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LETTER TO THE EDITOR

Magnetization behaviour of RCu_2Si_2 (R = Tb, Dy, Ho, Er and Tm) in the paramagnetic state

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Abstract. The result of DC magnetization (M) measurements on the compounds RCu_2Si_2 (R = Gd, Tb, Dy, Ho, Er and Tm) in the paramagnetic state up to 55 kOe are reported. In all cases, except that of Gd alloy, M is found to be a non-linear function of magnetic field tending to saturate at higher fields over a wide temperature range in the paramagnetic state. This magnetization behaviour is found to be essentially a single-ionic effect, presumably arising from strong 4f quadrupolar effects.

It is now recognized that 4f quadrupolar coupling effects play a major role not only in magnetic ordering [1–3], but also as regards the magnetic structure and the easy direction of magnetization in rare-earth compounds [4, 5]. However, very few attempts have been made to investigate seriously the existence of such quadrupolar effects on the magnetization behaviour in the paramagnetic state, as one tends to assume that isothermal magnetization (M) varies linearly with the magnetic field (H), as in any other paramagnetic material. In this respect the report of Morin *et al* [6] on the ferromagnetic-like isothermal magnetization behaviour in the paramagnetic state of *single-crystalline* TmZn gains importance. It has been assumed that such spectacular magnetization behaviour arises from cooperative quadrupolar effects [4]. In this article, the results of magnetization measurements above the Néel temperature for the ternary compounds RCu_2Si_2 (R = Tb, Dy, Ho, Er and Tm), crystallizing in the $ThCr_2Si_2$-type tetragonal crystal structure [7], are reported to demonstrate that such magnetization behaviour is widespread even among polycrystalline materials and that it is essentially a single-ionic effect. Initial results on the Tm sample have been reported earlier [8].

The polycrystalline samples, RCu_2Si_2 (R = Gd, Tb, Dy, Ho, Er and Tm), $Tm_{1-x}Y_xCu_2Si_2$ ($x = 0.5$ and 0.9), $Tm_{0.1}Lu_{0.9}Cu_2Si_2$ and $Ho_{0.1}Y_{0.9}Cu_2Si_2$ were prepared by arc melting followed by homogenization at 800 °C in an evacuated sealed quartz tube. The dependence of M on H up to 55 kOe was recorded at selected temperatures above T_N for all the alloys employing a superconducting quantum interference device (SQUID).

The results on the isothermal magnetization behaviour of RCu_2Si_2 (R = Gd, Tb, Dy, Ho, Er and Tm) are shown in figure 1. The values of T_N for R = Gd, Tb, Dy, Ho, Er and Tm have been reported to be about 13.5, 11.5, 11.0, 6.8, 4.8 and 6.9 K respectively [7]. One would therefore expect to find that the magnetization is a linear function of H above T_N. The experimental observations are in contrast to this expectation. For instance, in the case of Tm, as reported earlier [8], M tends towards saturation above 30 kOe at 10 K and this non-linear variation of M persists even at 30 K, though the field required to saturate M

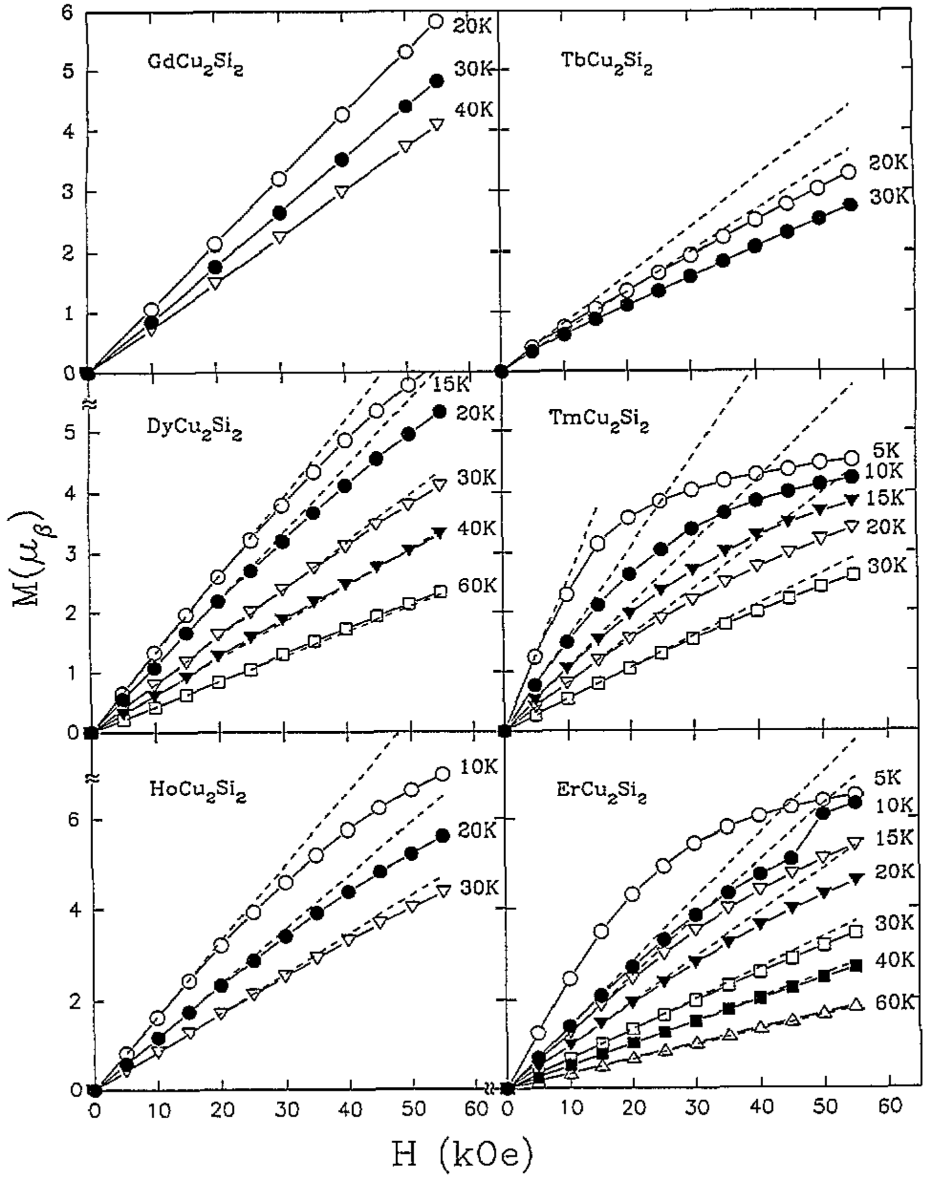

Figure 1. Isothermal magnetization behaviour of the compounds RCu_2Si_2 (R = Gd, Tb, Dy, Ho, Er and Tm). The continuous lines through the data points serve as guides to the eye. The dotted lines are in some cases drawn through the data below 10 kOe to highlight the non-linear behaviour.

increases with increasing temperature. This saturation behaviour of M persists over a wide temperature range for R = Tb, Dy, Ho and Er also (figure 1) and demonstrates that this ferromagnetic-like tendency is a more common phenomenon, but in the case of $GdCu_2Si_2$, M versus H is always linear above T_N. It is, therefore, clear that the non-linear behaviour is observed only for those rare earths for which the 4f quadrupole moment is non-zero, as if the alignment of the 4f quadrupole by the magnetic field [3, 9, 10] plays a vital role in the magnetization behaviour even in the paramagnetic state.

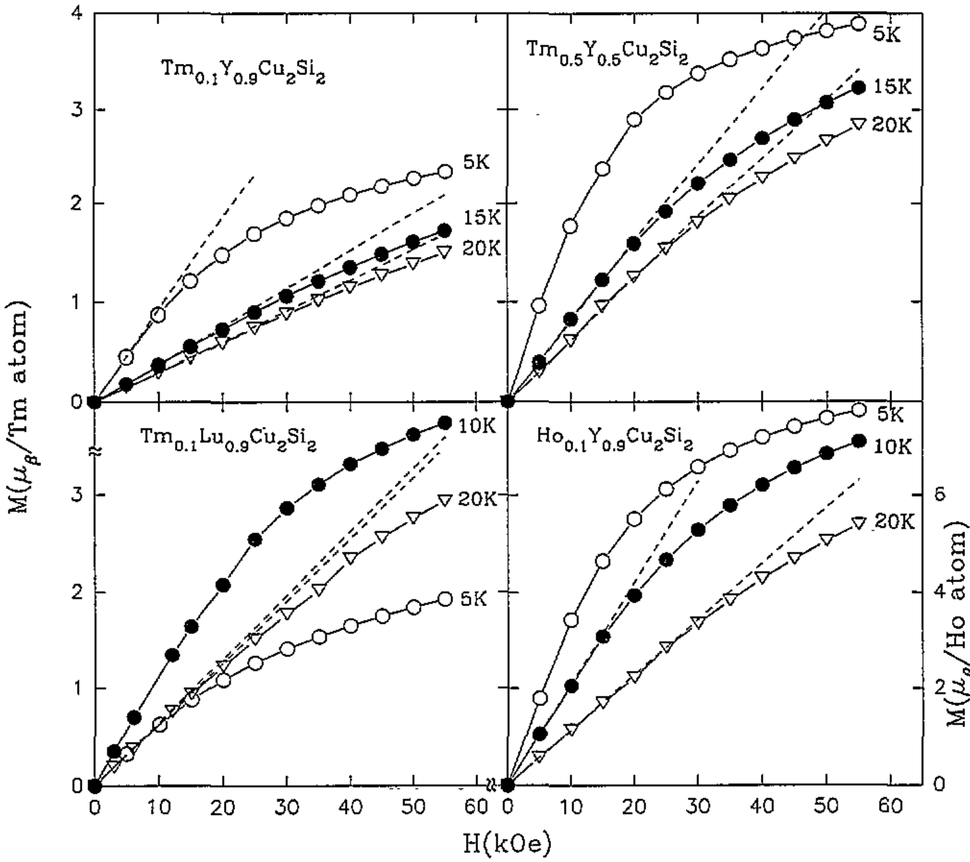

Figure 2. Isothermal magnetization behaviour as a function of external magnetic field for the alloys $Tm_{1-x}Y_xCu_2Si_2$ ($x = 0.5$ and 0.9) and $Tm_{0.1}Lu_{0.9}Cu_2Si_2$ and $Ho_{0.1}Y_{0.9}Cu_2Si_2$. The continuous lines drawn through the data points serve as guides to the eye. The dotted lines, in some cases, are drawn through the data below 10 kOe to highlight non-linear magnetization behaviour.

In order to address the question of whether the observed saturation behaviour of M is the result of cooperative quadrupole–quadrupole interaction, we have also performed the measurements on some of the alloys in which the R sublattice is diluted with non-magnetic ions, namely $Tm_{1-x}Y_xCu_2Si_2$ ($x = 0.5$ and 0.9), $Tm_{0.1}Lu_{0.9}Cu_2Si_2$ and $Ho_{0.1}Y_{0.9}Cu_2Si_2$. In the case of the Tm alloys, substitutions of both Y and Lu for Tm were undertaken to investigate whether there is any difference between the positive and the negative chemical pressure as regards the quadrupolar effects on the magnetization behaviour. The results are shown in figure 2. It is quite clear that the effect persists even when the R sublattice is diluted. The value of M scales approximately with the concentration of R ions; some discrepancy of course is noticed for the Tm alloys in the dilute limit ($x = 0.9$), which we attribute to the loss of Tm while arc melting. The results prove that the observed paramagnetic behaviour is not due to any cooperative effects, but is essentially a single-ionic effect. This finding tempts one to ask whether the observed findings are artefacts of crystal-field effects. However, it is difficult to believe that the crystal-field effects *always* result in the tendency of M to saturate at higher fields for *all heavy rare-earth members*, without any upward curvature in any of these alloys. Alternatively, strong 4f quadrupolar coupling effects can play a rule. In support of this, the observed magnetostriction behaviour [11] closely tracks the isothermal magnetization data reported here. The quadrupolar effects on the magnetization are expected to be primarily single-ionic, considering that [9, 10] the ionic magnetic moment of the Hund's rule ground state and the quadrupole moment of the ion have a common axis at all times, as a result of which the direction of magnetic moment is intimately connected with the orientation of the quadrupole dictated by the magnetic field.

To conclude, the results show that the isothermal magnetization need not be a linear function of H in the paramagnetic state for moderate values of H in those cases where the 4f orbital angular momentum is non-zero. The need to consider strong 4f quadrupolar coupling effects is suggested, as the quadrupoles are part and parcel of magnetism.

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