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Second-harmonic voltage responce for the magnetic Weyl semimetal $Co_3Sn_2S_2$

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Recent interest to the time-reversal-invariant non-linear Hall (NLH) effect [1] is a part of a broad research area of topological systems. In zero magnetic field, a non-linear Hall-like current arises from the Berry curvature, which can be 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 non-zero transverse second-harmonic voltage without magnetic field. Since Berry curvature concentrates in regions where two or more bands cross [2], topological systems are the obvious candidates to observe the NLH effect [1]. It has been experimentally demonstrated for monolayer transitional metal dichalcogenides [3, 4] and for three-dimensional Weyl and Dirac semimetals [5].

First experimentally investigated Weyl semimetals (WSMs) were non-centrosymmetric crystals with broken inversion symmetry. Even in this case, the magnetic field measurements allow to distinguish the NLH effect from the thermoelectric response [5].

Recently, giant anomalous Hall effect was reported for the kagome-lattice ferromagnet $Co_3Sn_2S_2$, 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 [6]. 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 $Co_3Sn_2S_2$ thermoelectric devices.

$Co_3Sn_2S_2$ single crystals were grown by the gradient freezing method, see [7, 8] for details. The kagome-lattice ferromagnet $Co_3Sn_2S_2$ can be easily cleaved, the Laue patterns confirm the hexagonal structure with (0001) as cleavage plane. Magnetoresistance measurements confirms high quality of our crystals: $Co_3Sn_2S_2$ samples demonstrate [7, 8] giant anomalous Hall effect and positive, non-saturating longitudinal magnetoresistance, which even quantitatively coincide with the previously reported ones.

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 $100\,\mu$m size and $1\,\mu$m thick) $Co_3Sn_2S_2$ flakes are transferred to the Au leads pattern, see [7–9] for details. We measure the second-harmonic longitudinal $V_{xx}^{2\omega}(I)$ and transverse $V_{xy}^{2\omega}(I)$ voltage components in standard four-point lock-in technique, see the principal circuit diagrams in Fig. 1a and b, respectively. The potential contacts are always situated along the sample edge, while the ac current I flows along the edge for $V_{xx}^{2\omega}(I)$ investigations in Fig. 1a, and normally to it in Fig. 1b for $V_{xy}^{2\omega}(I)$ ones.

We observe significant second-harmonic longitudinal voltage $V_{xx}^{2\omega}(I)$ for two different samples. $V_{xx}^{2\omega}(I)$ is strongly non-linear, it is proportional to the square of the applied current. This behavior strongly contradicts to zero $V_{xx}^{2\omega}(I)$ for non-magnetic monolayer transitional metal dichalcogenides [3, 4] and for three-dimensional Weyl and Dirac semimetals [5]. On the other hand, the inherent magnetization of a thick $Co_3Sn_2S_2$ flake is perpendicular to the flake's plane, so Nernst voltage appears as longitudinal $V_{xx}^{2\omega}(I) \sim I^2$ in our experimental setup, which is known as anomalous Nernst effect.

It has been demonstrated [5] for non-magnetic three-dimensional Weyl and Dirac semimetals, that magnetic field measurements are important to establish an origin of the second-harmonic voltage. The $V_{xx}^{2\omega}(B)$ dependence is a nearly odd function, there is also a significant jump in zero magnetic field, like it is expected for ANE.

For the transverse $V_{xy}(I)$ voltage component in zero magnetic field, the linear Hall voltage $V_{xy}^{1\omega}(I) \sim I$ is

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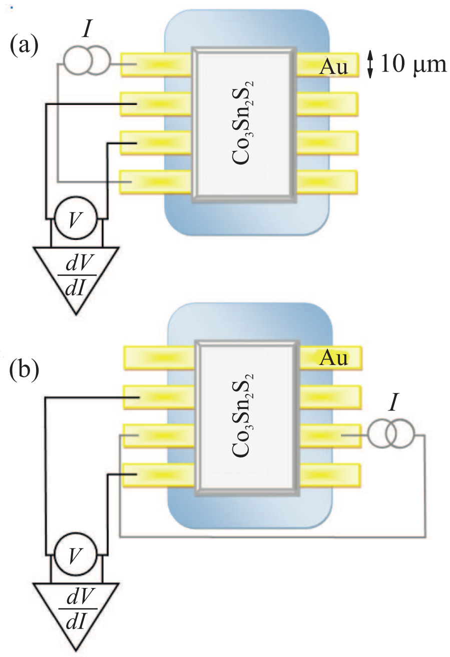

Fig. 1. (Color online) Sketch of the sample with electrical connections. Au leads are formed on a SiO_2 substrate, with 5 μm intervals between them. A thick $Co_3Sn_2S_2$ flake ($\approx$ 100 μm size) is transferred [7, 8] on the top of the leads, forming small Ohmic contacts ($\approx$ 10 μm overlap between the $Co_3Sn_2S_2$ flake and the leads). Current flows along the sample edge in (a) for $V_{xx}^{2\omega}(I)$ investigations, and normally to it in (b) for $V_{xy}^{2\omega}(I)$ ones. The second-harmonic (2ω) component of the longitudinal voltage $V_{xx}^{2\omega}(I)$ is measured in a standard four-point lock-in technique

due to the finite $Co_3Sn_2S_2$ magnetization in zero external field. We also obtain non-linear second-harmonic $V_{xy}^{2\omega}(I) \sim I^2$, which is one magnitude smaller than for the xx configuration. In principle, finite $V_{xy}^{2\omega}(I) \sim I^2$ can be produced [5] both by NLH and by the thermoelectric effects. In the latter case, the second-harmonic voltage reflects the Seebeck effect [5], because potential contacts are parallel to the temperature gradient.

We observe sophisticated magnetic field behavior for the second-harmonic xy component: $V_{xy}^{2\omega}(B)$ is always positive for both fields' directions, it demonstrates strong nonlinear increase in high magnetic fields. However, $V_{xy}^{2\omega}(B)$ is obviously not symmetric, there is also a linear region between -1.1 T and $+1.7$ T with a small zero-field jump. This behavior strongly contradicts to the known one both for NLH and for the Seebeck effects in non-magnetic materials [5].

In our experimental setup, the longitudinal thermal conductivity corresponds to the inverse $V_{xy}^{2\omega}(B)$ value, as described above, so the high-field behavior of $V_{xy}^{2\omega}(B)$ is in agreement with the theoretical predictions. 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] 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 [5] and also picks up the $Co_3Sn_2S_2$ magnetization. The latter should lead to the zero-field jump, since the $V_{xy}^{2\omega}(B)$ branches correspond to different magnetization directions. 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 [10].

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