Experimental Evidence for Ising Spin-Glass Transition in the YbCoGaO$_4$ Single Crystal

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Abstract. Magnetic measurements (ac and dc susceptibilities and magnetization) on YbCoGaO$_4$ single crystal as a function of frequency and temperature are presented. The studied system behaves like a three-dimensional Ising-like spin glass. A cusp is seen only in the longitudinal susceptibility, whereas a paramagnetic behavior is observed in the transverse susceptibility. Typical spin glass features are observed in the frequency and temperature dependence of the ac susceptibility. Dynamical scaling is performed and critical exponents are deduced and compared to those obtained for Ising and Heisenberg systems. The origin of the Ising anisotropy of Yb$^{3+}$ and Co$^{2+}$ ions is briefly discussed.

1. Introduction

In the present article, we report the magnetic properties of a single crystal of YbCoGaO$_4$ layered cobaltite. Its crystal structure is described by space group R-3m [1,2]. It consists of double layers of Co/GaO$_5$ triangular bipyramids and triangular layers of YbO$_6$ octahedra. In the YbCoGaO$_4$ structure Co$^{3+}$ and Ga$^{3+}$ cations are distributed randomly on the sites of the bilayers and consequently this crystal is both spin and charge frustrated. Moreover, this crystal is unusual in that it has two magnetic sublattices, both of which have triangular geometries. All these reasons are expected to lead to unconventional ground states such as a spin liquid. The main purpose of this paper is attempt to clarify this problem.

2. Experimental

The single crystal of YbCoGaO$_4$ was grown by floating zone image furnace techniques [1,2]. The crystal structure was investigated at room temperature by the powder X-ray diffraction method. It was found that samples investigated are rhombohedral (space group R-3m). The site symmetry of all the atoms is 3m. Magnetic measurements (dc and ac susceptibilities and magnetization) were performed using a commercial superconducting quantum interference device squid magnetometer MPMS-7 XL (Quantum Design). Figure 1 shows the temperature dependence of the zero-field-cooled (ZFC) and
field-cooled (FC) magnetization of YbCoGaO$_4$ crystal recorded in magnetic field $H=5$ kOe applied along the $c$ axis and perpendicularly to this axis. As is seen in this figure the longitudinal (with field applied along the $c$ axis) susceptibility shows a typical spin-glass-like cusp, whereas no cusp is observed in the transverse direction. Even stronger cusp is observed in lower magnetic fields (see figure 2) at temperature $T_\text{c}=20.3$ K. Above $T_\text{c}$ magnetic susceptibility $\chi$ is well described by the Curie–Weiss formula $\chi=C/(T+\theta)$ with the paramagnetic Curie–Weiss temperatures $\theta$ equal to: $\theta_c=33$ K for $H \parallel c$ and $\theta_c=56$ K for $H \perp c$. Despite high value of a Curie-Weiss temperature, no magnetic ordering is observed down to 2.4 K and a broad peak in the susceptibility is currently attributed to a spin glass transition.

![Figure 1](image1.jpg)  
**Figure 1.** Temperature dependence of the ZFC (squares) and FC (triangles) magnetization recorded in $H=5$ kOe applied along the $c$ axis and perpendicularly to this axis

![Figure 2](image2.jpg)  
**Figure 2.** Temperature dependence of the ZFC magnetization recorded in $H=50$ Oe applied along the $c$ axis

The observed difference between $\theta_c$ and $\theta_c$ arises due to the strong magnetocrystalline anisotropy of easy axis type in both sublattices. From the Curie-Weiss law one can determine magnetic moments $p_{\text{eff}}$ of the studied system per formula unit: $p_{\text{eff}}=6.6\mu_B$/f.u. for $H \parallel c$ and $p_{\text{eff}}=4.28\mu_B$/f.u. for $H \perp c$. The strong anisotropy of the paramagnetic Curie–Weiss temperatures and magnetic moments indicates on the Ising-like character of the system. The results of $p_{\text{eff}}$ measurements on polycrystalline magnets of the same structure: LuCoGaO$_4$, YbCuGaO$_4$ and LuCuGaO$_4$ [3] suggest that both Yb$^{3+}$ and Co$^{2+}$ are strongly anisotropic (probably Ising-like) ions in YbCoGaO$_4$ lattice. The mechanism responsible for this strong anisotropy is related to the crystal field effects. Because of strong trigonal crystal field the ground state of Yb$^{3+}$ is doublet with extremely anisotropic g-tensor in full analogy to the behavior of Tb$^{3+}$ in TbBaCo$_4$O$_{7}$ [4]. For Co$^{2+}$ ions it is sufficient to conclude that their ground state in YbCoGaO$_4$ lattice is orbital triplet $^4T_1$, and consequently their magnetic properties are determined mainly by orbital moment contribution to this state.

To further characterize the system near $T_\text{c}$ the real ($\chi'$) and imaginary ($\chi''$) components of ac susceptibility were measured using SQUID magnetometer with a small ac field ($h$) superimposed on a static field ($H$). Both magnetic fields were applied along the $c$ axis of the crystal. The susceptibility was measured in the frequency interval $1$ Hz $\leq \omega 2\pi \leq 1000$ Hz. Figure 3 presents the temperature dependences of the real part of the ac susceptibility with zero bias dc field. The different curves correspond to different frequencies $\omega 2\pi$ of the ac field. For fixed frequency $\omega 2\pi$ a sharp cusp is observed at $T_\text{c}$. The temperature dependence of the imaginary part of ac susceptibility presents figure 4.
Figure 3. The real part, \( \chi' \), of the ac susceptibility vs temperature at different frequencies

The positions of the cusps in \( \chi' \) correspond to the maximum slope in \( \chi'' \) and shift to higher temperature with increasing frequency, while the magnitude of \( \chi' \) decreases for higher frequency. Such behavior is a signature of spin-glass behavior. A quantitative measure of the observed frequency shift is introduced by Mydosh \[7\] parameter \( \Delta T_0/T_0 \ln \omega \). This parameter varies in the range 0.001÷0.02 for spin glasses and is considerably higher (of the order of 0.3) for superparamagnets. In the case of YbCoGaO\(_4\) this parameter is of about 0.02. This value confirms the spin-glass character of the studied crystal.

The most relevant support of this conclusion may be achieved by analyzing the frequency, field and temperature dependences of the magnetic susceptibility \( \chi(\omega,T) \). The fundamental question is whether the observed transition in \( T_0 \) temperature is a true phase transition or a gradual freezing of the magnetic moment.

According to \[5,6\] the following dynamic scaling equation should be valid for spin glass systems:

\[
\frac{\chi''(T)}{\omega^\beta z\nu} = f(\epsilon/\omega^\beta z\nu)
\]  

(1)

where \( \epsilon = T/T_0 - 1 \), \( \beta \) is a critical exponent, \( z\nu \) is a dynamic exponent and \( f(\chi) \) is a scaling function.

Using a fitting parameters, all \( \chi'(\omega) \) vs \( T \) curves should collapse onto the same curve. Figure 5 shows such scaling plot, performed with the parameters \( z\nu=18\pm1 \) and \( \beta=1.00\pm0.01 \). Interesting relation is obtained for \( \epsilon=0 \). In this case the relation for the maximum, \( \chi'_{\text{max}} \), of each curve in figure 4 is given:

\[
\chi'_{\text{max}}(T) = \omega^\beta z\nu
\]

(2)

Using the relation (2) the parameter \( \beta z\nu \) was determined to be \( \beta z\nu=0.055\pm0.02 \). This value is very comparable to that obtained using relation (1).

The critical exponents for the three-dimensional Ising-like spin glasses is expected \[8\] to be \( z\nu=10-12 \) and \( \beta=0.54 \) while for Heisenberg-like spin glasses these values are \( z\nu=6-8 \) and \( \beta=0.9 \). It means that the critical exponents of YbCoGaO\(_4\) essentially lie between those obtained for the Heisenberg and Ising spin glasses. The possible reason for the deviation from Ising model may be related to the twosublattices structure of YbCoGaO\(_4\) not taken into account in theoretical considerations.
3. Discussion and conclusion

It was shown that YbCoGaO$_4$ single crystal exhibits a strong uniaxial anisotropy observed in its dc and ac magnetic susceptibilities. A sharp cusp is seen in the longitudinal susceptibility measured with the field directed along the c axis, whereas a paramagnetic behavior is observed in transverse direction. Transverse susceptibility is considerably weaker than the longitudinal one because of Ising-like anisotropy. Considering all experimental data it was concluded that YbCoGaO$_4$ is Ising-like spin glass. The spin glass nature of this crystal is due to frustration arising from random distribution of Co$^{2+}$ and Ga$^{3+}$ ions. Yb$^{3+}$ subsystem exhibits also geometrical frustration. Ising anisotropy of both kinds of ions is due to crystal fields. The scaling analysis performed on YbCoGaO$_4$ single crystal and for magnetic field applied along the c axis proves that the relation (1) for spin-glass systems describes properly the frequency and temperature dependences of $\chi''(\omega, T)$.

The results obtained are in agreement with theoretical models based on mean-field calculations developed for uniaxial crystals[9, 10]. These models predict spin-glass ordering for the longitudinal spin components and not for the transverse ones under the condition that the uniaxial magnetic anisotropy is large enough with respect to the exchange interactions. In the case of YbCoGaO$_4$ crystals two such subsystems exist but only one cusp is observed in longitudinal susceptibility. It suggests strong Yb-Co exchange coupling inside the studied system.

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