Proceedings of the European Conference "Physics of Magnetism '99", Poznań 1999

MANIFESTATION OF SPIN-GLASS-LIKE BEHAVIOR IN THE ORGANOMETALLIC MAGNET

V.P. Dyakonov[a,c], M. Arciszewska[a], W. Dobrowolski[a], A. Szewczyk[a], H. Szymczak[a], E. Aksymentieva[b], V. Varyukhin[c], E. Zubov[c] and M.U. Gutowska[a]

[a]Institute of Physics, Polish Academy of Sciences
Al. Lotników 32/46, 02-668 Warszawa, Poland
[b]Lviv State University, Lviv, Ukraine
[c]Donetsk Physico-Technical Institute, Ukraine

Ac susceptibility (χ_{ac}) and dc magnetization (M_{dc}) of the organometallic magnet $Na[FeO_6(C_{10}H_8N)_3]$ have been studied in temperature range of 4.2–200 K, in an external magnetic field up to 16 kOe. Some peculiarities of the χ_{ac} and M_{dc} behavior, which are characteristic of spin-glasses, e.g. cusps in the susceptibility and zero-field-cooled magnetization, an irreversibility between the field-cooled and zero-field-cooled magnetization and dependences of the T_{cusp} temperature on the frequency and the intensity of the magnetic field, were observed at low temperatures.

PACS numbers: 75.30.Kz, 75.40.Cx

1. Introduction

The problem of the macroscopic magnetism in organic materials is a rather complex but perspective scientific problem [1]. One way to solve this problem is to dope the organic matrix with magnetic transition metal ions (organometallic magnets). Some of organometallic-based magnets have the Curie temperature close to room temperature [2, 3].

Investigations of magnetic properties of new metal-organic materials should favor the understanding of nature of magnetism in organic magnets.

In this paper the magnetic behavior of new organometallic magnet $Na[FeO_6(C_{10}H_8N)_3]$ has been studied over a temperature range of 4.2–200 K and in an external magnetic field up to 16 kOe. Preliminary results of investigations of dc magnetization (M_{dc}) and ac susceptibility (χ_{ac}) show that this insulating organometallic compound exhibits features of M_{dc} and χ_{ac}, which are characteristic of a spin-glass system.

2. Experimental

The nitrozo-β-naphthol with iron (II) complex $Na[FeO_6(C_{10}H_8N)_3]$ was synthesized by a procedure described in Ref. [4]. A molecular structure of $Na[FeO_6(C_{10}H_8N)_3]$ compound studied is shown in Fig. 1. In this compound the Fe ion is surrounded by six oxygen atoms belonging to three molecules of organic ligand.

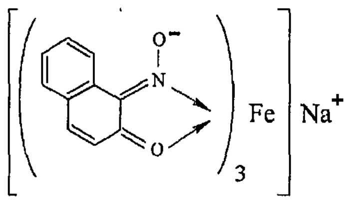

Fig. 1. Structure of the $Na[FeO_6(C_{10}H_8N)_3]$ complex.

According to data of the X-ray powder diffraction measured at room temperature (CuK_α-radiation) the crystal structure of the complex studied is monoclinic with the spatial group $P2_1$. The lattice parameters are: $a = 11.157$ Å; $b = 8.76$ Å; $c = 6.203$ Å. The monoclinic angle is $\beta = 92.4°$.

Magnetization, $M(T)$, measurements were performed using a VSM in the temperature range of 4.2–200 K. The zero-field-cooled (ZFC) and field-cooled (FC) magnetization was measured in magnetic fields from 0.1 up to 10 kOe. Real and imaginary parts of the ac susceptibility were measured using a mutual induction method with an excitation field $h_{ac} = 5.0$ Oe over the temperature range of 4.2–140 K. The frequency, f, was varied in the range 95 Hz$\leq f \leq$ 2000 Hz. The magnetic measurements were carried out on pressed powder pellets of cylindrical shape ($d = 3$ mm, $l = 6$ mm).

3. Results and discussion

Figure 2 presents the temperature dependences of the ac magnetic susceptibility, $\chi(T)$, measured at different magnetic field strengths $H = 0$, 0.5, and 5 kOe. The $\chi(T)$ curve measured at $H = 0$ shows a cusp at $T_f \approx 17$ K. As it is seen in Fig. 2, the value of susceptibility decreases sharply and the cusp in the $\chi(T)$ susceptibility becomes more rounded and disappears with increasing field strength.

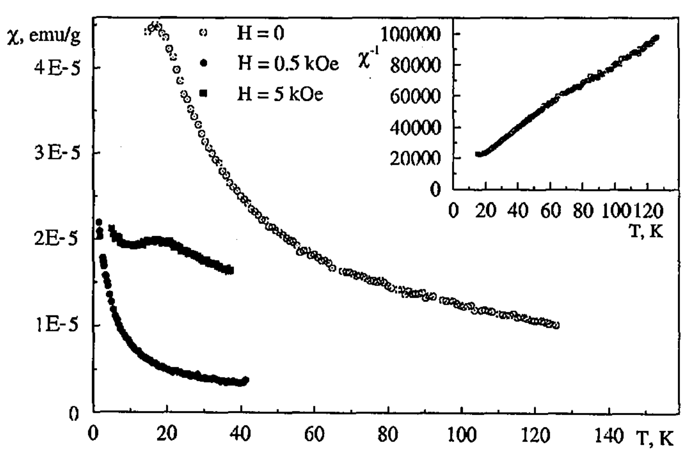

Fig. 2. Temperature dependence of the ac magnetic susceptibility at different magnetic field strengths: $H = 0$, 0.5, and 5 kOe. Inset: Temperature dependence of the inverse magnetic susceptibility for $H = 0$.

The inset to Fig. 2 shows the inverse susceptibility as a function of temperature. In the temperature range between 25 and 80 K the $\chi(T)$ dependence can be expressed by the Curie–Weiss law with a negative (antiferromagnetic) Weiss constant of $\theta_{\mathrm{AFM}} = -2.4$ K and the Curie constant $C = 0.46$ emu K/mol. It gives the effective moment m_{eff} equal to $1.9m_{\mathrm{B}}$ that is less than the magnetic moment of the Fe^{2+} ion. The latter is presumably due to the presence of superparamagetic particles in this complex.

The ac susceptibility as a function of temperature measured for different frequencies $f = 95 \div 2000$ Hz shows that a maximum of $\chi(T)$ dependence decreases and shifts to higher temperatures with increasing frequencies (Fig. 3). For analysis of $T_{\mathrm{cusp}}(f)$ we have used the theoretical dependence $T_f = T_{\mathrm{c}}[1 + (\tau_0 f)^{1/z\nu}]$ and the relative variation of T_f per decade of frequency $K = (\Delta T_f/T_f)/[\Delta \log(f)]$. Both the dynamic exponent ($z\nu = 4$) and K ($K = 0.09$) values are close to those reported for spin glasses [5].

These peculiarities of ac susceptibility versus the frequency and magnetic field are similar to those observed for ordinary spin-glass (SG) systems.

The cusp-like anomaly in the temperature dependence of M_{ZFC} of this system is also characteristic of SG systems. The FC magnetization increases with decreasing temperature showing no maximum. A large and clear irreversibility, seen in difference, between ZFC and FC magnetization is demonstrated in Fig. 4. Both cusp and irreversibility reflect a nonequilibrium character of thermodynamic behavior of the magnetic system.

The large difference between M_{FC} and M_{ZFC} below irreversibility temperature and the presence of frequency-dependent peak of $\chi(T)$ indicate the absence of magnetic long-range order in this compound up to 4.2 K. The magnetization of the $Na[FeO_6(C_{10}H_8N)_3]$ complex as a function of magnetic field does not reach its saturation value up to 16 kOe at 4.2 K.

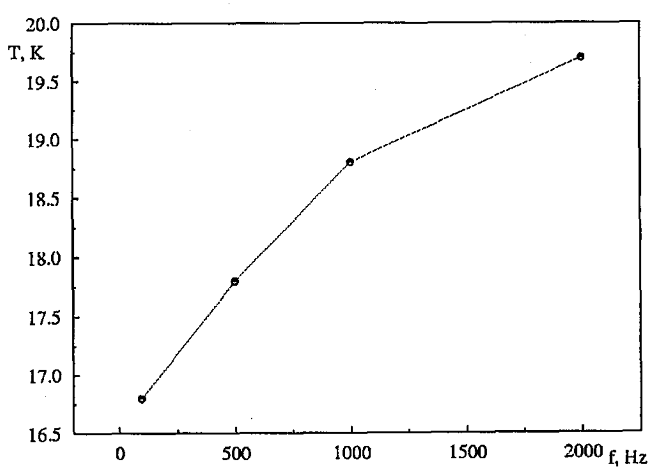

Fig. 3. Frequency dependences of T_{cusp} temperature of ac susceptibility.

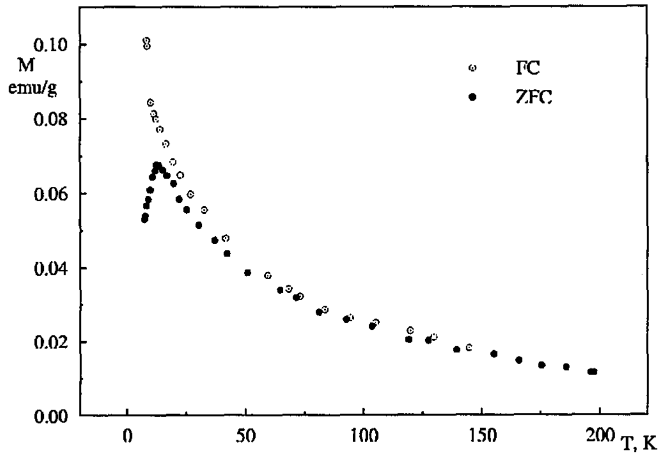

Fig. 4. Zero-field-cooled and field-cooled magnetization in a field of 0.1 kOe.

Thus, both types of magnetic measurements, ac susceptibility and dc magnetization, strongly support a disordered spin state, namely, a spin-glass-like one in the system studied.

It should be noted that the peculiarities of the χ_{ac} and M_{dc} behavior observed in the organometallic magnet $Na[FeO_6(C_{10}H_8N)_3]$ may also occur in systems other than spin-glass, such as superparamagnetic clusters. Therefore, the additional measurements will be required for further understanding of magnetic behavior in this system.

4. Conclusion

The experimental results and its analysis show that some peculiarities of the ac susceptibility and dc magnetization behavior in the organometallic magnet $Na[FeO_6(C_{10}H_8N)_3]$ observed at low temperatures and in an external magnetic field, e.g. cusps in the susceptibility and ZFC magnetization, an irreversibility between the FC and ZFC magnetization and dependences of the T_{cusp} temperature on the frequency and the intensity of the magnetic field, are characteristic of spin-glasses systems [5].

References

[1] O. Kahn, *Molecular Magnetism*, VCN, Weinheim 1993.

[2] J.S. Miller, W.M. Reiff, A.J. Epstein, *Chem. Rev.* **88**, 201 (1988).

[3] M. Wan, C. Zhang, C. Li, *Solid State Commun.* **87**, 379 (1993).

[4] A.R. Harutyunyan, A.A. Kuznetsov, H. Szymczak, R. Szymczak, M. Baran, A. Nabialek, *J. Magn. Magn. Mater.* **162**, 338 (1996).

[5] J.A. Mydosh, *Spin Glasses: An Experimental Introduction*, Taylor & Francis Ltd., London 1993, p. 256.