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Large Anomalous Hall Conductivity Derived from an f-Electron Collinear Antiferromagnetic Structure

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Appropriate symmetry breaking generates an anomalous Hall (AH) effect, even in antiferromagnetic (AFM) materials. Itinerant magnets with d electrons are typical examples that show a significant response. By contrast, the process by which a response emerges from f-electron AFM structures remains unclear. In this study, we show that an AFM material, $Ce₂C_uGe₆$, yields a large AH conductivity (AHC) of 550 Ω^{-1} cm⁻¹, which exceeds the values previously reported in d-electron AFM materials. Observed features, including the scaling relation against electrical conductivity, suggest that this AH transport is induced cooperatively by both intrinsic and extrinsic mechanisms derived from the AFM structure.

The symmetry argument has clarified that an anomalous Hall effect (AHE) occurs with specific antiferromagnetic (AFM) structures, irrespective of intrinsic and extrinsic mechanisms [1]. These types of AFM structures are represented by ferromagnetic (FM) point groups, which do not guarantee symmetric full compensation of spin configuration [1–3]. For the intrinsic mechanism, which is described by the Berry curvatures in momentum space [4–8], a major factor in obtaining large responses is the off-diagonal term of the Bloch states between different bands, which are affected strongly by spin–orbit interaction. This endows the conduction electrons with transverse anomalous velocity according to their spin directions [8, 9]. Therefore, itinerant magnets are naively considered suitable to generate this type of response. In fact, Fe is a typical example that exhibits a large intrinsic AH conductivity (AHC) of > 1000 Ω^{-1} cm⁻¹ for ferromagnets [10]. For AFM materials, Mn_3Z ($Z = Sn$, Ge) and NbMnP, in which the conduction electrons mainly originate from magnetic 3d electrons, exhibit large AHCs of $100 - 450 \Omega^{-1}$ cm⁻¹ by their intrinsic nature [11-15]. However, symmetrical consideration, which restricts the magnetic point group, does not indicate whether the magnetism originates from itinerant electrons or localized electrons. 4f electrons have strong spin–orbit coupling, which is advantageous for large Berry curvatures. However, they are generally not conductive. Whether the spin texture of localized 4f electrons can offer large Berry curvatures to the conduction electrons via exchange interaction is an open question. Another question is whether an extrinsic mechanism, which originates in skew- and sidejump scattering, contributes to AHC in AFM structures. In 5f-electron ferromagnets, the extrinsic contribution has been remarkably observed together with the intrinsic contribution [16]. The extrinsic mechanism should also be effective under an AFM structure, the symmetry of which is equivalent to ferromagnetism [1]. However, the magnitude of the AHC remains unclear. The manner in which a Hall response is generated by an f -electron AFM structure is intriguing. However, this type of example has

yet to be reported.

Here, we introduce $Ce₂CuGe₆$, whose AFM structure is described by the magnetic point group that allows an FM state [17]. $Ce₂CuGe₆$ crystalizes in an orthorhombic structure with the Cmce (or Cmca) space group (No. 64). This shows that the AFM transition at $T_N \simeq 15$ K is accompanied by a small net magnetization of ∼ 10[−]² μ_B/Ce along the c axis [17–20]. The magnetic structure with the propagation vector $Q = 0$ is represented by the Γ_3 irreducible representation, which corresponds to the B_{1q} representation or the magnetic point group $m'm'm$ [17]. This symmetry induces AFM components in the ab plane and FM components along the c axis, thereby generating the AHE even in an in-plane AFM structure. The actual magnetic structure of $Ce₂C_uGe₆$ is shown in

FIG. 1. Crystal and magnetic structure of $Ce₂C_uGe₆$ in the orthorhombic Cmce space group [17]. The $Q = 0$ magnetic structure is represented by a B_{1g} irreducible representation, which corresponds to the $m'm'm$ magnetic point group. This magnetic point group allows the FM component to emerge along the c axis, whereas the main components of 2 μ B/Ce are directed along the b axis to form a collinear AFM structure. The orange points in the left panel indicate the inversion centers of the crystal. This AFM alignment of the Ce moments breaks the \mathcal{PT} symmetry.

Fig. 1 [17], which is illustrated by VESTA [21]. The b-axis AFM components of 2 μ_B/Ce are dominant and consist of the AFM stacking of FM double layers. The Ce sites connected by the inversion symmetry, the center of which is located in the $z = 0$ and 0.5 planes, exhibit FM coupling. Accordingly, this magnetic structure preserves the space-inversion symmetry P , but the PT symmetry is broken. Here, $\mathcal T$ indicates time-reversal symmetry. The absence of the \mathcal{PT} symmetry is crucial for symmetry breaking to induce AHE. Note that the nonmagnetic atoms are not essential for the symmetry breaking, because the Ce atoms are located at a low-symmetric site (Wyckoff: 16g). The large ordered moment of $2\mu_B/Ce$ and the magnetic entropy of $\sim R \ln 2$ at T_N indicate the localized character of the $4f$ electrons [17, 19]. Therefore, $Ce₂CuGe₆$ is a suitable material to investigate how a Hall response is derived from the localized AFM structure. Another advantage in using this system is the large variety of similar crystal structures [18]. For example, $Ce₂PdGe₆$ and $Ce₂AuGe₆$ crystalize in the same structure as that of $Ce₂CuGe₆$. $Ce₂PdGe₆$ shows the AFM transition at $T_N \simeq 11.5$ K, where its magnetic structure has yet to be revealed. However, the same magnetic structure as that of $Ce₂CuGe₆$ can be expected from the similar magnetic character [20]. The physical properties of $Ce₂AuGe₆$ remain unknown.

In this study, we report Hall resistivity measurements for AFM materials Ce_2TGe_6 ($T = Cu$, Pd, and Au). We observe clear hysteresis in the field sweep of the Hall resistivities for $T = Cu$ and Pd, i.e., zero-field AHE. The estimated AHC at the lowest temperature is $\sim 550 \Omega^{-1}$ cm⁻¹ for Ce₂CuGe₆ and $\sim 100 \Omega^{-1}$ cm⁻¹ for Ce₂PdGe₆, which are comparable to those observed in ferromagnets. Our results show that the f-electron AFM structure generates a large Hall response cooperatively through both intrinsic and extrinsic mechanisms.

The platelike single crystals of Ce_2TGe_6 ($T = Cu$, Pd, and Au) are synthesized using a Bi-flux method from the ratio of Ce : T : Ge : Bi = $2 : 1 : 6 : 30$ [19, 20]. The single crystal of $Ce₂AuGe₆$ is grown for the first time. After generating electrical contacts for gold wires using the spot weld method, we measured the electrical and Hall resistivities using a standard four-probe method. The Hall resistivity was antisymmetrized against magnetic fields to remove the longitudinal component induced by contact misalignment. We measured magnetization in the temperature range of $2 - 300$ K and the field range up to ± 5 T using a Quantum Design magnetic property measurement system.

Figure $2(a)$ shows the temperature dependences of the electrical resistivities for Ce_2TGe_6 (T = Cu, Pd, and Au). They all exhibited a broad shoulder at approximately 80 K, most likely due to the crystal-field splitting of the 4f multiplet, which was followed by clear decreases below T_N . Anisotropy between ρ_{xx} and ρ_{yy} , which was checked for $Ce₂CuGe₆$, was weak, particularly when below T_N . Here, the $a, b,$ and c axes correspond to x , y , and z , respectively. The residual re-

FIG. 2. (a) Electrical resistivities for Ce_2TGe_6 ($T = Cu$, Pd, and Au). Clear AFM transitions at T_N can be observed. The T_N was estimated to be 15.0 K for $T = Cu$, 11.3 K for $T = Pd$, and 9.3 K for $T = Au$. Anisotropy between ρ_{xx} and ρ_{yy} was weak. (b) Temperature dependence of magnetic susceptibility for three compounds. For $H \parallel b$, typical AFM-like behaviors are exhibited, whereas the FM components were induced for $H \parallel c$ for all compounds. (c,d) Magnetization curves at 2 K for $H \parallel b$ and $H \parallel c$. The metamagnetic transitions at approximately 1 T along the b axis for three compounds suggests they have a similar magnetic structure. For $H \parallel c$, small spontaneous magnetizations were induced: $3 \times 10^{-2} \mu_B/\text{Ce}$ for Ce₂CuGe₆, 1×10^{-3} μ_B/Ce for Ce₂PdGe₆, and 4×10^{-2} μ_B /Ce for Ce₂AuGe₆.

sistivity of Ce₂CuGe₆ ($\rho_0 = 3 \mu \Omega$ cm) was comparable to that in the previous study [19], and the residual resistivities of other compounds were $\rho_0 = 19 \mu \Omega \text{cm}$ for T = Pd and $\rho_0 = 1.5 \mu \Omega$ cm for T = Au. As Fig. 2(b) shows, the magnetic susceptibility for $H \parallel b$ showed clear kinks at T_N , whereas increases below T_N were observed for $H \parallel c$ [20]. Figures 2(c) and 2(d) show magnetization curves at 2 K for $H \parallel b$ and $H \parallel c$, respectively. For $H \parallel b$, all the compounds showed metamagnetic transition at approximately 1 T, above which magnetization of \sim 1.7 μ_B/Ce was comparable to the ordered moment of $2 \mu_B/\text{Ce}$ for Ce₂CuGe₆ [17]. Under H | c, hysteresis appeared with all compounds with small spontaneous magnetization: $3 \times 10^{-2} \mu_B/{\rm Ce}$ for $T = {\rm Cu}, 1 \times 10^{-3} \mu_B/{\rm Ce}$ for $T = \text{Pd}$, and $4 \times 10^{-2} \mu_B/\text{Ce}$ for $T = \text{Au}$. These small net magnetizations along the c axis are allowed in the magnetic point group $m'm'm$ [17]. The similar magnetic properties among Ce_2TGe_6 (T = Cu, Pd, and Au) suggested that the magnetic structures for the three compounds similarly contained the $m'm'm$ component. The observed spontaneous magnetizations corresponded to 3.5 mT for $Ce₂CuGe₆$, 0.1 mT for $Ce₂PdGe₆$, and 5 mT for $Ce₂AuGe₆$, which were comparable to those in $Mn₃Z$ (Z = Sn, Ge); that is, they were too small to yield large AHCs [22, 23].

FIG. 3. (a)- (c) Field dependences of Hall resistivities $\rho_H = \rho_{yx} = -\rho_{xy}$ for $T = Cu$, Pd, and Au. In the PM state, a linear field dependence of ρ_H was observed, whereas a clear hysteresis appeared below T_N for $T = Cu$ and Pd. These zero-field AHEs show the opposite-sign values between $T = Cu$ and Pd. In $T = Au$, the green and orange curves show the field sweep in the opposite direction. Absence of the zero-field AHE in $T = Au$ with the largest spontaneous magnetization suggested that the AHE observed in $T = Cu$ and Pd did not arise from the net magnetization.

Figures $3(a)-(c)$ show the field dependences of the Hall resistivities $\rho_H = \rho_{yx} = -\rho_{xy}$, which could be nonzero from the magnetic point group $m'm'm$. To switch the AFM domains coupled with small net magnetization, magnetic fields were applied along the c axis. In the paramagnetic (PM) state above T_N , a linear field dependence of ρ_H was observed. Below T_N , a clear hysteresis emerged for $T=$ Cu and Pd, showing that a zero-field AHE was derived from their magnetic structure. For $T =$ Au, the zero-field AHE was not confirmed within the experimental resolution. A critical feature was observed at 10 K for $T = Cu$, where the domain selected by the positive magnetic field generated the positive field-induced component and the negative zero-field AHE. This clearly demonstrated that the zero-field AHE could not be explained by the magnetization origin.

Figure 4(a) shows the temperature dependence of the field-induced components in ρ_H measured at ± 1 T, where the zero-field components induced below T_N are not contained. In the PM state, the positive ρ_H increased with decreasing temperature for both $T =$ Pd and Cu compounds. These field-induced components most likely derived from incoherent skew-scattering, which has often been seen in f-electron systems [24, 25]. It generally follows $\rho_H \propto \chi \rho$, where χ is the magnetic susceptibility, but the observed data did not obey this relation probably because the ordinary Hall effect also depends on temperature. The itinerary of f-electrons has been known to induce a peak structure in the temperature variation of ρ_H at the onset of the coherent state [25]. Absence of this peak structure above T_N is qualitatively consistent with the remarkable localized character of the f electrons in

 $Ce₂TGe₆$. At low temperatures, the field-induced component was significantly suppressed for $T = Cu$, reflecting the significant decrease in electrical resistivity due to the high-quality sample. The zero-field AHE components were estimated by $\rho_{\rm AH} = [\rho_{\rm H}(+H \rightarrow 0) - \rho_{\rm H}(-H \rightarrow$ 0]/2 and are plotted against temperature in Fig. 4(b). They showed nonzero values below T_N for $T = Cu$ and Pd and exhibited the maximum at slightly below T_N , thus approximating a constant at low temperatures. Absence of the anomaly in $T = Au$ with the largest net magnetization indicated that these zero-field contributions did not originate simply in the net magnetization. The values of $\rho_{AH} \simeq -5 \times 10^{-3} \mu \Omega$ cm for Ce₂CuGe₆ and $\rho_{\rm AH} \simeq 4 \times 10^{-2} \mu \Omega$ cm for Ce₂PdGe₆ were much smaller than those observed in Mn_3Sn , Mn_3Ge , and low-quality NbMnP [11–14] but were comparable to that observed in high-quality NbMnP [15]. Note that a quantity determined by the Berry curvatures is not ρ_{AH} but σ_{AH} . The σ_{AH} is connected to ρ_{AH} through $\sigma_{\text{AH}} = \rho_{\text{AH}}/\rho^2$. Therefore, the intrinsic mechanism, where the dissipationless intrinsic σ_{AH}^{int} is independent of ρ , yields the relation of $\rho_{\text{AH}}^{int} \propto \rho^2$. Obviously, ρ_{AH}^{int} decreases in high-quality samples. Figure 4(c) shows the estimated $|\sigma_{\text{AH}}|$. The $|\sigma_{\text{AH}}|$'s increased toward low temperatures, reaching maximums of ∼ 550 Ω⁻¹cm⁻¹ for T = Cu and ~ 100 Ω⁻¹cm⁻¹ for $T = \text{Pd}$. Notably, σ_{AH} for $T = \text{Cu}$ showed a steep increase below ∼ 10 K.

The value of $Ce₂CuGe₆$ (~ 550 $\Omega^{-1}cm^{-1}$) was higher than those of d-electron AFM material Mn₃Sn (~ 140) Ω^{-1} cm⁻¹) and Mn₃Ge (~ 380 Ω^{-1} cm⁻¹) [11-13]. To identify the origin of this large AHC through the scaling relation, the σ_{AH} versus $\sigma = 1/\rho_{yy}$ is plotted in

FIG. 4. (a) Temperature dependence of the field-induced component of ρ_H , measured at ± 1 T. Below T_N , they were estimated from the field-sweep data by subtracting the zerofield components (open symbols). This contribution is considered to derive from incoherent skew-scattering, which has often been observed in many f-electron systems. (b,c) Temperature dependences of the contribution at zero-field AHE, ρ_{AH} and $|\sigma_{\text{AH}}|$. For $T = \text{Pd}$, σ_{AH} was estimated by assuming $\rho_{xx} \simeq \rho_{yy}$. The $|\sigma_{AH}| \sim 550 \Omega^{-1}$ cm⁻¹ for $T = Cu$ was higher than those of prototype d-electron AFM materials Mn3Sn and Mn3Ge. Red dotted and solid curves indicate the contributions estimated for the intrinsic and extrinsic mechanisms for $T = Cu$, respectively. (d) The scaling relation in $|\sigma_{\text{AH}}|$ versus σ . $M\sigma_{\text{AH}}$ is also plotted to correct the temperature dependence of the order parameter, where $\widetilde{M} = M_{\text{AFM}}(T=0)/M_{\text{AFM}}(T)$. The $\widetilde{M}\sigma_{\text{AH}}$ follows $\sigma_{\text{AH}}^{int} + \sigma_{\text{AH}}^{ext}$
(black curve), where σ_{AH}^{int} is independent of σ , and σ_{AH}^{ext} obeys σ^2 . It indicates that the large $|\sigma_{\text{AH}}|$ in Ce₂CuGe₆ originates in both intrinsic and extrinsic mechanisms.

Fig. 4(d), as σ_{AH}^{int} is independent of σ when the order parameter is fully grown [8, 26]. Here, we assume $\sigma_{\text{AH}} \propto M_{\text{AFM}}(T) \sigma^n$, where $M_{\text{AFM}}(T)$ indicates the temperature variation of the order parameter in $Ce₂C_uGe₆$ [17]. To identify the exponent n, we also plotted $M\sigma_{\rm AH}$, where $M = M_{\text{AFM}}(0)/M_{\text{AFM}}(T)$. This includes a correction term to correspond to the case of the fully grown order parameter, because the data shown in Fig. 4(d) include those slightly below T_N . The σ_{AH} and corrected $M\sigma_{\rm AH}$ show the obvious σ dependence, indicating that the extrinsic contribution was contained. Previous systematical investigations of Fe films have clarified that the extrinsic contribution σ_{AH}^{ext} , which includes both skewand side-jump scattering, follows $\sigma_{AH}^{ext} \propto \sigma^2$ in the temperature variation [27]. This corresponds to ρ_{AH}^{ext} being temperature independent. As the black curve shows in Fig. 4(d), $M\sigma_{\text{AH}}$ for Ce₂CuGe₆ followed the summation of two contributions, $\sigma_{\text{AH}}^{int} + \sigma_{\text{AH}}^{ext}$. The σ_{AH}^{int} was estimated to be $\sim 80 \Omega^{-1} \text{cm}^{-1}$ (dotted red line), whereas

 σ_{AH}^{ext} was $5 \times 10^{-9} \sigma^2$ (solid red line). This extrinsic term corresponds to $\rho_{\text{AH}}^{ext} = -5 \times 10^{-3} \mu \Omega \text{cm}$, which should be temperature-independent at low temperatures. This analysis suggests that ρ_{AH} shown in Fig. 4(b) was dominated by the extrinsic contribution at low temperatures. The intrinsic contribution in ρ_{AH} is significant just below T_N and is suppressed at low temperatures because of $\rho_{AH}^{int} \propto \rho^2$. In Fig. 4(c), the intrinsic and extrinsic contributions in σ_{AH} , which are estimated under the assumption of $\sigma_{\text{AH}} \propto M_{\text{AFM}}(T) \sigma^n$, are shown. The extrinsic contribution appears at low temperatures, being consistent with the two-step development of σ_{AH} .

Both the extrinsic and intrinsic mechanisms deriving from the AFM structure are allowed when their symmetries are equivalent to those of ferromagnets [1]. A study in the Fe films has suggested that ρ_{AH}^{ext} is determined by the residual resistivity ρ_0 [27]. For Ce₂CuGe₆, $ρ_{\text{AH}}^{ext} = -5 \times 10^{-3} \mu \Omega \text{cm}$ was obtained for $ρ_0 \sim 3 \mu \Omega \text{cm}$. For the Fe film, $\rho_0 = 3 \mu \Omega$ cm yielded $\rho_{AH}^{ext} = -4.8 \times 10^{-3}$ $\mu\Omega$ cm, which consists of skew-scattering contribution $\rho_{\text{AH}}^{skew} = -11.1 \times 10^{-3} \mu \Omega \text{cm}$ and side-jump contribution $\rho_{\text{AH}}^{side} = 6.3 \times 10^{-3} \mu \Omega \text{cm}$ [27]. This comparison means that the extrinsic contribution in $Ce₂C_uGe₆$ is comparable to that of the conventional itinerant ferromagnet, although the net magnetization is two orders smaller. The absence of the obvious AHE in $T = Au$ clearly shows that the AHE in $Ce₂CuGe₆$ is not relevant to the net magnetization. Therefore, the observed AHE was considered as deriving from the AFM structure through both intrinsic and extrinsic mechanisms. This study suggests that the conduction bands in these systems show spin splitting, which reflects the symmetry of the AFM structure, via the exchange interaction with the f electrons. The opposite sign in ρ_{AH} between $T = Cu$ and Pd indicates a difference in the band topology. Investigations of the band structure and its topology are crucial to understanding the mechanism of the observed AHE.

In conclusion, we investigated AHE for $Ce₂C_uGe₆$, whose AFM structure is described by the FM magnetic point group, together with isostructural $Ce₂PGGe₆$ and $Ce₂AuGe₆$. We observed the clear zero-field AHE accompanied by hysteresis loops for $Ce₂CuGe₆$ and $Ce₂PdGe₆$. Despite the weak net magnetization, the estimated AHC of \sim 550 Ω⁻¹cm⁻¹ was comparable to those of itinerant ferromagnets. Careful analysis suggested that both the intrinsic and extrinsic mechanisms contributed to the AHE in $Ce₂CuGe₆$, where the intrinsic AHC was estimated to be $\sim 80 \Omega^{-1} \text{cm}^{-1}$. The following two important findings were derived from this study. First, the relatively large intrinsic contribution emerged even from the localized f-electron AFM structure. Second, the extrinsic contribution from the AFM structure was confirmed to be comparable to that observed in ferromagnets. These findings can act as guidelines for obtaining a fuller understanding of the AHE and motivate further development of functional materials that generate FM-like responses in f-electron systems.

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