

# Heat transport of the quasi-one-dimensional Ising-like antiferromagnet $\text{BaCo}_2\text{V}_2\text{O}_8$ in longitudinal and transverse fields

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The very-low-temperature thermal conductivity ( $\kappa$ ) is studied for  $\text{BaCo}_2\text{V}_2\text{O}_8$ , a quasi-one-dimensional Ising-like antiferromagnet exhibiting an unusual magnetic-field-induced order-to-disorder transition. The nearly isotropic transport in the longitudinal field indicates that the magnetic excitations scatter phonons rather than conduct heat. The field dependence of  $\kappa$  shows a sudden drop at  $\sim 4$  T, where the system undergoes the transition from the Néel order to the incommensurate state. Another dip at lower field of  $\sim 3$  T indicates an unknown magnetic transition, which is likely due to the spin-flop transition. Moreover, the  $\kappa(H)$  in the transverse field shows a very deep valleylike feature, which moves slightly to higher field and becomes sharper upon lowering the temperature. This indicates a magnetic transition induced by the transverse field, which however is not predicted by the present theories for this low-dimensional spin system.

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## I. INTRODUCTION

Low-dimensional or frustrated quantum magnets are good candidates for studying the quantum phase transition (QPT), an interesting phenomenon in condensed matter driven by the strong quantum fluctuations. QPT can be accessed by varying such parameters as magnetic field, pressure, or doping concentration. Among them, the magnetic-field-induced long-range order in the spin-gapped systems has attracted much attention because it can be mapped into a Bose-Einstein condensation of magnetic excitations.<sup>1-6</sup> Contrary to the field-induced ordering, a peculiar order-to-disorder transition in the presence of magnetic field was recently discovered in  $\text{BaCo}_2\text{V}_2\text{O}_8$  (BCVO),<sup>7-9</sup> which is a quasi-one-dimensional (1D) antiferromagnetic (AF) insulator.

BCVO crystallizes in the tetragonal  $I41/acd$  space group, with the edge-shared  $\text{CoO}_6$  octahedra forming a four-step periodic screw chain along the  $c$  axis.<sup>10</sup> The low-temperature magnetic susceptibility indicated a large magnetic anisotropy and the  $c$  axis is the spin-easy axis.<sup>7-9</sup> The Hamiltonian of BCVO in the magnetic field can be described by a 1D  $S = 1/2$  ( $\text{Co}^{2+}$ )  $XXZ$  Heisenberg model:

$$H = J \sum_i \{S_{i,z}S_{i+1,z} + \epsilon(S_{i,x}S_{i+1,x} + S_{i,y}S_{i+1,y})\} - \mu_B \sum_i S_i g H, \quad (1)$$

where  $J/k_B = 65$  K,  $\epsilon = 0.46$ , and  $g = 6.2$  and  $2.95$  for longitudinal and transverse field, respectively, without considering the next-nearest-neighboring interaction.<sup>11</sup> BCVO is therefore regarded as a quasi-1D Ising-like spin-chain system. In zero field, owing to the inevitable interchain interaction, a long-range AF order is formed below  $T_N = 5.4$  K and the ordered  $\text{Co}^{2+}$  spins align antiferromagnetically along the screw chain.<sup>12</sup> When the longitudinal field ( $H \parallel c$ ) is applied, a gapless Tomonaga-Luttinger liquid (TLL) state is

formed as the Néel state is destroyed.<sup>10,11</sup> This unusual field-induced order-to-disorder transition is driven by the quantum fluctuations. Moreover, below 2 K, an incommensurate (IC) order is established above a temperature-independent critical field of  $H_c = 3.9$  T and is resulted from the development of the long-range correlation in the TLL spin chain.<sup>13,14</sup> In the IC ordered state, the longitudinal components of the adjacent  $\text{Co}^{2+}$  moments direct antiparallel along the chain.<sup>14</sup>

The phase diagram was believed to be much simpler for a transverse field ( $H \perp c$ ), in which a spin polarization transition occurs at very high field.<sup>9,11</sup> However, the high-field magnetic susceptibility in both longitudinal and transverse fields revealed some anomalies that could not fit to these phase boundaries. Another first-order phase boundary, which has never been detected in the thermodynamic quantities, was recently probed by the ultrasound measurement in the IC region and was supposed to originate from the orbital ordering.<sup>15</sup> Apparently, more careful experimental investigations using various techniques are necessary for revealing the precise phase diagrams of BCVO. Low-temperature heat transport has recently been found to be very useful for probing the ground states and phase transitions in the low-dimensional spin systems.<sup>6,16-21</sup> In this paper, we study the very-low-temperature heat transport of BCVO in both longitudinal and transverse fields. It is found that the magnetic excitations play an important role in the heat transport properties by scattering phonons, which causes drastic changes of thermal conductivity across the magnetic phase transitions. In particular, some anomalies of  $\kappa(H)$  associated with unknown phase boundaries are observed. Our results demonstrate that the existing theories based on the 1D  $S = 1/2$   $XXZ$  Heisenberg model<sup>22-27</sup> are still far from describing this spin system well.

## II. EXPERIMENTS

The high-quality BCVO single crystals were grown by a spontaneous nucleation method.<sup>7</sup> The crystals were accurately

oriented by using the x-ray back-scattering Laue photographs with a precision of  $1^\circ$ . Two samples with dimensions of  $1.2 \times 0.59 \times 0.16 \text{ mm}^3$  and  $1.0 \times 0.67 \times 0.38 \text{ mm}^3$  were used to measure the thermal conductivity along and perpendicular to the spin-chain direction ( $\kappa_c$  and  $\kappa_{ab}$ ), respectively. The thermal conductivities were measured using a conventional steady-state technique and two different processes: (i) using a “one heater, two thermometers” technique in a  $^3\text{He}$  refrigerator and a 14 T magnet at temperature regime of 0.3–8 K; (ii) using a Chromel-Constantan thermocouple in a pulse-tube refrigerator for the zero-field data above 4 K.<sup>6,20,21,28,29</sup>

### III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependencies of thermal conductivity of BCVO single crystals in the longitudinal magnetic field with the heat current parallel and perpendicular to the  $c$  axis, respectively. It is clearly seen that no matter which direction the heat flows,  $\kappa$  exhibits similar behaviors under the influence of longitudinal field. It is well known that in the strongly anisotropic spin systems, such as the spin-chain materials, the magnetic excitations are not able to propagate perpendicular to the chain direction since the interchain spin interaction is much weaker than the intrachain one. As a result,  $\kappa$  perpendicular to the spin-chain direction can only be the contribution from the phonons, but  $\kappa$  along the chain can be

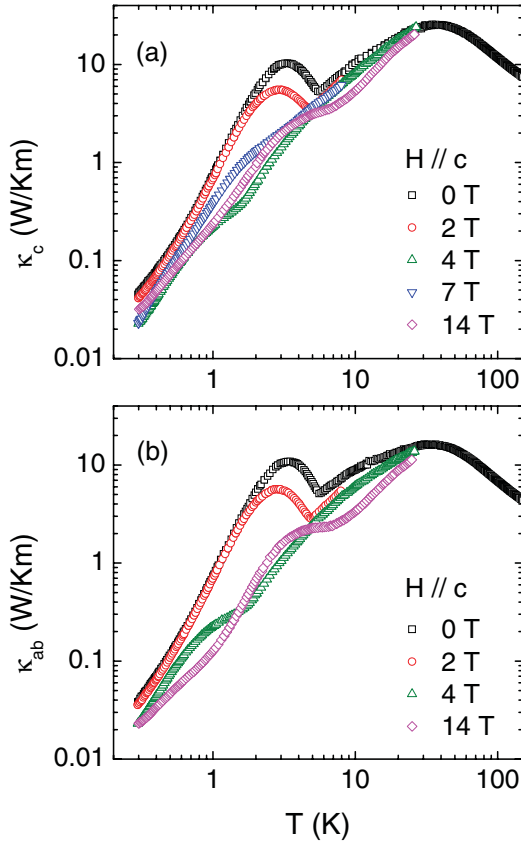


FIG. 1. (Color online) Temperature dependencies of thermal conductivity of  $\text{BaCo}_2\text{V}_2\text{O}_8$  single crystals in different magnetic fields applied parallel to the  $c$  axis with the heat current parallel (a) and perpendicular (b) to the  $c$  axis, respectively.

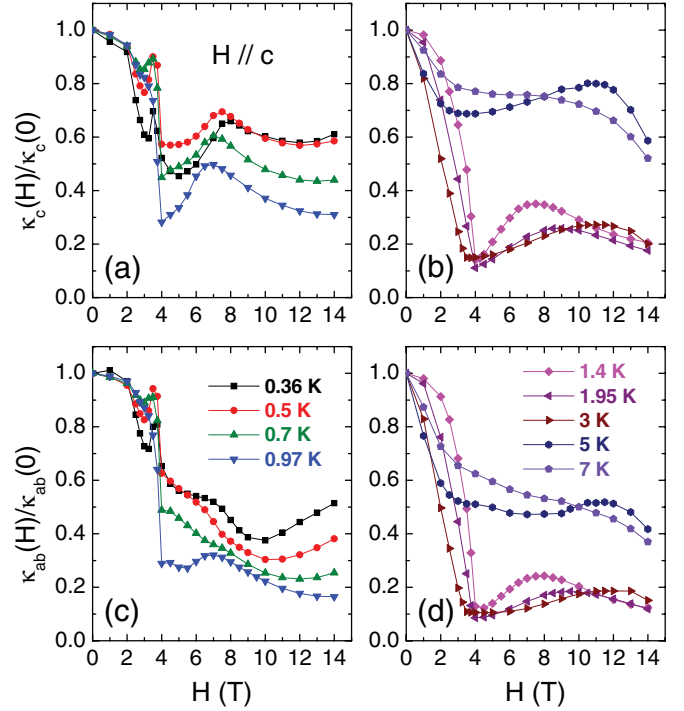


FIG. 2. (Color online) Magnetic-field dependencies of  $\kappa_c$  (a),(b) and  $\kappa_{ab}$  (c),(d) for field along the spin-chain direction.

the sum of phonons and magnetic excitations.<sup>30–32</sup> However, the nearly isotropic behavior of  $\kappa_c$  and  $\kappa_{ab}$  and much weaker temperature dependence than  $T^3$  at very low temperatures indicate that the heat carriers are mainly phonons for both directions of heat current. Therefore, the magnetic excitations in BCVO could act as a kind of phonon scatterers.

The zero-field  $\kappa(T)$  curves show a double-peak structure with a dip locating at  $\sim 5.4$  K, which is apparently related to the Néel order of  $\text{Co}^{2+}$  spins.<sup>12</sup> Upon increasing the magnetic field, the dip shifts to lower temperature, in agreement with the specific-heat results that the longitudinal field can suppress the AF order.<sup>8,9</sup> The double-peak structure is thus a result of strong phonon scattering by magnetic fluctuations, which are most significant at the phase transition. Moreover, the lower- $T$  peak weakens gradually due to the strengthened quantum fluctuation with increasing the field until 4 T, near the transition from Néel order to IC phase at  $H_c$ ,<sup>14</sup> at which field the lower- $T$  peak evolves into a shoulder. If the field is increased further, it seems that the shoulder moves to higher temperature. It can also be seen that  $\kappa$  in the magnetic field is never larger than the zero-field value even if a high field of 14 T is applied, indicating that the field-induced quantum fluctuation is dominant in the low temperature and high field region.

The magnetic-field dependencies of thermal conductivity with different heat-current directions in the longitudinal field are shown in Fig. 2. Similar to the  $\kappa(T)$  curves, the  $\kappa(H)$  isotherms also exhibit nearly isotropic transport behavior, again indicating the phonon scattering by magnetic excitations. In the presence of the longitudinal field,  $\kappa$  is always smaller than the zero-field value, and the strongest suppression of  $\kappa$  is down to  $\sim 10\%$ . The most remarkable feature of  $\kappa(H)$  is the steep drop at  $H_c \sim 4$  T, and this drop becomes broader

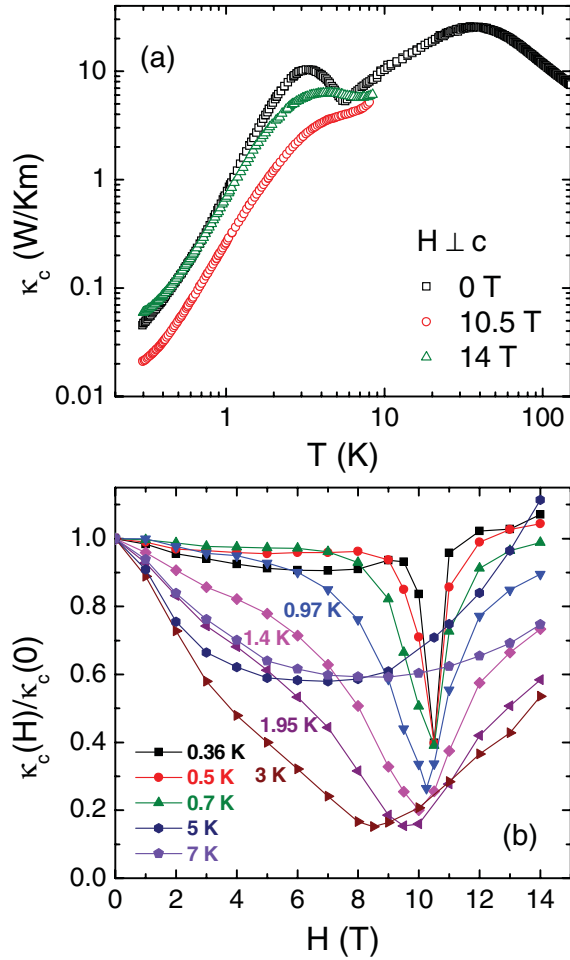


FIG. 3. (Color online)  $c$ -axis (intrachain) thermal conductivity of  $\text{BaCo}_2\text{V}_2\text{O}_8$  single crystal vs temperature (a) and magnetic field (b). The magnetic field is applied perpendicular to the  $c$  axis.

and deeper with increasing temperature. Clearly, if the dip of  $\kappa(H)$  corresponds to a magnetic transition, it is coincided with the phase transition from the Néel state to the IC state below 2 K or the one from the Néel state to the disordered state above 2 K,<sup>14</sup> respectively. Additionally, there is another dip around  $\sim 3$  T at sub-Kelvin temperatures, and the deepness of the dip is gradually reduced with increasing temperature; for example, at  $T = 0.97$  K the dip nearly disappears, as shown in Figs. 2(a) and 2(c). This kind of anomaly in a long-range AF state is most likely a consequence of the spin-flop transition for the magnetic field applied along the spin-easy axis.<sup>28,29</sup> However, as mentioned before, the spin system of BCVO was described by the XXZ model with an Ising-like anisotropy, which actually cannot allow the spin flop.<sup>10</sup> The present results therefore suggest that the spin anisotropy is not strong enough to prevent the occurrence of spin flop. As far as other experimental studies are concerned, one may note that the magnetization, which can probe the spin flop, has not yet been carried out down to sub-Kelvin temperatures.<sup>10,11</sup>

Nevertheless, the magnetic-field dependence of  $\kappa$  can be mainly ascribed to the scattering on phonons induced by the quantum fluctuations, which are strengthened with increasing field. The field-induced quantum fluctuations destroy the

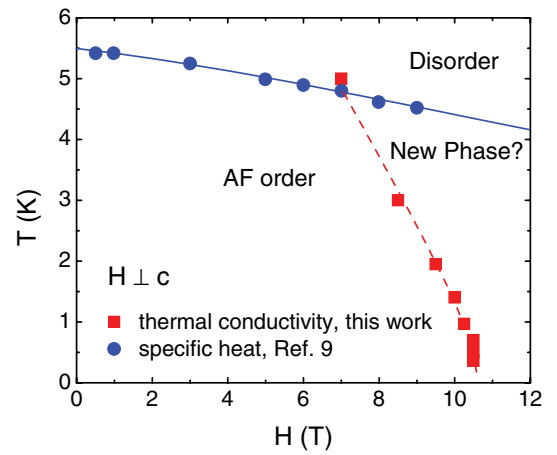


FIG. 4. (Color online) Magnetic phase diagram of  $\text{BaCo}_2\text{V}_2\text{O}_8$  for the transverse field  $H \perp c$ . The transitions of the specific heat data (Ref. 9) separate the long-range AF ordered state from the paramagnetic disordered state. The transitions of the  $\kappa_c(H)$  data indicate that there is a new phase transition inside the AF state. The lines are guides for the eyes.

long-range order but meanwhile establish the IC order. Across the phase transition, the scattering of magnetic excitations is pronounced and leads to a much more prominent feature in  $\kappa(H)$ . With further increasing field, the quantum fluctuations can even destroy the IC state and form a disordered phase. A broad-peak-like behavior of  $\kappa(H)$  in high fields and at high temperatures may result from the complicated interactions in the disordered phase.

By now, we have a relatively comprehensive understanding of the physical properties of BCVO in the longitudinal field based on many different measurements. The heat transport data are in general consistent with other results, whereas the experimental results about the effect of the transverse field are relatively rare<sup>9,11</sup> in spite of a lot of theoretical works.<sup>22–27</sup> The temperature dependencies of intrachain thermal conductivity in the transverse field are shown in Fig. 3(a). Different from the case of the longitudinal field, in which the conductivity in high field is much smaller than the zero-field value, the conductivity in the transverse field is first decreased, then followed by an enhancement with increasing field. In 14 T field, the low- $T$  thermal conductivity is nearly equal to the zero-field one, except for a weak reduction around the low- $T$  peak.

As shown in Fig. 3(b), the behavior of  $\kappa_c(H)$  in the transverse field is completely different from that in the longitudinal field. It can be seen that, at very low temperatures, the thermal conductivity is almost independent of the field except for a minimum at  $\sim 10$  T. Upon increasing temperature, the minimum becomes broader and deeper and shifts to lower field, and finally evolves into a shallow valley ranged over all the field region. First of all, although the phonon resonant scattering by paramagnetic moments could produce a similar diplike  $\kappa(H)$  behavior,<sup>33,34</sup> it is not likely the case in the present work. One clear discrepancy between the phenomenon of Fig. 3 and paramagnetic scattering is that in the latter case the minimum of  $\kappa(H)$  shifts to higher field as the temperature is increased.<sup>33,34</sup> Furthermore, the paramagnetic scattering is

closely related to the Zeeman effect and therefore should be essentially isotropic on the field direction, which is apparently different from what the BVCO data show. Therefore, it is more likely that there is some kind of magnetic transition responsible for the present results. The feature that the dip in Fig. 3(b) becomes sharper at  $T \rightarrow 0$  manifests that this transition is probably a QPT or spin-flop like. It is also notable that the transition associated with this transport behavior is different from the phase transition between the AF state and the paramagnetic state under the transverse field, which can be probed by the specific heat measurement.<sup>9</sup> In particular, the change of the AF transition temperature is not significant even if the field is increased up to 9 T, whereas the change of the minimum of  $\kappa$  is much more sizable. If we plot the AF transition points from the specific heat together with those from the heat transport in the  $H$ - $T$  phase diagram,<sup>35</sup> as shown in Fig. 4, it can be seen that they stand for two different phase boundaries. Apparently, the transitions of the  $\kappa(H)$  curves are likely associated with some field-induced magnetic transition inside the AF state.

Recently, Yamaguchi *et al.* found a similar magnetic transition using the ultrasound measurement (for  $H \perp c$ ), in which some “dip”-like anomalies appeared in the field dependence of the sound velocity at nearly the same critical fields as those in the  $\kappa(H)$  curves.<sup>36</sup> This possible phase transition demonstrated by the heat transport and ultrasound properties cannot, however, be explained by the existing theories based on the Heisenberg  $XXZ$  model. In this model, there is only one possible transition in the transverse field, that is, from the AF state to the paramagnetic state.<sup>23–26</sup> Note that the earlier high-field magnetization data were actually also difficult to understand.<sup>11</sup> On the one hand, the high-field magnetization at 1.3 K showed saturation at about 40 T, which is in good agreement with the theoretical results. On the other hand, it showed an anomaly at about

30 T, which is not expected from the theories. (In passing, the even lower- $T$  magnetization would be useful to reveal the anomaly of thermal conductivity at  $\sim 10$  T.) Therefore, these discrepancies between experiments and theories, as well as the spin-flop-like transition in the longitudinal field, demonstrate that the Heisenberg  $XXZ$  model may not be able to catch all the inherent properties of BCVO. Probably, either the finite interchain spin exchange or the deviation from the Ising anisotropy should be carefully considered in a more legitimate model to describe the spin system of BCVO.

#### IV. SUMMARY

We have studied the heat transport of  $\text{BaCo}_2\text{V}_2\text{O}_8$  single crystal, which is a quasi-1D Ising-like  $S = 1/2$  AF compound. In the longitudinal field, the nearly isotropic temperature dependencies of  $\kappa$  demonstrates the scattering of magnetic excitations on phonons. There is a distinct and sharp decrease in  $\kappa(H)$  isotherms, which is related to the transition from the Néel order to the IC state at  $H_c \sim 4$  T. Moreover, another dip of  $\kappa(H)$  at  $\sim 3$  T is supposed to result from the spin-flop transition, which has never been observed before. On the other hand, a possible magnetic transition is observed in the transverse field, which is evidenced as a minimum in the  $\kappa(H)$  curve. This novel transition is consistent with a recent ultrasound measurement, but its origin remained unclear and there is clear need for more detailed investigations.

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