Anisotropic heat transport via monopoles in the spin-ice compound $Dy_2Ti_2O_7$

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We report a study of the thermal conductivity κ of the spin-ice material $Dy_2Ti_2O_7$. From the anisotropic magnetic-field dependence of κ and by additional measurements on the phononic reference compounds $Y_2Ti_2O_7$ and $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$, we are able to separate the phononic and the magnetic contributions to the total heat transport, i.e., κ_{ph} and κ_{mag}, respectively, which both depend on the magnetic field. The field dependence of κ_{ph} arises from the noncollinear magnetic moments of the Dy ions, which tend to align along the field direction. For κ_{mag}, we observe a highly anisotropic magnetic-field dependence, which correlates with the corresponding magnetization data reflecting the different magnetic-field induced spin-ice ground states. The magnitude of κ_{mag} increases with the degree of the ground-state degeneracy. This anisotropic field dependence as well as various hysteresis effects suggest that κ_{mag} is essentially determined by the mobility of the magnetic monopole excitations in spin ice.

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I. INTRODUCTION

$Dy_2Ti_2O_7$ is a geometrically frustrated spin system with a degenerate, so-called spin-ice ground state. The magnetic Dy sites in $Dy_2Ti_2O_7$ form a pyrochlore lattice, consisting of corner-sharing tetrahedra. A strong crystal field results in an Ising anisotropy of the magnetic moments of the Dy ions, which align along their local easy axes in one of the {111} directions and point either into or out of the tetrahedra. Possible ground states in zero magnetic field are given by the "ice rule": two spins point into and two out of a tetrahedron. This behavior is analogous to the hydrogen displacement in water ice revealing a residual zero-temperature entropy.[1–5] Magnetic excitations can be created pairwise by flipping one spin resulting in two neighboring tetrahedra with configurations 1in-3out and 3in-1out, respectively. Such a dipole excitation can fractionalize into two individual monopole excitations that can freely propagate in zero magnetic field.[6–17]

Recently, we showed that these exotic magnetic excitations contribute to the heat transport in $Dy_2Ti_2O_7$.[18] The total heat transport can be decomposed into the sum of two contributions carried either by phonons or by magnetic excitations, that is,

$$\kappa(T,B) = \kappa_{ph}(T,B) + \kappa_{mag}(T,B)\,, \qquad (1)$$

and in zero field around ∼0.6 K, the magnetic contribution $\kappa_{mag}(T, B = 0)$ accounts for almost 50% of the total thermal conductivity κ. A magnetic field $\vec{B}\|[001]$ lifts the ground-state degeneracy of the magnetic system in $Dy_2Ti_2O_7$. This results in a complete suppression of κ_{mag} for rather small magnetic fields of ∼0.5 T. An alternative interpretation had been proposed earlier in Ref. 19, where the thermal conductivity was assumed to be purely phononic and the field dependence of κ was attributed to phonon scattering on magnetic excitations. Our data, however, do not support this interpretation (cf. discussion in Ref. 18). In Ref. 18, we studied κ_{mag} for a magnetic field applied parallel to [001]. By applying the magnetic field parallel to [111] or [110], one can realize more complex field-induced ground states with different degrees of degeneracy. For $\vec{B}\|[111]$, the field-induced ground state below 1 T (kagome-ice state) is, threefold degenerate consisting of different types of 2in-2out tetrahedra.[20,21] Above 1 T, the ground state changes into a nondegenerate ground state consisting of alternating 3in-1out and 1in-3out tetrahedra. For a magnetic field parallel to [110], two spins per tetrahedron are perpendicular to $\vec{B}$ and are, thus, not affected. This leads to a twofold-degenerate ground state, even for arbitrarily large magnetic fields. In this paper, we study the dependence of the magnetic heat transport on the degree of the field-induced ground-state degeneracy realized by the field directions [001], [111], and [110].

In the course of this study, we also extend the field-dependent thermal-conductivity measurements of $Dy_2Ti_2O_7$ to magnetic fields up to 7 T. An additional field dependence of κ is observed for higher fields (depending on the actual field direction), which we identify as a field-dependent phononic background $\kappa_{ph}(B)$ of $Dy_2Ti_2O_7$. The phononic background is essentially given by the field-dependent $\kappa(B)$ of the Y-doped $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$. This compound can be regarded as a magnetic reference system with strongly suppressed spin-ice features. Compared to the nonmagnetic $Y_2Ti_2O_7$, the half-doped reference compound can be utilized to study the magnetic-field dependence of the phononic contribution κ_{ph} of $Dy_2Ti_2O_7$. The field dependence of κ_{ph} can be explained by a magnetostrictive lattice distortion originating from small tilts of the noncollinear Dy momenta induced by the external magnetic field.

II. EXPERIMENTAL

Single crystals were grown from sintered bars of TiO_2 (3N, Sigma-Aldrich) and Y_2O_3 (4N, Alfa Aesar) and/or Dy_2O_3 (4N, REacton) in proper stoichiometry. The sintering processes were done in air at 1400 °C over night using corundum boats as crucibles. For the crystal growth, a floating-zone technique was applied inside a four-mirror image furnace to acquire centimeter-sized single crystals. To obtain crystals without cracks, the floating zone was run twice trough the bars: first at a rate of 25 mm/h and subsequently with 7 mm/h. Ambient pressure of pure oxygen was chosen as atmosphere during the growth. The pale yellow crystals exhibit high reflectance at a grazing angle but otherwise full transparency.

$Dy_2Ti_2O_7$ is slightly more yellow than $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ and $Y_2Ti_2O_7$. The resulting crystals were found to be single domain by extensive Laue-photo investigations and the bulk purity was checked by x-ray powder diffraction using a STOE D5000 diffractometer, Cu K_α radiation, and Bragg-Brentano reflection mode. The correct composition was confirmed by energy-dispersive spectroscopy inside a scanning electron microscope.

The thermal conductivity was measured using the standard steady-state method. The temperature difference was produced by a heater attached at one end of the sample and measured by a pair of matched RuO_2 thermometers. The thermal-conductivity measurements were performed on bar-shaped single crystals of approximately $3 \times 1 \times 1$ mm^3 with the long edge parallel to the $[1\bar{1}0]$ direction. The heat current was directed along the long edge of the crystal. In most cases, the magnetic fields applied along the different directions were oriented perpendicular to this long edge. Hence considerable demagnetization effects have to be taken into account. The demagnetization field is calculated on the basis of experimental magnetization data, which were measured with a home-built Faraday magnetometer on thin samples to minimize demagnetization effects within the magnetization measurements. The magnetostriction measurements were done with a home-built capacitance dilatometer, which allows to measure the uniaxial length changes either parallel or perpendicular to the applied magnetic field.

III. RESULTS

Figure 1 shows the relative change of the thermal conductivity $\kappa(B)/\kappa(0\,\mathrm{T})$ of $Dy_2Ti_2O_7$ for $\vec{B}\|[111]$ at temperatures between 0.35 and 1 K. The different data sets are shifted with respect to each other. With increasing field, $\kappa(B)$ decreases and exhibits a plateau within the kagome-ice phase below 1 T. For higher fields, $\kappa(B)$ further decreases and shows a kink around 1.5 T. The plateau feature is more pronounced at lowest temperature and broadens towards higher temperature. The inset of Fig. 1 displays the magnetization $M(B)$ of $Dy_2Ti_2O_7$ for $\vec{B}\|[111]$ at 0.4 K, which agrees within a few percent with previous studies, e.g., Refs. 4, 21, and 22. The magnetization as well as the thermal conductivity have been measured with slow sweep rates[23] to avoid thermal-runaway effects, cf. Ref. 11. The basic features of the $\kappa(B)$ data are reflected within the magnetization data, which also exhibit a pronounced plateau within the kagome-ice phase. At 1 T, $M(B)$ shows a sharp kink and increases towards the saturation value, which is essentially reached at ~1.5 T. This increase of $M(B)$ is accompanied by a decrease of κ between 1 and 1.5 T. The hysteresis in $M(B)$ at low temperature (~0.4 K) below ~0.3 T can also be observed in the thermal-conductivity data. Even the remnant zero-field magnetization observed in the measurement with decreasing field (open symbols), which results from slow relaxation processes of the spin ice, is reflected within the thermal-conductivity data. Below ~0.4 K, the κ data for decreasing field end in a reduced zero-field value, which slowly relaxes back to the initial zero-field value (after zero-field cooling, not shown). Such a correlation of magnetization and thermal conductivity has already been observed for $\vec{B}\|[001]$ (see Ref. 18). For $\vec{B}\|[111]$, however,

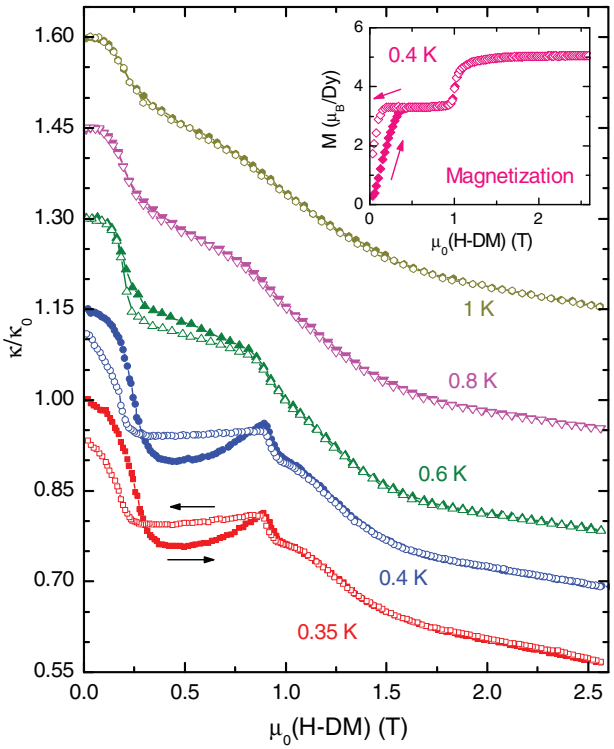

FIG. 1. (Color online) Relative change κ/κ_0 of $Dy_2Ti_2O_7$ for $\vec{B}\|[111]$ at various constant temperatures. The different curves are shifted by 0.15 with respect to each other. (Inset) Magnetization $M(B)$ of $Dy_2Ti_2O_7$ at 0.4 K. The arrows indicate the field-sweep direction. Demagnetization effects are taken into account.

clear differences between $M(B)$ and $\kappa(B)$ are also found. First of all, at lowest temperature, $\kappa(B)$ within the kagome-ice phase strongly depends on whether the measurement is performed with increasing field (after cooling in zero field) or with decreasing field (starting from high magnetic fields). Such a hysteresis is not present in the magnetization data. Even at lowest temperature, the magnetization within the kagome-ice phase shows no hysteresis with respect to the field-sweep direction. Secondly, $M(B)$ and $\kappa(B)$ show different high-field behaviors. At 0.4 K, the magnetization is almost saturated above 1.5 T, whereas the thermal conductivity $\kappa(B)$ further decreases with increasing field even above 1.5 T. The question arises whether these effects can be attributed to the phononic or to the magnetic contribution of κ.

In order to analyze this, we will first discuss the high-field dependence of κ for magnetic fields above ~1.5 T. Because the magnetization is essentially saturated above 1.5 T, the magnetic system in $Dy_2Ti_2O_7$ is fully polarized. Hence one would expect the field dependence of κ above 1.5 T to originate from a change of κ_{ph} rather than from κ_{mag}. By means of the data shown in Fig. 1, however, one cannot straightforwardly separate κ_{mag} and κ_{ph}. In Ref. 18, we showed that κ_{mag} vanishes for a rather small field of ~0.5 T applied parallel to [001]. Compared to $\vec{B}\|[111]$, the field-induced ground state for $\vec{B}\|[001]$ is less complex, as the ground-state degeneracy is lifted in one single step. Therefore, we start with the analysis of the high-field thermal conductivity of $Dy_2Ti_2O_7$ for $\vec{B}\|[001]$. The data published in Ref. 18 were only measured up to a

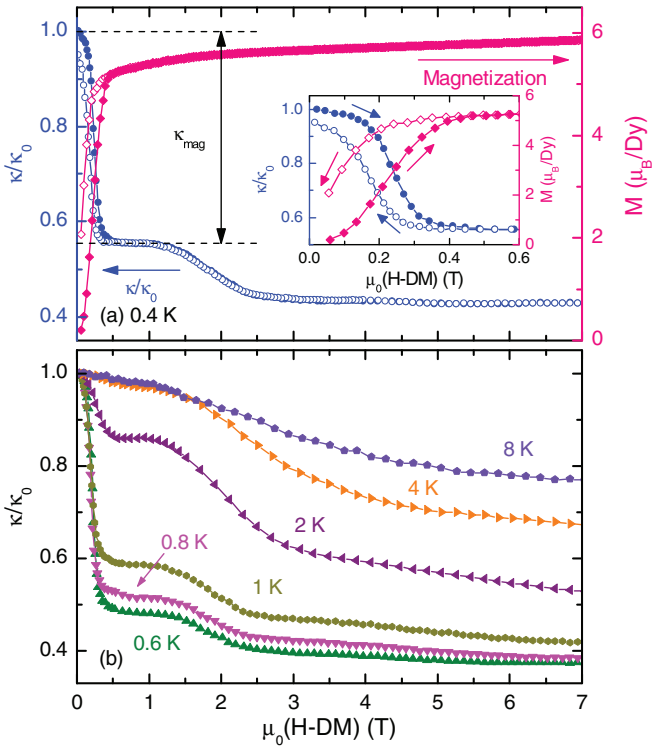

FIG. 2. (Color online) Relative change κ/κ_0 of $Dy_2Ti_2O_7$ for $\vec{B}\|[001]$ at various constant temperatures between 0.4 and 8 K. Demagnetization effects are taken into account. The field dependencies of magnetization and thermal conductivity at 0.4 K are compared in (a). The complete suppression of κ_{mag} below 1 T is tagged by the dashed lines. The inset is an enlargement of the low-field region, where the field-sweep directions are marked by arrows.

(demagnetization-corrected) maximum field of 0.5 T. Here, we extend the present κ data to magnetic fields up to 7 T. The results are displayed in Fig. 2, where the relative change $\kappa(B)/\kappa(0\,\mathrm{T})$ is shown for temperatures between 0.4 K and 8 K. Below 0.5 T, the data shown in Fig. 2 confirm the data previously published in Ref. 18. Figure 2(a) directly compares the field-induced change of κ with the magnetization $M(B)$ at the lowest measured temperature of 0.4 K. Below 0.5 T, $\kappa(B)$ shows a steplike decrease. As can be seen in the inset of panel (a), the sharp drop of $\kappa(B)$ below 0.5 T clearly correlates with $M(B)$ and can, thus, be attributed to κ_{mag}. Above ~1.5 T, $\kappa(B)$ further decreases. This additional field dependence $\kappa(B)$, however, cannot be attributed to κ_{mag} as all spins are fully polarized. This is shown exemplarily for 0.4 K, where the magnetization is essentially saturated above 1.5 T (apart from a slight linear increase, see below). As the excitation gap for flipping the magnetic moments monotonically increases with an increasing field $\vec{B}\|[001]$, the field-dependent decrease of κ also cannot be explained by such type of phonon scattering on magnetic excitations.

Figure 2(b) shows the field-induced relative change of $\kappa(B)$ at higher temperatures up to 8 K. Above ~4 K, κ_{mag} vanishes, whereas a distinct high-field dependence of κ above 1 T is still observed even at 8 K. As will be shown below, this high-field dependence of the thermal conductivity of $Dy_2Ti_2O_7$ can be attributed to a magnetic-field dependent phononic contribution $\kappa_{ph}(B)$.

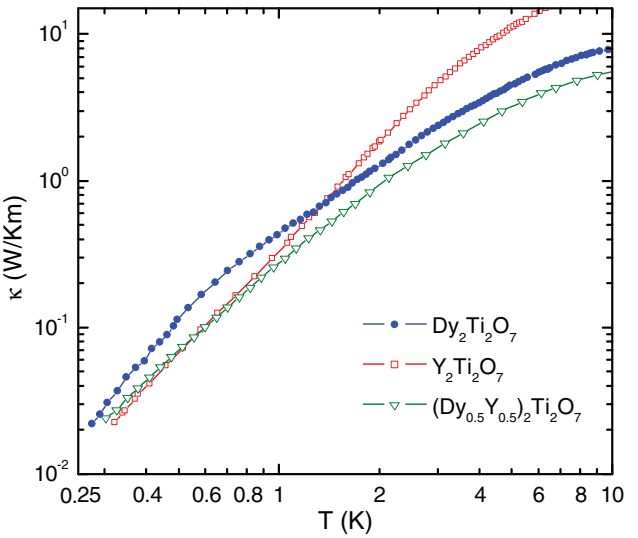

FIG. 3. (Color online) Comparison of the zero-field thermal conductivity of $Dy_2Ti_2O_7$ (closed blue circles) with the nonmagnetic reference $Y_2Ti_2O_7$ (open red squares) and the half-doped $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ (open green triangles).

In our previous work,[18] we studied the nonmagnetic $Y_2Ti_2O_7$ as a purely phononic reference compound to the magnetic spin ice $Dy_2Ti_2O_7$. At temperatures below ~3 K, the thermal conductivity of $Y_2Ti_2O_7$ shows a very similar power-law behavior as the phononic background of $Dy_2Ti_2O_7$, which was obtained by applying a magnetic field of 0.5 T parallel to [001]. For higher temperatures, however, the thermal conductivity of $Dy_2Ti_2O_7$ is significantly smaller (see Fig. 3). Most likely, this suppression originates from additional phonon scattering on the crystal-field excitations of the $4f$ electrons of the Dy ions. Here, we present an alternative approach in order to study the phonon background of κ by using the half-doped compound $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$. As every second Dy ion is replaced by a nonmagnetic Y ion, i.e., on average two of the four Dy ions per tetrahedron are replaced by Y ions, the spin-ice properties are supposed to be strongly suppressed. However, due to the similar ionic radii of Dy^{3+} and Y^{3+}, one can assume both compounds to have comparable phononic properties. Figure 3 shows the zero-field thermal conductivity of the spin-ice compound $Dy_2Ti_2O_7$ together with the thermal conductivity of the nonmagnetic reference $Y_2Ti_2O_7$ and the half-doped $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$. In all measurements, the heat current has been directed along the $[1\bar{1}0]$ direction. Below 1 K, both phononic reference compounds have very similar κ values well below κ of $Dy_2Ti_2O_7$. At higher temperature, the half-doped compound shows a behavior similar to the spin-ice compound, rather than to the nonmagnetic $Y_2Ti_2O_7$, which has significantly larger κ values. This supports our interpretation of an additional phonon scattering on the crystal-field excitations of the Dy ions, as given above. This additional scattering, indeed, can also be observed for the half-doped $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$, leading to κ values close to the values of $Dy_2Ti_2O_7$. The slightly smaller values of the half-doped compound can be explained by an enhanced defect scattering of phonons which originates from the partial Dy substitution.

IV. DISCUSSION

A. Phononic heat conductivity $\kappa_{ph}(B)$

The main advantage of the half-doped compound $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$, compared to $Y_2Ti_2O_7$, is the fact that it is still strongly magnetic and can, therefore, be utilized to study the magnetic-field dependence of the phononic background $\kappa_{ph}(B)$ of $Dy_2Ti_2O_7$. As shown exemplarily for $\vec{B}\|[111]$ in Fig. 4(d), κ of $Y_2Ti_2O_7$ (red squares) has no magnetic-field dependence, as expected for a nonmagnetic compound. Figures 4(a)–4(d) compare the thermal conductivity $\kappa(B)$ of $Dy_2Ti_2O_7$ (blue circles) and of $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ (green triangles) for various magnetic-field directions, where the heat current $\vec{j}$ is always directed parallel to $[1\bar{1}0]$. First of all, even at lowest temperature, $\kappa(B)$ of $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ is not hysteretic with respect to the field-sweep direction (not shown). This is contrary to the undoped $Dy_2Ti_2O_7$, where at lowest temperature ($\sim$0.4 K), a clear hysteresis in $\kappa(B)$ is observed below $\sim$0.3 T for all considered field directions, see, e.g., Figs. 1 and 5.

The data in Figs. 4(a) and 4(b) are measured with a magnetic field parallel to [001]. At 2 K [see Fig. 4(a)], $\kappa(B)$ of $Dy_2Ti_2O_7$ shows two steplike anomalies at $\sim$0.5 and $\sim$2 T and continuously decreases above the second anomaly. The step at $\sim$2 T and the subsequent continuous decrease are very well reflected by the $\kappa(B)$ data of $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$. The first anomaly (at $\sim$0.5 T), which we attribute to the suppression of κ_{mag} in $Dy_2Ti_2O_7$, is, however, absent in the $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ data. Figure 4(b) shows the data for the same field direction $\vec{B}\|[001]$ at a lower temperature of 0.4 K. Here again, the high-field behavior is very similar for both compounds, whereas the sharp decrease below $\sim$0.5 T is absent in the $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ data. The inset of Fig. 4(b) illustrates the different origins of the field dependencies of the half-doped compared to the undoped compound. One can very clearly see that for $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$, the change of $\kappa(B)$ does not correlate with the magnetization $M(B)$, which is essentially saturated at $\sim$1 T, whereas $\kappa(B)$ shows a distinct field dependence up to $\sim$3 T. This is different from the case of $Dy_2Ti_2O_7$, where the change of $M(B)$ at low field (below $\sim$0.5 T) directly correlates with the suppression of κ_{mag} [inset of Fig. 2(a)]. Thus we conclude that the high-field decrease of κ of $Dy_2Ti_2O_7$ can be identified as a field-dependent phononic background $\kappa_{ph}(B)$.

It is notable, however, that the zero-field κ values of $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ at lowest temperature are larger than the phononic background of $Dy_2Ti_2O_7$ obtained from the plateau value around 1 T [see Fig. 4(b)]. Most likely, this originates from a small remnant magnetic contribution κ_{mag} in $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$, which is more pronounced at lower temperature (0.4 K) than at higher temperature [2 K, Fig. 4(a)]. Hence one cannot directly identify the low-field (below $\sim$0.5 T) κ values of the half-doped compound as the phononic background κ_{ph} of the undoped mother compound. Nevertheless, the remnant magnetic contribution in $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ is strongly suppressed compared to κ_{mag} in $Dy_2Ti_2O_7$, as expected for half-doping.

In Fig. 4(c), the field dependence of κ is shown for a magnetic field parallel to [110]. For this particular field

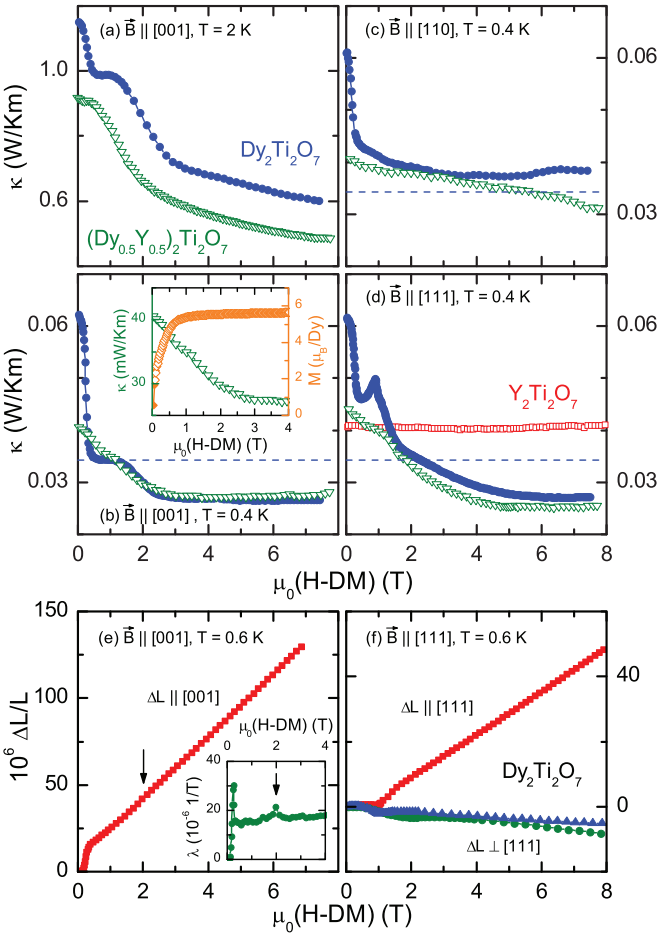

FIG. 4. (Color online) (a)–(d) Field-dependent $\kappa(B)$ of $Dy_2Ti_2O_7$ (blue circles) and of the half-doped phononic reference compound $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ (green triangles) for different magnetic-field directions; (d) also contains $\kappa(B)$ of $Y_2Ti_2O_7$. The inset in (b) compares $\kappa(B)$ with $M(B)$ for $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$. The dashed lines mark the low field $\kappa_{ph}(B < 1.5\,\mathrm{T}, T = 0.4\,\mathrm{K})$ estimated by the plateau in $\kappa(B)$ for $\vec{B}\|[001]$. (e) and (f) Field-induced length changes of $Dy_2Ti_2O_7$ for $\vec{B}\|[001]$ and $\vec{B}\|[111]$, respectively. The inset in (e) shows the corresponding field derivative of $\Delta L/L$, where the anomaly at 2 T (arrows) is better seen. In all cases, demagnetization effects are taken into account and, for clarity, only data obtained with increasing magnetic field are shown.

direction, two of the four spins per tetrahedron in $Dy_2Ti_2O_7$ are perpendicular to $\vec{B}$ and are, thus, not affected. This results in a twofold-degenerate ground state for this field direction[24] and we find that the decline of $\kappa(B)$ below 0.5 T is less pronounced as compared to $\vec{B}\|[001]$ [dashed line, cf. panel (b)]. This indicates that κ_{mag} cannot be completely suppressed by a field applied parallel to [110]. Furthermore, above $\sim$0.5 T, κ is hardly field dependent. This is different from the other considered field directions, where $\kappa(B)$ shows a distinct high-field dependence of κ. However, here also, the thermal conductivity of the half-doped reference compound closely resembles the high-field data of $Dy_2Ti_2O_7$ at least up to 4 T, whereas there are some deviations appearing at higher field. The data in Fig. 4(c) might suggest that the zero-field κ value of $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ essentially reflects the phononic background for $Dy_2Ti_2O_7$.

This is, however, misleading as both compounds exhibit a nonvanishing zero-field magnetic contribution to κ (as shown above for $\vec{B}\|[001]$) and, obviously, the zero-field κ is independent on the magnetic-field direction. Consequently, the field-induced twofold-degenerate ground state realized by $\vec{B}\|[110]$ leads to a considerable magnetic contribution $\kappa_{\rm mag}$, which is hardly field dependent up to highest fields of 7 T.

More complex ground states are realized by a magnetic field applied parallel to [111]. Within the kagome-ice phase below 1 T, the ground state is threefold degenerate. Above the transition at 1 T, the magnetic system changes to a nondegenerate, fully polarized ground state. The highly degenerate kagome-ice state results in a pronounced plateau within the field-dependent change of κ observed for $Dy_2Ti_2O_7$ at lowest temperature (see Fig. 1). The high-field dependence of κ for $\vec{B}\|[111]$ is shown in Fig. 4(d) together with the field-dependent data for the half-doped $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$ and the nonmagnetic $Y_2Ti_2O_7$. As mentioned above, $Y_2Ti_2O_7$ can only be used to estimate the zero-field phononic contribution, as κ of $Y_2Ti_2O_7$ shows no field dependence. In complete analogy to the field directions discussed above, the field dependence of κ above $\sim$2 T is very well described by κ of the half-doped reference compound, whereas the spin-ice features at lower field are almost completely suppressed. This, again, confirms our interpretation of $\kappa(B)$ of the half-doped reference compound as the field-dependent phononic background of $Dy_2Ti_2O_7$. However, it is more difficult to distinguish between the magnetic and the phononic contributions of $Dy_2Ti_2O_7$ for $\vec{B}\|[111]$. The field dependence well above $\sim$1.5 T can certainly be attributed to $\kappa_{\rm ph}$, but around 1.5 T, $\kappa_{\rm ph}(B)$ and the suppression of $\kappa_{\rm mag}$ due to the lifting of the ground-state degeneracy above the kagome-ice phase overlap. Consequently, the suppression of $\kappa_{\rm mag}$ appears as a kink in $\kappa(B)$ for $\vec{B}\|[111]$, rather than a pronounced plateau, as observed for $\vec{B}\|[001]$. The kink is located close to the zero-field phononic background obtained from the $\vec{B}\|[001]$ data [horizontal dashed line, cf. Fig. 4(b)]. This suggests that the field dependence $\kappa_{\rm ph}(B)$ below $\sim$1.5 T is much smaller than the suppression of $\kappa_{\rm mag}$. Hence, when we restrict the discussion to the low-field region below 1.5 T, we can assume a constant, i.e., field-independent, phononic background $\kappa_{\rm ph}$ of $Dy_2Ti_2O_7$. The magnetic contributions $\kappa_{\rm mag}$ extracted on the basis of this assumption are discussed in the following subsection.

A possible explanation of the field dependence of the phononic background $\kappa_{\rm ph}$ of $Dy_2Ti_2O_7$ arises from the fact that the different local easy axes are not collinear. Thus, for any field direction, at least three of the four local easy axes are not parallel to the magnetic field. A magnetic field that is not parallel to the easy axis partially mixes the higher-lying crystal-field states into the respective ground-state doublet. Moreover, such a field results in finite torques $\vec{\mu}\times\vec{B}$ of the Dy momenta $\vec{\mu}$, which tend to align the easy axis along the field direction. The combination of both effects can easily explain the weak, essentially linear high-field increase of the magnetization observed above about 1.5 T [cf. Fig. 2(a) and Ref. 25]. For example, a tilt of the Dy momenta of only $\sim$1.5° is already sufficient to induce the additional increase of $M(B)$ between 2 and 7 T. A further consequence of the noncollinear easy axes are field-induced lattice distortions, which can be visualized by magnetostriction measurements. Figures 4(e) and 4(f) show the field-induced length changes of $Dy_2Ti_2O_7$ for $\vec{B}\|[001]$ and $\vec{B}\|[111]$, respectively. In both cases, the $Dy_2Ti_2O_7$ crystals elongate parallel to the magnetic-field direction (red squares), where the effect is more pronounced for $\vec{B}\|[001]$. In the high-field region, i.e., for magnetic fields above the (almost) saturation of $M(B)$, this elongation is essentially linear in B and temperature independent in the temperature range between 0.25 and 2 K (not shown). Thus, the high-field magnetostriction cannot be attributed to the spin-ice system in $Dy_2Ti_2O_7$. For $\vec{B}\|[111]$ [see Fig. 4(f)], we also measured the field-induced length changes perpendicular to $\vec{B}$, namely, parallel to $[1\bar{1}0]$ and $[11\bar{2}]$, and we find that the crystal contracts in both perpendicular directions, but to a much smaller extent as compared to the elongation parallel to $\vec{B}\|[111]$. As the magnetic field causes a strong distortion of the lattice, it influences the phononic properties, in particular, the phononic thermal conductivity. The steplike anomaly observed in $\kappa(B)$ for $\vec{B}\|[001]$ around 2 T [see Figs. 4(a) and 4(b)] has, indeed, an equivalent within the magnetostriction data, which show a small anomaly at the same magnetic field. In Fig. 4(e), an arrow marks this anomaly, which is better seen in the field derivative of the length change [inset of Fig. 4(e)]. The origin of this anomaly is, however, unclear. In sum, the field dependence of the thermal conductivity of $Dy_2Ti_2O_7$ observed in the high-field region [see Figs. 4(a)–4(d)] can be attributed to a field-dependent phononic contribution $\kappa_{\rm ph}(B)$, which is not related to the spin-ice behavior.

B. Magnetic heat conductivity $\kappa_{\rm mag}(B)$

Now we proceed with the discussion of the magnetic contribution $\kappa_{\rm mag}$ of $Dy_2Ti_2O_7$. In the previous paragraphs, we showed that $\kappa_{\rm mag}$ can be suppressed by a rather small magnetic field, depending on the actual field direction. As $\kappa_{\rm ph}$ is only weakly field dependent for small magnetic fields, we can extract $\kappa_{\rm mag}$ by assuming a constant, i.e., field-independent, phononic background [dashed line, cf. Fig. 4(b)]. The field-dependent $\kappa_{\rm mag}$ for the different field directions are displayed in Figs. 5(a) and 5(b) at 1 and 0.4 K, respectively. For clarity, we only show the data measured with decreasing magnetic field, which are (for $B>0$) closer to thermal equilibrium than the data obtained with increasing field (see discussion below). The magnetic contribution is maximum in zero field (sixfold-degenerate ground state) and completely vanishes for $\vec{B}\|[001]$ (nondegenerate). For $\vec{B}\|[110]$ (twofold degenerate),[24] $\kappa_{\rm mag}$ cannot be completely suppressed, even for a rather large field of $\sim$7 T. For this particular field direction, the crystallographic directions [110] and $[1\bar{1}0]$ become inequivalent. The spins with a nonvanishing component parallel to $\vec{B}$ become fully polarized and form so-called α chains running parallel to $\vec{B}$. The remaining spins are perpendicular to $\vec{B}$ and form β chains that are oriented perpendicular to $\vec{B}$, i.e., $\alpha\|[110]$ and $\beta\|[1\bar{1}0]$, cf. Refs. 19, 26, and 27. For $\vec{B}\|[110]$, we measured $\kappa_{\rm mag}$ within two different measurement setups, either with the heat-current direction along the α chains, i.e., $\vec{j}\|\vec{B}$, or with the heat current along the β chains, i.e., $\vec{j}\perp\vec{B}$. At 1 K, both data

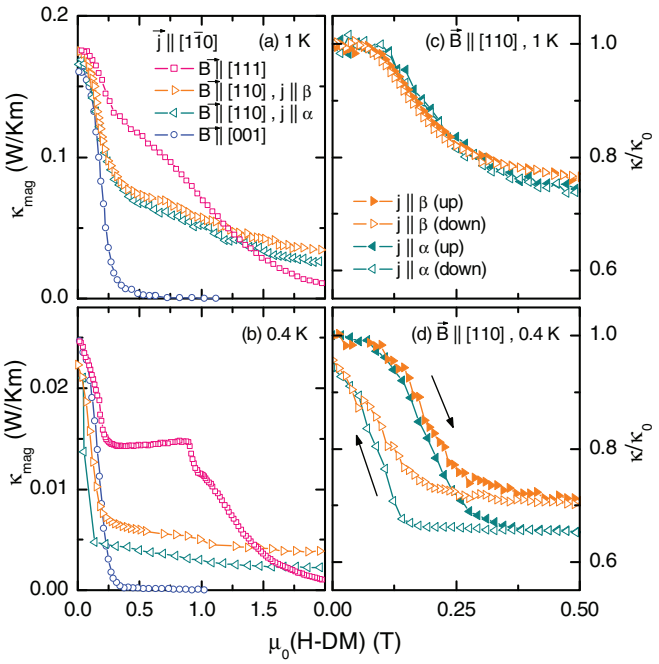

FIG. 5. (Color online) (a) and (b) Field-dependence of $\kappa_{\rm mag}$ of $Dy_2Ti_2O_7$ for different magnetic-field directions at 1 and 0.4 K, respectively. For clarity, only the data measured with decreasing field are shown. (c) and (d) Field-induced relative change $\kappa(B)/\kappa(0\,\mathrm{T})$ for $\vec{B}\|[110]$ and different directions of the heat current $\vec{j}$ at 1 and 0.4 K, respectively. The arrows indicate the different field-sweep directions. Demagnetization effects are taken into account.

sets agree with each other [see Fig. 5(a)], whereas at 0.4 K, $\kappa_{\rm mag}$ is significantly larger along the β chains [see Fig. 5(b)]. A direct comparison of the field-induced relative changes of κ for $\vec{j}\|\alpha$ and $\vec{j}\|\beta$ at 1 and 0.4 K is displayed in Figs. 5(c) and 5(d), respectively. We note that the data of Figs. 5(c) and 5(d) have been obtained in a different measurement run on another crystal as compared to the data displayed in panels (a) and (b). The low-temperature (0.4 K) data for both heat-current directions show a pronounced hysteresis with respect to the field-sweep direction and the $\kappa(B\to 0)$ data measured with decreasing field end in a reduced zero-field κ value, as it is also observed for $\vec{B}\|[001]$ and $\vec{B}\|[111]$. Moreover, the low-temperature data for $\vec{B}\|[110]$ clearly reveal that $\kappa_{\rm mag}$ is larger for a heat current along the β chains than along the α chains. Such an anisotropy of $\kappa_{\rm mag}$ with respect to $\vec{j}$ can be qualitatively explained within the microscopic model of monopole excitations propagating via single spin flips. The spins of the α chains are easily polarized by $\vec{B}\|[110]$, whereas the spins of the β chains are perpendicular to $\vec{B}$ and are, thus, not affected. As a consequence, the monopole propagation along the α chains should be suppressed compared to the β chains. Within the model of single spin flips, one would even expect that monopole excitations can only propagate along the β chains, which are separated by (fully polarized) α chains, whereas $\kappa_{\rm mag}$ should completely vanish for $\vec{j}\|\alpha$. A complete suppression of $\kappa_{\rm mag}$ along the α chains is, however, not observed [see Figs. 5(a) and 5(b)]. This shows that the single spin-flip formalism, which only accounts for nearest-neighbor interaction, is an oversimplification and the long-range dipolar interaction has to be taken into account.

For a magnetic field parallel to [111] within the kagome-ice phase below 1 T (threefold degenerate), $\kappa_{\rm mag}$ is significantly larger than for $\vec{B}\|[110]$ [see Figs. 5(a) and 5(b)]. Above 1 T, the ground-state degeneracy is lifted, resulting in a suppression of $\kappa_{\rm mag}$, which essentially vanishes above $\sim$1.5 T (indicated by a kink). As mentioned above, the field dependence of $\kappa_{\rm ph}$ cannot be neglected above $\sim$1.5 T. Hence the assumption of a field-independent phononic background is no longer justified, and $\kappa_{\rm mag}$ extracted here does not exactly approach zero. The hysteretic behavior below 1 T, however, can certainly be attributed to $\kappa_{\rm mag}$ because no hysteresis of $\kappa_{\rm ph}(B)$ is observed for the reference compound $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$. At the lowest temperature (below $\sim$0.4 K), $\kappa_{\rm mag}$ within the kagome-ice phase (below 1 T) depends on whether the measurement is performed with increasing field after zero-field cooling or with decreasing field starting from the fully polarized phase (above 1 T) (cf. Fig. 1). The hysteresis in $\kappa(B)$ below $\sim$0.3 T and the fact that the measurement with decreasing field does not end at the initial zero-field-cooled value is also present for $\vec{B}\|[110]$ [see Fig. 5(d)] and for $\vec{B}\|[001]$ (cf. Ref. 18), and there are corresponding hysteresis effects in the respective magnetization data (inset of Fig. 1).

The hysteresis within the kagome-ice phase, however, has no equivalent within the magnetization data. The absence of such a hysteresis in $M(B)$ can be explained by the single-tetrahedron model. The threefold-degenerate ground state consists of three different, energetically equivalent tetrahedron configurations. As these configurations have the same magnetization component parallel to $\vec{B}\|[111]$, they are indistinguishable for this particular field direction. The thermal conductivity, however, seems to be sensitive to the actually realized ground-state configuration. Above 1 T, the spin ice is fully polarized, and the thermal conductivity does not depend on the actual field-sweep direction. When entering the kagome-ice phase from higher field, the spins of the triangular planes, that separate the kagome planes, stay polarized, whereas the spins of the kagome planes arrange in such a way that the tetrahedra obey the ice rule 2in-2out. As this configuration can easily be achieved by flipping only one spin per tetrahedron, it can be assumed that this configuration is close to thermal equilibrium. As a consequence, $\kappa_{\rm mag}$ stays almost constant on the kagome-ice plateau. The situation is more complicated when approaching the kagome-ice phase from below, i.e., from the highly entropic ground state after zero-field cooling. The magnetic field $\vec{B}\|[111]$ initially polarizes the spins of the triangular planes. Flipping such a spin, however, produces a monopole/antimonopole pair in two neighboring kagome planes. To reach thermal equilibrium, these monopoles (and antimonopoles) have to be annihilated. This out-of-equilibrium state results in a reduced $\kappa_{\rm mag}$, and time-dependent measurements of $\kappa_{\rm mag}$ (not shown) reveal that it very slowly converges towards the larger equilibrium value. When approaching the transition at 1 T, the energy gap to the fully polarized state vanishes and, as a consequence, the kagome planes can easily reach thermal equilibrium. This results in an increase of $\kappa(B)$ towards 1 T. An interesting finding is the fact that in the field-decreasing run, i.e., in (or

at least close to) thermal equilibrium, $\kappa_{\rm mag}$ remains constant on the kagome-ice plateau although the energy gap to the (anti)monopole excitations increases with decreasing field.

In sum, our data for the different magnetic-field directions reveal that there is a magnetic contribution $\kappa_{\rm mag}$, whose magnitude correlates with the degree of the ground-state degeneracy of the magnetic spin ice.[24,28] At least qualitatively, we have evidence that $\kappa_{\rm mag}$ arises from the monopole excitations and that its magnitude mainly depends on the monopole mobility which is related to the degree of the ground-state degeneracy. However, on a more quantitative level, there are many open questions. Hence, a more fundamental theory is required.

V. CONCLUSION

In conclusion, we find that the low-temperature thermal conductivity of the spin-ice compound $Dy_2Ti_2O_7$ has a pronounced magnetic contribution $\kappa_{\rm mag}$ in zero magnetic field. This $\kappa_{\rm mag}$ has a strong magnetic-field dependence, which is highly anisotropic with respect to the magnetic-field direction. The magnitude of $\kappa_{\rm mag}$ correlates with the degree of the ground-state degeneracy. It is maximum in zero field, where the ground-state degeneracy is maximum (sixfold). Applying finite magnetic fields along different directions stabilize other (spin-ice) ground states with reduced degrees of degeneracy depending on the direction and magnitude of the magnetic field. For $\vec{B}\|[111]$, a threefold-degenerate kagome-ice phase occurs below 1 T, where $\kappa_{\rm mag}$ is reduced compared to the zero-field value, but remains larger than $\kappa_{\rm mag}$ in the twofold-degenerate ground state which is realized for $\vec{B}\|[110]$.[24,28] Nondegenerate ground states are reached either for $\vec{B}\|[001]$ or for $\vec{B}\|[111]$ above 1 T and in both cases, $\kappa_{\rm mag}$ vanishes completely. For $\vec{B}\|[110]$, we observe an additional anisotropy of $\kappa_{\rm mag}$ with respect to the direction of the heat current $\vec{j}$. For a heat current directed along the α chains, whose spins are easily polarized because of their finite components along the field direction, $\kappa_{\rm mag}$ is suppressed more strongly as compared to a heat current $\vec{j}$ directed along the perpendicular oriented β chains, whose spins are $\perp \vec{B}$ and are thus unaffected by the magnetic field. The anisotropy of $\kappa_{\rm mag}$ with respect to the direction of $\vec{j}$ is, however, too weak to be explained within a single spin-flip formalism that only accounts for nearest-neighbor interaction.

Independent of the direction of the magnetic field, there are pronounced hysteresis effects when the magnetic field is reduced from larger values towards zero field in the low-temperature region. In all these cases, $\kappa(\vec{B} \to 0)$ approaches reduced values compared to κ_0 obtained in the zero-field cooling process, and as a function of time, the reduced $\kappa(\vec{B} \to 0)$ value slowly relaxes towards κ_0. This low-field hysteresis of $\kappa(\vec{B} \to 0)$ correlates with the occurrence of a small, but finite remnant magnetization observed for all studied field directions at low temperature. For $\vec{B}\|[111]$, however, we also observe a hysteresis of $\kappa_{\rm mag}$ that has no analog in the magnetization data. Within the kagome-ice phase, $\kappa_{\rm mag}$ strongly depends on whether this phase is entered from zero or from high magnetic fields. Increasing the magnetic field from the degenerate zero-field ground state results in a $\kappa_{\rm mag}$ of the kagome-ice phase that is strongly reduced compared to the value of $\kappa_{\rm mag}$ which is obtained when the kagome-ice phase is entered from above, i.e., from the fully polarized state starting from higher fields. This larger $\kappa_{\rm mag}$ is practically field-independent within the kagome-ice phase and it neither depends on time, whereas the lower $\kappa_{\rm mag}$ slowly relaxes towards the larger plateau value.

Furthermore, we also identify a considerable field dependence of the phononic contribution $\kappa_{\rm ph}$ which emerges at higher field, i.e., above the (almost) saturation of the magnetization. Essentially, the same field-dependent $\kappa_{\rm ph}(B)$ is observed in the half-doped reference compound $(Dy_{0.5}Y_{0.5})_2Ti_2O_7$, where the spin-ice features are almost completely suppressed and all the above-described low-field features of $\kappa_{\rm mag}$ are essentially absent. The field dependence of $\kappa_{\rm ph}$ arises from the noncollinear Dy momenta, which tend to align along the field direction, and the resulting small tilts of the Dy momenta can also explain the continuous increase of the high-field magnetization.

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