Observation of Magnetic-Field-Induced Superconductivity

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A new superconducting state induced by an external magnetic field has been observed in the pseudoternary Eu-Sn molybdenum chalcogenides for different Eu concentrations. This phenomenon is explained in terms of the Jaccarino-Peter compensation effect which accounts correctly for the shape of the H_{c2}-T phase diagram.

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The study of the interplay of superconductivity and magnetism in ternary compounds has led to a number of new and exciting results.[1] Among the unusual phenomena that have been observed is the anomalous behavior of the critical field H_{c2} in the pseudoternary series $Eu_xSn_{1-x}Mo_6S_8$.[2] This observation has been interpreted in terms of the Jaccarino-Peter compensation mechanism,[3] which can account for both the enhancement of H_{c2} and its temperature anomalies around $x \simeq 0.8$. The negative sign of the exchange interaction necessary for this effect has been confirmed independently by both experiment[4,5] and theory.[6]

The compensation effect was originally proposed as a mechanism that would allow superconductivity to be induced in a weak ferromagnet by a high external magnetic field H. However, this field-induced superconductivity may occur equally well in the paramagnetic state if the exchange field H_J is larger than the paramagnetic limiting field $H_p(\sim T_c)$. In this case there will be two superconducting domains in the H-T diagram, one at low fields where the magnetization is sufficiently small and one at high fields, where superconductivity is possible when the polarization of the conduction-electron spins by the magnetic ions is compensated by the external field.[7] Thus a measurement of the resistivity as a function of field at low temperature should consecutively show a transition to the normal state, then to the superconducting state again, and finally to the normal state at very high fields (S-N-S-N behavior). If one applies this theory to the $Eu_xSn_{1-x}Mo_6S_8$ series it is found that such a situation may occur for certain values of the parameters that can be obtained by adjusting T_c. Around $x \simeq 0.8$, T_c of this series varies rapidly as a function of x because of a structural phase transformation appearing for $x > 0.8$.[8] As a result, close to $x \simeq 0.8$, T_c passes through a narrow interval consistent with field-induced superconductivity. A first indication of such a behavior has been reported by Isino, Kobayashi, and Muto[9] in a sample with nominal composition $Eu_{0.8}Sn_{0.2}Mo_6S_7$. More recently Wolf *et al.*[10] found a similar indication in $Eu_{1.2}Mo_6S_8$ at 14 kbar. Unfortunately, in both cases relatively broad superconducting transitions made the interpretation difficult. At this point we would like to stress that the range of composition that allows one to observe this effect is very narrow. This imposes very stringent conditions on the sample quality. One has to produce very homogeneous samples with narrow transitions at a very precise composition.

We have followed two different approaches to obtain samples with the necessary quality. In one series of samples, $Eu_xSn_{1-x}Mo_6S_8Br_\epsilon$, we added a small quantity ϵ of Br as a transport agency to favor the homogenization. The correct conditions were obtained by precisely adjusting x within the domain $0.78 \leq x \leq 0.82$. In the second series, $Eu_xSn_{1-x}Mo_6S_{8-y}Se_y$, we chose a somewhat lower Eu content ($x = 0.75$ and 0.70) so that T_c and the transition width were less influenced by the structural phase transformation. This gave samples with narrower transitions, but with too high values of T_c for $y = 0$. In order to adjust T_c to the optimum value we substituted small quantities of Se for S. It is indeed well known that such substitutions usually lead to a lowering of T_c in the ternary molybdenum sulfides.[11] The samples were all prepared by a hot pressing technique as usual.[8] X-ray analysis did not show any impurity phase. The critical field $H_{c2}(T)$ was determined by measuring the resistance of the sample as a function of field either in a ^{3}He-^{4}He dilution refrigerator combined with a superconducting magnet (12 T) or in a ^{3}He-refrigerator inserted into a polyhelix magnet (25 T). In the following we shall concentrate mainly on the behavior of one sample of the second series, $Eu_{0.75}Sn_{0.25}Mo_6S_{7.2}Se_{0.8}$ whose composition had been adjusted to place T_c in the narrow interval that allows the observation of the field-induced superconductivity.

The superconducting transition temperature of that sample in zero field, defined as the midpoint of the resistive transition, was 3.89 K with a transition width $\Delta T_c = 0.6$ K. Figure 1 shows the resistance R of the sample versus applied magnetic field H. At $T = 3.55$ K, the $R(H)$ curve is ordinary. The phenomenon of magnetic-field-induced superconductivity sets in below 1 K: Superconductivity is first destroyed by a weak magnetic field and R reaches more than 70% of its normal-state value. At higher fields, R starts to decrease again and disappears at about 10 T. The high-field state subsists up to fields of the order of 20 T, which can be seen from Fig. 2. At $T = 0.37$ K, the multiple transition *S-N-S-N* is clearly displayed. It should be emphasized that the field and temperature dependence of the resistance observed here cannot be explained by the well-known "peak effect" near H_{c2}.[12] The position of the resistance maximum at magnetic fields $H << H_{c2}$, the crucial composition dependence of the induced superconducting state, and the experimental conditions (field parallel to the measuring current, low current density $j \sim 10$ A/m^2) exclude an interpretation in terms of the peak effect. Note also, that in the low-field region (Fig. 1) a typical reentrant phenomenon is observed.

The upper critical field $H_{c2}(T)$, which is defined as the midpoint of the resistance transition, is presented in Fig. 3. The new field-induced superconducting domain is well defined and extends from about 4 to 22 T. A similar behavior was also observed in two samples of the first series, $Eu_{0.805}Sn_{0.195}Mo_6S_8Br_{0.1}$ ($T_c = 4.56$ K) and $Eu_{0.80}$-$Sn_{0.20}Mo_6S_{7.8}Br_{0.2}$ ($T_c = 4.58$ K), whereas a sample with the composition $Eu_{0.80}Sn_{0.20}Mo_6S_8Br_{0.1}$ ($T_c = 5.02$ K) showed a strongly anomalous H_{c2} with a nearly infinite slope at 2.6 K.

The solid line in Fig. 3 represents the critical field which was calculated by use of a multiple pair-breaking theory.[7] This theory deduces an implicit equation for $H_{c2}(T)$ containing as parameters the Maki parameter α, the spin-orbit scattering parameter $\lambda_{s.o.}$, a magnetic scattering parameter λ_m, the superconducting transition temperature T_c, the field- and temperature-dependent exchange field H_J, and the magnetization M. For the computation we used the measured T_c and took $\lambda_m = 0$, because magnetic scattering can be neglected in the series Eu_x-$Sn_{1-x}Mo_6S_8$.[8] The contribution of M to the mag-

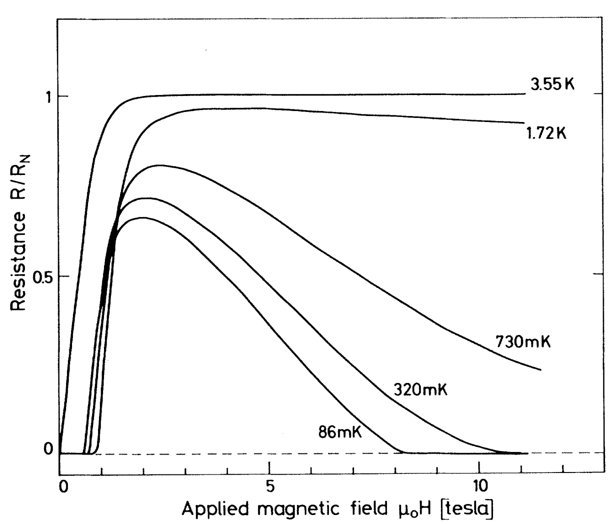

FIG. 1. Normalized resistance R/R_N of $Eu_{0.75}$-$Sn_{0.25}Mo_6S_{7.2}Se_{0.8}$ vs field ($H \leq 12$ T) at various temperatures. R_N is the resistance in the normal state.

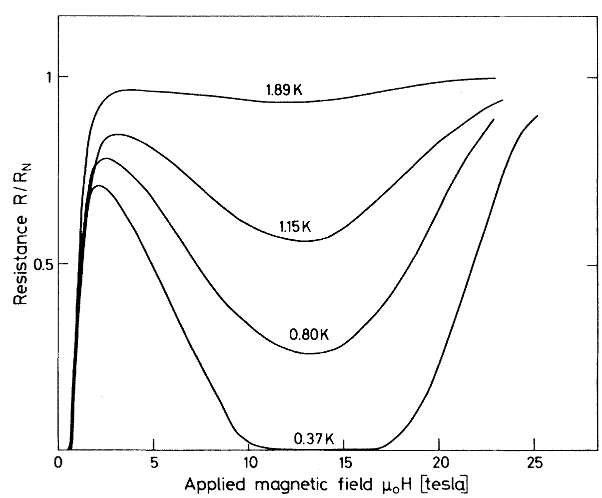

FIG. 2. Normalized resistance R/R_N of $Eu_{0.75}$-$Sn_{0.25}Mo_6S_{7.2}Se_{0.8}$ vs field ($H \leq 25$ T) at various temperatures.

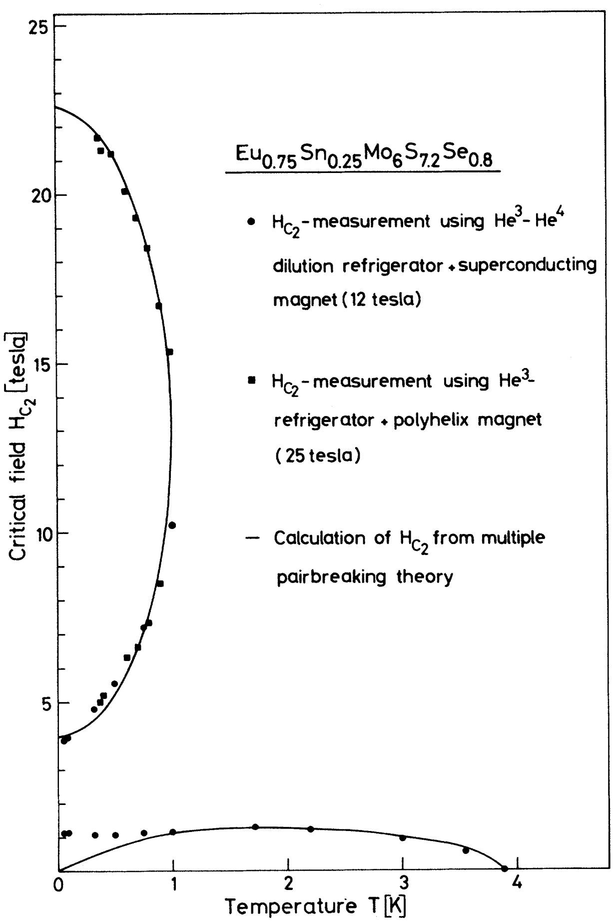

FIG. 3. Upper critical field H_{c2} vs temperature T.

netic induction B is negligible, too. Because of the strong paramagnetic limitation an accurate measurement of the initial slope was not possible so that there are three fitting parameters left. The best fit has been obtained with the following set of parameters: $\alpha = 4.75$ corresponding to an initial slope of 9 T/K, $\lambda_{s.o.} = 8$, and $H_{J0} = -30.2$ T at saturation. This fit is rather unique in that a small variation in any of the parameters will lead to an important change in $H_{c2}(T)$. The value for $\lambda_{s.o.}$ is in good agreement with the one reported for the pseudobinary system $Mo_6Se_{8-y}S_y$.[11] In the nonmagnetic reference compound $PbMo_6S_7Se_1$ an initial slope of nearly 8 T/K has been measured, so that our result for α does not seem unreasonable. For describing the temperature and field dependence of the exchange field we used the Brillouin function for Eu^{2+} ions. This leads to a saturation value of the exchange field, which is consistent with the value found in $EuMo_6S_8$ under pressure,[13] if one assumes a linear dependence of H_J on the Eu concentration. The departure observed in the low-field and low-temperature regime is not very surprising because of an antiferromagnetic ordering effect which is expected to appear below 1 K.[14] If we vary T_c in our calculation, keeping the other parameters fixed, we find that the field-induced state should appear only for samples $Eu_{0.75}Sn_{0.25}Mo_6S_{8-y}Se_y$ with T_c values in the interval 3.6 K $\leq T_c \leq$ 4.1 K. Applying the same analysis to the samples of the first series shows that T_c has to be in the interval 3.7 K $\leq T_c \leq$ 4.6 K, in agreement with our observations. The difference in the required T_c values between the two series is directly related to the difference in H_J. It should be pointed out that our interpretation does not depend at all on how the critical field is defined. Taking another definition, say, the endpoint of the resistive transition, simply means that one modifies correspondingly T_c, keeping the other parameters fixed.

Recently the critical field of the pressure-induced superconductivity in $EuMo_6S_8$ was investigated on a high-quality sample.[13] No field-induced superconductivity was observed. This is in accord with our result; in the investigated pressure range T_c is too high for this phenomenon to occur, and the compensation mechanism manifests itself only in an anomalous $H_{c2}(T)$ curve.

In conclusion, we have observed in the series $Eu_xSn_{1-x}Mo_6S_8$ containing small quantities of Br or Se, a field-induced superconducting domain in the H_{c2}-T diagram. The shape and size of this domain depend critically on the Eu concentration and T_c. In the sample $Eu_{0.75}Sn_{0.25}Mo_6S_{7.2}Se_{0.8}$, presented in detail here,[15] this domain extends from 4 to 22 T at $T = 0$ and from $T = 0$ to $T = 1$ K at $H \simeq 12$ T. This effect may be observed in samples with different Eu concentrations provided that T_c is adjusted correspondingly. We have interpreted this phenomenon in terms of the Jaccarino-Peter compensation effect and find a remarkably good agreement for reasonable values of the three fitting parameters α, $\lambda_{s.o.}$, and H_J. According to this theory the high-field domain exists only as a result of a delicate balance between the internal exchange field and the externally applied field. In spite of this good agreement several important questions relating to the exact nature of the field-induced state remain open: Investigations of properties like the superconducting gap, the behavior of the flux-line lattice, and the order of the field-induced transition are certainly necessary to get a complete understanding of this phenomenon.

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