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

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We experimentally investigate a non-linear Hall effect for three-dimensional WTe2 and Cd3As2 single crystals, representing Weyl and Dirac semimetals, respectively. We observe finite second-harmonic Hall voltage, which depends quadratically on the longitudinal current in zero magnetic field. Despite this observation well corresponds to the theoretical predictions, only magnetic field dependence allows to distinguish the non-linear Hall effect from a thermoelectric response. We demonstrate that second-harmonic Hall voltage shows odd-type dependence on the direction of the magnetic field, which is a strong argument in favor of current-magnetization effects. In contrast, one order of magnitude higher thermopower signal is independent of the magnetic field direction.

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I. INTRODUCTION

Non-linear Hall effect has been predicted in a wide class of time-reversal invariant materials1–2. In the linear response, there is no Hall current in the presence of time-reversal symmetry. It is argued in Refs. 1,2 that a non-linear Hall-like current can arise from the Berry curvature in momentum space. Since Berry curvature often concentrates in regions where two or more bands cross, three classes of candidate materials have been proposed3–6: topological crystalline insulators, two-dimensional transition metal dichalcogenides, and three-dimensional Weyl and Dirac semimetals. Another possible contribution to non-linear Hall effect is skew scattering with nonmagnetic impurities in time-reversal-invariant noncentrosymmetric materials6–9.

Recently, the time-reversal-invariant non-linear Hall (NLH) effect has been reported for layered transition metal dichalcogenides7,8. It stimulates a search for the Berry curvature dipole induced NLH effect in three-dimensional crystals, where Dirac and Weyl semimetals9,10 are excellent candidates, since there is symmetry-protected conic dispersion in the bulk spectrum10,11. This spectrum has been experimentally confirmed by angle-resolved photoemission spectroscopy (ARPES), e.g., for Cd3As2 Dirac material12,13, and for MoTe2 and WTe2 type II Weyl semimetals14,15. Because of low symmetry, MoTe2 and WTe2 are advantageous6–9 in a search for the NLH effect.

In the experiments6,16 on two-dimensional WTe2, the the second-harmonic Hall voltage depends quadratically on the longitudinal current. In the simplified picture, an a.c. excitation current generates sample magnetization, which leads to the anomalous Hall effect6–9 in zero external magnetic field. The latter appears as the second-harmonic Hall voltage, the amplitude is proportional to the square of the bias current. On the other hand, topological materials are characterized by strong thermoelectric effects16,17, which also appear as a second-harmonic quadratic signal18,19,20. 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 nonmagnetic materials.

Here, we experimentally investigate a non-linear Hall effect for three-dimensional WTe2 and Cd3As2 single crystals, representing Weyl and Dirac semimetals, respectively. We observe finite second-harmonic Hall voltage, which depends quadratically on the longitudinal current in zero magnetic field. Despite this observation well corresponds to the theoretical predictions, only magnetic

FIG. 1. (Color online) Top-view image of the sample with a small Cd3As2 single crystal and the sketch with electrical connections. 100 nm thick and 10 µm wide Au leads are formed on a SiO2 substrate. A Cd3As2 single crystal (∼100 µm size) is transferred on top of the leads, forming contacts S1-S8 in regions of ∼10 µm 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.

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field dependence allows to distinguish the non-linear Hall effect from a thermoelectric response. We demonstrate that second-harmonic Hall voltage shows odd-type dependence on the direction of the magnetic field, which is a strong argument in favor of current-magnetization effects. In contrast, one order of magnitude higher thermopower signal is independent of the magnetic field direction.

II. SAMPLES AND TECHNIQUE

$\text{Cd}_3\text{As}_2$ crystals were grown by crystallization of molten drops in the convective counterflow of argon held at 5 MPa pressure. For the source of drops the stalagmometer similar to one described$^{21}$ was applied. About one fifth of the drops were single crystals. The energy-dispersive X-ray spectroscopy (EDX) and X-ray powder diffractograms always confirmed pure $\text{Cd}_3\text{As}_2$ with $I4_1cd$ noncentrosymmetric group.

$\text{WTe}_2$ compound was synthesized from elements by reaction of metal with tellurium vapor in the sealed silica ampule. The $\text{WTe}_2$ crystals were grown by the two-stage iodine transport$^{22,23}$, that previously was successfully applied$^{22,23}$ for growth of other metal chalcogenides like $\text{NbS}_2$ and $\text{CrN}\text{b}_3\text{S}_6$. The $\text{WTe}_2$ composition is verified by EDX measurements. The X-ray diffraction confirms $Pmn2_1$ orthorhombic single crystal $\text{WTe}_2$.

The initial $\text{WTe}_2$ ingot is formed by a large number of small (less than 100 $\mu$m size) $\text{WTe}_2$ single crystals, which are weakly connected with each other. In contrast, small $\text{Cd}_3\text{As}_2$ single crystals are obtained by a mechanical cleaving method, somewhat similar to described in Ref.$^{24}$ we crush the initial 5 mm size $\text{Cd}_3\text{As}_2$ drop onto small fragments. This procedure allows to create a clean $\text{Cd}_3\text{As}_2$ surface without mechanical polishing or chemical treatment, see Ref.$^{25}$ for details.

Fig. 1 shows a top-view image of a sample. The leads pattern is formed by lift-off technique after thermal evaporation of 100 nm Au on the insulating SiO$_2$ substrate. The 10 $\mu$m wide Au leads are separated by 5 $\mu$m intervals, see Fig. 1. Then, a small (about 100 $\mu$m size) $\text{Cd}_3\text{As}_2$ or $\text{WTe}_2$ crystal is transferred to the Au leads pattern and pressed slightly with another oxidized silicon substrate. A special metallic frame allows to keep substrates parallel and apply a weak pressure to the piece. No external pressure is needed for a crystal to hold on a substrate with Au leads afterward.

We check by standard magnetoresistance measurements that our $\text{Cd}_3\text{As}_2$ samples demonstrate large magnetoresistance with Shubnikov de Haas oscillations in high magnetic fields$^{22}$. We estimate the concentration of carries as $n \approx 2.3 \times 10^{18}$ cm$^{-3}$ and low-temperature mobility as $\mu \approx 10^6$ cm$^2$/Vs, which is in the good correspondence with known values$^{22}$. Also, we check that our $\text{WTe}_2$ samples demonstrate large, non-saturating positive magnetoresistance $(\rho(B) - \rho(B = 0))/\rho(B = 0)$ in normal magnetic field, which goes to zero in parallel one, as it has been shown for $\text{WTe}_2$ Weyl semimetal$^{22}$. We do not see Shubnikov de Haas oscillations for $\text{WTe}_2$ samples due to lower mobility, see Ref.$^{25}$ for details of magnetoresistance measurements. Examples of the magnetoresistance curves are also shown for our samples in the

![FIG. 2. (Color online) Examples of $V(I)$ characteristics for a three-dimensional $\text{Cd}_3\text{As}_2$ crystal. Here, $V$ is the second-harmonic ($2\omega$) $xy$ voltage component, $I$ is the ac excitation current at frequency $\omega$. (a) In the case of the symmetric configuration, see Fig. 1 (a), the measured Hall voltage $V$ is obviously non-linear, $V \sim I^2$, as it can be seen from the inset. The $V(I)$ curve slightly (about 10%) depends on temperature in 1.4 K–4.2 K interval. (b) In the nonsymmetric configuration, depicted in Fig. 1 (b), the signal level is one order of magnitude higher, about 1 $\mu$V, but the curve is still non-linear $V \sim I^2$, see the inset. The curves are obtained in zero magnetic field.

![FIG. 3. (Color online) Second-harmonic voltage $V$ dependence on the magnetic field $B$ at fixed ac current $I$ for three-dimensional $\text{Cd}_3\text{As}_2$. (a) In the case of the symmetric voltage probe configuration, $\Delta V(B) = V(B) - V(B = 0)$ is nearly odd function, which is a strong argument in favor of current-magnetization effects. (b) $V(B)$ increases for both field directions for the nonsymmetric connection scheme, which allows to identify the thermoelectric response. Inset demonstrates usual (first-harmonic) $\text{Cd}_3\text{As}_2$ xx magnetoresistance $R(B)$ for our samples. All the curves are obtained at 4.2 K temperature. $I = 1.5$ mA is diminishing for (b) with respect to the $I = 3.5$ mA for (a), to avoid overheating effects for high signal in the (b) case.}
In this symmetric configuration, there is no temperature difference between contacts S2 and S3, while the Hall voltage is obtained at 4.2 K temperature.

FIG. 5. (Color online) Second-harmonic voltage V dependence on the magnetic field B at fixed ac current I for three-dimensional WTe$_2$. (a) In the case of the symmetric voltage probe configuration, $\Delta V(B) = V(B) - V(B = 0)$ is the odd function of B, similarly to the Cd$_3$As$_2$ case. (b) $V(B)$ is nearly even function for the nonsymmetric connection scheme. $I = 0.5$ mA is diminishing for (b) with respect to the $I = 3.5$ mA for (a), to avoid overheating effects for high signal in (b) case. Inset to (b) demonstrates usual (first-harmonic) WTe$_2$ xx magnetoresistance for our samples and nearly zero second-harmonic xx component. The curves are obtained at 4.2 K temperature.

III. EXPERIMENTAL RESULTS

Examples of $I - V$ characteristics are shown in Fig. 2 for symmetric (a) and nonsymmetric (b) configurations of the voltage probes. In the case of the symmetric configuration, like depicted in Fig. 1 (a), we obtain clearly non-zero Hall voltage $V^{2\omega}$ for the second harmonics of the ac excitation current $I$. The measured $V^{2\omega}$ is below 0.1 $\mu$V, it slightly (about 10%) depends on temperature in 1.4 K–4.2 K interval. The $I - V$ curve is obviously non-linear, $V^{2\omega} \sim I^2$, as it can be seen from the inset to Fig. 2 (a).

This behavior well corresponds 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, see Fig. 1 (b), we also obtain non-linear, $V^{2\omega} \sim I^2$, $I - V$ curve, as presented in Fig. 2 (b). In this case the signal level is one order of magnitude higher, about 1 $\mu$V, 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. Fig. 3 demonstrates second-harmonic voltage dependence on the magnetic field $V^{2\omega}(B)$ at fixed ac current values. In the case of the symmetric configuration, see Fig. 3 (a), $\Delta V^{2\omega}(B) = V^{2\omega}(B) - V^{2\omega}(B = 0)$ is nearly odd function, i.e. $V^{2\omega}(B)$ depends on the magnetic field direction: $V^{2\omega}(B)$ is diminishing for the positive fields, while it is increasing for the negative ones. In contrast, $V^{2\omega}(B)$ increases for both field directions for the nonsymmetric connection scheme, see Fig. 3 (b). In this case, $V^{2\omega}(B)$ even quantitatively resembles Cd$_3$As$_2$ magnetoresistance, which is depicted in the inset to Fig. 3 (b) for our samples.

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, see Figs. 4 and 5.

The measured nonlinear second-harmonic Hall voltage \( V^{2\omega} \) is also below 0.1 \( \mu V \) for the symmetric Hall probe connection scheme. Like for Cd₃As₂ samples, nonsymmetric connection leads to high (about 1 \( \mu V \)) nonlinear \( V^{2\omega} \sim I^2 \) voltage, see Fig. 4(b). We also check, that there is no significant second-harmonic signal for the voltage probes situated along the current line, i.e. \( xx \) voltage component is below 10 nV, see the inset to Fig. 4(b).

The specifics of NLH effects for layered WTe₂ is the strong signal dependence on the current direction. In our case of three-dimensional WTe₂, we obtain nearly the same \( V^{2\omega} \) for currents along both \( a \) and \( b \) directions at the liquid helium temperature 4 K, see Fig. 4(a). For lower (1.4 K) temperatures, the Hall voltage \( V^{2\omega} \) tends to zero for current along the \( a \) direction, as depicted in the inset to Fig. 4(a). This is the difference of our \( I - V \) curves from the layered two-dimensional WTe₂, where there was no strong temperature dependence.

The similarity between Cd₃As₂ and WTe₂ crystals can also be seen in the magnetic field behavior, see Fig. 5. For the symmetric connection scheme, \( V^{2\omega}(B) \) demonstrates strong odd-type behavior in respect to the magnetic field direction, as demonstrated in the (a) panel. In contrast, \( V^{2\omega}(B) \) is clearly even-type in Fig. 5(b), which well corresponds to the bulk WTe₂ non-saturating \( xx \) magnetoresistance, see the inset to the panel (b). We wish to note, that there is no noticeable field dependence for the second-harmonic \( (2\omega) \) \( xx \) tensor component, see also the inset to Fig. 5.

**IV. DISCUSSION**

As a result, we obtain non-linear second-harmonic \( xy \) signal \( V^{2\omega} \sim I^2 \), which demonstrates different magnetic field behavior, even- or odd-type, for nonsymmetric or strictly symmetric configurations of voltage probes, respectively.

The odd \( V^{2\omega}(B) \) 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 \(^{26}\) from the kinetic equation (in the spirit of Ref. [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).

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, \( V^{2\omega}(B) \) even quantitatively resembles \( R(B) \) magnetoresistance, see Figs. 4 and 5 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 \(^{30}\) 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 \( V^{2\omega} \) reflects NLH effect for the strictly symmetric ones.

For both connection schemes, some admixture of the effects is possible. We can not completely avoid an asymmetry of the potential contacts, so an admixture of \( R(B) \) produces distortions in high fields in Fig. 3(a). On the other hand, NLH effect should be present also in the nonsymmetric connection scheme, where the thermoelectric response dominates. Due to the strong odd field dependence of NLH voltage, it can be responsible for the observed \( V^{2\omega} \) branch asymmetry in Figs. 3(b) and 5(b).

While NLH effect was originally proposed \(^{13}\) for Weyl and Dirac semimetals, it can only be seen for noncentrosymmetric crystals. This requirement is obviously fulfilled for the type II Weyl semimetal WTe₂, but there is a discussion in the case of Cd₃As₂. Ref. [3] insists on the near-centrosymmetric structure with the space group \( I\bar{4}_{3} \). On the other hand, the previously established \(^{32}\) \( I\bar{4}_{1}cd \) noncentrosymmetric group is also confirmed in recent investigation \(^{33}\) and this crystal symmetry is in a reasonable correspondence with ARPES data on the Cd₃As₂ electronic structure \(^{34}\). This difference should originate from the Cd₃As₂ growth method, e.g. the X-ray diffraction confirms \( I\bar{4}_{1}cd \) noncentrosymmetric group for our samples. Also, in our case, strain may occur at SiO₂-Cd₃As₂ interface due to materials misfit, which affects the initial symmetry \(^{34}\). It is worth mentioning, that skew scattering is allowed in all noncentrosymmetric crystals, whereas Berry curvature dipole requires more strict symmetry conditions \(^{35}\). We still can not distinguish these two contributions to NLH effect in the present experiment.

**V. CONCLUSION**

We experimentally investigate a non-linear Hall effect for three-dimensional WTe₂ and Cd₃As₂ single crystals, representing Weyl and Dirac semimetals, respectively. We demonstrate finite second-harmonic Hall voltage, which depends quadratically on the longitudinal current in zero magnetic field. If the potential contact are perfectly symmetric in respect to the current line, the observed signal is in the nanovolt range. It shows odd-type dependence on the direction of the magnetic field, which is a strong argument in favor of current-magnetization effects. If the potential contact are strongly nonsymmetric, temperature gradient produces one order of magnitude higher thermopower signal with even-type magnetic field dependence.
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