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When a conduction electron couples with a non-coplanar localized magnetic moment, the real-space Berry curvature is exerted to cause the geometrical Hall effect, which is not simply proportional to the magnetization. So far, it has been identified in the case mostly where the non-coplanar magnetic order is present on the sublattice of conduction electrons. Here, we demonstrate that the geometrical Hall effect shows up even without long-range magnetic order of conduction electrons, as induced by non-coplanar exchange fields from the localized magnetic moments, in hole-doped pyrochlore molybdates $(\mathrm{Tb}_{1-x}\mathrm{Ca}_x)_2\mathrm{Mo}_2\mathrm{O}_7$. We find that the geometrical Hall effect is markedly anisotropic with respect to the applied magnetic field direction, which is in good accordance with the field-dependent magnitude and sign change of the real-space scalar spin chirality of local Tb moments. These results may facilitate the understanding of emergent electromagnetic responses induced by the Kondo-like coupling between conduction electrons and local spins in a broad material class.

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I. INTRODUCTION

Non-collinear or non-coplanar complex magnetic structures in solids are proven to play a fundamentally important role in their spin-related quantum transport and multiferroic properties [1, 2]. A representative example is the scalar spin chirality (SSC), which is defined by $\chi_{ijk} = \boldsymbol{S}_i \cdot (\boldsymbol{S}_j \times \boldsymbol{S}_k)$ for three neighboring-site spins $\boldsymbol{S}_i, \boldsymbol{S}_j$, and $\boldsymbol{S}_k$. As a conduction electron moves over a non-coplanar spin texture with finite SSC, it is endowed with the non-trivial quantum phase (Berry phase) and hence experiences the emergent magnetic field, which can be far beyond a real magnetic field [3–5]. One of the most common outcomes of SSC is the unconventional Hall effect, termed geometrical Hall effect (GHE) [5]. In the case where the mean free path is sufficiently longer than the magnetic period, both anomalous Hall effect and GHE are appropriately captured by the momentum space picture where the anti-crossing points (e.g. Weyl nodes) are associated with the Berry curvature due to the spin-orbit coupling (Karplus-Luttinger intrinsic mechanism) and the SSC [6, 7]. In contrast, as the magnetic period is longer than the mean free path, electrons hop around the spin triad in the real space and feel the emergent field $\boldsymbol{B}_{\mathrm{eff}} = h\boldsymbol{n}_{ijk}\chi_{ijk}/eS$, where $\boldsymbol{n}_{ijk}$ is the normal vector to the spin triad plane and S is the area of spin triad, as exemplified by the magnetic skyrmion lattice [8, 9].

Among a variety of non-coplanar magnets, pyrochlore molybdates $R_2\mathrm{Mo}_2\mathrm{O}_7$ (R being a trivalent rare-earth or Y ion) offer an ideal platform to study the correlation between charge transport and non-coplanar magnetism, because of a variety of magnetic/electronic phases [10–12].

The pyrochlore lattice consists of corner-linked tetrahedra with R ions and Mo ions, each of which is displaced by half a unit cell (Fig. 1(a)). The most well investigated is $\mathrm{Nd}_2\mathrm{Mo}_2\mathrm{O}_7$ with the possible largest-size R ion and hence with relatively small electron correlation. It is metallic in the whole temperature range and undergoes the ferromagnetic transition at 90 K, presumably due to the double-exchange-like mechanism of Mo-$4d$ electrons [13]. Additionally, the Nd-$4f$ magnetic moments, which host Ising anisotropy along the local $\langle 111 \rangle$ direction, begin to freeze below around 40 K. Therefore, the Mo spins are slightly tilted ($\sim$ 5-10°) via the f-d magnetic interaction at low temperatures, resulting in the Mo spins with finite SSC (Fig. 1(b)) [4]. Because of such anisotropy for R-$4f$ moments, various configurations are realized under the external magnetic field [14, 15]. For instance, as the field is applied along the [100] direction, two of four magnetic moments point out of the tetrahedron, while the other two inwards, termed the 2-in 2-out configuration. On the other hand, the field along [111] favors the 3-in 1-out state, as displayed in the bottom panel in Fig. 1(b). The anisotropy of Hall response in $\mathrm{Nd}_2\mathrm{Mo}_2\mathrm{O}_7$ is understood in terms of the 2-in 2-out or 3-in 1-out like habit of the Mo-$4d$ spins transmitted from the corresponding change of Nd-$4f$ configuration [16]. Such a case where the Mo-$4d$ ferromagnetic orders are slightly modulated by the R-$4f$ moments is termed the strong coupling, which has been extensively studied so far [4, 18]. On the other hand, little is known for the weak coupling regime, where the Mo conduction electrons exhibit no spontaneous long-range orders but are ~~moderately~~ weakly influenced by the exchange fields from the R-$4f$ moment configuration [20].

Previously, GHE in the weak-coupling regime was studied for the $(\mathrm{Tb}_{1-x}\mathrm{Cd}_x)_2\mathrm{Mo}_2\mathrm{O}_7$ polycrystals [19]. It is demonstrated that the Hall response systematically changes as a function of the "density" of SSC in the real space tuned by the doping level of magnetic Tb moments, in accord with the theoretical prediction [20]. However, the exchange-field induced GHE should be dominated by the R-$4f$ moment configuration; for instance, SSC is expected to change its sign and magnitude between 2-in 2-out and 3-in 1-out configuration, and further, diminish as the R-$4f$ moments are forced to align in a collinear manner. Thus, to obtain a deeper insight into GHE, the field-directional study on the single crystals is required.

In this study, we investigate the magnetotransport properties of $(\mathrm{Tb}_{1-x}\mathrm{Ca}_x)_2\mathrm{Mo}_2\mathrm{O}_7$ single crystals to see the role of the real-space SSC in the Hall effect. We successfully synthesize high-quality samples by using the state-of-the-art floating zone furnace equipped with high-power lasers and measure the transport and magnetization at high magnetic fields up to 31 T, which allows us to access a wide range of Tb magnetic states from non-coplanar spin textures to fully spin-aligned state. We observe the large anisotropy of GHE between the [100] and [111] field directions in the intermediate field range, which gradually diminishes at high fields. It can be reasonably explained in terms of the magnitude and sign change of the SSC of localized Tb moments that impose the f-d exchange field on the conductive Mo sublattice.

II. EXPERIMENTAL DETAILS

Single crystals were synthesized at $\sim$1880 °C under an argon atmosphere of 0.99 MPa in the laser floating zone furnace [21]. All of them were well characterized by x-ray powder diffraction and energy dispersive x-ray spectroscopy to check the lattice structure and composition. The transport measurement setup is shown in Fig. 2(a). The electric current flows along the [1$\bar{1}$0] crystalline direction. The magnetic field was rotated around the current direction, so that the transport measurements for different orientations can be performed on the same sample. $\theta = 0°$, which is perpendicular to the sample plane, is along the [100] crystalline direction, while $\theta = \pm54.7°$ is along the [111] and its equivalent direction. We obtained the Hall resistivity ρ_{yx} by normalizing the measured Hall signal ρ_H for $\theta = \pm54.7°$ divided by $\cos\theta$. We confirm that the deduced ρ_{yx} shows almost the same behavior as another sample with the [111] plane measured under the normal [111] field (see Appendix Figure 5). The transport properties and magnetization up to 14 T were measured by using DC magnets in Physical Property Measurement System (PPMS), Quantum Design. Higher-field measurements of magnetotransport and magnetization were performed by using non-destructive pulse magnets energized by capaci-
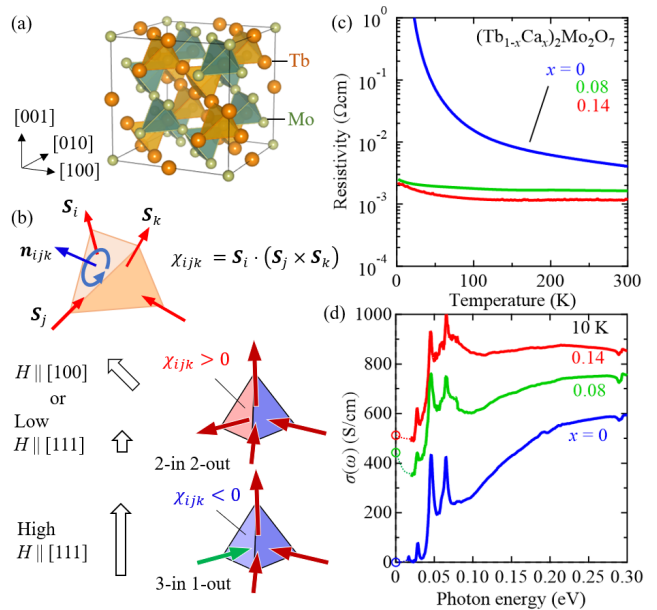

FIG. 1: (color online). (a) Crystal structure of pyrochlore type $\mathrm{Tb}_2\mathrm{Mo}_2\mathrm{O}_7$. (b) Definition of the SSC $\chi_{ijk} = \boldsymbol{S}_i \cdot (\boldsymbol{S}_j \times \boldsymbol{S}_k)$ in a tetrahedron with Tb spins $\boldsymbol{S}_i$, $\boldsymbol{S}_j$, and $\boldsymbol{S}_k$ on its vertices. The middle (bottom) figure shows χ_{ijk} in each triangle plane of a tetrahedron for 2-in 2-out (3-in 1-out) state. The color of the plane corresponds to the sign of χ_{ijk} when we take $\boldsymbol{n}_{ijk} \cdot \boldsymbol{H} > 0$. In these figures, $H \parallel$ [111] case is assumed. (c) Temperature dependence of resistivity for $(\mathrm{Tb}_{1-x}\mathrm{Ca}_x)_2\mathrm{Mo}_2\mathrm{O}_7$. (d) Optical conductivity spectra below 0.3 eV at 10 K for $(\mathrm{Tb}_{1-x}\mathrm{Ca}_x)_2\mathrm{Mo}_2\mathrm{O}_7$. ~~The inset shows the optical conductivity spectra up to 4 eV.~~

tor banks and a flywheel DC generator installed at International MegaGauss Science Laboratory of Institute for Solid State Physics (ISSP), University of Tokyo.

III. RESULTS AND DISCUSSION

Figure 1(c) shows the temperature dependence of resistivity for ~~several~~ three compositions ($x = 0, 0.08, 0.14$). The resistivity of $x = 0$ rapidly increases as the temperature decreases. Above $x = 0.08$, the resistivity significantly decreases down to the order of 10^{-3} Ωcm. The weak temperature dependence for higher doping reminds us of the high pressure effect on $\mathrm{Nd}_2\mathrm{Mo}_2\mathrm{O}_7$, which yields the anomalously diffusive metallic state ~~presumably~~ as a result of the strong competition between an antiferromagnetic (spin glass) insulator and ferromagnetic metal states [11]. The optical conductivity spectra at 10 K for several x ~~is~~ are displayed in Fig. 1(d). The optical conductivity for $x = 0$ gradually decreases below 0.2 eV, forming a clear charge gap of $\sim$0.05 eV. The observed magnitude of the charge gap is slightly different from the previous one [22], possibly due to the oxygen non-stoichiometry depending on the crystal-growth condition. With increasing x, the optical conductivity grad-
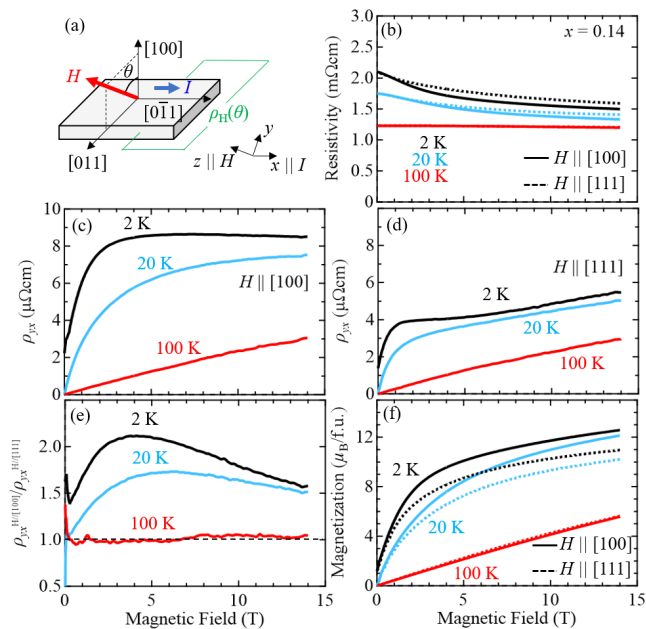
FIG. 2: (color online). (a) Measurement setup for magnetic transport properties. Magnetic field dependence of (b) resistivity for both $H \parallel [100]$ and $H \parallel [111]$, (c) Hall resistivity for $H \parallel [100]$, (d) Hall resistivity for $H \parallel [111]$, (e) anisotropic ratio of Hall resistivity between $H \parallel [100]$ and $H \parallel [111]$, and (f) magnetization for both $H \parallel [100]$ and $H \parallel [111]$ at several temperatures.

ually increases below $\omega_C \sim 1.5$ eV (see the ~~inset of Figure 1(d)~~ Fig. 5 in the Appendix) accompanied by the closing of the charge gap, in good accordance with the DC conductivity. The absence of the clear Drude peak confirms the diffusive metal state at $x = 0.08$ and 0.14. It is to be noted that the sharp peaks below 0.1 eV are infrared-active phonon modes allowed for cubic pyrochlore lattice [23–25], indicative of a good crystal quality even for the doped metallic samples.

In the following, we focus on the magnetotransport properties of the barely metallic $x = 0.14$ crystal. Figure 2(b) shows the magnetic field dependence of resistivity at several temperatures for $H \parallel [111]$ and [100]. The resistivity is almost independent of magnetic field at 100 K. Below 20 K, the resistivity gradually decreases with increasing field, likely due to the suppression of spin glass or antiferromagnetic spin fluctuation. Figures 2(c) and (d) show the Hall resistivity ρ_{yx} for $H \parallel [100]$ and $H \parallel [111]$, respectively. ρ_{yx} at 100 K is nearly proportional to the magnetic field with little difference between for $H \parallel [100]$ and $H \parallel [111]$. However, the anisotropy becomes evident at low temperatures. At 2 K, as the field increases, ρ_{yx} for $H \parallel [100]$ abruptly increases, reaches a maximum value of ~ 9 $\mu\Omega$cm at around 8 T, and then slightly decreases. On the other hand, ρ_{yx} for $H \parallel [111]$ shows a hump at around 2 T and gradually increases as the field is further increased. As can be seen in Fig. 2(e), the anisotropy ratio of ρ_{yx} between $H \parallel [100]$ and

$H \parallel [111]$ at 2 K exceeds 2 in the intermediate field region, and gradually decreases as the field increases. Figure 2(f) shows the magnetization curves for $H \parallel [100]$ and $H \parallel [111]$. At 100 K, the magnetization shows H-linear dependence and no anisotropy is observed as in ρ_{yx}. With lowering temperature, the magnetization for $H \parallel [100]$ becomes larger than that for $H \parallel [111]$. Such an anisotropy can be attributed to the different magnetic ~~configuration~~ arrangement of Tb moments. ~~Since each Tb moment shows single-ion anisotropy along the local $\langle 111 \rangle$ orientation, the sufficiently large, but not too large, magnetic field along [100] ([111]) favors 2-in 2-out (3-in 1-out) state (see Fig. 1(b)). In fact, the expected value of 2-in 2-out state is larger than that of 3-in 1-out state by ~ 1.2 μ_B/f.u..~~ In canonical spin-ice systems such as $Ho_2Ti_2O_7$ and its non-magnetic ion doped analogs, the sufficiently large, but not too large, magnetic field along [100] ([111]) favors 2-in 2-out (3-in 1-out) state (see Fig. 1(b)) [26, 27]. Because of the single-ion anisotropy of Tb-4f moments along the local $\langle 111 \rangle$ orientation, the similar magnetic arrangement can be realized in each field direction. In fact, the expected net moment of 2-in 2-out state is larger than that of 3-in 1-out state by ~ 1.2 μ_B/f.u. which is consistent with the magnetization observed in Fig. 2(f). ~~Such a difference of Tb magnetic configuration can give rise to the salient anisotropy of Hall effect.~~ Such a small difference of the net magnetization between $H \parallel [100]$ and $H \parallel [111]$ cannot explain the strong anisotropy in Hall resistivity. Instead, the non-coplaner magnetic arrangement, or equivalently the scalar spin chirality, is likely to play a key role in the observed Hall effect.

In order to elucidate the origin of Hall effect, we employ the higher magnetic field measurement which allows us to fully control the Tb-moment configurations. Figure 3(a) (3(b)) shows the magnetic field dependence of magnetization up to above 34 T at 2 K for $H \parallel [100]$ ([111]). The magnetization measured by the pulse magnet (red solid curve) perfectly overlaps with the one measured by the DC magnet (black curve). Remarkably, the magnetization for $H \parallel [100]$ monotonically increases, exceeds the expected value for the 2-in 2-out state (8.9 μ_B/f.u.) at 4 T, and nearly reaches 15.5 μ_B/f.u. at 34 T which is expected for fully-polarized Tb-4f moments, likely stemming from the competition between the magnetic anisotropy and Zeeman effect. Thus we can examine the magnetotransport properties for both collinear and non-collinear magnetic states on one sample. We also plot the magnetization for $x = 0$ in Figs. 3(a) and 3(b). To compare with that for $x = 0.14$, we ~~renormalize~~ normalize it by the density of Tb moments, namely multiply it by $1 - x = 0.86$. Notably, the ~~renormalized~~ normalized curve for $x = 0$ falls onto the same curve for $x = 0.14$ above 10 T, indicating that the Mo contribution to the whole magnetization is almost negligible not only for the spin-glass insulating $x = 0$ crystal but for the carrier-doped
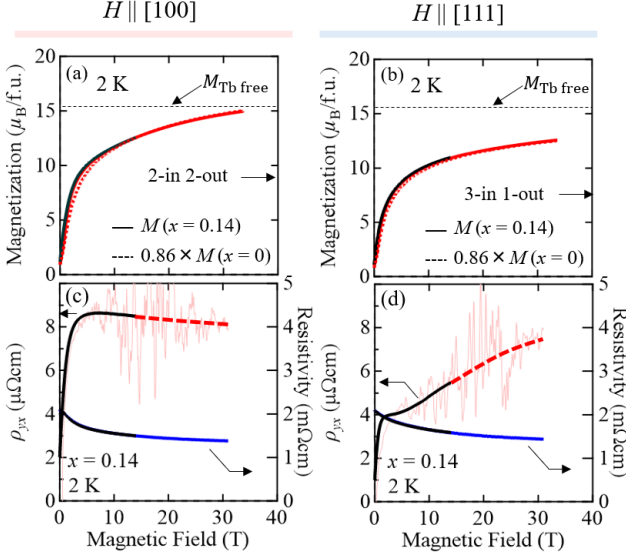
FIG. 3: (color online). Magnetic field dependence of magnetization up to 34 T for (a) $H \parallel [100]$ and (b) $H \parallel [111]$. Magnetic field dependence of Hall resistivity as well as resistivity for (c) $H \parallel [100]$ and (d) $H \parallel [111]$. The black curves indicate the results of DC field measurements up to 14 T. The red and blue curves show the results of high field measurements using pulsed fields up to 31 T.

metallic $x = 0.14$ one, in stark contrast to the Mo-spin ferromagnetic $Nd_2Mo_2O_7$ [4]. Figures 3(c) and 3(d) show the magnetic field dependence of resistivity and Hall resistivity for $H \parallel [100]$ and $H \parallel [111]$, respectively. The resistivity gradually decreases as the field increases up to 31 T for both field directions, as observed in the measurement in the DC magnet. The Hall resistivity data measured by the pulse magnet seem somewhat noisy, because of the small Hall signals compared to the longitudinal resistivity. Nevertheless, the overall field dependence and amplitude are reconciled with the DC data, and moreover the signal appears reliable again near the maximum field, since the data can be integrated during the relatively long time at high fields in the field pulse shape. As the field approaches to 31 T, ρ_{yx} for $H \parallel [100]$ gradually decreases down to ~ 8 $\mu\Omega$cm, while that for $H \parallel [111]$ explicitly increases up to ~ 7.5 $\mu\Omega$cm. Apparently, the anisotropy of ρ_{yx} disappears at high magnetic fields.

In general, the Hall resistivity is expressed as

$$\rho_{yx} = R_0 \mu_0 H + R_S \rho_{xx}^n M_z + \rho_{yx}^{G}. \quad (1)$$

where R_0 is the ordinary Hall coefficient, $R_S \rho_{xx}^n$ the anomalous Hall coefficient with the scaling factor n in case of diffusive metal [28], and ρ_{yx}^{G} the geometrical contribution. According to the previous study on Cd-doped $Y_2Mo_2O_7$, the ordinary Hall effect is quite small (less than 0.3 $\mu\Omega$cm at 14 T) [29]. Therefore, we neglect here the ordinary contribution for simplicity. To extract the geometrical contribution, we plot ρ_{yx} as a function
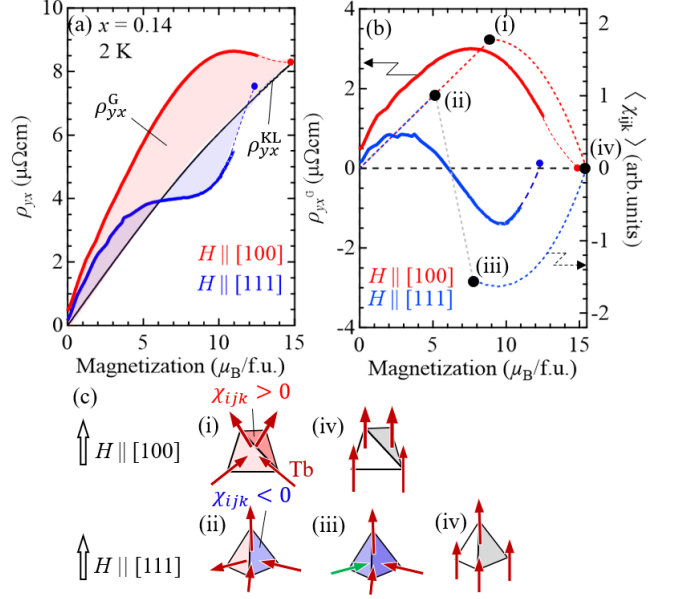
FIG. 4: (color online). (a) Magnetization dependence of the Hall resistivity. The solid lines denote the data obtained with the DC field, and the circles are the values at the maximum pulse field. The dotted lines are the smoothed data taken by the pulse field measurement. (b) Magnetization dependence of the GHE and simulated spin chirality. Red and blue dashed lines represent the simulated SSC of the single Tb tetrahedron based on the magnetization curves (see Appendix Note 3). (c) Possible magnetic structure and SSC at each point in the panel (b).

of magnetization in Fig. 4(a). The red and blue solid lines denote experimental data measured in DC magnetic field, red and blue circles are the data at the highest field 31 T, and red and blue dashed lines indicate the noise-smoothed connections between these two experimental data. Since the magnetization for $H \parallel [100]$ is almost saturated at 31 T (Fig. 3(a)), we can assume that the geometrical contribution is zero at 31 T. The black line in Fig. 4(a) indicates the Karplus-Luttinger type anomalous Hall resistivity, $\rho_{yx}^{KL} = R_S \rho_{xx}^n M_z$, where $n = 0.4$ in the carrier hopping regime, as confirmed in Appendix Figure 7[28]. As can be seen, ρ_{yx} for $H \parallel [100]$ is larger than ρ_{yx}^{KL} in the whole magnetization regime. Remarkably, for $H \parallel [111]$, ρ_{yx} shows non-monotonic magnetization dependence as opposed to ρ_{yx}^{KL}. Especially, ρ_{yx} crosses ρ_{yx}^{KL} at $M \sim 6$ μ_B/f.u., and eventually merges to ρ_{yx}^{KL} at the largest magnetization.

Figure 4(b) displays the extracted geometrical contribution, $\rho_{yx}^{G} = \rho_{yx} - \rho_{yx}^{KL}$, as a function of magnetization. As the magnetization increases, ρ_{yx}^{G} for $H \parallel [100]$ gradually increases, reaches maximum at ~ 8 μ_B/f.u., and eventually decreases towards zero at the full moment of 15.5 μ_B/f.u.. ~~We speculate that the observed~~ This envelope-shaped magnetization dependence of ρ_{yx}^{G}, which is also observed in several materials [3, 30], ~~reflects~~ may
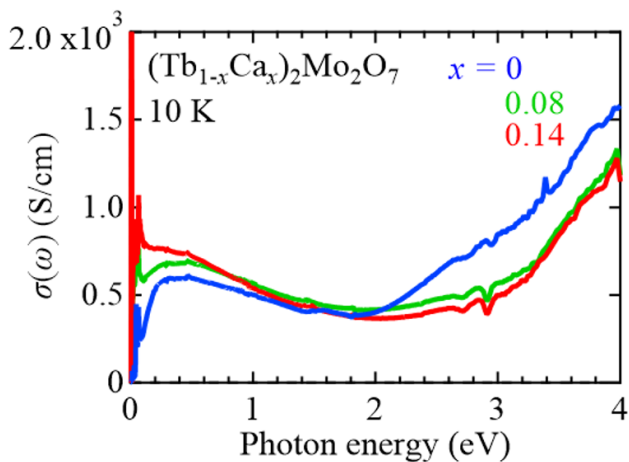
FIG. 5: (color online). Expanded energy dependence of optical conductivity speactra up to 2 eV for three compounds ($x = 0, 0.08, 0.14$).
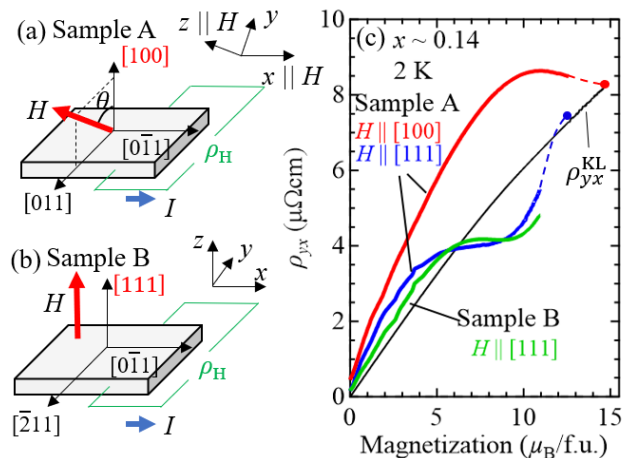

FIG. 6: (color online). Measurement setups for (a) sample A which is mainly used for the anisotropic measurement and (b) sample B for comparison. Each out-of-plane axis is along the [100] and [111] crystalline orientation, respectively. (c) Magnetization dependence of Hall resistivity for (a) and (b) setups. The red and blue curves are measured for sample A, and the green curve is measured for sample B. The black curve is Karplus-Luttinger term of Hall resistivity. For more detail, see the main text.

measured along [011] axis which is perpendicular to both [100] and [0$\bar{1}$1] axes.

Let us take the magnetic field direction as the z-axis and the current direction as the x-axis, as depicted in Fig. 6(a). When the field is tilted by θ off [100] direction, Hall resistivity ρ_H is expressed as

$$\rho_H(\theta) = \rho_{yx}(\theta)\cos\theta - \rho_{zx}(\theta)\sin\theta. \qquad (2)$$

The first term is the usual Hall effect perpendicular to both magnetic field and current, which should be extracted from the measured value ρ_H. Considering the symmetry of the setup of Fig. 6(a), we obtain $\rho_{yx}(\theta) = \rho_{yx}(-\theta)$ and $\rho_{zx}(\theta) = \rho_{zx}(-\theta)$, while $\sin\theta$ is odd. Therefore, we can extract $\rho_{yx}(\theta)$ by averaging $\rho_H(+\theta)$ and $\rho_H(-\theta)$ and then dividing by $\cos\theta$. Fig. 6(c) displays the magnetization dependence of ρ_{yx} for both [100] and [111] field directions, in which one can clearly see the anisotropy, as discussed in the main text.

To double-check ρ_{yx} for $H\|$[111], we prepare another sample B, whose out-of-plane axis is along [111] crystalline direction while the current direction is the same as that in sample A ([0$\bar{1}$1]). In this geometry, we can obtain $\rho_{yx} = \rho_H$ for $H\|$[111] without any calculation, as plotted in Fig. 6(c). Although the chemical composition of sample B ($x = 0.134$) is slightly different from that of sample A ($x = 0.140$), ρ_{yx} of sample B is remarkably similar to that of sample A for $H\|$[111]. Thus, the anisotropy discussed in the main text is ubiquitous for the present system.

A3. Scaling Law for Hall Conductivity

In the hopping region, the anomalous Hall conductivity σ_{xy} is known to follow the empirical scaling relation $\sigma_{xy} \propto \sigma_{xx}^{1.6}$ where σ_{xx} is longitudinal conductivity [28].

Note that ρ_{yx} is proportional to $\rho_{xx}^{0.4}$ when the Hall conductivity is much smaller than longitudinal conductivity, since $\rho_{yx} = \sigma_{xy}/(\sigma_{xx}^2 + \sigma_{xy}^2)$. Fig. 7 shows σ_{xy} normalized by magnetization as a function of σ_{xx} for several different samples at 2 K and 14 T, at which the anomalous Hall contribution is dominant. One can see that $\sigma_{xy} \propto \sigma_{xx}^{1.71}$ holds for in a wide range of σ_{xx} from 10^2 S/cm to 10^3 S/cm. Therefore, we use $n = 0.4$ in eq. (1) in the main text to estimate the Karplus-Luttinger term ρ_{yx}^{KL}. However, the H-dependence of σ_{xx} is not large, and even if there were a slight discrepancy in n, the conclusion shown in Fig.4 of the main text would be hardly affected.

A4. Calculation of the scalar spin chirality

Since the Mo magnetization in $(\text{Tb}_{1-x}\text{Ca}_x)_2\text{Mo}_2\text{O}_7$ is negligible, as demonstrated in Fig. 2 of the main text, we can speculate assume the magnetic structure of the Tb tetrahedra from the magnetization and hence calculate the scalar spin chirality χ_{ijk}.

First we consider the case for $H\|$[100]. At zero magnetic field, Tb magnetic moments are disordered but host 2-in 2-out (2I2O) like habit because of the ferromagnetic interaction between nearest-neighbor Tb moments. As the magnetic field increases, the number of 2I2O Tb tetrahedra, whose net magnetizations are parallel to the field direction, increases and hence the magnetization reaches the expected value of M_{2I2O} =
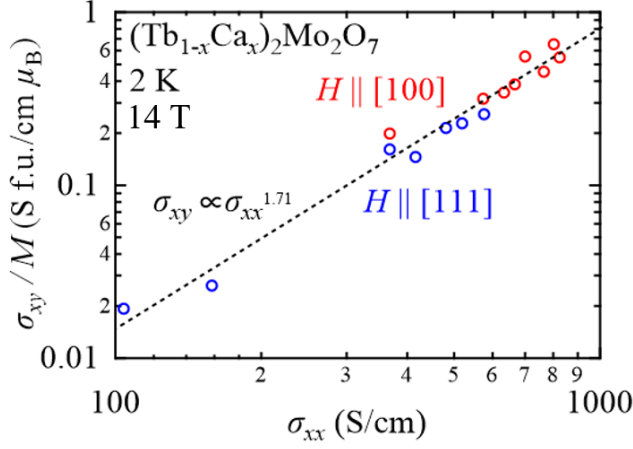
FIG. 7: (color online). Scaling plot of σ_{xy} versus σ_{xx} in the logarithmic scale. The red (blue) marks denote the data for $H\|[100]$ ($H\|[111]$). The dashed line indicate the scaling relation $\sigma_{xy} \propto \sigma xx^{1.71}$.
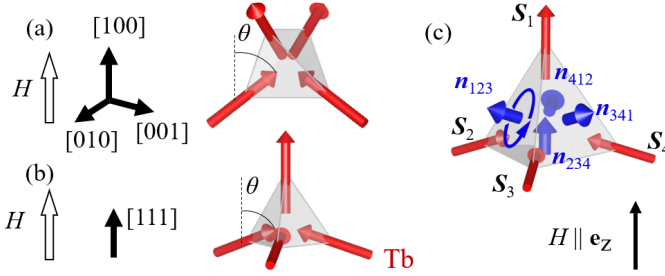

FIG. 8: (color online). Tb moments modulation for (a) $H\|[100]$ at $M \geq M_{2I2O}$ and (b) $H\|[111]$ at $M \geq M_{3I1O}$. (c) Schematic picture of the tetrahedron for the calculation of the scalar spin chirality. Red arrows indicate the spins at each site ($\boldsymbol{S}_1$, $\boldsymbol{S}_2$, $\boldsymbol{S}_3$, and $\boldsymbol{S}_4$), and blue arrows indicate the unit vector parallel to the emergent field ($\boldsymbol{n}_{123}$, $\boldsymbol{n}_{234}$, $\boldsymbol{n}_{341}$, and $\boldsymbol{n}_{412}$).

$2(1-x) \times (\frac{4\mu_{Tb}}{\sqrt{3}})/4 = 10.4(1-x)$ [μ_B/f.u] where $\mu_{Tb} = 9$ [μ_B/Tb] (state (i) in Fig. 4(c) of the main text). With increasing field above 2 T, we speculate that the Zeeman energy overcomes the single-ion anisotropy and hence the Tb moments gradually approach the field direction. We define θ as the angle between the Tb moments and the magnetic field direction (Fig. 8(a)), so that the magnetization is given as $M_{100} = 2(1-x)\mu_{Tb}\cos\theta$. In this way, we estimate the tilt angle of the Tb moment from the measured magnetization to calculate χ_{ijk} as discussed later.

Magnetization process for $H\|[111]$ is more complex. Similar to spin-ice materials, the magnetization for [111] field direction increases with increasing field towards $M_{KI} = 2 \times (1 - x) \times (\mu_{Tb} + \frac{\mu_{Tb}}{3})/4 = 6(1 - x)$ [μ_B/f.u], at which the kagomé ice state is realized (the state (ii)

in Fig. 4(c) in the main text). In other words, the 2-in 2-out state is preserved up to M_{KI}, resulting in the nearly the same curve as the case for $H\|[100]$. Above M_{KI}, the crossover occurs from (ii) 2-in 2-out state to (iii) 3-in 1-out state, at which the magnetization becomes $M_{3I1O} = 2(1-x) \times (\mu_{Tb} + \frac{\mu_{Tb}}{3} \times 3)/4 = 9(1-x)$ [μ_B/f.u]. Eventually, as the field increases further, Tb moments deviate from the local $\langle 111 \rangle$ axes and get aligned collinearly (the state (iv) in Fig. 4(c) of the main text). Between (iii) and (iv) states, the magnetization is written as $M_{111} = 2(1-x) \times (\mu_{Tb} + 3\mu_{Tb}\cos\theta)/4$ where θ is the angle between Tb moments and the field, as shown in Fig. 8(b).

Making use of the information of ~~Tb magnetic configurations~~ configurations of Tb magnetic moments as extracted above, we calculate the spin chirality of the Tb tetrahedron averaged over four Tb sites on the vertices of one tetrahedron. According to the theoretical calculation in the weak coupling region [20], the emergent magnetic field H_{eff} acting on the Mo-conducting electrons is given as

$$
\begin{aligned}
H_{eff} &\propto \langle \chi_{ijk} \rangle \\
&= \frac{1}{6N} \sum_{i,j,k \in \text{all sites}} \boldsymbol{S}_i \cdot (\boldsymbol{S}_j \times \boldsymbol{S}_k) \boldsymbol{F}(r_i, r_j, r_k) \cdot \boldsymbol{e}_z
\end{aligned}
$$
$$(3)$$

where

$$
\begin{aligned}
\boldsymbol{F}(\boldsymbol{r_i}, \boldsymbol{r_j}, \boldsymbol{r_k}) &= (\boldsymbol{a} \times \boldsymbol{b}/ab)I'(a)I'(b)I(c) \\
&+ (\boldsymbol{b} \times \boldsymbol{c}/bc)I(a)I'(b)I'(c) \\
&+ (\boldsymbol{c} \times \boldsymbol{a}/ca)I'(a)I(b)I'(c) \quad (4)
\end{aligned}
$$

$\boldsymbol{e}_z$ is the unit vector parallel to the field direction, $\boldsymbol{r}_i$, $\boldsymbol{r}_j$, and $\boldsymbol{r}_k$ are the positions of local spins at site i,j, and k, $\boldsymbol{a} = \boldsymbol{r}_i - \boldsymbol{r}_j$, $\boldsymbol{b} = \boldsymbol{r}_j - \boldsymbol{r}_k$, $\boldsymbol{c} = \boldsymbol{r}_k - \boldsymbol{r}_i$ are the distances between each site, $I(\boldsymbol{r})$ is the rapidly decreasing function of $\boldsymbol{r}$, and $I' = \frac{dI(r)}{dr}$. Thus we take into account only the nearest neighbor sites 1, 2, 3, and 4 in a single tetrahedron shown in Fig. 8(c)

$$
\langle \chi_{ijk} \rangle \propto \sum_{i,j,k \in 1,2,3,4} \boldsymbol{S}_i \cdot (\boldsymbol{S}_j \times \boldsymbol{S}_k) \boldsymbol{n}_{ijk} \cdot \boldsymbol{e}_z. \quad (5)
$$

where $\boldsymbol{n}_{ijk}$ is the unit vector defined by right hand screw rule when orbiting the site i, j, and k. The calculated results are plotted in Fig. 4(c) in the main text. We note that the spin chirality is not calculated but just a connecting straight line drawn between M_{KI} and M_{3I1O} in $H\|[111]$ (the state (ii) and (iii) in Figs. 4(b) and 4(c)), because the detailed magnetic state is not trivial. The calculated curves of the SSC based on the above simple assumption reasonably reproduce the experimental results of geometrical Hall components.

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