

PRESSURE DEPENDENT MAGNETIZATION STUDIES  
OF PYROCHLORE  $(\text{Gd}_{1-x}\text{Dy}_x)_2\text{Mo}_2\text{O}_7$   
( $x = 0, 0.1, 0.2$  AND  $0.4$ )\*

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We have measured temperature dependent magnetization of pyrochlore molybdates  $(\text{Gd}_{1-x}\text{Dy}_x)_2\text{Mo}_2\text{O}_7$  ( $x=0, 0.1, 0.2, 0.4$ ) under pressures up to 9.5 kbar. With increasing pressures, the magnetization of all four samples decreases substantially and the ferromagnetic transition seen in  $\text{Gd}_2\text{Mo}_2\text{O}_7$  is suppressed and not found above 9 kbar; which is very similar to the effects of Dy substitutions. Interestingly enough, we observed a clear difference in magnetization measured after zero field cooling (ZFC) and field cooling (FC). From these results we have confirmed that the lattice constant of  $\text{R}_2\text{Mo}_2\text{O}_7$  is crucial in determining the magnetic state and demonstrated that the ferromagnetic ordering coexists with metastable behavior of a spin glass type.

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## 1. Introduction

Pyrochlore compounds  $\text{R}_2\text{M}_2\text{O}_7$  (R=rare earth, M=transition metal) have both R and M ions sitting at the vertices of corner-sharing tetrahedra so antiferromagnetic interaction between nearest neighbors gives rise to so-called geometrical frustration effects in the 3D lattice. Interestingly, the transport and magnetic properties of pyrochlore molybdates  $\text{R}_2\text{Mo}_2\text{O}_7$  are found to be very sensitive to the ionic size of the rare earth element. For example, ferromagnetic(FM) metallic behavior is observed in compounds with

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larger rare earth elements such as Nd, Sm and Gd while spin-glass(SG) behavior with high resistivity values is observed for smaller rare earth elements such as Tb, Dy and Y [1].

Recent studies near the magnetic phase boundary of  $R_2Mo_2O_7$  showed that the magnetic transition is accompanied by a metal-insulator transition and the FM state coexists with the SG state near the phase boundary [2]. Through structural analysis of the compounds, O-Mo-O bond angle is found to increase with increasing the ionic radius of rare earth elements too [3]. This then pointed to possible importance of the systematic studies of magnetization with controlling lattice parameters by external pressures. In this work, we have measured ac susceptibility at ambient pressure and dc magnetization of pyrochlore compounds  $(Gd_{1-x}Dy_x)_2Mo_2O_7$  under pressures up to 9.5 kbar. We discuss our data measured as varying pressures with respect to the effect of Dy substitutions.

## 2. Experimental details

Samples studied in this work are single phase polycrystals of  $(Gd_{1-x}Dy_x)_2Mo_2O_7$  ( $x=0, 0.1, 0.2$  and  $0.4$ ). Details of sample preparation and characterizations were previously reported in Ref [2]. DC magnetization measurements were made by using a commercial SQUID magnetometer (MPMS-7, Quantum Design). Samples were cooled to 5 K in zero magnetic field and the magnetization was measured at 100 Oe while warming the sample up to 120 K for zero-field-cooling (ZFC) measurements and then subsequently cooling down to 5 K in the presence of the magnetic field for field-cooling (FC) measurements. To generate hydrostatic pressures up to 9.5 kbar, we used a Be-Cu pressure cell and the 1:1 mixture of FC70 and FC77 as a pressure transmitting medium [4]. We also measured ac susceptibility at ambient pressure with an ac driving field of 10 Oe in the frequency range from 100 Hz to 10 KHz using a commercial ac susceptometer (PPMS-9, Quantum Design).

## 3. Results and discussion

The dc magnetization of  $(Gd_{1-x}Dy_x)_2Mo_2O_7$  at ambient pressure shows a systematic decrease of ferromagnetic signals with Dy substitutions (Fig. 1(a)). As Dy with a smaller ionic size replaces Gd with a larger ionic size, magnetization decreases gradually and the FM transition seen at 65 K in the  $x = 0$  sample becomes reduced and eventually the FM transition is found to disappear at the  $x=0.4$  sample. For example, if we take the magnetization value at an irreversibility point where the ZFC and FC data meet, it decreases from 4609 emu/mol for  $x = 0$  to 364 emu/mol for  $x = 0.4$ . We also note that in all samples there is a clear difference in the magnetization taken after ZFC and FC.

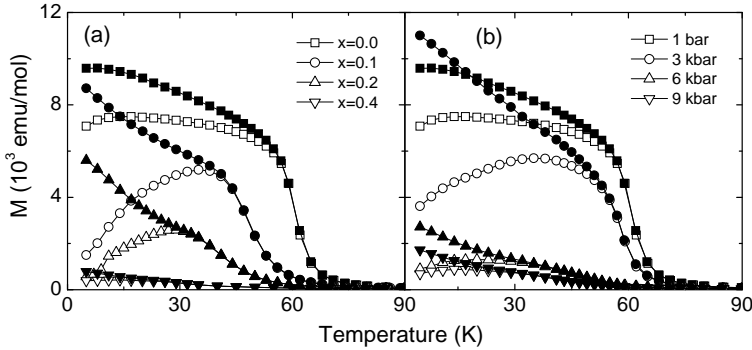


Fig. 1. ZFC (open symbols) and FC (closed symbols) dc magnetization of (a)  $(\text{Gd}_{1-x}\text{Dy}_x)_2\text{Mo}_2\text{O}_7$  ( $x = 0, 0.1, 0.2$  and  $0.4$ ) measured at 100 Oe and ambient pressure, and (b)  $\text{Gd}_2\text{Mo}_2\text{O}_7$  measured at 100 Oe and under different pressures up to 9 kbar.

By applying hydrostatic pressures, we can see very similar behavior with those produced by Dy substitutions (see Fig. 1(b)). Magnetization decreases drastically with increasing pressures, and the FM transition seen at ambient pressure gets suppressed and eventually almost disappears at 9 kbar. The Curie temperature also decreases considerably with pressures. The irreversibility point ( $T_{\text{irr}}$ ) at different pressures decreases from 59 to 35 K with increasing pressures from 1 bar to 9 kbar while the magnetization at  $T_{\text{irr}}$  decreases from 4609 to 632 emu/mol. It is also interesting to note that the pressure effect on the magnetization is more pronounced at relatively lower pressures up to 6 kbar.

Irreversible behavior in dc magnetization is generally believed to originate from some kind of a metastable nature like spin glass. From the ac susceptibility results, we have found that  $\chi'_{\text{ac}}$  and  $\chi''_{\text{ac}}$  exhibit frequency dependence for all our samples. This frequency dependence becomes clearer in the  $x = 0.4$  sample, where the temperature shift of a cusp in  $\chi'_{\text{ac}}$  ( $\Delta T_{\text{cusp}}$ ) is about 6 K with varying frequency from 100 Hz to 10 KHz (see Fig. 2). This indicates that the low temperature state of  $(\text{Gd}_{0.6}\text{Dy}_{0.4})_2\text{Mo}_2\text{O}_7$  has a spin glass character.

Similarity between Dy substitutions and pressures in the magnetization of  $\text{Gd}_2\text{Mo}_2\text{O}_7$  clearly suggests that lattice constant of  $\text{R}_2\text{Mo}_2\text{O}_7$  plays an important role in determining the magnetic state of pyrochlore molybdates. With decreasing lattice constant by substitutions or pressures, the FM state becomes unstable and the magnetic state transforms into a metastable state of SG type, which is also consistent with the previous studies of the same samples [2]. It should also be noted that FM correlation seems to coexist even with the SG phase. The Curie–Weiss temperature,  $\Theta_{\text{CW}}$ , deduced from

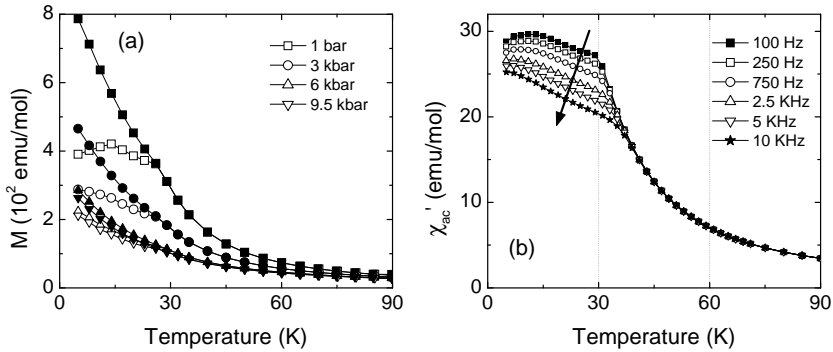


Fig. 2. (a) ZFC (open symbols) and FC (closed symbols) dc magnetization of  $(\text{Gd}_{0.6}\text{Dy}_{0.4})_2\text{Mo}_2\text{O}_7$  measured at 100 Oe and under different pressures up to 9.5 kbar. (b) Real part of the ambient pressure ac susceptibility of  $(\text{Gd}_{0.6}\text{Dy}_{0.4})_2\text{Mo}_2\text{O}_7$  measured with ac driving field of 10 Oe and frequencies from 100 Hz to 10 KHz. Arrow indicates the direction of increasing frequency.

the magnetization measurement up to 350 K is 71, 59, 55 and 31 K for  $x=0, 0.1, 0.2$  and  $0.4$ , respectively. Since the FM transition is due to Mo moments ordering as shown by the neutron scattering experiment of  $\text{Nd}_2\text{Mo}_2\text{O}_7$  [5], we propose that the possible origin of the SG behavior lies in the antiferromagnetic coupling between the rare earth moments and Mo moments and resultant magnetic frustration on the pyrochlore lattice. Saturated moment obtained from the magnetization measurement of  $\text{Gd}_2\text{Mo}_2\text{O}_7$  at 2.3 K (not shown here) also shows that with decreasing lattice constant by pressures AFM coupling between  $\text{Gd}^{3+}$  and  $\text{Mo}^{4+}$  ions develops gradually and consequently the magnetic properties become more metastable as expected.

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