Anomalous temperature dependence of the upper critical field of the magnetic superconductor NdMo₆S₈

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(Submitted 28 March 1984)

Pis'ma Zh. Eksp. Teor. Fiz. 39, No. 10, 456-458 (25 May 1984)

The $H_{c2}(T)$ curve of NdMo₆S₈ has a maximum at $T \sim 1.3$ K.

Stoichiometric ternary compounds containing magnetic rare-earth ions have recently attracted considerable research interest. Among these compounds are the ternary chalcogenides of molybdenum, ^{1,2} REMo₆X₈ (X is S or Se, and RE is a rare earth), and compounds of the composition³ RERh₄B₄. Although about 10% of the ions in the lattice are magnetic, many compounds of these groups are superconductors (see Ref. 4, for example), apparently because the exchange interaction is quite weak in these compounds.

Research on the electrical, magnetic, and thermal properties of these superconductors has revealed several anomalous features in some of them. It has been suggested that a magnetic ordering occurs at temperatures below T_c (Refs. 3, 5, and 6). Neutron-scattering measurements have revealed that the compounds ${\rm ErRh_4B_4}$ (Ref. 7) and ${\rm HoMo_6S_8}$ (Ref. 8) do in fact exhibit a ferromagnetic ordering of the rare-earth ions, which disrupts the superconductivity at $T_m < T_c$. In the compounds ${\rm DyMo_6S_8}$, ${\rm TbMo_6S_8}$, and ${\rm ErMo_6S_8}$ (Ref. 9), on the other hand, an antiferromagnetic ordering which occurs at $T_m < T_c$ does not disrupt the superconductivity, simply giving rise to an anomalous feature on the temperature dependence of the upper critical field $H_{c2}(T)$.

In this letter we report observing ananomalous temperature dependence $H_{c2}(T)$ for the compound NdMo₆S₈. Several measurements were carried out with samples of the compound DyMo₆S₈, which has been studied previously.⁶ The samples were prepared by direct synthesis of the necessary amounts of Mo, S, and NdS (the composition of the original material was Nd_{1.2}Mo₆S₈) at T = 900 °C. The samples were rendered homogeneous by pulverizing them in an agate mortar, compression, and annealing at T = 1100 °C for about 20 h.

The measurements in the temperature interval 0.5–1.5 K were taken in an apparatus with He-3 vapor pumping at the International Laboratory of Strong Magnetic Fields and Low Temperatures, Wroclaw, Poland. Temperatures down to 0.05 K were produced by adiabatic demagnetization of yttrium aluminum garnet with a partial substitution of erbium¹⁰ in an apparatus with a cold plate similar to that described in Ref. 11.

Figure 1 shows the resistance r(T, H) of an NdMo₆S₈ sample versus the temperature for various magnetic fields. At H = 0 the sample goes superconducting at $T_c = 3.4$ K. In magnetic fields up to 1 kOe, the sample remains completely superconducting at temperatures below 2 K. At higher magnetic fields, an incomplete superconducting transition is observed, and the resistance increases as the temperature is lowered below 1 K. At $T \sim 0.3$ K, the r(T) curve has a slight maximum, at a position

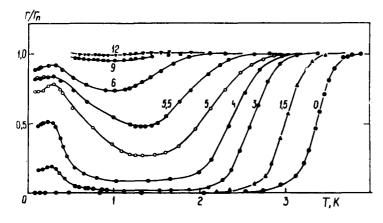


FIG. 1. Superconducting transitions of NdMo₆S₈. The curves are labeled with the magnetic field H in kilo-oersteds.

which does not depend on the magnetic field. In contrast, the temperature at which the resistance begins to rise before the maximum on the r(T) curve depends strongly on the magnetic field. It should be noted that the measurements were taken at a constant value of H; the increases in H were carried out systematically after heating above T_c .

The results of the measurements of r(T, H) for the compounds $DyMo_6S_8$ agree well with the data of Ref. 6. At fields above 1 kOe, lowering the temperature below T_c produces a sharp maximum on the r(T) curve, at a position which does not depend on the magnetic field. In contrast with the results for $NdMo_6S_8$, in the compound $DyMo_6S_8$ the temperature at which the resistance increases in front of the maximum is essentially independent of the magnetic field.

If we plot $H_{c2}(T)$ from the centers of the transitions, we find curves with a maximum. For DyMo₆S₈, the $H_{c2}(T)$ curve has a characteristic minimum at $T\sim0.2$ K. In contrast with the results of Ref. 6, at T<0.2 K we observe a further increase in $H_{c2}(T)$ with decreasing temperature. For NdMo₆S₈, we observe a far fainter structural feature on the $H_{c2}(T)$ curve at $T\sim0.2$ K.

Our results on r(T, H) and $H_{c2}(T)$ for NdMo₆S₈ are basically similar to those obtained in Ref. 6 on ErMo₆S₈. For ErMo₆S₈, an antiferromagnetic ordering has been

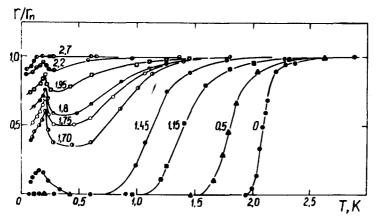


FIG. 2. Superconducting transitions of DyMo₆S₈. The curves are labeled with the magnetic field H in kilo-oersteds.

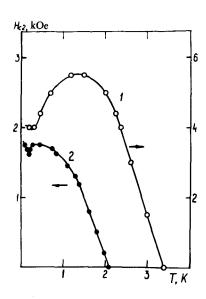


FIG. 3. Temperature dependence of the upper critical magnetic field H_{c2} . 1—NdMo₆S₈; 2—DyMo₆S₈.

observed in neutron-scattering measurements in a field of 250 Oe at $T\sim0.2$ K (Ref. 9). In Ref. 6, an $H_{c2}(T)$ curve with a maximum, similar to that which we found in the present measurements for NdMo₆S₈, was attributed to an increase in the effective internal "depairing" field with decreasing temperature. This field is associated with the increase in the magnetic susceptibility of the sublattice of Er^{3+} ions, which precedes the magnetic ordering. The $H_{c2}(T)$ dependence observed for NdMo₆S₈ in the present experiments can apparently be explained in a similar way.

The neutron-scattering measurements of Ref. 9 showed that the compound DyMo_6S_8 goes into an antiferromagnetically ordered state at $T \sim 0.4$ K.

In summary, we have observed an anomalous $H_{c2}(T)$ dependence (with a maximum) for the compound NdMo₆S₈. This dependence is similar to that observed previously⁶ for the compound ErMo₆S₈. The apparent reason for this anomalous structural feature is an antiferromagnetic ordering at $T \sim 0.3$ K in the system of Nd³⁺ ions, with the compound remaining in the superconducting state.

We wish to thank Yu. A. Deniskin, E. P. Khlybov, A. S. Rudenko, and Kh. Medge for assistance in the experiments.

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Translated by Dave Parsons

Edited by S. J. Amoretty