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The thermomagnetic irreversibility and metamagnetic behaviour of $DyCu_2Si_2$

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Abstract. The results of magnetization measurements on $DyCu_2Si_2$, which orders antiferromagnetically ($T_N \approx 11$ K), show interesting thermomagnetic irreversibility and metamagnetic behaviour. The possible origins of such effects are discussed.

Thermomagnetic irreversibility in the temperature dependence of magnetization (M), i.e. $M_{\mathrm{ZFC}} \neq M_{\mathrm{FC}}$, where ZFC and FC stand for the zero-field-cooled and the field-cooled state respectively, is traditionally thought to be the hallmark of spin-glass systems. In recent times such a characteristic has been used to establish the existence of spin-glass, re-entrant spin-glass and ferromagnet–spin-glass mixed states in quite a few ordered intermetallic compounds, e.g. U_2TSi_3, T = Fe, Co, Ni and Cu [1] and and UPd_2Ge_2 [2]. We have shown recently that it is possible to have thermomagnetic irreversibility in ordered compounds with long-range ferromagnetic or antiferromagnetic order, without invoking the idea of the spin-glass state [3–5]. To address this subject further, we shall present here the results of our magnetization study on $DyCu_2Si_2$. This compound, with $ThCr_2Si_2$ structure, orders antiferromagnetically at $T_N = 10.9$ K [6] and has been the subject of detailed investigation in connection with the question of the 4f–quadrupolar coupling effect on the magnetism of $ReCu_2Si_2$ compounds (Re = rare earth) [7].

Our polycrystalline sample of $DyCu_2Si_2$ was prepared by argon arc melting followed by homogenization at 800 °C in vacuo. For the magnetic measurements we used a commercial SQUID magnetometer (Quantum Design MPMS5). To eliminate the effect of anisotropy (if any, since anisotropy seems to be an intrinsic property of the compounds with $ThCr_2Si_2$ structure) we maintained the same sample position and orientation with respect to the external magnetic field throughout our measurements. We used a scan length of 4 cm, and for each measurement an average was taken over three scans, each scan containing 32 data points. This relatively short scan length minimizes the variation of the magnetic field in which the sample travels.

In figures 1 to 3, we present plots of the magnetization (M) versus temperature (T) for $DyCu_2Si_2$, obtained with the external field (H) kept fixed at 1 mT, 0.5 T or 5 T. Data also exist for $H = 5$ mT, 50 mT and 0.1 T but are not shown here for the sake of clarity and conciseness. For $H = 1$ mT (figure 1) there is a distinct peak at 11.5 K accompanied by a remarkable thermomagnetic hysteresis, i.e. $M_{\mathrm{ZFC}} \neq M_{\mathrm{FC}}$, which started well above the peak temperature (T_{peak}) and became very prominent below T_{peak}. Here ZFC (FC) stands for zero-field-cooled (field-cooled) measurements. This peak in the magnetization is indicative of antiferromagnetic ordering in this system [8]; however, for a simple antiferromagnet no

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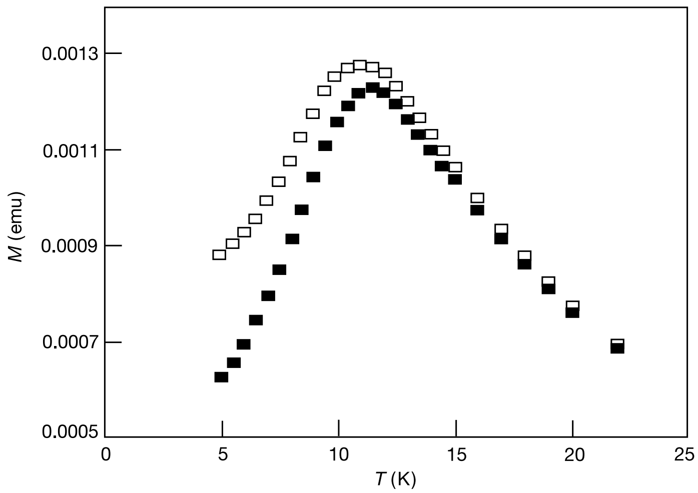

Figure 1. A plot of magnetization versus temperature for $DyCu_2Si_2$ with an external field being kept constant at 1 mT. ■ stands for ZFC and □ for FC measurements.

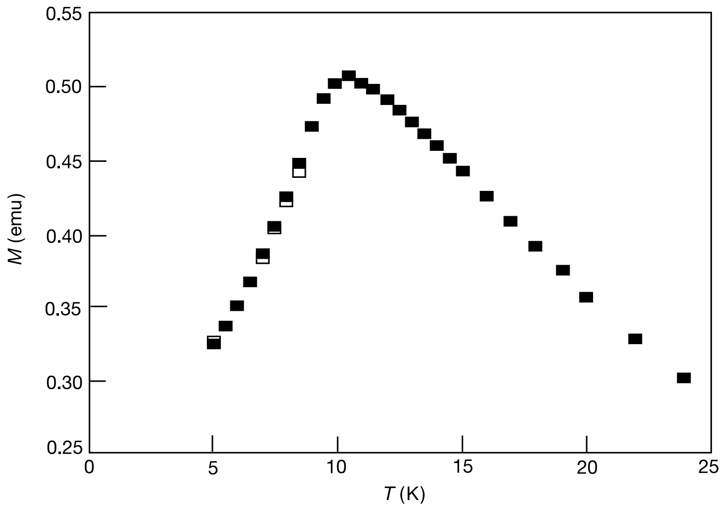

Figure 2. A plot of magnetization versus temperature for $DyCu_2Si_2$ with an external field being kept constant at 0.5 T. ■ stands for ZFC and □ for FC measurements.

thermomagnetic hysteresis is expected. This latter effect, although traditionally accepted as a hallmark of spin-glass systems, has also been observed recently in other antiferromagnetic systems with the $ThCr_2Si_2$ structure [3]. The onset temperature of irreversibility (T_{irrev}) decreases with the increase of the applied field, and for $H \geqslant 0.1$ T we failed to observe any hysteresis down to 4.5 K (see figure 2). There is initially a slight suppression of T_{peak} with the increase in H; T_{peak} went down to 10.5 K for $H = 0.5$ T. But quite dramatic changes start occurring for $H \geqslant 1.5$ T. An extreme example is shown in figure 3 where for $H = 5$ T no peak is observed down to 4.5 K. To elucidate this matter further we have measured M versus H at temperatures below T_{peak}. Figure 4 shows that at $T = 5$ K, a distinct change in the character of the M–H curve takes place at $H = 2$ T indicating the onset of a

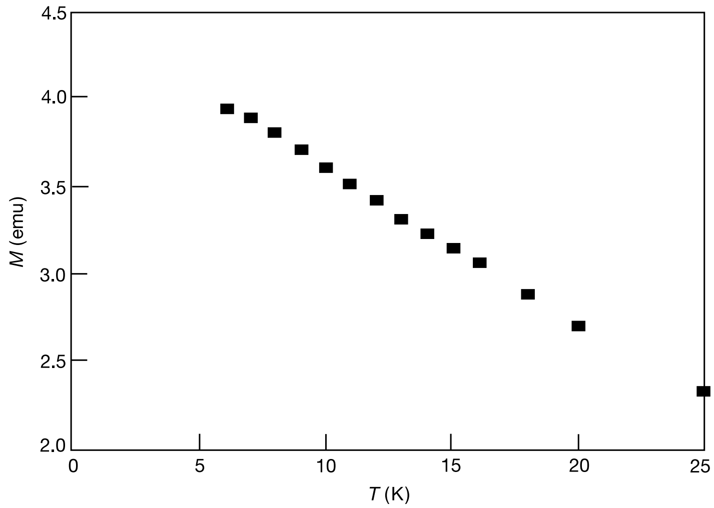

Figure 3. A plot of magnetization versus temperature for $DyCu_2Si_2$ with an external field being kept constant at 5 T. ■ stands for ZFC and □ for FC measurements.

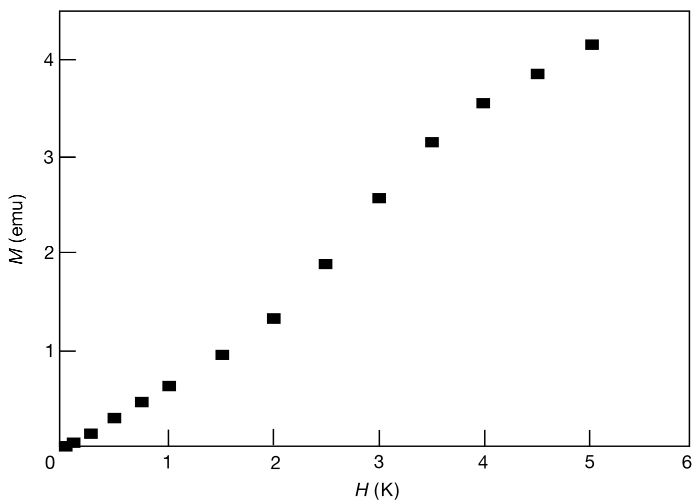

Figure 4. A plot of magnetization versus field for $DyCu_2Si_2$ at $T = 5$ K.

metamagnetic transition; this is similar to an earlier report [9]. A distinct non-linearity is observed above the metamagnetic transition field (H_M) but there is no marked hysteresis as a function of H. With the increase in temperature, H_M is decreased and the non-linearity is observed over a broader field regime (figure 5). As one enters the paramagnetic regime, the non-linearity seems to encompass the entire field regime [7].

The main finding of interest here is that the plot of M versus T at low fields mimics the behaviour normally encountered among spin-glass systems, although it is clear that there is no obvious source of frustration in this system, which orders antiferromagnetically [6]. The question now arises of what can give rise to such a behaviour. In $DyCu_2Si_2$ the magnetic ordering is described as ferromagnetic [101] planes of Dy^{3+} ions coupled antiferromagnetically with the sequence $+-+-$. It is quite possible for the presence of

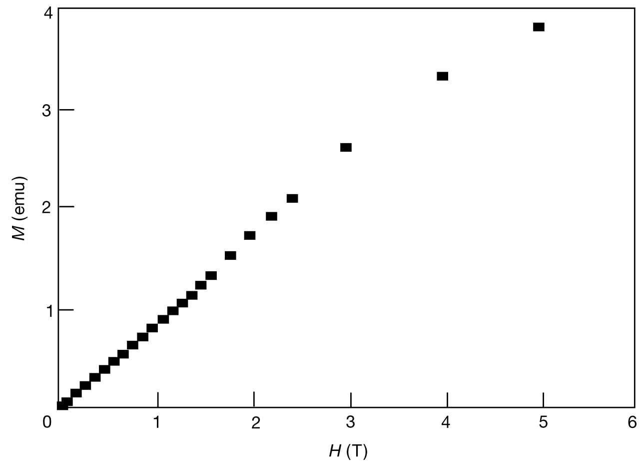

Figure 5. A plot of magnetization versus field for $DyCu_2Si_2$ at $T = 8$ K.

stacking faults to introduce a small degree of disorder in such a sequence; this would give rise to spin-glass-like behaviour. Another interesting consequence of there being stacking faults, particularly in the presence of anisotropic character (related to the $ThCr_2Si_2$ structure of $DyCu_2Si_2$), may arise in the following manner. Huse and Henley [10] argued that randomly placed impurities or defects that alter the local exchange couplings, but do not generate random fields or destroy the long-range order, can roughen the domain walls in Ising systems for a dimensionality in the range $5/3 < d < 5$. These defects pin the domain walls in energetically favourable positions and drastically slow down the kinetics of ordering. On the other hand anisotropic exchange interactions of Dzyaloshinski–Moriya (DM) type can also play an important role. It is believed in some quarters that the presence of such DM anisotropy is essential for the occurrence of macroscopic irreversibility in metallic spin glasses [11] and re-entrant spin glasses [12]. Also weak ferromagnetism (or canted antiferromagnetism) can arise from a combination of RKKY and DM interactions in certain crystal symmetries—for example, 2-Fe_2O_3, MnSi and $CrFe_3$ [13]. The observed metamagnetic behaviour of $DyCu_2Si_2$ provides support in this direction. However, whether the $ThCr_2Si_2$ structure satisfies the relevant conditions is not known to the present authors. The occurrence of weak ferromagnetic behaviour and thermomagnetic irreversibility (similar to what we have described above) has been reported recently for another planar compound: Y_2CuO_4 [14] with antiferromagnetic ordering.

There also remains the question of the role of the 4f–quadrupolar coupling effect in the observed magnetic properties. However, at this stage it is not clear whether this thermomagnetic irreversibility can be associated with the locking in of the magnetization directions due to the interactions [15] between the magnetization, the 4f charge cloud and the applied magnetic field.

In conclusion we can say that our study on $DyCu_2Si_2$ presents a good example of where a random orientation of crystallites, with the right combination of inherent anisotropy and intrinsic disorder (e.g. stacking faults), show features which are traditionally associated with spin glasses, while retaining distinct long-range antiferromagnetic ordering. These results highlight the difficulties encountered in trying to identify a few specific macroscopic magnetic characteristics which can be used to distinguish a spin glass unequivocally from an antiferromagnet.

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