Comment on "Phase diagram of reentrant and magnetic-field-induced superconducting states with Kondo impurities in bulk and proximity-coupled compounds"

Guy Deutscher,[1] Shay Hacohen-Gourgy,[1] and A. C. Mota[2]

[1]*Raymond and Beverly Sackler School of Physics and Astronomy, Tel-Aviv University, 69978 Tel-Aviv, Israel*

[2]*Solid State Laboratory, ETH-Zurich, CH-8093 Zurich, Switzerland*

(Received 10 August 2012; published 1 April 2013)

The Meissner effect induced in a normal metal in close contact with a superconductor is known to weaken at very low temperatures, and to be restored by the application of a moderate magnetic field [Visani *et al.*, Phys. Rev. Lett. **65**, 1514 (1990)]. This reentrant behavior was recently interpreted as resulting from the presence of Kondo impurities in the normal metal, with calculated concentrations on the order of 100 ppm [Simons *et al.*, Phys. Rev. B **86**, 064509 (2012)]. We show here that in fact the presence of magnetic impurities in the normal metal at such concentrations is ruled out by the absence of a minimum of resistivity in the same samples down to 2 K.

DOI: 10.1103/PhysRevB.87.136501 PACS number(s): 74.62.Dh, 74.25.Dw, 74.45.+c, 75.20.Hr

Discovered in the 1960's, the occurrence of a Meissner effect in a normal metal in close contact with a superconductor[1] has continued attracting attention following measurements at lower temperatures on samples made of a cylindrical superconducting core embedded in a normal metal matrix of Cu, Ag, or Au.[2] The early experiments by the Orsay group in films extended only down to about 1 K and showed Meissner screening currents extending progressively in the normal metal further and further away from the superconductor/normal metal interface.[3] But the later work of the Zurich group showed that at very low temperatures the Meissner effect, after saturating, became, in fact, weaker.[2] Furthermore, it was also found that the application of a moderate magnetic field restored the full Meissner effect.

So far, this reentrant behavior is not well understood. Bruder and Imry tried to explain it in terms of glancing states,[4] however, Belzig *et al.* later found the effect to be too small to fit the experiment.[5,6]

Recently, Simons *et al.*[7] presented a simple approach for understanding reentrant and magnetic-field-induced superconducting behavior in Kondo systems and have used it to explain the paramagnetic reentrant effect measured in proximity cylinders.[2] Spin-flip scattering off impurities weakens superconductivity, and therefore could trigger a weakening of the proximity effect at low temperatures. But this scattering can be suppressed by the application of a magnetic field that aligns the impurity spins, an effect that gives rise to the well known negative magnetoresistance of Kondo alloys. Hence the idea that a magnetic field large enough to align the spins, but small enough so that it does not destroy induced superconductivity, can restore the full Meissner effect. This can in principle happen at low enough temperatures where the field required to align the spins becomes smaller than the field necessary to break down superconductivity, known as the breakdown field.[8] Simons *et al.* calculated that the concentration of magnetic impurities necessary for their Kondo interpretation of reentrance to apply falls in the range of several 100 ppm up to 1000 ppm. They claim that this concentration is small enough to have passed unnoticed by Visani *et al.*,[2] Mota *et al.*,[9] Muller-Allinger, and Mota,[10] and Muller-Allinger and Mota.[11]

However, this interpretation is contradicted by the fact that the reentrance phenomenon is more pronounced in the cleaner samples (Nb/Ag)[11] than in the less clean ones (Nb/Au).[10] It is well known that it is extremely difficult to eliminate impurities such as Fe from Au, but not from Ag where 1 ppm impurity levels can be achieved. In Nb/Ag samples the breakdown field was indeed found to fit the clean limit value, while the reentrance effect was most prominent. In Nb/Au samples the breakdown field value did not fit the clean limit value, and reentrance was weaker. If Kondo impurities were at the origin of reentrance, the opposite behavior would have been expected. Based on the results of Mota *et al.*,[9] it is therefore unlikely that the Kondo interpretation of reentrance fits the experiments.

Nevertheless, we felt that a direct check of the presence or absence of magnetic impurities in the samples studied by Mota and co-workers was called for. To this effect we have measured the temperature dependence of the resistance of Nb/Ag and Nb/Au composite wires from room temperature down to the superconducting critical temperature of Nb (9.2 K) in zero applied field, and down to 2 K under an applied field of 1.5 T, sufficient to quench superconductivity in the Nb core.

The Nb/Au sample used here was from the same batch as that used by Muller-Allinger and Mota,[10] having a Nb core with a diameter of 23 μm and a Au cladding 3.2 μm thick. The Nb/Ag wire had a Nb core 70 μm thick and a Ag cladding 10 μm thick. The Nb/Ag sample used by Muller-Allinger and Mota[11] was drawn from that wire, made with 99.9999 pure Ag. Four point measurements were performed in a physical property measurement system (PPMS), using a 1 K/min cooling rate and a 200 μA measuring current.

$R(T)$ measurements on Nb/Ag and Nb/Au samples under zero applied field are shown Figs. 1(a) and 1(b). The $R(T)$ curves show no minimum above 9 K. In fact, the resistances of both samples decrease slowly while they approach saturation until the critical temperature of Nb is reached.

There remains, however, the possibility that a minimum of resistance could exist below the critical temperature of Nb.

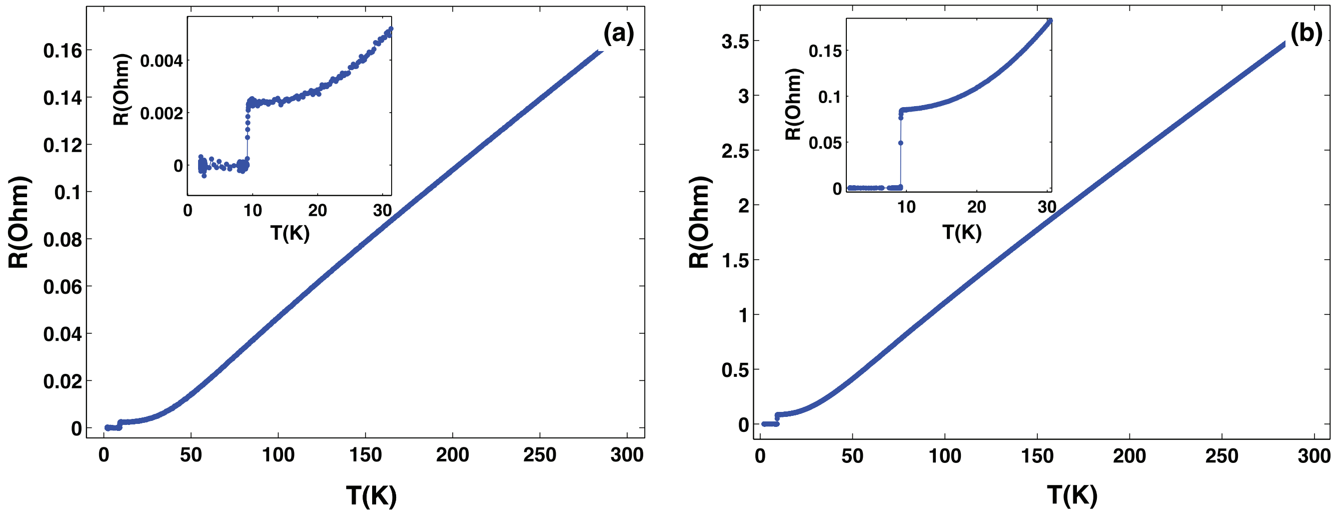

FIG. 1. (Color online) Temperature dependence of the resistance of (a) Nb/Ag and (b) Nb/Au wires in zero applied field. Insets are a detailed view of the data at low temperatures.

To check whether this is the case, we have measured $R(T)$ under a magnetic field large enough to quench superconductivity in the Nb core. Its critical field was found to be of about 1 T at 2 K. A field of 1.5 T was used for the measurements shown Fig. 2 for the Nb/Ag sample. Similar results were obtained for the Nb/Au sample. In both cases $R(T)$ flattens out towards a constant value. There is no evidence of any Kondo-like increase in resistance down to 2 K. The magnetoresistance appears to be weak and positive at low temperatures.

In order to establish an upper limit to the possible concentration of magnetic impurities in the Ag and Au claddings, we have calculated the value of the residual resistivity of the normal metal based on the resistance ratio of the composite wire between room temperature and the constant value reached at low temperatures. In this calculation we have neglected the contribution of the Nb core to the total conductance of the wires. This is justified by the fact that this core is evidently in the dirty limit, based on the fact that its critical field (1 T) is large compared to that of pure Nb (0.2 T).

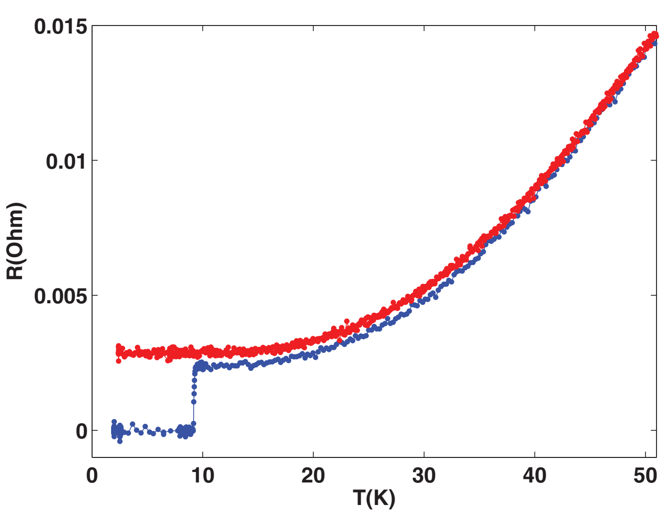

FIG. 2. (Color online) Temperature dependence of the resistance of a Nb/Ag wire under an applied field of 1.5 T (red), compared to that in zero applied field (blue). Down to 2 K, there is no indication of an increase of resistance.

For the Nb/Ag wire the resistance ratio is about 60. The Ag cladding is therefore pure enough to ascribe to its room temperature resistivity the value of pure Ag, equal to 1.6 $\mu\Omega$ cm, from which we obtain for the residual resistivity the value 0.027 $\mu\Omega$ cm. This is of the same order as the residual resistivity of a 70 ppm AgMn alloy.[12] But in fact this alloy has a minimum of resistance at 8 K. If a minimum exists in our Ag cladding, it occurs at a temperature smaller than 2 K. The concentration of Mn or similar impurities in our Ag cladding is therefore extremely small, at the ppm level according to the law that the temperature of minimum resistivity varies as the concentration of magnetic impurities to the 1/5 power.

In AuFe Kondo alloys, the residual resistivity is given by Ford *et al.*[13] as 7.6 $\mu\Omega$ cm/at. %. The resistance ratio in our Nb/Au wires is equal to 40, and based on the room temperature resistivity of pure Au of 2.4 $\mu\Omega$ cm, this corresponds to a residual resistivity of 0.06 $\mu\Omega$ cm. Assuming that it is entirely due to Fe impurities, this corresponds to a 80 ppm concentration. But, in fact, for a 100 ppm AuFe alloy the temperature of minimum resistance is equal to 9 K. Again, since in our Nb/Au wire the temperature of minimum resistance is less than 2 K, the concentration of Fe must be in the few ppm range.

In conclusion, the absence of a minimum of resistance around 10 K for the Nb/Ag and Nb/Au composite wires from the same batch as used previously by Mota and co-workers[2,9] rules out the presence of magnetic impurities at the level of 100 ppm estimated for the Kondo interpretation of reentrance proposed by Simons *et al.*[7] to apply. In fact, based on the absence of a minimum down to 2 K, the concentration of Kondo impurities can be no more than a few ppm.

[1] J. P. Burger, G. Deutscher, E. Guyon, and A. Martinet, Phys. Lett. **17**, 180 (1965).
[2] P. Visani, A. C. Mota, and A. Pollini, Phys. Rev. Lett. **65**, 1514 (1990).
[3] G. Deutscher and P. G. de Gennes, in *Treatise on Superconductivity*, edited by R. D. Parks (Dekker, New York, 1969), p. 1005.
[4] C. Bruder and Y. Imry, Phys. Rev. Lett. **80**, 5782 (1998).
[5] A. L. Fauchere, W. Belzig, and G. Blatter, Phys. Rev. Lett. **82**, 3336 (1999).
[6] W. Belzig, C. Bruder, and Y. V. Nazarov, J. Low Temp. Phys. **147**, 95 (2007).
[7] Y. B. Simons, O. Entin-Wohlman, Y. Oreg, and Y. Imry, Phys. Rev. B **86**, 064509 (2012); In this article, a more detailed assessment of the impurity concentrations needed to explain reentrance was given as ranging from 70 ppm up to 430 ppm.
[8] Orsay Group on Superconductivity, in *Quantum Fluids*, edited by D. Brewer (North-Holland, Amsterdam, 1965), p. 26.
[9] A. C. Mota, P. Visani, A. Pollini, and K. Aupke, Physica B **197**, 95 (1994).
[10] F. B. Müller-Allinger and A. C. Mota, Phys. Rev. B **62**, R6120 (2000).
[11] F. B. Müller-Allinger and A. C. Mota, Phys. Rev. Lett. **84**, 3161 (2000).
[12] D. Jha and M. H. Jericho, Phys. Rev. B **3**, 147 (1971).
[13] P. J. Ford, T. E. Whall, and J. W. Loram, Phys. Rev. B **2**, 1547 (1970).