Phononic thermal Hall effect in diluted terbium oxides

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We observe thermal Hall conductivity for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$, which is even larger than the previously observed thermal Hall conductivity for $Tb_2Ti_2O_7$ [Science **348**, 106 (2015)]. The robustness against the dilution of the Tb magnetic moment indicates the Hall effect of phonons is the origin of thermal Hall conductivity in Tb pyrochlore oxide. The temperature and magnetic field dependences are observed to follow the empirical relation that the Hall angle is proportional to the magnetization. We also investigate thermal Hall conductivity for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$. We observe barely finite thermal conductivity at low temperatures. This confirms the thermal Hall conductivity in Tb-garnet oxide. The origins of the empirical relation in the pyrochlore system and the magnitude difference between the two systems are discussed comparatively with previously proposed theories of the phonon Hall effect.

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I. INTRODUCTION

The thermal Hall effect is the induction of thermal current perpendicular to both the magnetic field and the thermal gradient. In dielectrics, charge-neutral excitations, such as phonons and magnetic excitations, carry the heat current. The Lorentz force, which is the origin of the ordinary Hall effect, does not act on these charge-neutral particles. However, similarly to the anomalous Hall effect in itinerant ferromagnets, it is expected that the charge-neutral heat current is deflected by a spin-orbit interaction in magnetic dielectrics. Strohm *et al.* first reported the thermal Hall effect in a paramagnetic dielectric $Tb_3Ga_5O_{12}$ (TGG) [1]. At the measured temperature, there is no magnetic order on TGG, and therefore the origin of the thermal Hall effect is ascribed to the Hall effect of phonons. Nevertheless, the thermal Hall effect measurement was performed at one temperature, and seems quite sample dependent [2], and several microscopic theories were proposed but remain under discussion [3–5]. Subsequently, the thermal Hall effect has been also observed in a number of magnetic dielectrics [6–13]. While some of them can be explained by the Berry-phase-induced magnon Hall effect due to the Dzyaloshinskii-Moriya interaction [6,7], the mechanisms for the other observations were not well understood. In particular, the thermal Hall effect observed in the paramagnetic state of some frustrated magnets [8,10–13] has two possible mechanisms, i.e., the phonon Hall effect and the Hall effect due to exotic magnetic excitation, which are quite difficult to be distinguished. Here, we propose the dilution of the magnetic moment is useful for elucidating this issue. In the case of exotic magnetic excitation, the magnetic dilution should largely suppress the thermal Hall effect because the heat current is carried by a magnetic interaction. On the other hand, the phonon Hall effect is induced by the deflection of the phonon current originating from the local magnetoelastic coupling. The effect of magnetic dilution is more gradual.

In this paper, we study the dilution effect of the thermal Hall effect in $Tb_2Ti_2O_7$. Previously, Hirschberger *et al.* observed a large thermal Hall effect in $Tb_2Ti_2O_7$ below 100 K [8]. They suggested that conventional phononic mechanisms cannot explain it because the temperature and magnetic field dependences contradict the phononic mechanism and the magnitude is much larger than that observed in TGG. Alternatively, they speculated that it is induced by some exotic magnetic excitations. However, we have found that the thermal Hall effect is unchanged or rather slightly enhanced by the 70% substitution of Tb^{3+} by Y^{3+}. This observation strongly contradicts the scenario of magnetic excitation, and therefore supports the phononic origin. Incidentally, we study the thermal Hall effect in a similarly diluted magnetic dielectric, $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$. We have observed a finite thermal Hall effect at low temperature, which supports the first report of the thermal Hall effect by Strohm *et al.* [1].

II. EXPERIMENTAL METHOD

We grew single crystals of $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ and $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ by means of the floating zone method. The atmospheres were the air for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ and 1 atm O_2 gas for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$. We confirmed the absence of the impurity phase and their single crystallinity by powder x-ray diffraction and Laue photography, respectively. Detailed data are shown in the Supplemental Material [14]. For the measurements, we used rectangular samples. The size of the $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ $[(Tb_{0.3}Y_{0.7})_2Ti_2O_7]$ sample is $3.05 \times 1.15 \times 0.3$ mm^3 ($3.25 \times 1.43 \times 0.3$ mm^3) with the widest plane parallel to (100) [(111)]. Magnetic susceptibility was measured with the use of a superconducting quantum interference device (SQUID). Thermal conductivity measurements were performed by means of the steady state method in a superconducting magnet. The measurement atmosphere was evacuated down to 3×10^{-3} Pa. Heat current was generated

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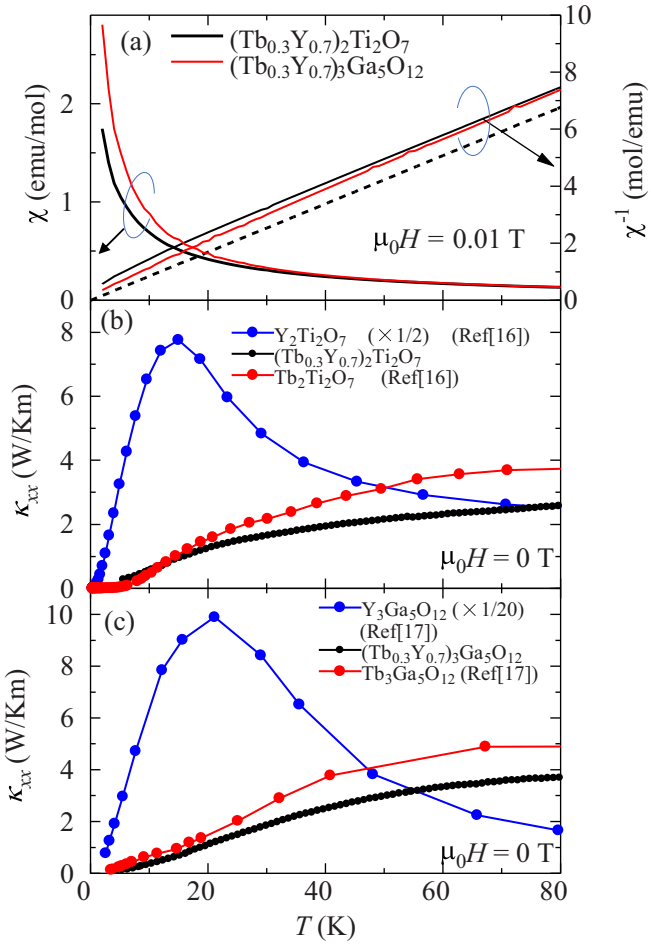

FIG. 1. (a) Temperature dependence of magnetic susceptibility χ and the reciprocal χ^{-1} for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ and $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$. The dashed line indicates paramagnetic susceptibility deduced by the Curie law with the free Tb^{3+} moments. (b) Temperature dependence of longitudinal thermal conductivity κ_{xx} for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$. Previously reported data of $Tb_2Ti_2O_7$ and $Y_2Ti_2O_7$ are also shown for comparison [16]. (c) Temperature dependence of longitudinal thermal conductivity κ_{xx} for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$. Previously reported data of $Tb_3Ga_5O_{12}$, and $Y_3Ga_5O_{12}$ are also shown for comparison [17].

by chip resistance. The thermal gradient was measured by two thermometers (CERNOX, Lake Shore Cryotronics, Inc.). Details of the measurement are shown in the Supplemental Material [14].

III. RESULTS AND DISCUSSIONS

In Fig. 1(a), we show the temperature dependence of the magnetic susceptibility χ for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ and $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ at 0.01 T. For both samples χ steeply increases with decreasing temperature and χ^{-1} shows a linearlike temperature dependence with small intercepts at least above 20 K. A similar temperature dependence of χ was reported for $(Tb_{0.02}Y_{0.98})_2Ti_2O_7$ in the literature [15]. The effective magnetic moments deduced from the high-temperature slope of χ^{-1} are $9.71\mu_B$ and $9.78\mu_B$ for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ and $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$, respectively, where μ_B is the Bohr magneton. These are almost equal to the value of the free Tb^{3+} moment ($9.72\mu_B$). When we assume that the effective moment is equal to that of free Tb^{3+}, we can confirm that the Tb concentration is close to the prescribed value. This also shows the magnetization is approximately described by the Curie paramagnetism of free Tb^{3+} ions [dashed line in Fig. 1(a)]. The difference between the measured magnetization and that expected for Curie paramagnetism is caused by the crystal field splitting of Tb $4f$ states, as discussed in the literature [15].

In Fig. 1(b), we show the temperature dependence of longitudinal thermal conductivity κ_{xx} for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ compared with the $Y_2Ti_2O_7$ and $Tb_2Ti_2O_7$ data in the literature [16]. The thermal conductivity for $Y_2Ti_2O_7$ shows a typical phonon-mediated-type temperature dependence. It increases with phonon population as the temperature is increased from 0 K, shows a peak at a temperature, and then decreases with temperature due to the increase of phonon-phonon scattering. On the other hand, in $Tb_2Ti_2O_7$, the thermal conductivity is remarkably suppressed and monotonically decreases with decreasing temperature, similarly to glassy compounds. It suggests that the phonons are strongly scattered by Tb magnetic moments. When the Tb ions are partially substituted by Y ions, the disorder due to the solid solution is introduced but the number of Tb ions is decreased. As a result, the thermal conductivity for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ becomes quite similar to that of $Tb_2Ti_2O_7$. As shown in Fig. 1(c), a similar rare-earth site dependence is observed for garnet oxides [17]. The magnetic field dependence of longitudinal thermal conductivity is shown in the Supplemental Material [14].

We plot in Figs. 2(a) and 2(b) the magnetic field dependence of thermal Hall conductivity divided by temperature κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ at various temperatures. As is shown in these figures, we certainly observed finite κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ below 80 K. In the high-temperature region, κ_{xy}/T increases linearly with increasing magnetic field. As the temperature is lowered, κ_{xy}/T is increased and tends to saturate in the high-field region. However, as the temperature is further decreased below 11 K, the magnitude of κ_{xy}/T decreases, as indicated in Fig. 2(a). The origin of the temperature and magnetic field dependences will be discussed later.

In Figs. 3(a)– 3(e), we show the magnetic field dependence of κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ at various temperatures. We observed a finite thermal Hall effect for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ around 7.2 K. κ_{xy}/T increases almost linearly with magnetic field but it shows kinklike behavior around 2 T, which seems to be related to the change of the Tb $4f$ state. The ground state is a quasidoublet with an energy difference of 3.7 K, which is comparable to the Zeeman energy [18]. Therefore, the magnetic field change of the Tb $4f$ state is expected in this material. Similar magnetic field dependences are observed at 5.6 and 8 K. Above 12 K, the data of κ_{xy}/T become scattered and the signal cannot be resolved. κ_{xy} is proportional to the square of κ_{xx} as well as the measured transverse temperature gradient (see Supplemental Material [14]). The longitudinal thermal conductivity increases and the thermal Hall angle $\tan\Theta_H = \kappa_{xy}/\kappa_{xx}$ decreases with increasing temperature, as shown in Figs. 3(f)–3(j). In the high-temperature region, the noise magnitude in the unit of κ_{xy} is enhanced and the signal becomes difficult to be resolved.

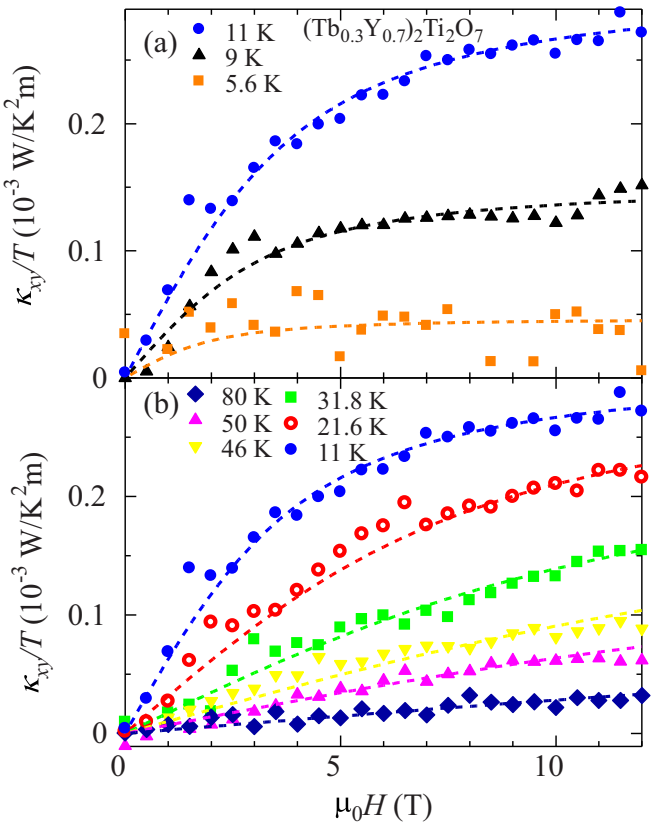

FIG. 2. (a), (b) Magnetic field dependence of thermal Hall conductivity divided by temperature κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ at various temperatures. The dashed lines indicate the result of fitting with the Brillouin function (see text).

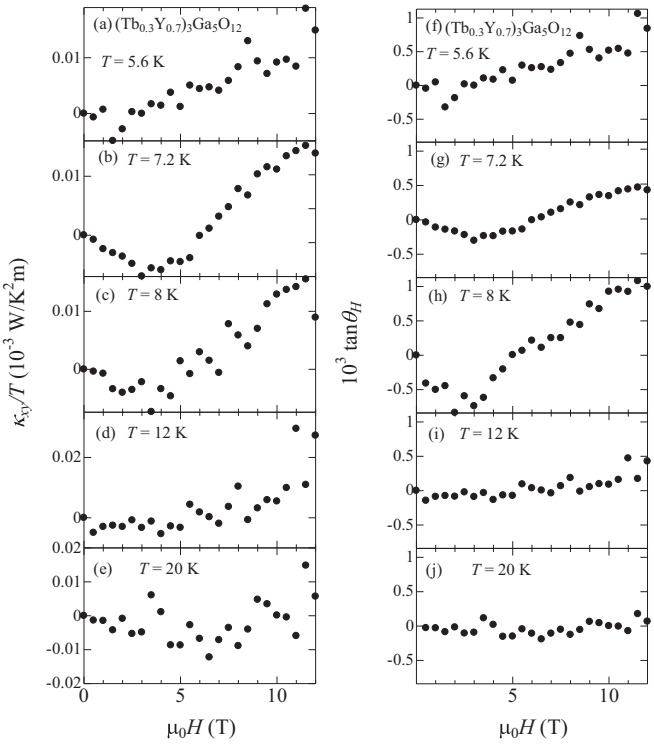

FIG. 3. Magnetic field dependence of (a)–(e) κ_{xy}/T and (f)–(j) the thermal Hall angle $\tan\Theta_H = \kappa_{xy}/\kappa_{xx}$ for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ at various temperatures.

We plot the temperature dependence of κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ and $Tb_2Ti_2O_7$ [8] at 6 T in Fig. 4(a). This clearly shows that κ_{xy}/T is unchanged or rather increased by the substitution of Tb with Y. As discussed above, the observation of the finite thermal Hall effect in a diluted system contradicts the magnetic excitation mechanism of the thermal Hall effect. Therefore, the thermal Hall effect in this system is mostly induced by the Hall effect of phonons. Nevertheless, the existence of an additional magnetic excitation component in $Tb_2Ti_2O_7$ cannot be completely excluded in this work. In order to study the microscopic mechanism, let us discuss the details of the temperature and magnetic field dependence. The magnetic field dependences shown in Figs. 2(a) and 2(b) are quite similar to that of magnetization in a Curie-type paramagnet, in which the temperature and magnetic field dependences of magnetization are described by the Brillouin function. In fact, the magnetic field dependence can be scaled by paramagnetic magnetization derived from the Brillouin function $M = g_L\mu_B JN/VB_J(\frac{g_L\mu_B H}{k_B T})$ with $J = 6$ and $g_L = 3/2$ (dashed lines in Fig. 2), where k_B is the Boltzmann constant, N is the number of magnetic ions, and V is the volume of specimen, respectively. Therefore, the temperature and magnetic field dependences of κ_{xy}/T can be expressed as

$$\kappa_{xy} \approx AM, \tag{1}$$

where A is a temperature-dependent scaling parameter. We plot A obtained by the scaling in Fig. 4(b), compared with the longitudinal thermal conductivity κ_{xx}. A seems to be proportional to κ_{xx}, indicating the relationship

$$\kappa_{xy}/\kappa_{xx} \approx C' B_J\left(\frac{g_L\mu_B H}{k_B T}\right) \approx CM, \tag{2}$$

where C is a constant. As discussed above, the crystal field splitting of the Tb $4f$ state gives rise to a small but finite modulation of magnetization. In order to examine the influence of this modulation on the κ_{xy} analysis, we did the analysis based on the observed low-field (<5 T) magnetization (see Supplemental Material [14]), and confirmed that the same conclusion was obtained. This relation suggests that κ_{xy} is proportional to κ_{xx} or relaxation time, similarly to the case of the skew scattering mechanism of the electronic anomalous Hall effect [19], and consistent with the picture that the thermal Hall effect is induced by the deflection of phonon current due to polarized Tb magnetic moments. Several theories for the phonon Hall effect have been proposed so far [3–5]. Some theories study the Berry phase effect of the phononic band [3] but the solid solution of Y and Tb should smear out the Berry curvature in momentum space. Therefore, the scattering affected by the spin-orbit interaction seems to be the origin of the present phononic thermal Hall effect. Mori *et al.* proposed the theory for the thermal Hall effect induced by resonant skew scattering due to Tb $4f$ excitation [5]. In this theory, the thermal Hall effect is expected to be largely

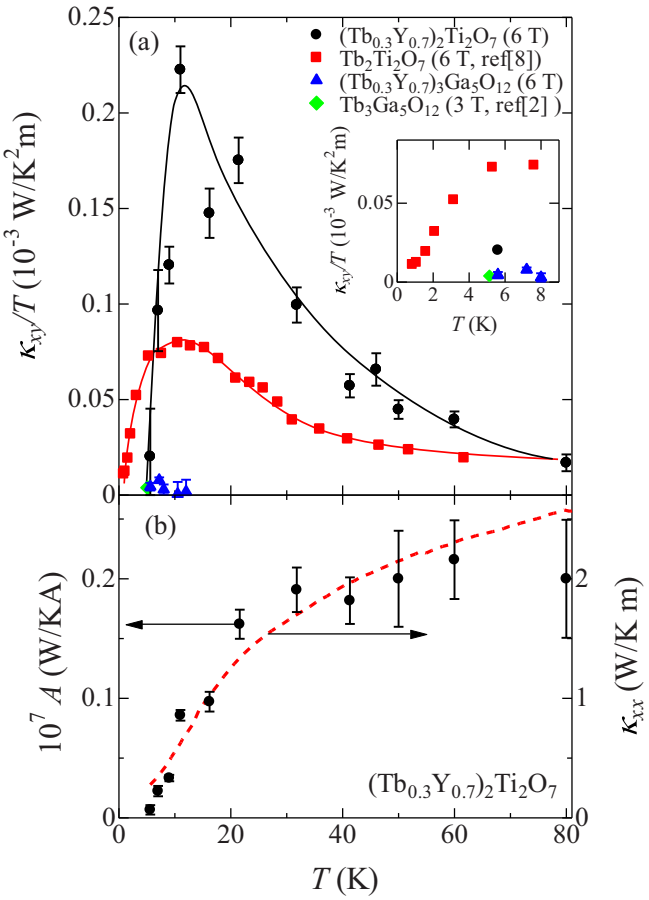

FIG. 4. (a) Temperature dependences of κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ and $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ at 6 T. κ_{xy}/T at 6 T for $Tb_2Ti_2O_7$ and that at 3 T for $Tb_3Ga_5O_{12}$ are reproduced from the literature [2,8]. The inset magnifies the low-temperature and small κ_{xy}/T region. The error bars are estimated as the standard deviation of magnetic field dependence of κ_{xy} from the Brillouin function for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$, and the standard deviation from the linear relation above 5 T for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$. Solid lines are merely guides for the eyes. (b) Temperature dependence of the scaling constant $A(T)$ and longitudinal thermal conductivity. The error bars for $A(T)$ are estimated as the standard deviation of κ_{xy} divided by the calculated magnetization at 6 T.

enhanced around the temperature whose energy scale is of the Tb $4f$ excitation of the electronic state. Nevertheless, such a resonance structure was not observed in the temperature dependence of κ_{xy}/T. Another class of theory is related to the scattering of phononic current that is polarized owing to Raman-type spin-phonon coupling [4]. This theory cannot be well examined at present because the relation between κ_{xx} and κ_{xy} was not clearly discussed and it is uncertain whether the experimentally obtained relation of $\kappa_{xy}/\kappa_{xx} \sim M$ can be reproduced with this theory. In any case, the relation should be the experimental constraint for the theories of the phononic thermal Hall effect in this system.

We also plot κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ at 6 T and $Tb_3Ga_5O_{12}$ at 3 T [2] in Fig. 4(a). κ_{xy}/T for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$ has the same order as that of $Tb_3Ga_5O_{12}$, which was reported in 2005 [1]. The present observation seems to confirm the thermal Hall effect in the Tb garnet system after 14 years. The magnitude is much smaller than that in the pyrochlore system. This difference seems to be related to the electronic structure of Tb. Tb $4f$ electrons in a pyrochlore system have a doublet ground state and excited states around 20 K [20]. On the other hand, in a garnet system the ground state of Tb is a quasidoublet, and the excitation energy is around 50 K [18]. The energy difference of the excited state affects the magnitude of phononic thermal Hall conductivity in any case of microscopic mechanisms. The Raman-type spin-phonon interactions are induced by the transition between the doublet states via high-energy excited states [21]. Resonant skew scattering is also affected by the energy of the excited states.

IV. CONCLUSION

In conclusion, we observed the thermal Hall effect for pyrochlore $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ and garnet $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$. The thermal Hall conductivity for $(Tb_{0.3}Y_{0.7})_2Ti_2O_7$ is even larger than the previously observed thermal Hall conductivity for $Tb_2Ti_2O_7$. This clearly shows the thermal Hall conductivity in this system is robust against magnetic dilution, which indicates that the phononic mechanism is dominant for thermal Hall conductivity. The magnetic field and temperature dependences follow the empirical relation of $\kappa_{xy}/\kappa_{xx} \sim M$, which is reminiscent of the skew scattering mechanism of the electronic anomalous Hall effect [19] and should be a constraint on the theories of the phononic thermal Hall effect in the system. We have also observed small thermal Hall conductivity for $(Tb_{0.3}Y_{0.7})_3Ga_5O_{12}$. The magnitude difference between the two systems seems to be caused by the energy scale of the excited states.

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