

Non-linear Hall effect in three-dimensional Weyl and Dirac semimetals

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Non-linear Hall effect has been predicted in a wide class of time-reversal invariant materials [1], like topological crystalline insulators, two-dimensional transition metal dichalcogenides, and three-dimensional Weyl and Dirac semimetals [2]. Recently, the time-reversal-invariant non-linear Hall (NLH) effect has been reported for two-dimensional layered dichalcogenides [3, 4]. It stimulates a search for the Berry curvature dipole induced NLH effect in three-dimensional crystals, where Dirac and Weyl semimetals are excellent candidates.

In the experiments [3, 4] on two-dimensional WTe₂, the the second-harmonic Hall voltage depends quadratically on the longitudinal current. On the other hand, topological materials are characterized by strong thermoelectric effects, which also appear as a second-harmonic quadratic signal. For this reason, it is important to experimentally distinguish between the Berry curvature dipole induced NLH effect and a thermoelectric response while searching for the NLH effect in non-magnetic materials.

Cd₃As₂ crystals were grown by crystallization of molten drops in the convective counterflow of argon held at 5 MPa pressure. The energy-dispersive X-ray spectroscopy (EDX) and X-ray powder diffractograms always confirmed pure Cd₃As₂ with *I4₁cd* noncentrosymmetric group. WTe₂ compound was synthesized from elements by reaction of metal with tellurium vapor in the sealed silica ampule. The X-ray diffraction confirms *Pmn2₁* orthorhombic single crystal WTe₂.

Figure 1 shows a top-view image of a sample. We ensure, that the measured voltage is antisymmetric with respect to the voltage probe swap and it is independent of the ground probe position. We check, that the lock-in signal is also independent of the modulation frequency (about 110 Hz). The measurements are performed in a standard 1.4–4.2 K cryostat equipped with superconducting solenoid.

In the case of the symmetric configuration, like depicted in Figure 1a, we obtain clearly non-zero Hall volt-

age $V_{xy}^{2\omega}$ for the second harmonics of the ac excitation current I . The measured $V_{xy}^{2\omega}$ is below 0.1 μV , it slightly (about 10%) depends on temperature in 1.4–4.2 K interval. The curve is obviously non-linear, $V_{xy}^{2\omega} \sim I^2$. This behavior well corresponds [3, 4] to the expected for NLH effect. However, this interpretation can not be accepted without additional arguments. For example, if the potential contacts are not symmetric in respect to the current line, we also obtain non-linear, $V_{xy}^{2\omega} \sim I^2$ curve. In this case the signal level is one order of magnitude higher, about 1 μV , which better corresponds to typical thermopower values.

To experimentally determine the origin of the effect in every of these two cases, we apply an external magnetic field. In the case of the symmetric configuration, $\Delta V_{xy}^{2\omega}(B) = V_{xy}^{2\omega}(B) - V_{xy}^{2\omega}(B=0)$ is nearly odd function, i.e. $V_{xy}^{2\omega}(B)$ depends on the magnetic field direction: $V_{xy}^{2\omega}(B)$ is diminishing for the positive fields, while it is increasing for the negative ones. In contrast, $V_{xy}^{2\omega}(B)$ increases for both field directions for the non-symmetric connection scheme. In this case, $V_{xy}^{2\omega}(B)$ even quantitatively resembles Cd₃As₂ magnetoresistance [5].

The observed behavior can be reproduced not only for different samples in different cooling cycles, but also can be demonstrated for another three-dimensional material, like WTe₂ Weyl semimetal. As a result, we obtain non-linear second-harmonic xy signal $V_{xy}^{2\omega} \sim I^2$, which demonstrates different magnetic field behavior, even- or odd-type, for nonsymmetric or strictly symmetric configurations of voltage probes, respectively.

The odd $V_{xy}^{2\omega}(B)$ dependence is a good argument for NLH origin of the non-zero second-harmonic Hall voltage: if the ac excitation current generates sample magnetization, the latter should be sensitive to the direction of external magnetic field. More precisely, it is possible to demonstrate from the kinetic equation (in the spirit of [1]), that second – order response is absent in classical Hall effect, while it is an odd function of magnetic field for the spectrum with Berry curvature (Weyl semimetals).

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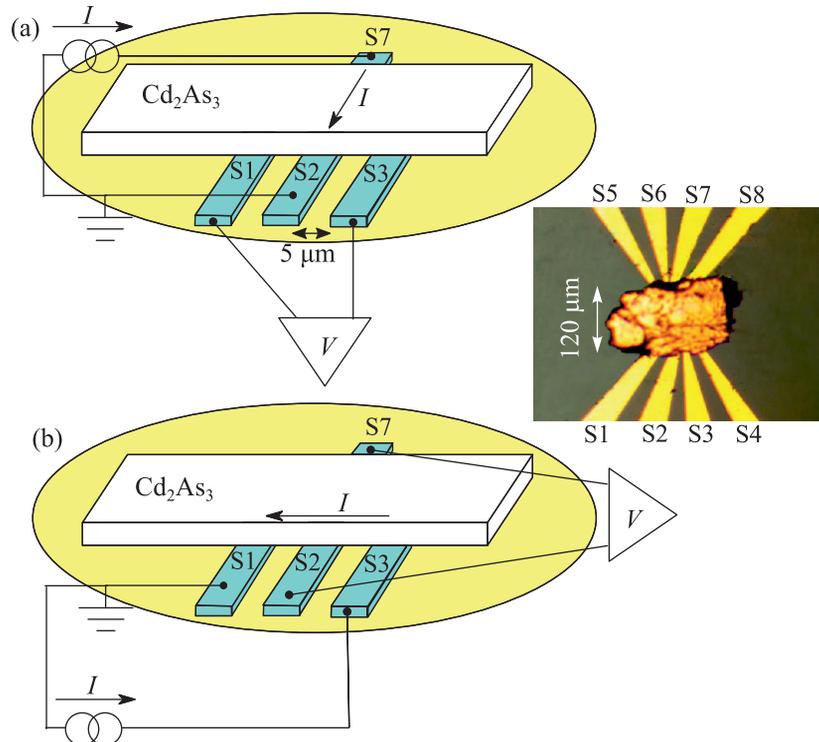


Fig. 1. (Color online) Top-view image of the sample with a small Cd_3As_2 single crystal and the sketch with electrical connections. 100 nm thick and $10 \mu\text{m}$ wide Au leads are formed on a SiO_2 substrate. A Cd_3As_2 single crystal ($\approx 100 \mu\text{m}$ size) is transferred on top of the leads, forming contacts S1–S8 in regions of $\approx 10 \mu\text{m}$ overlap between the crystal and the leads. The second-harmonic (2ω) component of the Hall voltage V is investigated in a standard four-point lock-in technique in symmetric (a) and nonsymmetric (b) connection of the Hall voltage probes in respect to the current line (denoted by arrows), which mostly flows along the sample edge between S1 and S3 in the (b) case

In contrast, thermoelectric effects are defined by the sample heating, which is proportional to RI^2 in our case, i.e., they also produce the second-harmonic response. The magnetic field dependence should be mainly defined by the magnetoresistance $R(B)$, since it is extremely strong in Weyl and Dirac semimetals. Thus, the thermoelectric response can not be sensitive to the magnetic field direction. In the experiment, $V_{xy}^{2\omega}(B)$ even quantitatively resembles $R(B)$ magnetoresistance for our samples. Note, that Nernst effect can not contribute to the measured xy voltage, since the temperature gradient is along the y axis in the geometry of the experiment. On the other hand, the Seebeck effect is also characterized [6] by even, $R(B)$ -like magnetic field dependence.

Thus, we can identify high second-harmonic signal as thermoelectric voltage for nonsymmetric connection schemes, while low $V_{xy}^{2\omega}$ reflects NLH effect for the strictly symmetric ones.

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