Superconductivity and Magnetic Interactions in $\text{Yb}_{1.2-x}\text{Eu}_x\text{Mo}_6\text{S}_8$ Pseudoternary Compounds

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The superconducting and magnetic properties of the $\text{Yb}_{1.2-x}\text{Eu}_x\text{Mo}_6\text{S}_8$ series of compounds have been investigated by means of ac magnetic susceptibility, electrical resistivity, and upper critical magnetic field $H_{c2}$ measurements. The superconducting transition temperature at ambient pressure is depressed by Eu additions, until superconductivity disappears completely at $x \sim 0.7$. For Eu concentrations $x < 0.45$, $H_{c2}$ is enhanced at low temperatures and exceeds the values for pure $\text{Yb}_{1.2}\text{Mo}_6\text{S}_8$. The temperature dependence of $H_{c2}$ for the various $\text{Yb}_{1.2-x}\text{Eu}_x\text{Mo}_6\text{S}_8$ compounds is analyzed using a multiple pair-breaking theory that includes the compensation of the applied magnetic field by the negative exchange field produced by antiferromagnetic interactions between the conduction electron spins and the Eu$^{2+}$ spins (Jaccarino–Peter effect). Hydrostatic pressure was found to induce superconductivity in all Eu-rich compositions that were not superconducting at ambient pressure.

1. INTRODUCTION

The superconducting transition temperatures $T_c$ of the ternary rare earth R molybdenum chalcogenides with general composition $\text{R}_x\text{Mo}_6\text{X}_8$ (where $x = 1.0-1.2$; $\text{X} = \text{S}, \text{Se}$) show a systematic variation with $\text{R}$ (ref. 1) that can be accounted for in terms of the theory of Abrikosov and Gor'kov (AG). 2 Exceptions to this systematics include $\text{Ce}_{1.2}\text{Mo}_6\text{X}_8$ and $\text{Eu}_{1.2}\text{Mo}_6\text{X}_8$ ($\text{X} = \text{S}$ and Se), which do not exhibit superconductivity, and $\text{Yb}_{1.2}\text{Mo}_6\text{S}_8$, which has a $T_c$ much higher than expected. 1 While most R ions in the ternary $\text{R}_x\text{Mo}_6\text{X}_8$ compounds are trivalent, Eu (ref. 3) and Yb (ref. 4) are in nearly divalent states. Baillif et al. 5 found that a structural phase transition at $\sim 109$ K prevents superconductivity from occurring in $\text{EuMo}_6\text{S}_8$ at ambient pressure. This structural transformation can be suppressed by hydrostatic pressure, 6 and pressure-induced superconductivity in $\text{EuMo}_6\text{S}_8$ occurs at a $T_c$ (refs. 6–8) which is also much higher than what one would

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expect from the AG theory. The high $T_c$ values of $\sim 7.3$ K for Yb$_{1.2}$Mo$_6$S$_8$ (sintered chunk) and $\sim 14$ K for Eu$_{1.2}$Mo$_6$S$_8$ (zero-pressure extrapolated value for a melted sample) are probably associated with the large unit cell volume due to the divalent character of the Yb and Eu ions, as suggested by Sergent et al. Thus, it is not surprising that these two compounds do not follow AG systematics for trivalent R in RMo$_6$S$_8$ compounds.

One of the most striking effects due to Eu in ternary molybdenum chalcogenides was discovered by Fischer et al., who reported that the upper critical magnetic fields $H_{c2}$ of Sn$_{1.2}$Mo$_{6.35}$S$_8$ and PbMo$_{6.35}$S$_8$ are significantly enhanced by partial substitution of Eu for Sn or Pb. The enhancement of $H_{c2}$ was attributed to compensation of the applied magnetic field by a negative exchange field due to an antiferromagnetic exchange interaction between the conduction electron spins and the Eu$^{2+}$ magnetic moments, via a mechanism first suggested by Jaccarino and Peter (JP). Nuclear magnetic resonance and Mössbauer effect studies by Fradin et al. on Sn$_{0.5}$Eu$_{0.5}$Mo$_6$S$_8$ showed a negative s-band spin polarization at the Mo sites, thus supporting the interpretation of the enhancement of $H_{c2}$ in terms of the JP compensation effect. Partial substitution of Eu for La (ref. 13) or Yb (ref. 14) has also been found to enhance $H_{c2}$ in La$_{1.2}$Mo$_6$S$_8$ and Yb$_{1.2}$Mo$_6$S$_8$ through the same magnetic field compensation mechanism. The JP compensation effect is also responsible for magnetic field-induced superconductivity at very low temperatures ($T < 1$ K) in Eu$_{0.75}$Sn$_{0.25}$Mo$_6$S$_{7.2}$Se$_{0.8}$, and, possibly, in Ho$_{1-x}$Eu$_x$Mo$_6$S$_8$ compounds.

Reported herein are the results of an investigation carried out on Yb$_{1.2-x}$Eu$_x$Mo$_6$S$_8$ pseudoternary compounds by means of measurements of low-frequency ac magnetic susceptibility $\chi_{ac}$ to determine $T_c$ and $H_{c2}$ at atmospheric pressure and $T_c$ under pressure up to 20 kbar, and low-frequency ac electrical resistivity $\rho$.

2. EXPERIMENTAL DETAILS

An impedance bridge operating at a frequency of 16 Hz was employed to determine $\chi_{ac}$ and $\rho$ using standard mutual inductance and four-lead techniques, respectively. Pressures up to 20 kbar were attained in Be–Cu clamped piston-cylinder devices using a 50:50 mixture of $n$-pentane and isoamyl alcohol as the nearly hydrostatic pressure-transmitting medium and a superconducting Pb manometer to determine the pressure. Magnetic fields up to 100 kG were applied with a NbTi superconducting solenoid. Preliminary results were discussed in ref. 14.

The Yb$_{1.2-x}$Eu$_x$Mo$_6$S$_8$ compounds were prepared by sintering. Commercially available powders of Yb$_2$S$_3$, EuS, Mo, and S were ground together, pressed into pellets, and reacted in an inert atmosphere at 800°C for 24 h.