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LETTER TO THE EDITOR

Novel magnetization behaviour of $TmCu_2Si_2$ in the paramagnetic state

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Abstract. We report here the results of DC magnetization (M) and heat-capacity measurements on the compound $TmCu_2Si_2$. Although the results show that this compound is paramagnetic above 3 K, M in the paramagnetic state tends to saturate with the application of magnetic field. This spectacular behaviour of M mimics that known for single-crystal specimens of TmZn and we attribute this ferromagnetic-like tendency to strong 4f quadrupole couplings.

During the course of investigation of 4f quadrupolar effects in TmZn, Morin *et al* [1] observed a spectacular magnetization versus magnetic field (H) behaviour in the paramagnetic state of the single-crystal specimens. This behaviour was explained [2] in terms of 4f quadrupolar coupling effect above the Néel temperature (T_N), which is strong enough to induce a transition from the paramagnetic state to a ferromagnetic-like state with the application of H.

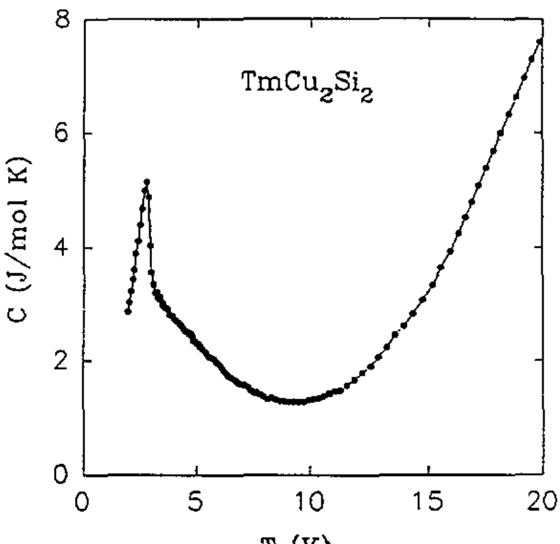

Figure 1. The heat-capacity (C) as a function of temperature (T) for $TmCu_2Si_2$.

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This transition occurs for a critical field which depends on the strength of the quadrupolar interactions. In view of the novelty of this concept, there is a need to look for such a behaviour of quadrupole-induced ferromagnetic tendency in various other compounds. In this respect, we thought it worthwhile to look for such anomalies in $TmCu_2Si_2$, as it appears [3, 4] that the Néel temperature (T_N) of this compound gets enhanced by a strong coupling between the 4f quadrupole and the magnetic moment. Interestingly, the M behaviour reported here is similar to that known for TmZn. These experimental observations, *noted even in the polycrystalline samples*, are of significant relevance to the field of magnetism, as the magnetization is traditionally expected to be proportional to a reasonable magnitude of the applied field in the paramagnetic state at temperatures above magnetic-ordering temperatures.

The polycrystalline sample, $TmCu_2Si_2$, was prepared by arc melting followed by homogenization at 800 °C in an evacuated sealed quartz tube. The magnetization behaviour of the compounds, $DyCu_2Si_2$ and $GdCu_2Si_2$ above T_N (10.9 K and 13.5 K respectively, [5]), was investigated for comparison and the samples are the same as those employed earlier [4]. The dependence of M on H was recorded up to 55 kOe employing a superconducting quantum interference device at various temperatures above 4.2 K. Heat-capacity (C) measurements on $TmCu_2Si_2$ in the temperature interval 2–100 K were also performed by a semi-adiabatic heat-pulse method.

The results of C measurements on $TmCu_2Si_2$ below 20 K are shown in figure 1. There is a distinct peak at 3 K in C of $TmCu_2Si_2$ indicating the existence of long-range magnetic order. In addition, C rises smoothly below 8 K; the origin of this rise is not clear at the moment. The electrical resistivity was shown to start decreasing at 7 K as the temperature is lowered, as if there is a phase transition [3] at this temperature, though the possible existence of a Schottky peak due to the crystal field effects was not discussed. There is no evidence for any phase transition in the data above 20 K (not shown in the figure). The C data therefore prove that this compound is paramagnetic above 8 K.

One therefore expects that the magnetization is a linear function of the magnetic field above 8 K. In contrast to this expectation, interestingly, M tends towards saturation above 30 kOe at 5 K and 10 K (figure 2). This non-linear variation of the magnetization persists even at 30 K, though the field range in which M tends to saturate increases with increasing temperature.

For comparison, we show the data for $GdCu_2Si_2$ (T_N = 13.5 K) and $DyCu_2Si_2$ (T_N = 10.9 K). Interestingly, even in the case of $DyCu_2Si_2$, there is a tendency for non-linear magnetization behaviour in a wide temperature range well above T_N. But in the case of $GdCu_2Si_2$, M versus H is always linear above T_N. This comparison suggests that the non-linear magnetization behaviour is observed only in those rare earths for which the 4f quadrupole moment is non-zero. This naturally implies that strong 4f quadrupolar coupling effects are responsible for the spectacular magnetization behaviour of $TmCu_2Si_2$, even in *the polycrystalline specimens. This means that the alignment of the 4f quadrupole caused* by the application of H forces the alignment of the magnetic moment as there is a stong coupling between the axes of the magnetic moment and quadrupole moment [2–4, 6–8].

To conclude, the polycrystalline samples of $TmCu_2Si_2$ exhibit novel magnetization behaviour in the paramagnetic state, which we attribute to strong 4f quadrupole coupling effects.

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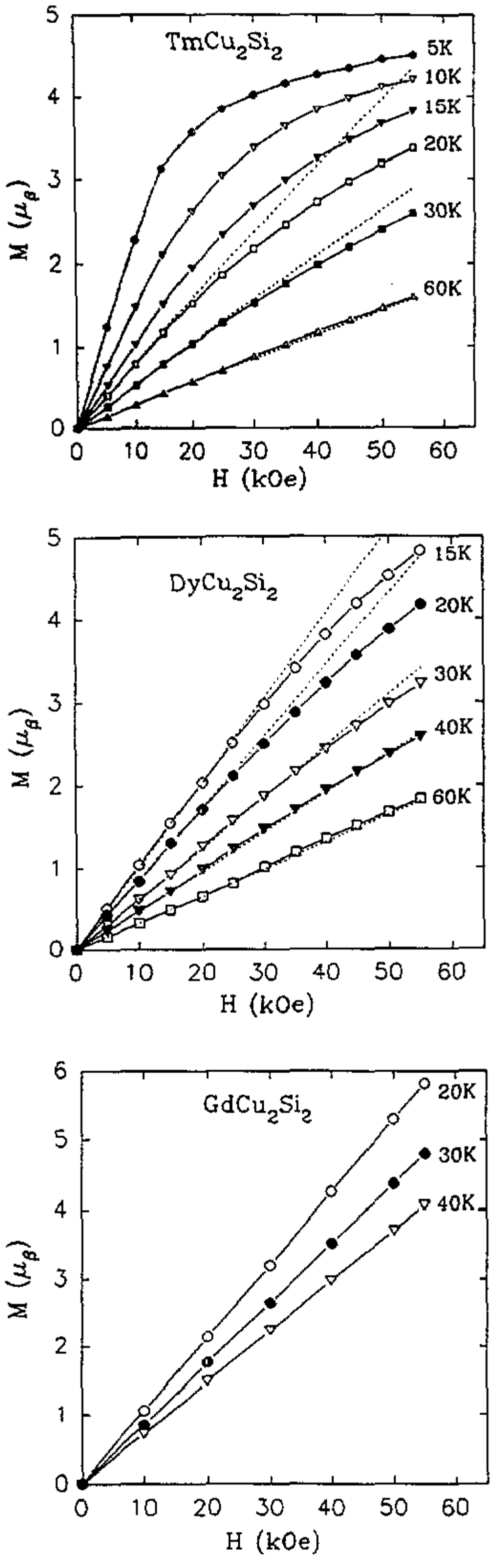

Figure 2. The magnetization (M) as a function of magnetic-field (H) at various temperatures for $TmCu_2Si_2$, $DyCu_2Si_2$ and $GdCu_2Si_2$. The solid lines drawn through the data points serve as guides to the eyes. The broken lines for $TmCu_2Si_2$ (only for $T > 20$ K) and $DyCu_2Si_2$ are linear extrapolations from the data below 10 kOe.

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