Observation of Spin-Glass Behavior in Spinel Compound CoGa2O4

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Research Article

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Abstract

In this work, the synthesis process, crystal-structure, and comprehensive physical properties of spinel compound CoGa$_2$O$_4$ have been investigated. The competition between antiferromagnetism (AFM) and ferromagnetism (FM) are considered to be the crucial elements for resulting in spin-glass (SG) behavior due to magnetic frustration. The observed SG behavior is determined by the temperature dependence of magnetization $M(T)$ curves under the ZFC (zero-field-cooled) and FCC (field-cooled) processes, where form the intense irreversibility divergence. Moreover, the corresponding fitting parameters (the freezing temperature $T_0 = 9.32$ K, the flipping time $\tau_0 = 4.49 \times 10^{-10}$ s, and the dynamical exponent $z = 4.46$) strongly indicate the existence of the SG behavior. Meanwhile, as another specific characteristic for SG, in our present work, frequency ($f$) and magnetic field ($H$) have a strong influence on the peaks of AC susceptibility. From where, with the increase of $f$ and $H$, the freezing temperature follows a corresponding peak shift. All the above phenomena and relevant analyses of magnetic frustration behavior confirm the typical SG behavior in CoGa$_2$O$_4$ system.

1. Introduction

Since the typical spin glass (SG) behavior has been found in magnetic materials, the sub-stable magnetic alloy material and magnetic compounds have been extensively studied due to its interesting structure and properties [1, 2]. The research of SG plays a crucial role in many comprehensive study of interdisciplines, meanwhile, it also provides mathematical tools to solve practical problems in memory elements, microwave components, computing machinery, functional materials, and biology science [3–5]. As is known to all, the SG behavior is usually existed in the antiperovskite compounds with a formula AXM$_3$ (A = Ga, Al, Cu, In, Sn, Zn, Ge etc.; X = C, N; M = Ni, Mn, Fe, etc) such as SnCFe$_3$ [6] and GaNMn$_3$ [7]. However, to date, there are no report on the SG behavior in CoGa$_2$O$_4$ spinel compound, and thus the work seems more necessary and meaningful.

In this paper, the spinel structure CoGa$_2$O$_4$ has been prepared by solid state method. The distribution of the cations at the tetrahedral and the octahedral sites can be reflected by the structural formula $(\text{Co}_{1-x}\text{Ga}_x)[\text{Co}_x\text{Ga}_{2-x}]\text{O}_4$ [8, 9]. It can be seen that, Co$^{2+}$ ions and Ga$^{3+}$ ions are both divided into A and B-sites. We report and verify the spin glass behavior of the prepared sample, from where, the SG behavior in CoGa$_2$O$_4$ is confirmed to be resulted from the magnetic frustration and atomic disorders according to the systematical measurements. The temperature dependence of magnetization $M(T)$ and magnetic susceptibility $\chi(T)$ are measured at several fixed frequencies. The isothermal remanent magnetization ($M_{IRM}$) are also performed and described in the following sections. We illustrate a detailed investigation of the magnetic property and structural characteristic of the sample. As all results shown, the competition between AFM and FM interactions should be responsible for the SG behavior of CoGa$_2$O$_4$ [10, 11].

2. Experimental Details
CoGa$_2$O$_4$ with the chemical formula of \((\text{Co}_{1-x}\text{Ga}_x)[\text{Co}_x\text{Ga}_{2-x}]\text{O}_4\) was analyzed by standard solid state reaction method in air atmosphere. The powder of materials \(\text{Co}_2\text{O}_3\) (100%) and \(\text{Ga}_2\text{O}_3\) (99.8%) were used as starting materials, and homogeneous mixed by the ball milling. The mixture was sintered at 1300 °C in the muffle for 12 h. When the sample is cooled to room temperature at a rate of 3 K/min, the sample is basically prepared. After quenching to room temperature, the products were pulverized, mixed, pressed into pellets, and annealed again in order to obtain homogeneous samples.

The phase identification of the prepared sample was measured by X-ray diffraction at room temperature (XRD, Rigaku D/max-2550V/PC, Cu Kα (\(λ = 1.5406 \text{ Å}\))) with the angle from 20° to 90°. Scanning electron microscope (SEM) was used to characterize the microstructure, and Energy Dispersive X-Ray spectroscopy (EDX) was used to analyze the element composition. All the magnetic measurements were performed by a Quantum Design superconducting quantum interference device based on magnetic property measurement system (SQUID-MPMS 3).

### 3. Results And Discussion

Figure 1. (a) XRD spectra and Rietveld refinement at room temperature for CoGa$_2$O$_4$. Inset shows the Rietveld refinement XRD pattern. (b) The crystal structure of CoGa$_2$O$_4$.

In order to further gain the insight of element composition and state of CoGa$_2$O$_4$, XPS spectrum of the sample is illustrated in Fig. 3. Figure 3(a) presents the survey XPS spectrum of CoGa$_2$O$_4$ sample, where the survey XPS spectrum contains the peaks of O 1s, Ga 2p, and Co 2p, and confirms the existence of Co, Ga, and O in the CoGa$_2$O$_4$ composite. As shown in Fig. 3(b), the Co 2p spectrum is exhibited, in which the binding energies of the Co 2p$_{1/2}$ and Co 2p$_{3/2}$ have four peaks at 803.2 eV, 797 eV, 786.6 eV, and 781.3 eV. Meanwhile, two specific peaks at 781.3 eV and 797 eV are attributed to Co$^{3+}$, and the other two peaks at 786.6 eV and 803.2 eV are associated with Co$^{2+}$ [18–20]. Figure 3(c) shows the XPS spectrum of Ga 2p. The observed energy peaks of Ga 2p$_{1/2}$ and Ga 2p$_{3/2}$ are located at 1144.5 eV and 1117.7 eV respectively [21, 22]. In the case of Fig. 3(d) for O 1s spectra, the peak with binding energy of 531.05 eV is related to oxygen bonding [23, 24]. Through the analyses above, all of the measurement results of XPS spectrum show that the prepared sample contains all the elements, which are well consistent with other related investigations.

Figure 4(a) shows the temperature dependence of magnetization \(M(T)\) curves for CoGa$_2$O$_4$ at different magnetic fields under the processes of zero field cooled (ZFC) and field cooled (FC). It is not difficult to identify that the compound adopts a characteristic of spin glass behavior since the \(M_{ZFC}\) and \(M_{FC}\) curves exhibit significant divergence at lower temperatures. The observed spin glass behavior of the sample may be due to the disorder of atoms and the competition of different magnetic spin-orders [25]. Meanwhile, with increasing the applied magnetic field, both the peak position of weak irreversibility \(T_{wi}\) (defined as the temperature where \(M_{ZFC} = M_{FC}\)) and strong irreversibility \(T_{si}\) (defined by the maximum value of magnetization) curves shift to lower temperature. It is shown that the spin glass state is gradually
destroyed under a large external magnetic field [26]. As plotted in Fig. 4(a), an obvious irreversibility appears below $T_{wp}$ which are the typical characteristic of spin glass behavior. Below $T_{wp}$ the value of $M_{FC}$ remains almost a constant, meanwhile, $M_{ZFC}$ almost drops to zero as the temperature decreases.

According to previous reports, at low temperature, the difference between $M_{FC}$ and $M_{ZFC}$ curves may be resulted from the competition between magnetic couplings [27]. As shown in Fig. 4(b), the magnetic hysteresis loops ($M$-$H$) for CoGa$_2$O$_4$ sample are performed at 300 K and 5 K respectively. All $M(H)$ curves have a linear relationship with magnetic field in high field, which is consistent with spin glass system. According to the obtained results, neither of them reach saturation even up to 50 kOe. For another prominent case, $M$ is bigger than 300 K at 5 K because of thermal disturbance has a susceptible effect on the magnetic moment.

In order to further confirm the spin-glass behavior, we characterized the temperature dependence of AC magnetic susceptibility under the changing frequency and magnetic field process. Figure 5(a) and 5(b) display AC magnetic susceptibility $\chi(T)$ for CoGa$_2$O$_4$ at AC field of $H_{AC} = 2$ Oe under different fixed frequencies ($f = 1, 10, 100, 500,$ and 1000 Hz), respectively. It can be found that both $\chi'(T)$ and $\chi''(T)$ present strongly frequency-dependent peaks, obviously, the positions of these peaks shift to higher temperatures and the magnitudes decrease with increasing $f$, which indicate a typical spin glass behavior. Generally, $\Delta T_f/\Delta (\log_{10} f)$ can determine the dependence of peak shift on frequency, and the typical value for spin-glass system is between 0.0045 and 0.08. Under the AC magnetic field, the energy of the applied magnetic field becomes smaller in one direction, and $H_{AC}$ changes rapidly in the opposite direction with the increase of $f$. Meanwhile, the system needs a higher temperature field $T_{f}(f)$ to reach a stable state. In fact, the value of relaxation time $\tau$ around the transition temperature can be written as follows [28]:

$$\tau = \tau_0[T_{f}(f)/T_0 - 1]^{zv}, \quad T_f > T_0 \quad (1)$$

Here, $T_0$ represents the freezing temperature, $\tau$ stands for the relaxation time $\tau = 1/(2\pi f)$, $\tau_0$ stands for the characteristic flipping time of the magnetic moments, $T_f$ is the frequency dependent of the peak position in $\chi'(T)$, and $zv$ is the dynamical critical exponent. Acknowledged from the previous reports on spin glass, the parameters of $\tau_0$ and $zv$ for typical spin glass are located as follows, where $\tau_0$ is in the range of $10^{-10}$-$10^{-13}$ s and $zv$ is 4–13, respectively. In this paper, the parameters of $T_0 = 9.32$ K, $\tau_0 = 4.49 \times 10^{-10}$ s, and $zv = 4.64$ obtained by fitting formula (1) further suggest the typical spin-glass behavior [6]. Figure 5(c) and 5(d) present the $\chi'(T)$ and $\chi''(T)$ for CoGa$_2$O$_4$ under several bias DC magnetic fields respectively, with AC magnetic field $H_{AC} = 2$ Oe and $f = 10$ Hz. It is clearly shown that the values of $T_f$ transfer to a lower temperature and the value of peak decreases with $H_{DC}$ increase. There is a typical dependence and linear relationship between the value of $T_f(H)$ and $H^{2/3}$, which is expected for the 3D-Heisenberg SG behavior [29].
The isothermal remanent magnetization $M_{IRM}(t)$ was measured under ZFC process with different magnetic fields from 300 to 5 K. It is necessary to emphasize the measurement process that under the premise of cooling the sample to the required temperature in the zero field, then apply a magnetic field for about 600 s and measure the remanent magnetization with decaying time. It is obviously seen from the Fig. 6 that $M_{IRM}$ is mainly a straight line and is nonzero in different fields, which proves the distinct existence of spin frustration in CoGa$_2$O$_4$ and also a typical SG behavior. The data of experiment under different fields can be obtained by fitting the following formula [28, 30]:

$$M_{IRM}(t) = M_0 - \alpha \ln(t), \quad (2)$$

As shown in the inset of Fig. 6, the relationship between fitting parameters $M_0$ and $\alpha$ is depicted. Obviously both parameters increase rapidly and then tend to saturate, which further proves the SG behavior of CoGa$_2$O$_4$ [31, 32].

In Fig. 7, we have exhibited PODS and total DOS for spin-up (↑) and spin-down (↓) states for CoGa$_2$O$_4$ and the figure is formed by the s, p, d and Total states. It can be concluded from the figure that the conduction band in CoGa$_2$O$_4$ is about 15 eV wide and is formed by the Co 3d states, Ga 4s, 4p states and O 2p states. The width of the valence band is about 15 eV and consists of two sub-band, it is also clear in the band structure: the one between −8 and 0 eV is the O 2p states and Co 3d states. The other one is between −15 and −13 eV is created by Co 3d states. The band between −20 and −18 eV is due to the Ga 4s states [33]. The spin magnetic moments of the spin up and spin down with asymmetric distribution imply the net magnetization in the sample [28]. The calculated magnetic characteristics of CoGa$_2$O$_4$ are consistent with the measured magnetic properties.

We note that the CoGa$_2$O$_4$ sample also behaves a SG behavior with the reliable parameters:

$$\Delta T_f/\Delta T_0 \Delta (\log_{10} f) \sim 0.025, \quad T_0 = 9.32 K, \quad \tau_0 = 4.49 \times 10^{-10} s, \quad \text{and} \quad zv = 4.64.$$ As the result shown that the characteristic parameter varying with the different fixed frequencies: the magnitude of $\Delta T_f/\Delta T_0 \Delta (\log_{10} f)$ decreases with increasing $f$, which indicates a strong dependence on frequency and associates with a typical SG behavior in CoGa$_2$O$_4$. Generally, the existence of SG is closely associated with the competition between FM and AFM interactions which results from the multiconfiguration of spins or spin frustration. The similar case have been reported in other analogous spinel ferrites as well as antiperovskite compounds [Zn$_{0.8-x}$Ni$_x$Cu$_{0.2}$Fe$_2$O$_4$ and SnNCo$_3$] [1, 28], where the local FM cluster and atomic disorder caused by the atomic deficiency were observed. And thus, based on these discussions, the origin of SG behavior in our present work should be closely dependent upon the spin frustrations and disorder occupations of the mixed sites. That is to say, the observed SG behavior in CoGa$_2$O$_4$ may be attributed to the atomic disorders introduced by the Ga/Co deficiency which affects the characteristic parameters of the SG state remarkably.

**4. Conclusion**
In summary, the structure and magnetic properties of the compound CoGa$_2$O$_4$ were investigated systematically. The results of XRD, EDX, and XPS confirmed that the pure CoGa$_2$O$_4$ sample was prepared, and exhilaratingly, SG was also observed in CoGa$_2$O$_4$ from magnetic measurements. To further elaborate the SG, the following work is completed, for instance, $M(T)$ curves with different applied field/frequency, field dependent AC susceptibility, and $M_{IRM}$ at fixed magnetic fields. The characteristic parameters $T_0 = 9.32$ K, $\tau_0 = 4.49 \times 10^{-10}$ s, and $z\nu = 4.64$ obtained from AC magnetic susceptibility indicate that CoGa$_2$O$_4$ is a SG system. Meanwhile, the curves of $M(T)$, $\chi'(T)$, and $\chi''(T)$ change regularly with the variation of magnetic and frequency. These phenomena can be explained by different magnetic competition and disorder. In other words, the competition between FM and AFM leads to the existence of SG, which is characterized by spin frustration and disorder. On this basis, the spin glass behavior of CoGa$_2$O$_4$ may be also caused by the competition between competing magnetic orders.

**Declarations**

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**Data Availability**

All data generated or analyzed during this study are included in this published article.

**Compliance with ethical standards**

**Conflict of interest** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Figures

(a) XRD spectra and Rietveld refinement at room temperature for CoGa2O4. Inset shows the Rietveld refinement XRD pattern. (b) The crystal structure of CoGa2O4.

Figure 1
**Figure 2**

(a) EDX spectroscopy for the prepared sample. (b) SEM image of CoGa2O4. (c)-(f) EDX element maps for the sample.

| Element | Weight% | Atomic% |
|---------|---------|---------|
| O       | 17.67   | 47.19   |
| Ga      | 61.31   | 37.57   |
| Co      | 21.02   | 15.24   |
Figure 3

XPS spectrum of CoGa2O4: (a) The survey XPS spectra; (b) Co 2p; (c) Ga 2p; (d) O 1s.
Figure 4

(a) M(T) curves for CoGa2O4 under ZFC/FC processes with different applied magnetic fields. (b) Magnetic hysteresis loops of CoGa2O4 at 5 and 300 K. The above and below insets show the enlarged M(H) region at 5 K and 300 K respectively.
Figure 5

(a) and (b) Temperature dependence of $\chi'(T)$ and $\chi''(T)$ measured at several fixed frequencies for CoGa2O4. Inset of Fig. 5(a) stands for $\tau(s)$ vs $T(K)$ plot. (c) and (d) Temperature dependence of $\chi'(T)$ and $\chi''(T)$ at several fixed DC fields. Both insets of Fig. 5(c) and 5(d) show the $T_f$ plot as a function of $H^{2/3}$. 
Figure 6

The plots of isothermal remanent magnetization (MIRM) at several fixed magnetic fields for CoGa2O4. The solid lines are obtained by fitting the experimental data with formula $M_{\text{IRM}}(t) = M_0 - \alpha \ln(t)$. The inset shows the magnetic field dependence fitting parameters $M_0$ and $\alpha$. 
Figure 7

exhibited PODS and total DOS for spin-up (↑) and spin-down (↓) states for CoGa$_2$O$_4$ and the figure is formed by the s, p, d and Total states.