Paramagnetic ground state with field-induced partial order in Nd$_3$Ga$_5$SiO$_{14}$ probed by low-temperature heat transport

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We study the low-temperature heat transport of Nd$_3$Ga$_5$SiO$_{14}$, which is a spin-liquid candidate, to probe the nature of ground state and the effect of magnetic field on the magnetic properties. The thermal conductivity ($\kappa$) shows a purely phononic transport in zero field. The external magnetic field along the $c$ axis induces a dip-like behavior of $\kappa(H)$, which can be attributed to a simple paramagnetic scattering on phonons. However, the magnetic field along the $ab$ plane induces another step-like decrease of $\kappa$. This kind of $\kappa(H)$ behavior is discussed to be related to a field-induced partial order, which yields low-energy magnetic excitations that significantly scatter phonons. These results point to a paramagnetic ground state that partial magnetic order can be induced by magnetic field along the $ab$ plane, which is also signified by the low-$T$ specific heat data.

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I. INTRODUCTION

Geometrically frustrated magnetic materials with competing spin interactions give rise to macroscopic degeneracy of ground state. They have displayed abundant exotic magnetic phenomena and introduced important concepts to condensed matter physics.$^{1-9}$ The recently discovered rare-earth Langasites, $R_2$Ga$_5$SiO$_{14}$ ($R$ = Nd, Pr), with two-dimensional geometrical frustration have attracted much attention due to their possible spin-liquid state.$^{8,9}$

Nd$_3$Ga$_5$SiO$_{14}$ (NGSO) crystallizes in the trigonal space group $P321$, and Nd$^{3+}$ spins ($S = 9/2$) occupy a network of corner-sharing triangles, forming a distorted kagomé-like lattice in the $ab$ plane stacked along the $c$ axis, which is topologically equivalent to a perfect one when only the nearest-neighboring interaction is considered.$^8$ The magnetic anisotropy associated with crystal field effect was confirmed from the magnetic susceptibility and nuclear magnetic resonance (NMR) measurements, which displayed a distinct crossover from the easy plane (kagomé plane) to the easy axis ($c$ axis) below 33 K.$^8,10$ The experimental investigations on the ground state of NGSO seemed to be rather confusing. The muon spin relaxation ($\mu$SR) and neutron scattering experiments did not detect any phase transition or long-range magnetic order down to milliKelvin temperatures in zero field.$^7,9,10$ In some earlier works, the magnetic susceptibility showed that the Curie-Weiss temperature is about -52 K or -62 K and therefore the frustration factor is large ($\sim 1300$), indicating a spin-liquid ground state with rather strong spin interactions.$^8,9$ However, by measuring the low-lying crystal-field levels of Nd$^{3+}$ via the magneto-optical spectroscopy, it was found that the exchange interaction of Nd$^{3+}$ spins is surprisingly small, leading to a revised frustration factor of $\sim 1.1$. This finding is essentially consistent with the single-ion quantum relaxation at low temperatures revealed by the neutron scattering.$^{9,12}$ These results ruled out NGSO as a spin liquid at least down to 100 mK. So the ground state of NGSO remains to be fully understood. In addition, the effect of magnetic field on the ground state of NGSO has been probed by magnetization and neutron scattering measurements.$^8,11-13$ At 1.6 K, the magnetization for $H \parallel c$ and $H \perp c$ showed a simple spin polarization, but the saturated moments are only half of the free Nd$^{3+}$ moment, which is still an open question.$^8,10,12$

Low-temperature heat transport has recently been found to be an effective way to probe the magnetic excitations and the field-induced quantum phase transitions.$^{14-16}$ In this work, we study the heat transport properties of NGSO at low temperatures down to 0.3 K and in magnetic field up to 14 T. The thermal conductivity ($\kappa$) shows a typical phononic heat transport behavior in zero field and at very low temperatures, while it exhibits remarkably anisotropic magnetic-field dependence. The magnetic field along the $c$ axis causes a low-field dip in $\kappa(H)$ isotherms and the high-field $\kappa$ are the same as the zero-field values, which can be simply understood as a paramagnetic scattering on phonons. Whereas the $\kappa(H)$ isotherms in magnetic field along the $ab$ plane show sharp and step-like decreases, which indicates a field-induced magnetic transition. These transport results point to a paramagnetic ground state of NGSO but a field-induced partial order for $H \parallel ab$, which is consistent with what the low-$T$ specific heat data suggest.
II. EXPERIMENTS

High-quality NGSO single crystals are grown by the floating-zone technique in flowing argon and oxygen mixture with the ratio of 10:1.8,9 To reduce the bubble during the growth, the sintered feed rod is firstly pre-melted with a rate of 25 mm/h. Then the single-crystal rods (5 mm in diameter and 9 mm in length) are successfully obtained with a low growth rate of 1 mm/h. It should be noted that this fast-scan procedure is very useful to improve the quality of NGSO crystals in the aspects of both the size and the crystallinity. The samples for thermal conductivity measurements are cut into parallelepiped shape with a typical size of $3 \times 0.6 \times 0.15$ mm$^3$ along either the $a$ axis or the $c$ axis. The thermal conductivity is measured at low temperatures down to 0.3 K using a conventional steady-state technique, which has been described in details elsewhere.15,17–19 The specific heat is measured by the relaxation method in the temperature range from 0.4 to 30 K using a commercial physical property measurement system (PPMS, Quantum Design).

III. RESULTS AND DISCUSSION

Before presenting the heat transport results of NGSO single crystals, we study the low-$T$ specific heat in details. Figure 1 shows the temperature dependencies of the specific heat in zero field and in magnetic fields for $H \parallel c$ and $H \perp c$. The zero-field specific heat shows a upturn at low temperatures below 2 K, which has been observed in previous work and is apparently of magnetic origin. The distinct peaks develop in various magnetic fields both for $H \parallel c$ and $H \perp c$ with the peak position shifting to higher temperatures with increasing field. This is a characteristic behavior of the Schottky anomaly, which commonly exists in many magnetic materials. The phononic specific heat can be estimated by fitting the high-$T$ data in zero field using the low-frequency expansion of the Debye function $C_p = \beta T^3 + \beta_5 T^5 + \beta_7 T^7$ as shown by the dashed lines in Fig. 1. The best fitting gives $\beta = 1.5 \times 10^{-3}$ J/K$^3$mol, $\beta_5 = 1.29 \times 10^{-6}$ J/K$^5$mol, and $\beta_7 = -1.93 \times 10^{-9}$ J/K$^7$mol. The magnetic specific heat $C_m$ can be obtained by subtracting the lattice contribution from the total $C_p$, as shown in Fig. 2. The Schottky anomaly in NGSO can be simply interpreted as the splitting of a ground-state doublet of Nd$^{3+}$ with the effective spin $S = 1/2$. Therefore, the data in magnetic fields are tried to fitted by using the two-level Schottky anomaly.20,21

$$C_{sch} = N \left( \frac{\Delta}{k_B T} \right)^2 \frac{e^{\Delta/k_B T}}{(1 + e^{\Delta/k_B T})^2}, \quad (1)$$

where $\Delta$ is the gap value for the Zeeman splitting of the Nd$^{3+}$ Kramers doublet. The concentrations of free spins is $N/R$, with $R$ the universal gas constant.

![FIG. 1: (Color online) Temperature dependencies of the low-temperature specific heat of NGSO single crystals in the magnetic field applied along (a) and perpendicular (b) to the $c$ axis. The dashed black lines show the fitting of the high-$T$ zero-field data by using the Debye model of lattice specific heat.](image-url)
field along the $ab$ plane induces some magnetic excitations or spin fluctuations, which indicate some kind of field-induced partial or short-range magnetic order (no sign for long-range order in the specific heat). As we can see below, such kind of in-plane field induced order has remarkable impact on the low-$T$ heat transport.

Thermal conductivity as a bulk measurement technique is a sensitive probe to all elementary excitations that carry heat or scatter heat carriers. Figure 3 shows the temperature dependencies of $\kappa$ with heat flowing along the $a$ and $c$ axes in zero field. Both curves show phonon peaks at about 7 K, a typical feature of insulating crystals, and exhibit rather weak anisotropy. At subKelvin temperatures, a rough $T^{2.4}$ dependence is observed, indicating that $\kappa(T)$ behaviors are close to the boundary scattering limit. In order to judge whether the phonons are free from microscopic scattering at sub-Kelvin temperatures, we make an estimation of the mean free path $l$ of phonons at low temperatures. The phononic thermal conductivity can be expressed by the kinetic formula $\kappa_{ph} = \frac{1}{3}C v_p l$, where $C = \beta T^3$ is the phonon specific heat at low temperatures and $v_p$ is the average velocity. With $\beta$ value obtained from the above fitting of the specific-heat data, the average sound velocity $v_p = 2410$ m/s and the Debye temperature $\Theta_D = 310$ K are calculated by using the expression $\Theta_D = \frac{\hbar v_p}{k_B (6\pi^2 N_s V)^{1/3}}$ and $\beta = \frac{12\pi^4}{5} \frac{R s}{\Theta_D^3}$, where $V$ is the volume of crystal, $R$ is the universal gas constant, $N$ is the number of molecules per mole, and $s$ is the number atoms in each molecule. The inset to Fig. 3 shows the temperature dependencies of the ratio $l/W$ for $\kappa_a$ and $\kappa_c$, where $W$ is the averaged sample width. It is found that both the $l/W$ ratios are rather close to 1 with lowering temperature down to 0.3 K, indicating that the phonon heat transport of NGSO indeed approaches the boundary scattering limit at very low temperatures and the microscopic scatterings on phonons are nearly wiped out.

For NGSO, the $\mu$SR and neutron spectroscopy revealed that the magnetic fluctuations slow down below 10 K but keep persistent down to milliKelvin temperatures in zero field. However, the $\kappa(T)$ curves do not show any clear anomalies around 10 K, which suggests that the magnetic

![Figure 2](image1.png)

**FIG. 2:** (Color online) Temperature dependencies of magnetic specific heat $C_m$ in field applied along (a) and perpendicular to (b) the $c$ axis, respectively. The solid lines correspond to the fitting results by a simple two-level Schottky model. (c) The relationship between the fitting parameter $\Delta$ and magnetic field. The thin lines show linear dependences $\Delta = 0.175H$ and $\Delta = 0.125H$ for $H \parallel c$ and $H \perp c$, respectively.

![Figure 3](image2.png)

**FIG. 3:** (Color online) Temperature dependencies of thermal conductivity along the $a$ axis and the $c$ axis of NGSO single crystals in zero field, respectively. The solid lines are the fitting results using the Debye model. The inset shows the temperature dependencies of the ratio of the phonon mean free path $l$ to the averaged sample width $W$. 

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excitations do not couple with phonons. Therefore, it is possible to try a more quantitative analysis on the zero-field data by using a classical Debye model of phonon thermal conductivity:

\[
\kappa_{ph} = \frac{k_B}{2\pi^2 v_p (\hbar/k_B)^2 T^3} \int_0^{\Theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \tau(\omega, T) dx, \tag{2}
\]

in which \(x = \hbar \omega/k_B T\) is dimensionless, \(\omega\) is the phonon frequency, and \(\tau(\omega, T)\) is the mean lifetime of phonon. The phonon relaxation is usually defined as

\[
\tau^{-1} = v_p/L + A \omega^4 + BT\omega^3 \exp(-\Theta_D/bT) + \tau_{res}^{-1}, \tag{3}
\]

where the four terms represent the phonon scattering by the grain boundary, the point defects, the phonon-phonon Umklapp scattering, and the resonant scattering, respectively. Since the \(\kappa(T)\) curves do not show clear signature of resonant scattering, the last factor can be neglected. The parameters \(v_p\) and \(\Theta_D\) are obtained from the specific-heat data and other parameters \(L\), \(A\), \(B\) and \(b\) are free ones. It is turned out that the zero-field thermal conductivity below 25 K is well simulated as the blue lines shown in Fig. 3. The best fitting parameters are \(L = 3.7 \times 10^{-4}\) m, \(A = 8.15 \times 10^{-41}\) s\(^3\), \(B = 9.8 \times 10^{-29}\) K\(^{-1}\)s\(^2\), \(b = 8.7\), and \(L = 4.8 \times 10^{-4}\) m, \(A = 5.5 \times 10^{-41}\) s\(^3\), \(B = 5.0 \times 10^{-29}\) K\(^{-1}\)s\(^2\), \(b = 9.0\) for \(\kappa_a\) and \(\kappa_c\), respectively. The parameters \(L\) are found to be in good consistent with the averaged sample widths of these two samples, which are \(3.70 \times 10^{-4}\) m (\(\kappa_a\)) and \(3.95 \times 10^{-4}\) m (\(\kappa_c\)), respectively. Note that the failure of fitting at \(T > 25\) K can be related to either the magnetic fluctuations or the contributions of optical phonons. However, these complexities at relatively high temperatures are not involved in the following discussion about the magnetic-field effect on low-\(T\) heat transport.

Detailed magnetic-field dependencies of the low-\(T\) thermal conductivity can provide useful information on the nature of the ground state. Figure 4(a) shows the detailed magnetic field dependencies of \(\kappa_a\) for \(H \parallel c\). The \(\kappa(H)\) are strongly suppressed down to about 50\% at low magnetic field and are recovered at high magnetic field, yielding a dip-like feature. Note that the position of the minimum moves to high magnetic field as increasing temperature. Based on the Schottky contribution to the low-\(T\) specific heat, the dip-like behaviors of \(\kappa(H)\) can be attributed to the phonon scattering by the paramagnetic moments, that is, the field-induced splitting of Nd\(^{3+}\) Kramers doublet can cause a resonant scattering on phonons, as we previously observed in Pr\(_{1.3}\)La\(_{0.7}\)CuO\(_4\) and GdBaCo\(_2\)O\(_{5+y}\). The Zeeman splitting of the spin doublet is known from the above fitting of Schottky anomaly for \(H \parallel c\), which gives \(\Delta(H) \approx 0.175H\). We can simulate the magnetic-field dependencies of \(\kappa(H)\) by using the Debye model for phonon thermal conductivity as shown in Fig. 4(b). One can see that most of \(\kappa(H)\) isotherms are in good agreement with the calculated ones except for some deviations for the high-\(T\) data, which might be due to the excitations of the higher-energy crystal field levels. Therefore, the \(c\)-axis magnetic field seems to affect the heat transport mainly by introducing some paramagnetic scattering on phonons. This is in good correspondence with what the specific-heat data suggest. It is worthy of pointing out that the \(\kappa_a(H)\) isotherms for \(H \parallel c\) are the same as those of \(\kappa_a(H)\) shown in Fig. 4.

The above paramagnetic scattering effect is known to be qualitatively isotropic for magnetic field along different directions. However, the heat transport behaves very differently if the magnetic field is applied in the \(ab\) plane. Figure 5 shows the magnetic-field dependencies of both \(\kappa_a\) and \(\kappa_c\) for \(H \perp c\). It is easily found that the \(\kappa(H)\) behaviors are essentially the same for different directions of heat current. All the \(\kappa(H)\) isotherms show significant suppression down to 10–20\% of the zero-field value. With lowering temperature, the decrease of \(\kappa\) becomes sharper at low fields. If a dip-like \(\kappa(H)\) term from the paramagnetic scattering effect is also included, the field dependence of Fig. 5 apparently has another contribution. This can be clarified by a simple analysis. As some examples shown in Fig. 6 (similar analysis can be done for other data), the changes of \(\kappa_a\) induced by para-
magnetic scattering are calculated in the same way as that for $H \parallel c$, considering $\Delta(H) \approx 0.125H$. Then subtracting this part of change from the raw data of $\kappa_a(H)$, the other contribution to the field dependence can be roughly estimated. It can be seen that at low temperatures, this additional term is actually a step-like suppression, which suggests that the $ab$-plane field induces some magnetic transition. This is compatible with what the specific-heat data signify, as discussed above. Although the quantitative description of the phonon scattering by the magnetic excitations in such a broad temperature region is not available now, the qualitative understanding can be easily figured out. Apparently, the field-induced partial or short-range order causes strong dynamic spin fluctuations that can strongly scatter phonons.

It should be pointed out that the in-plane field induced partial order, suggested by the specific heat and thermal conductivity data in the present work, however, has not been evidenced in the earlier works. It is known that the low-$T$ magnetization showed a simple spin polarization above $\sim 2$ T for both $H \parallel c$ and $H \perp c$\textsuperscript{8,12} and the neutron scattering also confirmed this phenomenon for $H \parallel c$.\textsuperscript{2} But the moments of the high-field $M(H)$ plateau are only half of the free Nd$^{3+}$ moment, which is an open question for the existing theories on the kagomé antiferromagnet.\textsuperscript{8,10,12} There have been no other experimental investigation about the details for $H \perp c$. Here, we propose one possibility that when the Nd$^{3+}$ moments are gradually aligned along the in-plane field, the $c$-axis component of Nd$^{3+}$ moments may form some kind of AF ordering. They could be anti-parallel either for the neighboring sites or for the neighboring layers. This picture is not inconsistent with the experimental results that the high-field $M(H)$ plateau is lower for $H \perp c$ than that for $H \parallel c$.\textsuperscript{8,12} Nevertheless, the validity of this canted AF order and the underlying mechanism, which should be related to the spin anisotropy, the crystal field effect, and the spin frustration, calls for a deeper investigation.
IV. CONCLUSION

In summary, the temperature and magnetic field dependencies of the thermal conductivity and specific heat are studied for NGSO single crystals. The zero-field thermal conductivity shows a purely phononic heat transport behavior at low temperatures, without showing the strong coupling between phonons and magnetic excitations. A dip-like field dependence of $\kappa$ for magnetic field along the $c$ axis can be explained by a simple paramagnetic scattering on phonons; whereas a step-like field dependence of $\kappa(H)$ for $H \perp c$ indicates a field-induced magnetic transition at low temperatures. These behaviors are consistent with the field dependencies of low-$T$ specific heat. The present results demonstrate a paramagnetic ground state rather than the spin liquid. However, the partial magnetic order induced by the field along the $ab$ plane seems to be quite unusual and remains to be further investigated both experimentally and theoretically.

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