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# Magnetic properties of $GdMn_2$ from μSR

Eve M. Martin[a], E. Schreier[a], G.M. Kalvius[a,*], A. Kratzer[a], O. Hartmann[b], R. Wäppling[b], D.R. Noakes[c], K. Krop[d], R. Ballou[e], J. Deportes[e]

[a]Physics Department, TU Munich, James-Franck, Strasse D-85747 Garching, Germany
[b]Physics Department, University of Uppsala, S-75121 Uppsala, Sweden
[c]Physics Department, Virginia State University, Petersburg, VA 23806, USA
[d]University of Mining and Metallurgy, PL-30059 Krakow, Poland
[e]Laboratoire Louis Néel, CNRS, F-38042 Grenoble, France

**Abstract**

$GdMn_2$ has a Néel transition around 100 K and a second magnetic transition near 40 K, which is considered a Curie point. The μSR data show that both the Gd and the Mn sublattice order at $T_N$, in contrast to a published model. Using the signal from muons stopped in a diamagnetic surrounding (high-purity silver) it was found that a ferromagnetic component exists already below $T_N$. At 40 K only a spin reorientation takes place. The magnetic Gd sublattice relieves some of the geometrical frustration of the Mn sublattice, but one still observes the presence of a dynamic short-range correlated fraction above $T_N$, similar to findings made previously in $YMn_2$. High-pressure studies gave a change of Néel temperature $dT_N/dp \approx -5$ K/kbar, which is nearly an order of magnitude smaller than in $YMn_2$. The temperature dependence of spin fluctuations just above $T_N$ follows a critical law with little change on volume reduction. Pressure influences the spatial arrangement of ordered spins slightly as revealed by changes in the ferromagnetic response. 

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$YMn_2$ is a prime example of an infinitely frustrated antiferromagnet among the $RMn_2$ type (R = rare earth) cubic Laves phase compounds. Its Néel transition is of first order and connected to distortion of cubic symmetry as well as to a substantial change in unit cell volume, which results in a pronounced thermal hysteresis. It was shown in previous μSR studies [1] that roughly 25 K above $T_N$ the compound exhibits magnetic short-range order in part of its volume as a consequence of frustration.

The other $RMn_2$ compounds differ from $YMn_2$ by the presence of a second magnetic sublattice, which is not frustrated. They also possess a large rare-earth single-ion anisotropy and are quite sensitive to crystalline electric field (CEF) effects. $GdMn_2$ plays a special role within the $RMn_2$ intermetallics since $Gd^{3+}$ as an S-state ion is devoid of single-ion anisotropy and does not feel the CEF. Yet, its magnetic properties are not well established in detail. A first-order transition takes place at

* Corresponding author. Tel.: + 49-89-2891-2501; fax: + 49-89-320-6780.
*E-mail address:* kalvius@ph.tum.de (G.M. Kalvius).

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$T_N \approx 100\,K$ but the volume change is small and a tetragonal distortion absent. Specific heat sees a broad anomaly around 40 K and magnetization data show ferromagnetic behavior below 40 K. For this reason the transition is often referred to as the Curie point $T_C$. Unfortunately, the high neutron absorption of Gd renders neutron diffraction measurements very difficult and little direct information on ordered spin structures is available at present. Whether both the Gd and Mn sublattices order simultaneously has been a matter of debate.

The measurements were carried out at the μE1/4 beamlines of PSI (decay muons, $\sim 80$ MeV/$c$) and the M13 beamline of TRIUMF (surface muons, $\sim 30$ MeV/$c$). In both cases powder samples were used. No fundamental difference was seen between the two data sets. The high-pressure measurements used the apparatus at PSI described elsewhere [2].

Fig. 1 shows the μSR signal amplitude as a function of temperature, clearly establishing the Néel transition. As in bulk magnetic measurements [3], no hysteresis is observed. The muon spin relaxation rate close to $T_N$ is an order of magnitude larger ($\sim 5\ \mu s^{-1}$) than in $YMn_2$. This establishes definitely that both sublattices order at $T_N$. In addition, the whole sample volume participates in the magnetic transition. That excludes a model [4] claiming that two spatially separate phases exist, one showing antiferromagnetic (AFM) order of Gd and Mn ions at $T_N$, the other remaining paramagnetic down to $T_C$ where only the Gd ions order ferromagnetically.

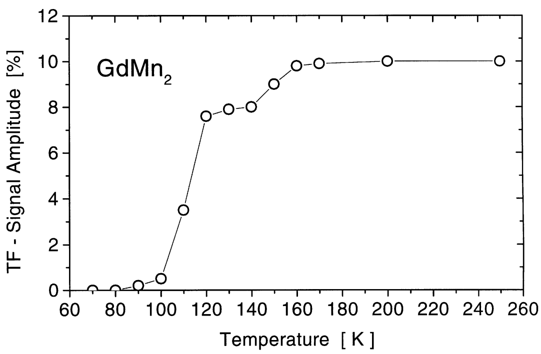


Fig. 1. TF sample-signal amplitude as a function of temperature. The signal originating from muons stopped in the sample holder is subtracted. The line is a guide to the eye. The appearance of three magnetic states can be distinguished. At high temperatures all of the sample signal comes from the paramagnetic state. The first slight reduction in amplitude is due to the development of a short-range ordered fraction. When passing $T_N$ the sample signal is completely lost.

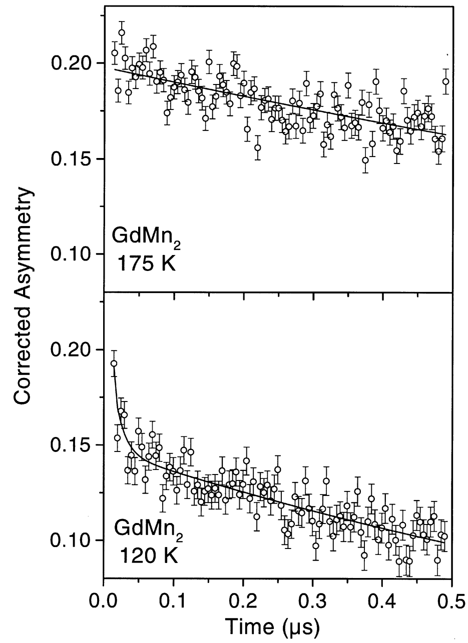


Fig. 2. ZF spectra taken well above and close to the Néel point showing the appearance of the rapidly decaying signal portion near $T_N$, which is ascribed to a short-range ordered portion.

Just above the onset of strong amplitude reduction a small loss of the TF signal strength is seen. A similar observation had been made in $YMn_2$, which was connected to the appearance of a rapidly depolarizing signal. This was interpreted as the formation of short-range order in part of the material. In $GdMn_2$ the presence of a rapidly depolarizing signal was also visible in zero-field (ZF) spectra. An example is shown in Fig. 2 that depicts the initial portion of two ZF-μSR spectra, one taken well above, the other just above $T_N$. The fast depolarization rate is $\sim 50\ \mu s^{-1}$ and thus about one order of magnitude faster than in $YMn_2$ or $Y_{0.9}Tb_{0.1}Mn_2$ [1]. It is conceivable that the sublattice of paramagnetic $Gd^{3+}$ ions speeds up depolarization. The application of a longitudinal field of 1 kG had no influence on spectral shape, meaning that we deal with a highly dynamic spin-glass-like state. The presence of a second magnetic sublattice (Gd) probably relieves some (no hysteresis) but not all (spin-glass-like state still present) of the frustration of the Mn tetrahedral AFM sublattice.

By recording the signal from muons implanted in a diamagnetic material (Ag) surrounding the sample one can detect the presence of ferromagnetic (FM) spontaneous magnetization. The results of this type of measurement establish the presence of an FM component already below $T_N$. It becomes, however, more pronounced below $T_C$. This makes it evident that the spin structure changes at 40 K and that the so-called Curie point is rather a spin reorientation transition. A similar conclusion can be drawn from the Mössbauer spectroscopy [5].

High-pressure measurements were also carried out. Fig. 3 shows high-pressure data analogous to Fig. 1. One observes a downshift of $T_N$ with a pressure coefficient of $\sim -5$ K/kbar. This is nearly an order of magnitude smaller than the coefficient in $YMn_2$ [6,7]. The presence of the large moment Gd magnetic sublattice reduces the effect of the Mn moment destabilization.

The paramagnetic relaxation just above $T_N$ follows a critical power law (Fig. 4):

$$\lambda \propto [(T - T_N)/T_N]^{-w}. \quad (1)$$

Basically, such a behavior is expected on approaching a second-order phase transition. For $GdMn_2$ (and also for $YMn_2$, see Ref. [1]) this means that the system tends towards a normal second-order transition, but the frustration in the spin lattice prevents this type of transition. The system rather performs a sudden switch to a first-order transition where the lattice expansion (and distortion) reduces frustration. The critical exponent $w$ of Eq. (1) exhibits a small increase under applied pressure that is at the limit of data accuracy. The value at ambient pressure ($w = 0.34$) is somewhat small, but even lower values have been seen. For example, in Er metal the exponent was only $w = 0.15$, which remains unexplained [8]. In the case of $GdMn_2$ the low value of $w$ may be caused by the switch-over to first order before the second-order transition can develop fully. If this is accepted, then the increase of $w$ would indicate that the second-order nature of the Néel transition becomes more pronounced under pressure.

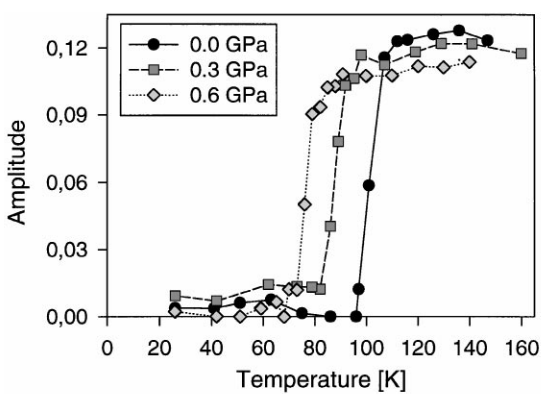


Fig. 3. Temperature dependence of the TF sample-signal amplitude of $GdMn_2$ at different pressures. The sudden drop of amplitude constitutes the magnetic transition (see also Fig. 1), which is clearly pressure dependent. The lines are guides to the eye.

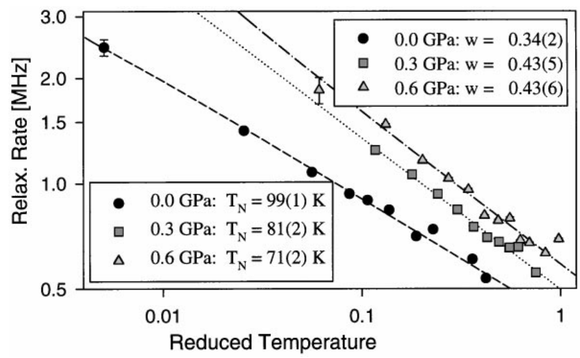


Fig. 4. Muon spin relaxation rate in the paramagnetic regime as a function of reduced temperature $(T - T_N)/T_N$ at different pressures (1 GPa = 10 kbar). The lines are fits to a critical power law as discussed in the text. The fit parameters are given in the inset.

Finally we mention, without going into details, that the remanence signal from the sample surroundings also changes under pressure. In these measurements the background signal is the μSR response from muons stopped in the wall of the CuBe high-pressure cell. The result means that the spin structure is sensitive to volume reduction. A slight positive shift of $T_C$ with pressure is indicated.

The μSR data confirm that the presence of a second magnetic sublattice (Gd) relieves some of the magnetic frustration of the Mn sublattice, leading to a much more sharply defined Néel transition without hysteresis. Other effects of frustration are still visible, in particular, as in $YMn_2$, the formation of a short-range ordered state in part of the compound just above $T_N$. The data refute the model of Ref. [4] that claimed the existence of a mixture of ordered and paramagnetic states between $T_N$ and $T_C$. The presence of a ferromagnetic component is confirmed, but in contrast to other magnetic measurements it is shown to be present up to $T_N$ and, in contrast to neutron data [9], even at very low applied fields. The transition at 40 K, usually referred to as $T_C$, is rather a spin reorientation transition between two magnetic states, both having a ferromagnetic component. The simplest model would be canted antiferromagnetic spin structures. Applied pressures up to 0.6 MPa cause a downshift of $T_N$ nearly one order of magnitude smaller than observed in $YMn_2$, meaning that the presence of strong paramagnetic ions ($Gd^{3+}$) makes the Mn moment less sensitive to volume changes. The critical behavior of paramagnetic spin fluctuations indicates the presence of an underlying second-order transition at $T_N$. Critical spin dynamics is only slightly affected by pressure. As indicated by high-pressure remanence data, the long-range ordered spin structures are sensitive to volume changes.

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## References

[1] M. Weber et al., Hyperfine Interactions 85 (1994) 265.
[2] A. Kratzer et al., Hyperfine Interactions 87 (1994) 1055.
[3] H. Wada et al., J. Magn. Magn. Mater. 70 (1987) 134.
[4] M. Ibarra et al., J. Magn. Magn. Mater. 128 (1993) L249.
[5] J. Przewoznik et al., J. Magn. Magn. Mater. 119 (1993) 150.
[6] R. Hauser et al., J. Magn. Magn. Mater. 140–144 (1995) 134.
[7] G.M. Kalvius et al., Hyperfine Interactions, submitted.
[8] R. Wäppling et al., J. Magn. Magn. Mater. 119 (1993) 123.
[9] B. Ouladdiaf, Ph.D. Thesis, University of Grenoble, 1986, unpublished.