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Magnetic behaviour of the rare earth molybdenum silicides $RE_2Mo_3Si_4$ with the $U_2Mo_3Si_4$ structure type

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Abstract

Ternary rare earth molybdenum silicides $RE_2Mo_3Si_4$ have been synthesized by the arc melting technique for RE = Gd, Tb, Dy, Ho, Er, Tm and Y. From X-ray powder diffraction analysis, all the compounds were found to crystallize in the $U_2Mo_3Si_4$ structure type (space group: $P2_1/c$). Magnetic susceptibilities were generally measured in the temperature range from 2 to 300 K. $Y_2Mo_3Si_4$ is a temperature-independent paramagnet. Except for $Er_2Mo_3Si_4$ and $Tm_2Mo_3Si_4$, the studied compounds show antiferromagnetic ordering at low temperature. $Tb_2Mo_3Si_4$ is the only one which shows two antiferromagnetic transitions (19 and 2.3 K). Moreover $Gd_2Mo_3Si_4$, $Tb_2Mo_3Si_4$ and $Dy_2Mo_3Si_4$ exhibit metamagnetic type behaviour.

Keywords: Ternary silicides; Rare earths; Uranium; Antiferromagnetism

1. Introduction

Intensive studies have been devoted to the physics of strong electron correlation in metallic systems involving 5f or 4f electron metals. In 1984, Bodak et al. [1] announced the existence of a series of ternary silicides $RE_2Mo_3Si_4$ (RE = Tb→Tm, Y) which are isotypic with $U_2Mo_3Si_4$ [2,3]. The discovery in 1986 by Aliev et al. [4] of the occurrence of antiferromagnetic ordering (T_N = 19 K) and superconductivity at lower temperature (between 0.5 and 1.2 K) in the compound $Tb_2Mo_3Si_4$ has revealed the large interest of this structure type. To our knowledge no magnetic studies had been performed at low temperature on the full series of isotypic rare earth compounds. In this paper we describe the preparation and the magnetic behaviour of $RE_2Mo_3Si_4$ (RE = Gd, Tb, Dy, Ho, Er, Tm and Y).

2. Experimental details

The alloys $RE_2Mo_3Si_4$, each with a total mass of about 1 g, were synthesized by argon arc-melting ingots of the elements in a stoichiometric amount. The materials used were commercially available as high-purity elements: rare earths were in the form of chips (Aldrich and Auer-Remy GmbH, purity 99.9%), as well as silicon (Johnson-Matthey, purity 99.9%); molybdenum was in powder (Ventron, purity 99.9%). In order to ensure maximum homogeneity, the arc-melted pellets were ground before new arc-melting. Nevertheless, it was difficult to avoid small but detectable amounts of the binary Mo_5Si_3 in the final products. Ternary compounds could be formed only with RE = Gd, Tb, Dy, Ho, Er, Tm and Y. Attempts to prepare similar phases with Sm, Yb and Lu were unsuccessful.

3. Structural chemistry

The X-ray powder patterns of $RE_2Mo_3Si_4$ could be indexed on the basis of a primitive monoclinic cell with $P2_1/c$ as space group. By using the atom parameters as determined for $U_2Mo_3Si_4$ [3], excellent agreement is obtained between observed and calculated X-ray powder intensities, confirming isotypism among these compounds.

Lattice parameters and standard deviations (Table

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Table 1
Crystallographic and magnetic data for $RE_2Mo_3Si_4$ compounds

RE	a (Å)	b (Å)	c (Å)	β(°)	V (Å^3)	$\mu_{eff.}$ (μ_B/RE)	$\mu_{theo.}$ (μ_B/RE^{3+})	θ_p(K)	T_N (>2 K)	H_c (kOe)
Gd	6.872(1)	6.921(2)	6.759(1)	109.08(2)	303.7	8.29	7.94	4.9	10.0	8.0
Tb	6.883(1)	6.929(1)	6.781(1)	109.06(1)	305.5	9.55	9.72	11.0	19.0 and 2.3	8.0
Dy	6.826(2)	6.881(3)	6.722(1)	109.04(2)	298.5	10.42	10.63	5.2	12.0	2.0
Ho	6.822(2)	6.881(2)	6.714(1)	108.98(1)	298.0	10.49	10.60	3.0	4.8	—
Er	6.817(1)	6.875(1)	6.699(2)	109.00(2)	296.9	9.51	9.57	1.2	—	—
Tm	6.804(1)	6.859(1)	6.690(1)	109.01(1)	295.2	7.18	7.63	1.7	—	—
Y	6.843(2)	6.901(2)	6.735(1)	108.98(1)	300.8	TIP				

1) were obtained by a least squares refinement of room temperature diffractometer X-ray powder data, using CuKα_1 radiation with an external standard of silicon. They are in good agreement with previously reported values [1]. The variation of the volume as a function of the atomic number of the lanthanide element (Table 1) follows the lanthanide contraction.

4. Magnetic properties

The magnetic properties of $RE_2Mo_3Si_4$ compounds have been studied using a superconducting quantum interference device magnetometer in the temperature range 2–300 K and in magnetic fields up to 30 kOe. The results are summarized in Table 1 and in Figs. 1–6.

The values of the effective moments calculated from the thermal variation of the reverse susceptibility are in good agreement with the theoretical values for RE^{3+} ions.

Except for $Er_2Mo_3Si_4$, $Tm_2Mo_3Si_4$ and $Y_2Mo_3Si_4$ the compounds studied show an antiferromagnetic ordering of the rare earth sublattice above 2 K.

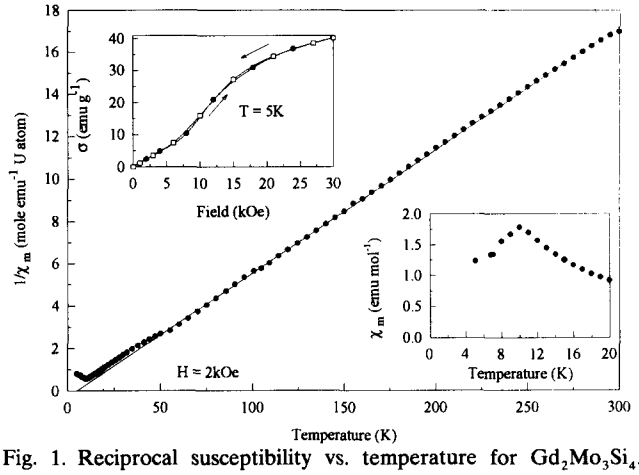

Fig. 1. Reciprocal susceptibility vs. temperature for $Gd_2Mo_3Si_4$. Insets: low-temperature susceptibility and magnetization vs. field.

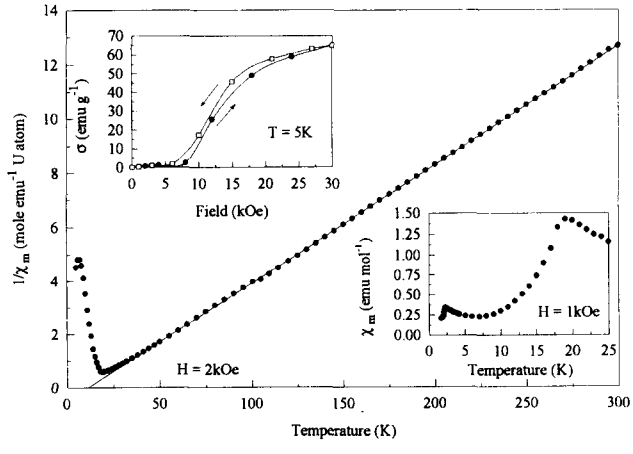

Fig. 2. Reciprocal susceptibility vs. temperature for $Tb_2Mo_3Si_4$. Insets: low-temperature susceptibility and magnetization vs. field.

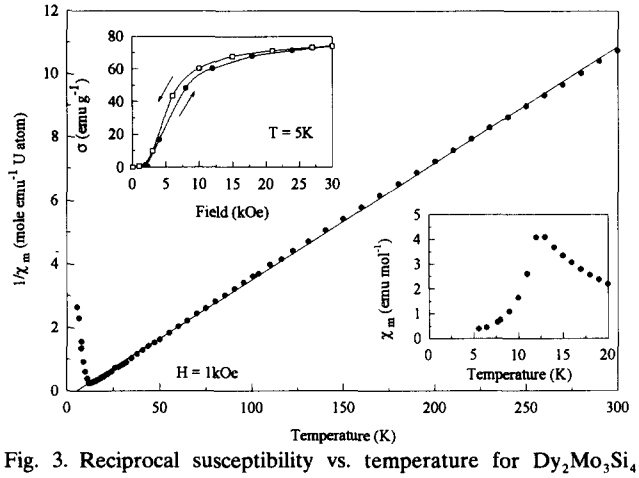

Fig. 3. Reciprocal susceptibility vs. temperature for $Dy_2Mo_3Si_4$. Insets: low-temperature susceptibility and magnetization vs. field.

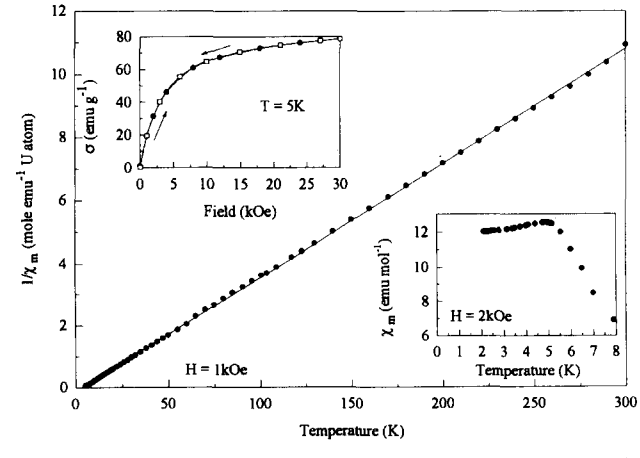

Fig. 4. Reciprocal susceptibility vs. temperature for $Ho_2Mo_3Si_4$. Insets: low-temperature susceptibility and magnetization vs. field.

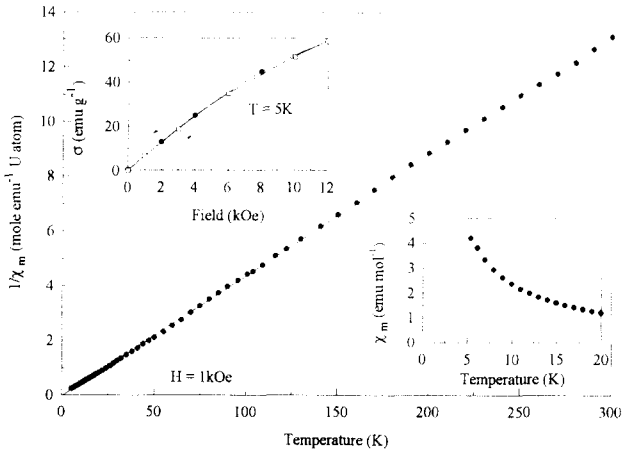

Fig. 5. Reciprocal susceptibility vs. temperature for $Er_2Mo_3Si_4$. Insets: low-temperature susceptibility and magnetization vs. field.

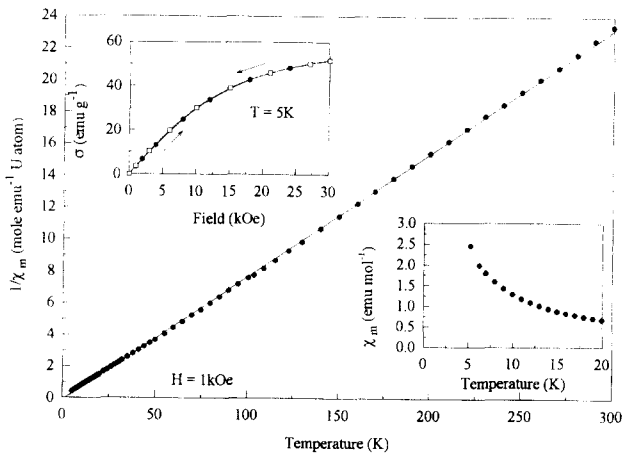

Fig. 6. Reciprocal susceptibility vs. temperature for $Tm_2Mo_3Si_4$. Insets: low-temperature susceptibility and magnetization vs field.

$Tb_2Mo_3Si_4$ exhibits quite an unusual behaviour, showing two antiferromagnetic transitions ($T_{N1} = 19$ K and $T_{N2} = 2.3$ K) and was also reported to exhibit superconductivity below 1 K [5]. The magnetic transitions were recently confirmed by specific heat measurements [5] and during the study of magnetic structure by neutron diffraction [6].

As expected from the positive values of the paramagnetic Curie temperature θ_p, metamagnetism was observed for $Gd_2Mo_3Si_4$, $Tb_2Mo_3Si_4$ and $Dy_2Mo_3Si_4$, the critical field H_c depending on the rare earth (Table 1). Concerning $Er_2Mo_3Si_4$ and $Tm_2Mo_3Si_4$, their paramagnetic Curie temperature is significantly lower, but it is probable that a transition exists at a temperature below 2 K.

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