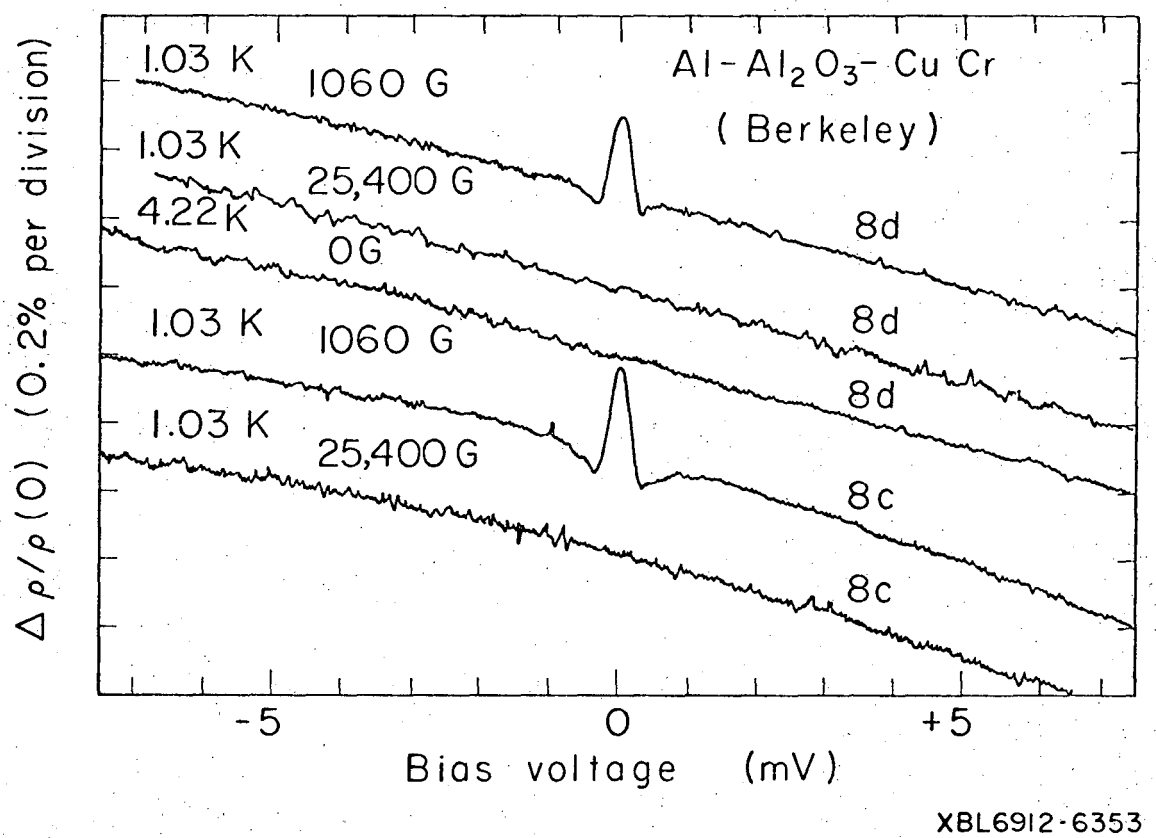
Fig. 1. Expanded scale plot of resistivity versus bias voltage for Al/oxide/CuFe junctions at 4.2°K for several different impurity concentrations. A_5 is the pure Cu sample; the others are described in Table I.

Fig. 2. Expanded scale plot of resistivity versus bias voltage for two similar Al/oxide/CuCr junctions (8 ppm Cr) as a function of temperature and magnetic field. The small peak at the center is due to residual superconductivity at the edges of the Al film.



XBL6912-6354

Fig. 1



XBL6912-6353

Fig. 2

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