We report the observation of the higher-order thermoelectric conversion based on a magneto-Thomson effect. By means of thermoelectric imaging techniques, we directly observed the temperature change induced by the Thomson effect in a polycrystalline Bi$_8$Sb$_{12}$ alloy under a magnetic field and found that the magnetically enhanced Thomson coefficient can be comparable to or even larger than the Seebeck coefficient. Our experiments reveal the significant contribution of the higher-order magnetothermoelectric conversion, opening the door to “nonlinear spin caloritronics.”

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The thermoelectric Thomson effect was predicted in the middle of the 19th century by William Thomson, known as Lord Kelvin [1,2]. When a charge current with the density $j_c$ and a temperature gradient $\nabla T$ are applied to a conductor, the Thomson effect induces heat release or absorption proportional to $j_c$ and $\nabla T$ [Fig. 1(a)]; the heat production rate per unit volume due to the Thomson effect is described as

$$ q = -\tau j_c \cdot \nabla T, \tag{1} $$

where $\tau$ is the Thomson coefficient. The Thomson effect originates from the simultaneous operation of the Seebeck and Peltier effects, and appears when the temperature $T$ dependence of the Seebeck coefficient $S$ in a conductor is finite. If the charge current flows through the spatial gradient of $S$, the self-induced Peltier effect modulates the temperature in response to the charge current. Thus, the Thomson coefficient is related to $S$ and the Peltier coefficient $\Pi$ as

$$ \tau = \frac{d\Pi}{dT} - S = T \frac{dS}{dT}. \tag{2} $$

This is the first Thomson (or Kelvin) relation, which is derived from the energy conservation and the second Thomson relation, i.e., the Onsager reciprocal relation between the Seebeck and Peltier effects: $\Pi = ST$ [3–6].

The Seebeck and Peltier coefficients are known to be dependent on a magnetic field or a magnetization direction. Such magnetothermoelectric effects are one of the central topics in the field of spin caloritronics [7,8]. However, the magnetothermoelectric effects have been investigated only in a linear response regime, and their nonlinear effects remain to be observed. In nonlinear spin caloritronics, the temperature derivative of magnetothermoelectric and/or thermostop conversion coefficients plays a key role. In fact, Eq. (2) suggests that, if a conductor exhibits the magneto-Seebeck and Peltier effects [9] with finite $T$ dependence, $\tau$ may also depend on a magnetic field or magnetization (note that $\Pi = ST$ holds under a magnetic field if the asymmetric field dependence of the Seebeck coefficient, known as the Umkehr effect, is absent [3]). This is a nonlinear magnetothermoelectric conversion phenomenon that should be called a magneto-Thomson effect (MTE).

In this study, we report the observation of the MTE in a nonmagnetic conductor under a magnetic field and reveal its significant contribution. If the MTE appears, the magnitude of the Thomson-effect-induced temperature change is modulated by a magnetic field [Fig. 1(b)]. To demonstrate this effect, we use a polycrystalline Bi$_8$Sb$_{12}$ alloy since it shows the large temperature and magnetic field dependences of the Seebeck coefficient [10–16], which fills the requirements for the appearance of the MTE.

To achieve the direct observation of the MTE, it is important to establish a versatile measurement method for the Thomson effect. We realized highly sensitive pure detection of the temperature change induced by the Thomson effect by means of the thermoelectric imaging technique based on the lock-in thermography (LIT) [17–20]. In the LIT measurements, we measure thermal images of a sample surface while applying a square-wave-modulated ac charge current with the amplitude $J_c$, frequency $f$, and zero dc offset to the sample and extract temperature change oscillating with the same frequency as the current through Fourier analysis. Here, the obtained thermal images are transformed into the lock-in amplitude $A$ and phase $\phi$ images. This analysis allows us to separate the contribution of thermoelectric effects ($\alpha J_c$) from that of

$$ \text{CMAP} : 0031-9007/125(10)/106601(6) \text{ DOI: 10.1103/PhysRevLett.125.106601} \text{ © 2020 American Physical Society} \text{ PHYSICAL REVIEW LETTERS 125, 106601 (2020)} \text{ Observation of the Magneto-Thomson Effect} \text{ Ken-ichi Uchida},^{1,2,3,*} \text{ Masayuki Murata},^{4} \text{ Asuka Miura},^{1} \text{ and Ryo Iguchi}^{1} \text{ 1National Institute for Materials Science, Tsukuba 305-0047, Japan } \text{ 2Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan } \text{ 3Center for Spintronics Research Network, Tohoku University, Sendai 980-8577, Japan } \text{ 4National Institute of Advanced Industrial Science and Technology, Tsukuba, 305-8568, Japan } \text{ (Received 20 June 2020; accepted 29 July 2020; published 2 September 2020)} \text{ We report the observation of the higher-order thermoelectric conversion based on a magneto-Thomson effect. By means of thermoelectric imaging techniques, we directly observed the temperature change induced by the Thomson effect in a polycrystalline Bi$_8$Sb$_{12}$ alloy under a magnetic field and found that the magnetically enhanced Thomson coefficient can be comparable to or even larger than the Seebeck coefficient. Our experiments reveal the significant contribution of the higher-order magnetothermoelectric conversion, opening the door to “nonlinear spin caloritronics.” DOI: 10.1103/PhysRevLett.125.106601} \text{ The thermoelectric Thomson effect was predicted in the middle of the 19th century by William Thomson, known as Lord Kelvin [1,2]. When a charge current with the density } j_c \text{ and a temperature gradient } \nabla T \text{ are applied to a conductor, the Thomson effect induces heat release or absorption proportional to } j_c \text{ and } \nabla T \text{ [Fig. 1(a)]; the heat production rate per unit volume due to the Thomson effect is described as } \dot{q} = -\tau j_c \cdot \nabla T, \tag{1} \text{ where } \tau \text{ is the Thomson coefficient. The Thomson effect originates from the simultaneous operation of the Seebeck and Peltier effects, and appears when the temperature } T \text{ dependence of the Seebeck coefficient } S \text{ in a conductor is finite. If the charge current flows through the spatial gradient of } S, \text{ the self-induced Peltier effect modulates the temperature in response to the charge current. Thus, the Thomson coefficient is related to } S \text{ and the Peltier coefficient } \Pi \text{ as } \tau = \frac{d\Pi}{dT} - S = T \frac{dS}{dT}. \tag{2} \text{ This is the first Thomson (or Kelvin) relation, which is derived from the energy conservation and the second Thomson relation, i.e., the Onsager reciprocal relation between the Seebeck and Peltier effects: } \Pi = ST [3–6]. \text{ The Seebeck and Peltier coefficients are known to be dependent on a magnetic field or a magnetization direction. Such magnetothermoelectric effects are one of the central topics in the field of spin caloritronics [7,8]. However, the magnetothermoelectric effects have been investigated only in a linear response regime, and their nonlinear effects remain to be observed. In nonlinear spin caloritronics, the temperature derivative of magnetothermoelectric and/or thermostop conversion coefficients plays a key role. In fact, Eq. 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To achieve the direct observation of the MTE, it is important to establish a versatile measurement method for the Thomson effect. We realized highly sensitive pure detection of the temperature change induced by the Thomson effect by means of the thermoelectric imaging technique based on the lock-in thermography (LIT) [17–20]. In the LIT measurements, we measure thermal images of a sample surface while applying a square-wave-modulated ac charge current with the amplitude } J_c, \text{ frequency } f, \text{ and zero dc offset to the sample and extract temperature change oscillating with the same frequency as the current through Fourier analysis. Here, the obtained thermal images are transformed into the lock-in amplitude } A \text{ and phase } \phi \text{ images. This analysis allows us to separate the contribution of thermoelectric effects (} \alpha J_c \text{) from that of } \text{ }}$
During the LIT measurements, an external magnetic field $H_{\text{ext}}$ was applied to the slab along the $x$ direction, where $H_{\text{ext}}$ is the magnitude of the external magnetic field. The Thomson signal is reversed across the center of the slab, while the direction of the current is reversed between $j_{\text{LIT}}$. The temperature gradients increase in proportion to both the temperature gradient along the $y$ direction and the Joule heating generated by the ac charge current $J_{\text{LIT}}$. The sign of the current-induced temperature modulation at $R_1$ is opposite to that at $R_2$ because the $\phi_{\text{TE}}$ difference between $R_1$ and $R_2$ is approximately $180^\circ$ [Fig. 2(g)]. We confirmed that the temperature distribution is as designed; the temperature distribution is little affected by the Joule heating due to the charge current in the Bi$_{12}$Sb$_{12}$ slab. We start with demonstrating the validity of our LIT-based method by performing the measurements of the Thomson effect in the absence of a magnetic field. Figure 2(e) shows the steady-state temperature profiles along the $y$ direction for the Bi$_{12}$Sb$_{12}$ slab at $J_{\text{c}} = 100.0$ mA and $f = 1.0$ Hz. The clear charge-current-induced temperature modulation was observed at $P = 30$ mW [Fig. 2(b)]. By subtracting the Peltier background from the signals in Fig. 2(b), we obtained the $A_{\text{TE}}$ and $\phi_{\text{TE}}$ images in Fig. 2(d); hereafter, we focus on the subtracted images. As shown in the $A_{\text{TE}}$ profiles along the $y$ direction in Fig. 2(f), the magnitude of the current-induced temperature modulation exhibits the maximum values around $R_1$ and $R_2$. We confirmed that the magnitude of the temperature modulation increases in proportion to both the charge current and temperature gradients applied to the Bi$_{12}$Sb$_{12}$ slab [Figs. 2(h) and 2(i)], where the temperature gradients along the $y$ direction were estimated by fitting the steady-state temperature profiles in $R_1$ and $R_2$ with linear functions [Fig. 2(e)]. Importantly, the sign of the current-induced temperature modulation at $R_1$ is opposite to that at $R_2$ because the $\phi_{\text{TE}}$ difference between $R_1$ and $R_2$ is approximately $180^\circ$ [Fig. 2(g)]. These behaviors are consistent with the features of the Thomson effect. We note that the spatial distribution of the temperature modulation in Figs. 2(d), 2(f), and 2(g) appears as a consequence of the facts that the sign of the Thomson signal is reversed across the center of the Bi$_{12}$Sb$_{12}$ slab and both the ends of the slab are thermally connected to the heat baths.

Now, we are in a position to investigate the MTE by means of the LIT-based method. In Figs. 3(a) and 3(b), we compare the $A_{\text{TE}}$ and $\phi_{\text{TE}}$ images at $[\mu_0 H] = 0.0$ T with those at $[\mu_0 H] = 0.9$ T. Here, we focus on the temperature modulation showing $H$-even dependence because the asymmetric field dependence of the Seebeck coefficient due to the Umkehr effect is negligibly small in polycrystalline Bi-Sb alloys around room temperature [16]; the extraction of temperature modulation signals with $H$-even

![FIG. 1. Thomson and magneto-Thomson effects. (a) Schematic of the conventional Thomson effect. When a charge current $J_j$ and a temperature gradient $\nabla T$ are applied to a conductor, the Thomson effect induces heat absorption or release depending on the scalar product of $J_j$ and $\nabla T$. (b) Schematic of the magneto-Thomson effect. The heat production rate due to the Thomson effect can be modulated by applying a magnetic field $H$ to a conductor.](image-url)
dependence is necessary for separating the Thomson signal from the contribution of the Ettingshausen effect with $H$-odd dependence (see Fig. S1 in the Supplemental Material [26] for the details of background subtraction procedures in the presence of a magnetic field). The Thomson signal in the Bi$_{88}$Sb$_{12}$ slab was observed to be enhanced by applying the magnetic field. Figure 3(c) shows the $\eta_{\text{TE}}$ values as a function of $|\mu_0 H|$ at $f = 1.0$ Hz, where $\eta_{\text{TE}} = |A_{\text{TE}}/J_c \nabla y T|$ with $J_c$ and $\nabla y T$, respectively, being the square-wave amplitude of the charge current density and the temperature gradient along the $y$ direction is proportional to $\tau$ [see Eq. (1) and Sec. S3 in the Supplemental Material [26,30], where the relation between $\eta_{\text{TE}}$ and $\tau$ is analytically derived]. We found that the magnitude of the Thomson signal monotonically increases with increasing $|\mu_0 H|$ and the $|\mu_0 H|$ dependence of $\eta_{\text{TE}}$ is independent of $f$, as shown in Fig. S2 in Supplemental Material [26]. Surprisingly, the enhancement ratio of the Thomson signal for the Bi$_{88}$Sb$_{12}$ slab reaches $|\eta_{\text{TE}}(0.9 \ T) - \eta_{\text{TE}}(0.0 \ T)|/\eta_{\text{TE}}(0.0 \ T) = 90.3 \pm 8.3\%$ at room temperature. This is much greater than the field dependence of the linear-response transport coefficients: $|\sigma(0.9 \ T) - \sigma(0.0 \ T)|/\sigma(0.0 \ T) = -19.4\%$ for the electrical conductivity $\sigma$ [Fig. S3(a) in Supplemental
Material [26], \([\kappa(0.9 \text{ T}) - \kappa(0.0 \text{ T})]/\kappa(0.0 \text{ T}) = -8.3\%\) for the thermal conductivity \(\kappa\) [Fig. S3(b)], and \([S(0.9 \text{ T}) - S(0.0 \text{ T})]/S(0.0 \text{ T}) = 20.5\%\) for the Seebeck coefficient [Fig. 4(a)] of the Bi\(_{88}\)Sb\(_{12}\) slab at \(T = 300\) K. We also note that, in isotropic polycrystalline alloys, the MTE properties are not changed when the \(H\) direction is rotated in the \(z\)-\(x\) plane, where \(H\) is perpendicular to the charge current. In contrast, the magnetic field dependence of the Thomson signal is expected to be reduced when \(H\) is applied along the charge current (\(y\) direction), since the MTE in a nonmagnetic conductor originates from the Lorentz force acting on charge carriers.

To further verify the giant MTE, we systematically measured the \(H\) and \(T\) dependences of \(S\) for the same Bi\(_{88}\)Sb\(_{12}\) slab, where \(|\mu_0H| < 1.0\) T and \(270\) K < \(T < 350\) K. As shown in Figs. 4(a) and 4(b), \(S\) is negative in the \(\mu_0H\) and \(T\) ranges and \(|S|\) increases (decreases) with increasing \(H\) (increasing \(T\)). The \(H\)-even dependence of \(S\) is consistent with the expected behavior of the magneto-Seebeck effect, where the asymmetric component is negligibly small. The systematic data on the magneto-Seebeck effect allows us to estimate the \(|\mu_0H|\) dependence of \(\tau\) based on Eq. (2). We found that \(\tau\) monotonically increases with increasing \(|\mu_0H|\), which is similar to the \(|\mu_0H|\) dependence of \(\eta_{\text{TE}}\) [compare Fig. 4(c) with Fig. 3(c)]. This good consistency between the directly observed MTE and the magneto-Seebeck effect confirms the validity of the first Thomson relation under a magnetic field. The observed Thomson coefficient of our polycrystalline Bi\(_{88}\)Sb\(_{12}\) reaches \(\tau = 98.3 \times 10^{-6} \text{ V K}^{-1}\) at \(|\mu_0H| = 0.9\) T and \(T = 300\) K, which is comparable to its Seebeck coefficient. Our finding indicates that the Thomson coefficient enhanced by the MTE can even be larger than the Seebeck coefficient under stronger magnetic fields, highlighting the significant contribution of the higher-order magnetothermoelectric conversion.

As shown above, we have investigated the MTE in a nonmagnetic conductor under a magnetic field. It is also interesting to measure the Thomson effect in magnetic materials. The observation of the anisotropic magneto-
Seebeck and Peltier effects in ferromagnetic metals [20,31–36], in which the Seebeck and Peltier coefficients depend on the direction of spontaneous magnetization, suggests the possible existence of an anisotropic MTE; however, this phenomenon is yet to be observed. To investigate the anisotropic MTE, we performed the same LIT measurements using ferromagnetic Ni$_{50}$Pt$_5$ and Ni slabs, which exhibit the substantially large anisotropic magneto-Seebeck and Peltier effects at room temperature [20,36]. As shown in Figs. S4(a)–S4(c) in Supplemental Material [26], we observed clear temperature modulation signals due to the Thomson effect in the Ni$_{50}$Pt$_5$ and Ni slabs and found that the sign of the temperature modulation for the ferromagnets is opposite to that for the Bi$_{88}$Sb$_{12}$ slab [compare Figs. S4(b) and S4(c) in Ref. [26] with Figs. 3(a) and 3(b)]. This behavior can be explained by the fact that the $T(dS/dT)$ values for Ni$_{50}$Pt$_5$ and Ni are opposite in sign to those for Bi$_{88}$Sb$_{12}$ (Figs. 4 and S5 in Supplemental Material [26]). However, we found that the Thomson signals for Ni$_{50}$Pt$_5$ and Ni are an order of magnitude smaller than those for Bi$_{88}$Sb$_{12}$ and almost independent of the magnetization [Figs. S4(d) and S4(e) [26]], indicating that no anisotropic MTE signals appear in these ferromagnets within the margin of experimental errors. To realize the observation of the anisotropic MTE, further physics research for understanding its microscopic mechanism and materials science research for exploring ferromagnets with large anisotropy of the Thomson coefficient are necessary.

In summary, we have realized the direct observation of the MTE in the polycrystalline Bi$_{88}$Sb$_{12}$ slab under a magnetic field by means of the thermoelectric imaging technique based on the LIT. The temperature change induced by the Thomson effect in the Bi$_{88}$Sb$_{12}$ slab was found to be strongly enhanced with increasing the magnetic field; the enhancement ratio at room temperature reaches 90.3 ± 8.3% under a relatively low magnetic field of 0.9 T. The giant MTE clarifies the importance of the nonlinear magnetothermoelectric conversion and provides an unconventional concept for thermal energy engineering. The establishment of the direct measurement method for the MTE is also important for further development of spin caloritronics. Our technique is directly applicable to the measurements of the anisotropic MTE in magnetic materials, as discussed above. In addition to the anisotropic MTE, various higher-order thermoelectric and thermospin conversion phenomena, e.g., Thomson effects for spin currents and spin waves [37,38], should exist in magnetic and spintronic materials if the temperature derivative of magnetothermoelectric and/or thermospin conversion coefficients is finite. This work is the first step for investigating physics and applications of such unexplored phenomena.

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