We experimentally investigate longitudinal and transverse second-harmonic voltage response to alternating electric current for a magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$. In contrast to the previously observed Berry-curvature induced nonlinear Hall effect for non-magnetic Weyl and Dirac semimetals, the second-harmonic transverse voltage demonstrates sophisticated interplay of different effects for Co$_3$Sn$_2$S$_2$. In high magnetic fields, it is of Seebeck-like square-B law, while the low-field behavior is found to be linear and sensitive to the direction of sample magnetization. The latter can be expected both for the nonlinear Hall effect and for the surface state contribution to the Seebeck effect in Weyl semimetals. Thus, thermoelectric effects are significant in Co$_3$Sn$_2$S$_2$, unlike non-magnetic Weyl and Dirac materials.

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Recent interest to the time-reversal-invariant nonlinear Hall (NLH) effect [1–14] is a part of a broad research area of topological systems. In zero magnetic field, a nonlinear Hall-like current arises from the Berry curvature, which can be regarded as a magnetic field in momentum space. It leads to a quadratic response to ac excitation current, so NLH effect should appear as a nonzero second-harmonic voltage without magnetic field. Since Berry curvature concentrates in regions where two or more bands cross [15], topological systems are the obvious candidates to observe the NLH effect [1]. It has been experimentally demonstrated for monolayer transitional metal dichalcogenides [16, 17] and for three-dimensional Weyl and Dirac semimetals [18].

Dirac semimetals host special points of Brillouin zone with three-dimensional linear dispersion [15]. In Weyl semimetals, by breaking time reversal or inversion symmetries, every Dirac point splits into two Weyl nodes with opposite chiralities. First experimentally investigated Weyl semimetals (WSMs) were noncentrosymmetric crystals with broken inversion symmetry. Even in this case, a second-harmonic quadratic signal can also originate from the thermoelectric Seebeck effect [19, 20]. When the magnetic field is perpendicular to the temperature gradient, it leads to quadratic-B correction in the Seebeck coefficient [21, 22]. In contrast to these calculations, a second-harmonic NLH voltage shows [18] odd-type dependence on the direction of the magnetic field. Thus, the magnetic field measurements allow distinguishing the NLH effect from the thermoelectric response.

There are only a few candidates [23–26] of magnetically ordered WSMs with broken time-reversal symmetry. Recently, giant anomalous Hall effect was reported [27, 28] for the kagome-lattice ferromagnet Co$_3$Sn$_2$S$_2$, as an indication for the existence of a magnetic Weyl phase. Sophisticated regimes of second-harmonic response should also be expected in Weyl semimetals with broken time reversal symmetry. In addition to the expected Berry curvature contribution to the Hall-like currents, the chiral anomaly contribution to second-harmonic generation in the lowest order is linearly proportional to the applied magnetic field [29]. Moreover, in magnetic materials, Nernst voltage can be generated normally to the temperature gradient even without an external magnetic field, which is known as anomalous Nernst effect (ANE). ANE was reported for different Co$_3$Sn$_2$S$_2$ thermoelectric devices [30–32].

Here, we experimentally investigate longitudinal and transverse second-harmonic voltage response to alternating electric current for a magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$. In contrast to the previously observed Berry-curvature induced nonlinear Hall effect for non-magnetic Weyl and Dirac semimetals, the second-harmonic transverse voltage demonstrates sophisticated interplay of different effects for Co$_3$Sn$_2$S$_2$. In high magnetic fields, it is of Seebeck-like square-B law, while the low-field behavior is found to be linear and sensitive to the direction of sample magnetization. The latter can be expected both for the nonlinear Hall effect and for the surface state contribution to the Seebeck effect in Weyl semimetals. Thus,
thermoelectric effects are significant in $\text{Co}_3\text{Sn}_2\text{S}_2$, unlike non-magnetic Weyl and Dirac materials.

$\text{Co}_3\text{Sn}_2\text{S}_2$ single crystals were grown by the gradient freezing method, see [33, 34] for details. The kagome-lattice ferromagnet $\text{Co}_3\text{Sn}_2\text{S}_2$ can be easily cleaved, the Laue patterns confirm the hexagonal structure with (0001) as cleavage plane. Magnetoresistance measurements confirm high quality of our crystals; $\text{Co}_3\text{Sn}_2\text{S}_2$ samples demonstrate [33, 34] giant anomalous Hall effect and positive, non-saturating longitudinal magnetoresistance, which even quantitatively coincide with the previously reported ones [27, 28].

Despite it is possible to form contacts directly on the cleaved $\text{Co}_3\text{Sn}_2\text{S}_2$ crystal plane [35], it is not desirable for transport investigations: the leads to the bonding pads would also participate in current distribution. To obtain a definite sample geometry, the leads pattern is formed on the insulating SiO$_2$ substrate by lift-off technique after thermal evaporation of 100 nm Au, see Fig. 1. Small (about 100 μm size and 1 μm thick) $\text{Co}_3\text{Sn}_2\text{S}_2$ flakes are obtained by a mechanical cleaving method. The most plane-parallel flakes with clean surface are transferred to the Au leads pattern, see [33, 34] for details. This procedure provides reliable Ohmic contacts, stable in multiple cooling cycles, which has been successfully demonstrated for different layered materials [33–36].

We measure the second-harmonic longitudinal $V^{2\omega}_{xx}(I)$ and transverse $V^{2\omega}_{xy}(I)$ voltage components in standard four-point lock-in technique, see the principal circuit diagrams in Figs. 1a and 1b, respectively. The potential contacts are always situated along the sample edge, while the alternating current $I$ flows along the edge for $V^{2\omega}_{xx}(I)$ investigations in Fig. 1a, and normally to it in Fig. 1b for $V^{2\omega}_{xy}(I)$ ones.

We ensure that the measured second-harmonic $V^{2\omega}_{xx}$ voltage is antisymmetric with respect to the voltage probe swap and it is also independent of the ground probe position. We check that the lock-in signal is not sensitive to the alternating current frequency in the range 1100 Hz$^{-1}$ kHz, which is defined by applied filters. For $\text{Co}_3\text{Sn}_2\text{S}_2$, magnetic moments order ferromagnetically [27] below 175 K. For this reason, the measurements are performed in a usual He4 cryostat equipped with a superconducting solenoid. We do not obtain noticeable temperature dependence in the interval 1.4–4.2 K, so all the results below are obtained at 4.2 K and for the normal (to the flake plane) magnetic field orientation.

Examples of the first-harmonic longitudinal voltage $V^{\omega}_{xx}$ are shown as function of alternating current $I$ in Fig. 2a for two different samples in zero magnetic field. The curves are strictly linear and they correspond to a sample resistance of 6 and 2 Ω. In this case, second-harmonic signal $V^{2\omega}_{xx}(I)$ cannot be produced by $I-V$ nonlinearity. In contrast to this expectation, we observe significant second-harmonic longitudinal voltage for these two samples, see Fig. 2b. $V^{2\omega}_{xx}(I)$ is strongly nonlinear, it is proportional to the square of the applied current, as it is demonstrated in the inset to Fig. 2b. The obtained $V^{2\omega}_{xx}(I)$ values are in four times higher for the resistive (6 Ω) sample in comparison with the other (2 Ω) one.

This behavior strongly contradicts to zero $V^{2\omega}_{xx}(I)$ for non-magnetic monolayer transitional metal dichalcogenides [16, 17] and for three-dimensional Weyl and Dirac semimetals [18]. The inherent magnetization of a thick $\text{Co}_3\text{Sn}_2\text{S}_2$ flake is perpendicular to the flakes plane [27], so nonzero $V^{2\omega}_{xx}(I)$ should be connected with the anomalous Nernst effect [30–32] for the magnetic Weyl semimetal $\text{Co}_3\text{Sn}_2\text{S}_2$ in Fig. 2b. Temperature gradient is created in the $\text{Co}_3\text{Sn}_2\text{S}_2$ flake due to the Joule heating by alternating current $I$, the gradient is perpendicular to the current line in Fig. 1a.
Nernst voltage appears as longitudinal in our experimental setup, since it is normal both to the temperature gradient and to the magnetization direction. The experimental observation also well corresponds to the Joule heating effect. It has been demonstrated [18] for non-magnetic three-dimensional Weyl and Dirac semimetals that magnetic field measurements are important to establish an origin of the second-harmonic voltage. First- and second-harmonic dependences are shown in Figs. 3a and 3b, respectively. The first-harmonic component is an even function of \( B \), it demonstrates usual non-saturating longitudinal magnetoresistance [27, 28, 33] in normal magnetic field. The dependence is a nearly odd function; there is also a significant jump in zero magnetic field, as expected for ANE [30–32].

The zero-field jump can be attributed to sample magnetization switching. To work with a definite magnetic state of \( \text{Co}_3\text{Sn}_2\text{S}_2 \), the curves are obtained in Fig. 3 by sweep from highest field values both for the positive and for the negative fields. Because \( \text{Co}_3\text{Sn}_2\text{S}_2 \) demonstrates a giant anomalous Hall effect [27, 28, 33], the magnetization directions are opposite in zero field for these two curves in Fig. 3b. Thus, the curves correspond to ANE signals of different sign, which we observe as the zero-field \( V_{xx}^{2\omega} (I) \) jump. On the other hand, any imaginary \( I–V \) nonlinearity effects should be symmetric in magnetic field, due to the even \( V_{xx}^{1\omega} (B) \) magnetoresistance dependence in Fig. 3a, so the magnetic field behavior confirms thermoelectric origin of finite longitudinal second-harmonic \( V_{xx}^{2\omega} (I) \) voltage.

Fig. 2. (Color online) Examples of the (a) first- \( V_{xx}^{1\omega} (I) \) and (b) second-harmonic \( V_{xx}^{2\omega} (I) \) longitudinal characteristics for two different samples of a magnetic Weyl semimetal \( \text{Co}_3\text{Sn}_2\text{S}_2 \). For strictly linear first-harmonic \( V_{xx}^{1\omega} (I) \) in (a), one cannot expect [16–18] nonzero second-harmonic \( V_{xx}^{2\omega} \) signal in (b). The significant \( V_{xx}^{2\omega} (I) \) should be connected with the anomalous Nernst effect [30–32], since it is normal both to the temperature gradient and to the \( \text{Co}_3\text{Sn}_2\text{S}_2 \) magnetization direction in Fig. 1. Inset confirms the quadratic dependence \( V_{xx}^{2\omega} (I) \sim I^2 \), which well corresponds to the Joule heating effect. The curves are obtained at a temperature of 4.2 K in zero magnetic field.

Fig. 3. Magnetic field dependence of the (a) first- \( V_{xx}^{1\omega} (B) \) and (b) second-harmonic \( V_{xx}^{2\omega} (B) \) at fixed alternating current \( I = 1 \) mA. To avoid magnetization switchings [27, 28, 33] in low fields, the curves are obtained by sweep from highest field values for both field directions. In (a), \( V_{xx}^{1\omega} (B) \) is an even function of \( B \), it demonstrates usual non-saturating longitudinal magnetoresistance [27, 28, 33] in \( \text{Co}_3\text{Sn}_2\text{S}_2 \). In (b), two \( V_{xx}^{2\omega} (B) \) branches reflect anomalous Nernst voltage of different sign due to the opposite magnetization directions for these branches. All the curves are obtained at a temperature of 4.2 K.
Similar behavior can be demonstrated for the transverse $V_{xy}(I)$ voltage component in zero magnetic field, see Fig. 4a. The linear Hall voltage $V_{xy}^{(1)}(I) \sim I$ is due to the finite Co$_3$Sn$_2$S$_2$ magnetization in zero external field. We also obtain nonlinear second-harmonic $V_{xy}^{(2)}(I) \sim I^2$, which is one magnitude smaller than for the $xx$ configuration in Fig. 2b, despite of similar first-harmonic values in Figs. 2a and 4a. In principle, finite $V_{xy}^{(2)}(I) \sim I^2$ can be produced [18] both by NLH and by the thermoelectric effects. In the latter case, the second-harmonic voltage reflects the Seebeck effect [18], because potential contacts are parallel to the temperature gradient in Fig. 1b.

For non-magnetic Weyl and Dirac semimetals, $V_{xy}^{(2)}(B)$ behaves as an odd function of magnetic field due to NLH effect, which has been demonstrated for the exactly same experimental geometry [18]. In contrast, we observe sophisticated magnetic field behavior in Fig. 4b, despite the samples are prepared in the same technique and the contact configuration is also the same [18]. $V_{xy}^{(2)}(B)$ is always positive for both field directions, it demonstrates strong nonlinear increase in high magnetic fields. However, $V_{xy}^{(2)}(B)$ is obviously not symmetric, there is also a linear region between $-1.1$ and $+1.7$ T with a small zero-field jump.

As a result, the longitudinal second-harmonic voltage $V_{xx}^{(2)}$ behavior well corresponds to the expected one for the anomalous Nernst effect in our experimental setup. In contrast, the magnetic field behavior $V_{xy}^{(2)}(B)$ is quite sophisticated, it strongly contradicts to the known one both for NLH and for the Seebeck effects in non-magnetic materials [18].

Let us start from the thermoelectric effects in Weyl and Dirac semimetals. When the magnetic field is perpendicular to the temperature gradient, the longitudinal thermal conductivity (Seebeck effect) is expected [21, 22] to pick up a negative contribution that goes as in the square of the magnetic field. In our experimental setup, the longitudinal thermal conductivity corresponds to the inverse $V_{xy}^{(2)}(B)$ value, as described above, so the high-field behavior in Fig. 4b is in agreement with the theoretical predictions [21, 22].

On the other hand, the low-field linear behavior with the zero-field jump demands another explanation. On the one hand, Berry curvature dipole NLH effect [1−14] should also be seen in magnetically ordered WSMs. Berry curvature acts analogously to a magnetic field in the momentum space, so NLH voltage is linear in external magnetic field [18] and also picks up the Co$_3$Sn$_2$S$_2$ magnetization. The latter should lead to the zero-field jump, since the $V_{xy}^{(2)}(B)$ branches correspond to different magnetization directions in Fig. 4b. On the other hand, one can expect some contribution from Fermi arcs to magnetothermal transport in Weyl semimetals. For topological insulators the latter effect is known to produce large and anomalous Seebeck effects with an opposite sign to the Hall effect [37]. Fermi arcs were directly demonstrated for Co$_3$Sn$_2$S$_2$ by scanning tunneling spectroscopy [38].

These two possibilities could be distinguished by dependence of $V_{xy}^{(2)}(B)$ on the magnetic field orientation [21, 22]. However, Co$_3$Sn$_2$S$_2$ magnetic properties arise from the kagome-lattice cobalt planes, whose magnetic moments order ferromagnetically [27] out of plane below 175 K. We have tested that even comparatively small parallel magnetic field component results in the force, which is large enough to detach the Co$_3$Sn$_2$S$_2$ flake from the substrate.

As a conclusion, we experimentally investigate longitudinal and transverse second-harmonic voltage response to alternating electric current for a magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$. We find that the longitudinal component depends quadratically on the alternating current in zero magnetic field, which reflects strong anomalous Nernst effect, well known for Co$_3$Sn$_2$S$_2$ material. In contrast to the previously observed Berry-curvature induced nonlinear Hall effect for non-magnetic Weyl and Dirac semimetals, the second-harmonic transverse voltage demonstrates sophisticated interplay of different effects for Co$_3$Sn$_2$S$_2$. In high magnetic fields, it is of Seebeck-like
square-B law, while the low-field behavior is found to be linear and sensitive to the direction of sample magnetization. The latter can be expected both for the nonlinear Hall effect and for the surface state contribution to the Seebeck effect in Weyl semimetals. Thus, thermoelectric effects are significant in Co₃Sn₂S₂, unlike non-magnetic Weyl and Dirac materials.

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