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AC susceptibility of a novel 2D rare-earth boron-cluster spin glass system

T Mori[1,2,3] **and H Mamiya**[1]

[1] National Institute for Materials Science, Namiki 1-1, Tsukuba, 305-0044, Japan
[2] PRESTO, Japan Science and Technology Agency, 4-1-8 Honcho, Kaweguchi, Saitema, Japan

E-mail: MORI.Takao@nims.go.jp

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Abstract

The AC susceptibility of a novel rare-earth boron-cluster-containing spin glass was measured. $REB_{17}CN$, $REB_{22}C_2N$, and $REB_{28.5}C_4$ (RE = Er, Ho) are a newly discovered class of crystalline, non-doped spin glass system in which the rare-earth atoms have a two-dimensional configuration. Well defined maxima in the in-phase linear AC susceptibility of $HoB_{22}C_2N$ were observed with corresponding behaviour of the out-of-phase susceptibility as expected for a spin glass. It was found that the dynamical scaling theory of a 3D system, $\tau/\tau_0 = [(T - T_0)/T]^{-z\nu}$, could not describe the data well. More reasonable parameters could be extracted in terms of a generalized Arrhenius law $\ln(\tau_{max}/\tau_0) \propto T_f^{-2}$ which describes 2D spin glass systems. This result is in agreement with the previous studies which indicate the two-dimensionality of this system.

1. Introduction

Spin glasses have been extensively researched throughout the years as an exciting field of magnetism [1–3] with recent intriguing discoveries made in a myriad of new systems [4, 5]. Magnetic properties of rare earth B_{12} icosahedral cluster-containing compounds have also been the focus of growing interest because a number of magnetic transitions have been discovered at moderate temperatures despite their being insulating, magnetically dilute, f-electron systems (see [6] for example). A particularly interesting aspect of these transitions is that the B_{12} icosahedral boron clusters are indicated to mediate the magnetic interaction, which is a novel phenomenon.

For boron systems it has also been reported that such compounds as doped CaB_6 [7] and CaB_2C_2 [8] exhibit high temperature ferromagnetism. However, it has been demonstrated that the observed ferromagnetic properties are indicated to be of non-intrinsic origin [9], with the

[3] Author to whom any correspondence should be addressed.

magnetism in the CaB_6 system indicated to be due to Fe impurities electrochemically plated onto the crystal surface during the Al flux removal procedure.

In a recent development, a homologous series of non-doped crystalline B_{12} compounds, $REB_{17}CN$, $REB_{22}C_2N$, and $REB_{28.5}C_4$ (REB = Er, Ho), was found to exhibit spin glass behaviour [10–12].

AC susceptibility measurements were carried out on $HoB_{22}C_2N$ to investigate dynamical properties of this new spin glass system.

$HoB_{22}C_2N$ is rhombohedral (space group $R\bar{3}m$) with lattice constants of $a = 5.614$ Å, $c = 44.625$ Å. The structure is shown in figure 1. The compound has a layered structure along the c-axis with holmium and B_6 octahedral layers separated by three B_{12} icosahedral and C–B–C chain layers. Looking at this series of homologous compounds, the number of B_{12} and C–B–C layers separating the rare-earth layers increases successively from two for the $REB_{17}CN$ compound to four for the $REB_{28.5}C_4$ compound. The configuration of the rare-earth atoms in respect to each other (figure 1(b)) is the same for all the homologous compounds. Two flat triangular rare-earth atom layers are closely stacked on top one another in AB configuration. From the previous studies on extensive measurements using a DC SQUID magnetometer [10] and comparison between the homologous compounds [12] it has been indicated that the magnetic interaction is dominant within the two-dimensional triangular layer. This is a reasonable result since we can see from the structure depicted in figure 1 that the B_{12} icosahedra (which have been indicated to mediate the interaction) are adjacent to the planes of the triangular layers, while there are no B_{12} to mediate between the layers. The separation of rare-earth atoms within the triangular layers is 5.62 Å.

2. Experimental details

$HoB_{22}C_2N$ was synthesized as described previously by a two-step sintering process at 1700 °C using BN crucibles [10]. The sample was characterized by high resolution powder x-ray diffractometry and chemical analysis. AC susceptibility measurements were made using a commercial Quantum Design SQUID magnetometer in which the residual field was reduced below 1.0 Oe by resetting the magnet. The amplitude of the applied AC field was 3.0 Oe while frequencies were varied over four decades of time (8×10^{-2} to 8×10^{2} Hz).

3. Results and discussion

The real part χ'_{ac} and imaginary part χ''_{ac} of the AC susceptibility of $HoB_{22}C_2N$ are plotted in figures 2(a) and (b), respectively. A well defined peak is observed in χ'_{ac} corresponding to the maximum slope of χ''_{ac} as expected for a spin glass. The cusp temperature is defined as T_f.

We first attempt to analyse our results in terms of the dynamical slowing down of the spin fluctuations above the glass transition temperature T_0 ($\neq 0$) for a three-dimensional system [2, 13]. The well known result of dynamical scaling gives the relation $\tau \sim \xi^z$, where τ is the measured relaxation time, ξ the correlation length, and z the dynamic exponent [2, 13]. The correlation length ξ diverges as $\xi \sim [T/(T - T_0)]^\nu$ with ν the critical exponent. From this, the relation

$$\tau/\tau_0 = [(T - T_0)/T]^{-z\nu} \tag{1}$$

can be obtained, where τ_0 is the shortest relaxation time of the system. Expressing this in a form for analysis of the experimental results it can be written as

$$T_f = T_0[1 + (\tau_0 f)^{1/z\nu}]. \tag{2}$$

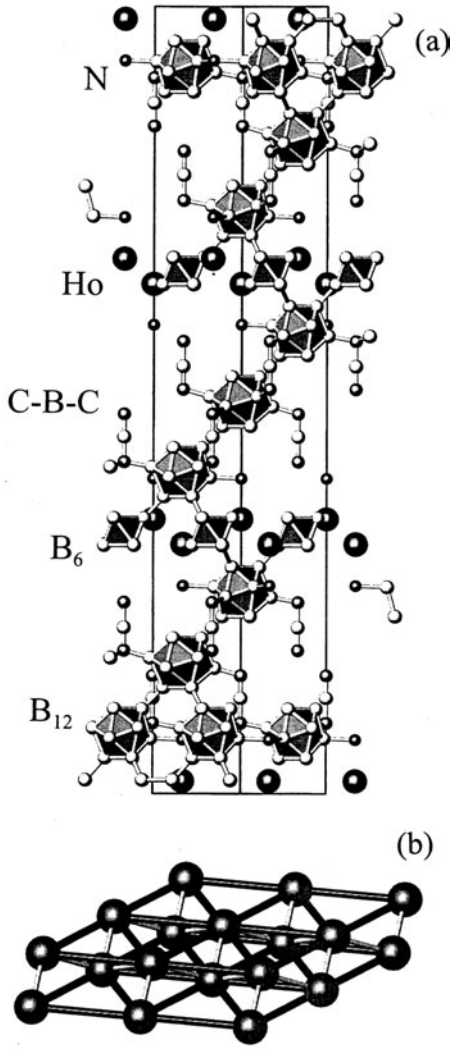

Figure 1. Crystal structure of $HoB_{22}C_2N$: (a) a view perpendicular to the *c*-axis and (b) a view of the configuration of the holmium atoms only. Small white circles indicate boron atoms, with the large polyhedra being B_{12} icosahedra, and smaller polyhedra B_6 octahedra. Small dark and grey circles indicate nitrogen and carbon atoms, respectively, with the three bonded atoms along [0 0 1] being C–B–C chains. The large dark circles indicate holmium atoms. The rare-earth atoms form pairs of flat triangular layers stacked closely on top of one another in AB configuration.

Since the least squares fitting to our data in this case is largely dependent on the starting values assigned to the three parameters and therefore not completely satisfactory, we assume a $T_0 = 22.5$ K assigned from the DC measurements [10]. Then we obtain a fit with $\tau_0 = 2.5 \times 10^{-4}$ s, $z\nu = 8.3$ as depicted in the curve in figure 3(a). The value of $z\nu$ agrees with the range of $z\nu \sim 7.9 \pm 1$ determined by the simulations of Ogielski [14] for 3D spin glasses with short range magnetic interactions. However, the obtained value of $\tau_0 = 2.5 \times 10^{-4}$ s as the shortest relaxation time seems to be too long to be reasonable, since values typically in the range of 10^{-11}–10^{-13} s have been obtained for canonical metallic spin glasses such as

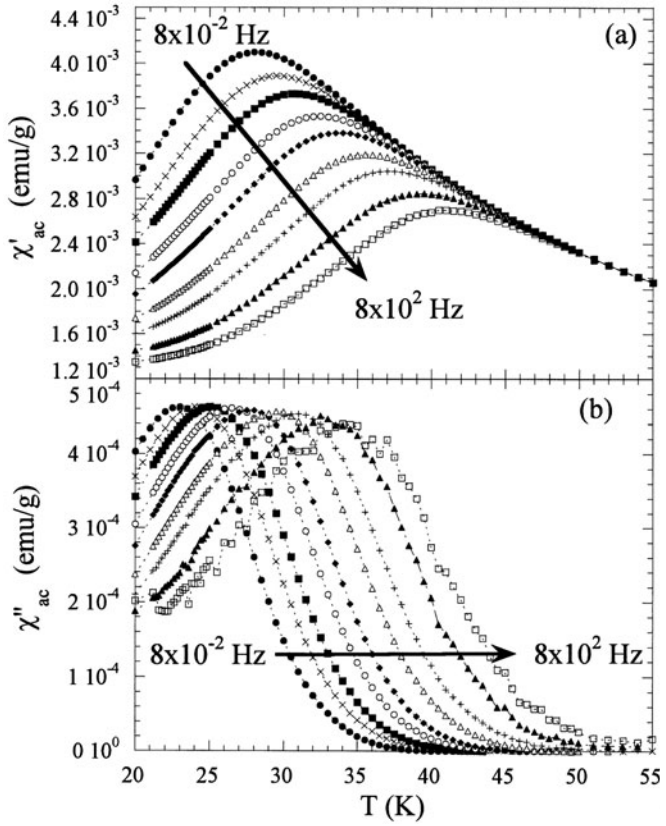

Figure 2. Temperature dependence of the (a) real part χ'_{ac} and (b) imaginary part χ''_{ac} of the ac magnetic susceptibility of $HoB_{22}C_2N$. Measuring frequencies are 8×10^{-2} Hz (closed circles), 3×10^{-1} Hz (×), 8×10^{-1} Hz (closed squares), 3 Hz (open circles), 8 Hz (closed diamonds), 3×10^{1} Hz (open triangles), 8×10^{1} Hz (crosses), 3×10^{2} Hz (closed triangles), and 8×10^{2} Hz (open squares).

CuMn [2]. Even for insulating spin glasses, the largest values of τ_0 have been $\sim 2 \times 10^{-7}$ s for $Eu_{0.6}Sr_{0.4}S$ [15].

It has been pointed out by Bontemps *et al* [15] that simply using the maximum of χ'_{ac} is not adequate to properly analyse the critical dynamics of a spin glass system. χ'_{ac} has a dependence on the isothermal susceptibility χ_0 and τ which both depend on T. They set a criterion

$$\tan\phi = \chi''/\chi' = \omega\tau, \tag{3}$$

where $\tan\phi \sim \phi$ is set to a constant small value. From this condition a set of temperatures $T_i(f)$ is defined for a specific response time τ. $T_i(f)$ is plotted in figure 3(b). A fit to equation (2) yields, as a best fit, parameters of $T_0 = 24.4$ K, $\tau_0 = 2.0 \times 10^{-3}$ s and $z\nu = 9.3$. As a result of this analysis we obtain even larger values of τ_0 which is unsatisfactory.

Sandlund *et al* [16] have previously systematically investigated Cu–Mn films and found a dimensional crossover of the dynamical behaviour. The dynamical behaviour of two-dimensional films was successfully analysed in terms of a generalized Arrhenius law

$$\log(\tau_{max}/\tau_0) \propto T^{-(1+\psi\nu)}, \tag{4}$$

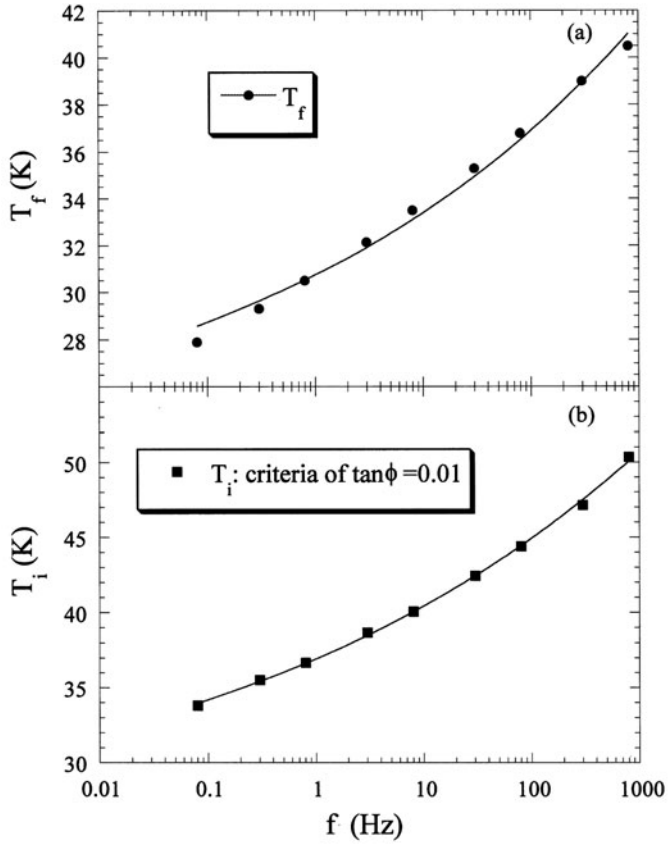

Figure 3. Frequency dependence of (a) cusp temperatures T_f of χ'_{ac} (closed circles) and (b) T_i which is determined from the criteria in equation (3), $\tan\phi \sim \phi = 10^{-2}$ (closed squares). Curves indicate the best fit to equation (2) while parameters are given in the text.

with $1 + \psi\nu = 2.6$ [16]. The generalized Arrhenius dependence is derived from the droplet scaling theory of Fisher and Huse [17] and Monte Carlo simulations on 2D systems have given values of $1 + \psi\nu \sim 2$ [18, 19]. As we noted above, analysis of our data in terms of the dynamical scaling of a three-dimensional system has not been successful. Investigation of the homologous series of $REB_{17}CN$, $REB_{22}C_2N$, $REB_{28.5}C_4$ has yielded basically the same spin glass behaviour, indicating that the configuration of the 2D triangular rare-earth layers dictates the physics of this system [12]. The number of data points is not particularly large versus the number of free parameters in equation (4), and in such cases setting one of the parameters to a fixed reasonable value is a method typically used. Since a wide range of values for τ_0 have been observed for insulating systems [2, 15, 20], we fit our data with a set value of exponent, using the theoretical form for 2D systems of $\log(\tau_{max}/\tau_0) \propto T^{-2}$. As a result, we are able to obtain a good fit with $\tau_0 = 5 \times 10^{-7}$ s as is indicated by the bold line in figure 4. Thus, using the generalized Arrhenius form for two-dimensional systems, we are satisfactorily able to obtain a shorter τ_0 which is close to that determined for the $Eu_{0.6}Sr_{0.4}S$ system, for example [15]. This result is consistent with the previous conclusions which indicated the two-dimensionality of this rare-earth boron-cluster system.

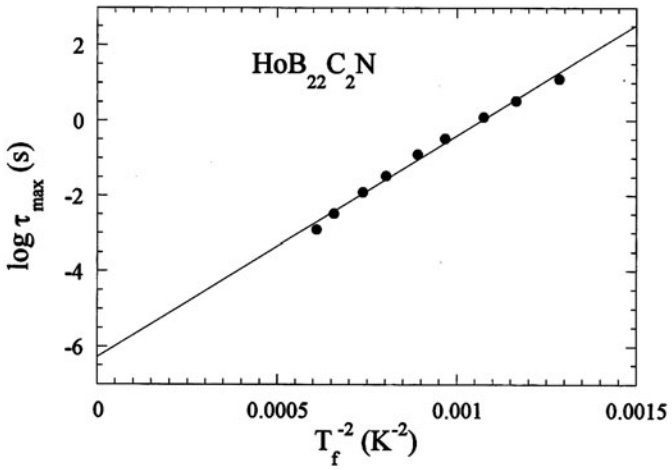

Figure 4. $\log(\tau_{\max})$ plotted versus T_{f}^{-2}. The bold line in (b) represents a fit to $\log(\tau_{\max}/\tau_0) \propto T^{-2}$ with $\tau_0 = 5 \times 10^{-7}$ s.

4. Conclusions

AC susceptibility of the new spin glass system $HoB_{22}C_2N$ was investigated. $HoB_{22}C_2N$ is taken to be representative of the new class of crystalline non-doped rare-earth boride spin glasses $REB_{17}CN$, $REB_{22}C_2N$, and $REB_{28.5}C_4$, where it has been indicated that magnetic interaction is dominant in the two-dimensional rare-earth layers.

It was found that the dynamical scaling theory for three-dimensional systems did not give satisfactory descriptions for our results. However, applying the generalized Arrhenius law of a two-dimensional spin glass system to the data yielded more reasonable parameters, which is consistent with the previous results indicating the two-dimensionality of this system.

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