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Spin-Hall magnetoresistance in multidomain helical spiral systems

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Abstract

We study the spin-Hall magnetoresistance (SMR) in multidomain helical spiral magnet $\text{Cu}_2\text{OSeO}_3/\text{Pt}$ heterostructures. We compare the SMR response of Cu_2OSeO_3 at 5 K, when the magnetic domains are almost frozen, to that at elevated temperatures, when domain walls move easily. At 5 K the SMR amplitude vanishes at low applied magnetic fields, while at 50 K it does not. This phenomenon can be explained by the effect of the magnetic field on the domain structure of Cu_2OSeO_3 . At elevated temperatures the system can reach the thermodynamic equilibrium state, in which a single domain that has a minimal energy for a given field direction occupies the whole sample and gives rise to a nonzero SMR signal. In contrast at 5 K, the three types of domains with mutually orthogonal spiral wave vectors have equal volumes independent of the field direction, which leads to the cancellation of the SMR signal at low fields. In the single-domain conical spiral and collinear ferrimagnetic states, the angular and field dependence of the SMR is found to be same at all temperatures ($T \leq 50$ K). This behavior can be understood within the framework of the SMR theory developed for collinear magnets.

Keywords: spin currents, spin-Hall effect, magnetoresistance

(Some figures may appear in colour only in the online journal)

1. Introduction

The recently discovered magnetoresistance phenomenon, known as the spin-Hall magnetoresistance (SMR) [1–3], has attracted considerable attention because of the possibility to electrically detect the magnetization of an insulating magnetic layer [4–7]. To measure the SMR, a simple device structure consisting of a heavy normal metal (NM) with a finite spin-Hall angle [8–10] on top of a ferromagnetic or ferrimagnetic (FM) insulator is used. In the SMR, the resistance of the NM contact changes with the absorption/reflection of spin currents at the NM/FM interface [11]. The SMR has been mostly studied for collinear magnetic insulators including the prototype material, yttrium iron garnet (YIG), recently in some non-conventional magnetic insulators, e.g. LaCoO_3 [12] and $\text{Tm}_3\text{Fe}_5\text{O}_{12}$ [13] and, in more complex magnets, e.g. CoFe_2O_4 [5, 7], CoCr_2O_4 [4] and $\text{YGd}_2\text{Fe}_4\text{InO}_{12}$ [14]. Among magnetic

materials with complex magnetic orders, the noncollinear magnets with a broken space-inversion symmetry, known as chiral magnets, have the most fascinating physical properties. Chiral magnets show numerous phases with complex spin structures, such as the multidomain helical spiral state, the single-domain conical spiral state and the skyrmionic crystal state [15]. In these magnets, the relativistic Dzyaloshinskii–Moriya interaction between spins [16, 17] results in a twist of the collinear ferromagnetic spin ordering, which stabilizes noncollinear chiral magnetic structures [18–21]. Examples of such magnets are: MnSi [15, 22], $\text{Mn}_{1-x}\text{Fe}_x\text{Ge}$ [23], FeGe [24–26], $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ [27] and Cu_2OSeO_3 [28–32]. Among these chiral magnets, Cu_2OSeO_3 is the first magnetic insulator ($T_c = 58$ K) in which magnetic skyrmions are experimentally observed [28–30]. To detect non-collinear chiral magnetic structures in these systems, sophisticated experimental techniques, such as Lorentz transmission electron microscopy, are used [28]. Recently, it has been shown that the SMR is an effective tool for all-electric detection of canted spin states in

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Figure 1. (a) Schematic illustration of the types of field-induced magnetic order in the Cu_2OSeO_3 single crystal. Here $\mathbf{H}$, $\mathbf{Q}$ and $\mathbf{m}$ represent the applied magnetic field, the spiral wave vector and magnetization unit vector of Cu_2OSeO_3 , respectively. H_{c1} and H_{c2} represent the fields at which the magnetic transitions occur in Cu_2OSeO_3 . (b) Device configuration for the transverse resistance measurement of the Pt film on top of Cu_2OSeO_3 . (c) Angular dependence of the SMR signal (V_{SMR}) at 5 K in ferrimagnetic state of Cu_2OSeO_3 . Here, the solid line shows a $\sin(2\alpha)$ fit.

frustrated magnets [14] and complex spin structures in chiral magnetic insulators [33].

In this paper, we report a systematic study of the excitation and detection of spin currents in the multidomain helical spiral system Cu_2OSeO_3 using the SMR. Cu_2OSeO_3 is a cubic chiral magnet showing a variety of noncollinear magnetic phases (see figure 1(a)). At zero and low applied magnetic fields the magnetic state of Cu_2OSeO_3 is the helical spiral state with a period of 50 nm [28]. This is a multidomain magnetic state, as the spiral wave vector $\mathbf{Q}$ can be oriented along any of the three equivalent crystallographic directions: (100), (010) or (001). When the external magnetic field exceeds the lower critical field ($H \geq H_{c1}$), the multidomain state transforms into a single-domain conical spiral state with the spiral wave vector $\mathbf{Q}$ oriented along the applied field $\mathbf{H}$. The cone angle θ of the spiral decreases with increasing magnetic field and becomes zero above the second-critical field ($H \geq H_{c2}$), which corresponds to the field-induced collinear ferrimagnetic (FM) state (see figure 1(a)). We reported previously the field dependence of the SMR in different magnetic phases of Cu_2OSeO_3 at 5 K [33]. Here, we study the angular and field dependence of the SMR at elevated temperatures (25 K and 50 K) focusing on effects of the transformations from the multidomain helical spiral state into the single-domain conical spiral state and then to the collinear ferrimagnetic state. We find interesting differences between the SMR responses of Cu_2OSeO_3 at low and high temperatures.

2. Experimental procedure

Cu_2OSeO_3 single crystals of typical dimensions $\approx 4 \text{ mm} \times 4 \text{ mm} \times 2 \text{ mm}$ were grown by a chemical vapor transport method [34, 35]. The structure was characterized by a Bruker D8 Venture single crystal x-ray diffractometer. The same diffractometer was used to orient the crystals along (111) direction, followed by the polishing step on (111) surfaces to obtain the surface roughness around 2 nm. The Hall-bar device structures were patterned on the polished (111) crystal surface of Cu_2OSeO_3 by using e-beam lithography. In these patterned Hall-bars 5 nm thick Platinum (Pt) was deposited by dc sputtering. To check the surface roughness and thickness of deposited metallic layers, atomic force microscopy was used.

All electrical measurements were performed in the transverse Hall-bar configuration (see figure 1(b)), in a quantum design Physical Properties Measurement System (PPMS). A Stanford SR-830 lock-in amplifier is used to measure the first harmonic signal at a reference frequency of 17 Hz. The current (1 mA) was sent to the device using a custom-built current source and the response signal was pre-amplified, before sending it back to the lock-in amplifier. The angular dependence of the sample response was recorded by rotating it in the superconducting magnet of the PPMS with the magnetic field varying in the xy -plane of the device, as shown in figure 1(b).

3. Results and discussion

When current I is sent through the Pt Hall-bar, a transverse spin current is generated due to the spin-Hall effect (SHE) [36–38], resulting in a spin accumulation, μ_s , at the Pt/ Cu_2OSeO_3 interface. When μ_s is perpendicular to the magnetization $\mathbf{M}$ of Cu_2OSeO_3 , the spin current from the Pt contact to Cu_2OSeO_3 will be partially absorbed at the interface because of the spin torque effect, resulting in a higher resistance of the Pt contact. When $\mu_s \parallel \mathbf{M}$, the spins arriving at the interface reflect back into the Pt contact. These reflected spins generate an additional charge current via the inverse spin-Hall effect (ISHE), resulting a smaller resistance. When $\mathbf{M}$ makes an angle with μ_s , the additionally generated charge current due to the ISHE results in both longitudinal and transverse responses. The latter gives rise to the SMR signal presented here.

When the applied magnetic field $\mathbf{H}$ is rotated in the Pt film plane and the magnetization $\mathbf{M}$ fully aligns along $\mathbf{H}$, a periodic angular dependence $\sin(2\alpha)$ of the transverse Pt resistance is observed, where α is the angle between the applied current and $\mathbf{H}$, as shown in figure 1(b). The SMR is measured by detecting the angular dependence of the first-order response of the system, for which it is necessary to measure the first and third harmonics by the lock-in amplifier in the transverse configuration generated by the current I [4, 39]. As the third harmonic response is negligible, only the first harmonic signal of the lock-in amplifier is considered for the SMR. For all measurements ($B \geq 30 \text{ mT}$), the SMR signal is defined as $V_{\text{SMR}} = V_1 - V_{\text{HE}}$, where V_{SMR} , V_1 and V_{HE} represent the SMR signal, the first harmonic response and the contribution due to the ordinary Hall effect resulting from a slight misalignment of

Figure 2. ((a)–(f)) Angular dependence of the SMR signal (V_{SMR}) at 50 K at different applied magnetic field strengths. The solid lines represent $\sin(2\alpha)$ fits.

the sample. An example of such a measurement is presented in figure 1(c). At low fields ($B < 30$ mT), the Hall contributions are calculated by the linear interpolation of the Hall contributions at high field (as the V_{HE} scales linearly with applied magnetic field) and then subtracted from the observed signals. After subtraction of the Hall contributions from the observed signal at low-fields, the amplitude of the SMR signal $V_{\text{SMR}}^{\text{ampl}}$ is defined as $\frac{V_{\text{SMR}(\alpha=45^\circ)} + V_{\text{SMR}(\alpha=135^\circ)}}{2}$. At high field the SMR signals show a $\sin(2\alpha)$ angular dependence.

Figure 1(c) shows a clear $\sin(2\alpha)$ angular dependence at 5 K in a magnetic field of 100 mT, as expected for the SMR. The angular dependence of the SMR in the ferrimagnetic state at temperatures higher than 5 K is also given by $\sin(2\alpha)$, as shown in figure 2(a). As the magnetic field decreases and the system undergoes a transition into the conical spiral state, the angular dependence of the SMR is still $\sin(2\alpha)$ (see figures 2(b)–(d)). The notable feature observed in the conical spiral state is the sign change of the amplitude of the SMR signal, $V_{\text{SMR}}^{\text{ampl}}$ (see figures 2(a) and (b) with figures 2(c) and (d)): $V_{\text{SMR}}^{\text{ampl}}$ decreases with decreasing field from a positive value at H_{c2} to a negative value at H_{c1} . In the helical spiral state, the sign of the SMR amplitude remains negative upon further decrease of the applied magnetic field, as shown in figures 2(e) and (f). In contrast, the angular dependence of the SMR in the helical state strongly deviates from the $\sin(2\alpha)$ (see figure 2(f)), consistent with the observations reported in literature [33]. The presence of domain walls in the helical state is manifested through these strong deviations from the sinusoidal angular dependence of the SMR. We observe sharp discontinuities in the angular dependence only at low magnetic fields and these discontinuities are present in both trace and retrace curves (see figures A1(a) and (b) in appendix).

A similar behavior is observed at 25 K in the conical phase: $V_{\text{SMR}}^{\text{ampl}}$ decreases with decreasing field from a positive value at H_{c2} to a negative value at H_{c1} (see figure 3(a) with figures 3(b) and (c)). The angular dependence of the SMR deviates from $\sin(2\alpha)$ dependence at the conical spiral to helical spiral transition (see figure 3(d)). The angular dependence in the conical state (figures 3(b) and (c)) deviates from $\sin(2\alpha)$ dependence. This deviation may be related to fact that close to the transition between the conical and helical state, the three helical domains transform into the conical spiral state at different values of the magnetic field, which broadens the helical-to-spiral transition. In the helical state, sharp discontinuities and deviations from the $\sin(2\alpha)$ dependence are observed (see figures 3(e)–(i)). The SMR signal vanishes at zero field, as shown in figure 3(j).

The sign reversal of the SMR signal in the conical phase at 25 K and 50 K (shown in figures 3(b), (c) and 2(c)) can be explained by using equation (1) from [33]:

$$V_{\text{SMR}} \propto \langle m_x m_y \rangle = \frac{1}{4} \sin 2\alpha \left(3 \left(\frac{H}{H_{c2}} \right)^2 - 1 \right) \quad (1)$$

where m_x and m_y represents the parallel and orthogonal in-plane components of the magnetization unit vector $\mathbf{m}$ with respect to the applied current direction, respectively. $V_{\text{SMR}}^{\text{ampl}}$ remains constant for $H > H_{c2}$ and decreases for $H < H_{c2}$ by changing sign at $H = \frac{H_{c2}}{\sqrt{3}}$ in good qualitative agreement with the experimental observations at different temperatures (see figures 4(a)–(c)).

The spin configuration in the helical spiral state is complicated by the presence of three types of magnetic domains with different orientations of the spiral wave vector $\mathbf{Q}$. Moreover, in each domain the helix deforms under the applied magnetic

Figure 3. ((a)–(j)) Angular dependence of the SMR signal (V_{SMR}) at 25 K at different applied magnetic field strengths.

field. Therefore, one can imagine three possible situations for a given H :

- (i) three domains with three different orientations of $\mathbf{Q}$ occupy the same volume and equally contribute to the SMR;
- (ii) The domains with different orientations of $\mathbf{Q}$ occupy different volumes and contribute to the SMR unequally;
- (iii) The SMR signal comes from a single magnetic domain that has the lowest energy and occupies the whole sample.

All these possible situations in the helical state are different from the conical state. In the conical state the spiral wave vector $\mathbf{Q}$ is parallel to the applied magnetic field, whereas in the helical state domains (including the domain with the lowest energy in third scenario) the direction of $\mathbf{Q}$ depends on the direction and strength of the applied magnetic field, which results in deviations from the $\sin(2\alpha)$ angular dependence (see figures 2(f) and 3(e)–(i)). At low fields $\mathbf{Q}$ is parallel to one of the cubic axis ((100), (010) or (001)), while at higher fields it starts aligning with the field direction. The field dependence of $\mathbf{Q}$ is not always continuous, which gives rise to discontinuities in the angular dependence of SMR.

In first scenario when all three domain occupy same volume and contribute equally to the SMR, the SMR signal vanishes in zero field due to cancellation of the contributions from the three types domains with mutually orthogonal spiral wave vectors $\mathbf{Q}$. Such a cancellation of the SMR signal in the multidomain state is observed at 5 K (see figure 4(c)). It is plausible that at low temperatures the domains are ‘frozen’ and that the domain boundaries do not propagate in the rotating magnetic field, resulting in equal volumes of all three types of domains,

which leads to cancellation of the SMR signal. When the magnetic field is increased above 20 mT at 5 K, the domain boundaries start to move and the helical spirals in each domain become distorted, resulting in a linear increase of the SMR signal until H_{c1} , at which the material undergoes a transition into the conical spiral state (see figure 4(c)). At 25 K the SMR also vanishes in low applied magnetic fields. Nevertheless, it vanishes at much lower fields ($H < 15$ mT) than at 5 K (see figures 4(b) and (c)). The sample at 25 K may follow the second scenario in which the three types of domains occupy different volumes, which leads to a non-zero SMR signal. Only close to zero field the contributions from the three types of domains become equal and the cancellation occurs (see figure 4(b)).

At 50 K the SMR signal does not vanish in the helical phase. The situation corresponds to third scenario, in which the magnetic domain with the lowest energy occupies the whole sample, resulting in a finite SMR signal V_{SMR} in the helical phase. This can be explained by the fact that at higher temperatures (50 K), thermal fluctuations facilitate the propagation of domain boundaries. If the domain boundaries move fast on the time scale of measurements of the angular dependence of the SMR signal ($\sim 2^\circ/\text{min}$), the sample will always be in thermal equilibrium. In thermal equilibrium, the domain with the minimal energy occupies the whole sample, resulting in a nearly constant SMR signal in the helical phase, as observed in figure 4(a). Helical single domain state is different from the conical state. In the conical state the spiral wave vector $\mathbf{Q}$ is parallel to the applied magnetic field, whereas in the helical state domains (including the domain with the lowest energy) the direction of $\mathbf{Q}$ depends on the direction and strength of the magnetic field, which results in deviations from the $\sin(2\alpha)$

Figure 4. Magnetic field dependence of the SMR signal (V_{SMR}) at (a) 50 K, (b) 25 K and (c) 5 K. (d) Field dependence of the V_{SMR} at 5 K with the transition between different magnetic states of the Cu_2OSeO_3 marked by vertical lines. The dark blue curve indicates the calculated amplitude of the SMR signal by considering equal contribution from three magnetic domains and the light blue curve is calculated by assuming that the system is in thermal equilibrium.

angular dependence. At low fields $\mathbf{Q}$ is parallel to one of the cubic axis ((100), (010) or (001)), while at higher fields it starts aligning with the field direction. The field dependence of $\mathbf{Q}$ is not always continuous, which gives rise to discontinuities in the angular dependence of SMR. This dependence is further complicated by the deformation of the helical in the magnetic field as explained in [33]. Figure 4(d) shows the calculated fits at 5 K by considering the first scenario of frozen domains with equal contribution to the SMR signal and the third scenario with thermal equilibrium (details of the fitting procedure can be found in [33]). Both calculated fits are same at higher applied magnetic fields, in the conical and field induced collinear ferrimagnetic states of Cu_2OSeO_3 . The prominent differences appear in the multidomain helical state where the calculated amplitude of the SMR does not goes to zero by considering the system in the thermal equilibrium, consistent with the observation at 50 K (see figures 4(a) with (d)).

The critical fields H_{c1} and H_{c2} decrease with increasing temperature as observed in the susceptibility measurements [40] and H_{c2} varies very little below 40 K. This behavior is reproduced by numerical simulations of a cubic chiral magnet [41]. The difference between the [40] and the measurements shown for the SMR in this paper is the field orientation. For the SMR measurements we rotate the applied magnetic field in the (1 1 1) plane. To confirm that the SMR signals are measured below H_{c1} in the helical phase of Cu_2OSeO_3 , the measurements are done at 5 mT (see figure A2 in appendix).

In conclusion, we demonstrated that the SMR can be used to electrically detect the inhomogeneous spin structures, including helical and conical spirals, in the multidomain helical magnet Cu_2OSeO_3 . This technique is sensitive to the orientation of the spiral wave-vector in the single domain conical and the multidomain helical states. The observation of the non-sinusoidal angular dependence in the helical state shows its sensitivity to domain structure of this state. We showed that the SMR can be used as an all-electrical tool to study temperature dependence of the domain structure. This technique can also be used to detect and, perhaps, to manipulate

more complex nanomagnetic spin textures, such as domain walls and skyrmions. As the SMR signal is sensitive to the angle of the magnetization vector and the spin accumulation. It may lead to identify the sense of rotation of the skyrmion lattice [42, 43] and therefore the sign of the underlying DM interaction [44]. We believe that these findings constitute an important step forward in the emerging field of insulator spintronics, which holds promise of energy-efficient magnetic memory devices.

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Appendix. SMR in $\text{Pt|Cu}_2\text{OSeO}_3$ in the helical state

Figures A1(a), (b) and A2 show the SMR signal in the helical state of Cu_2OSeO_3 at different temperatures. The SMR signal deviates strongly from the $\sin(2\alpha)$ dependence, as explained in the main text that the helical (multidomain or single domain) state is different from the single domain conical state. Therefore, we observe a strong deviation from $\sin(2\alpha)$ behavior in the helical state. We observed a hysteresis in the trace and retrace curves and the reason for this behavior is still unclear. Nevertheless, the features and trend of the discontinuities are observed to be same in both trace and retrace curves (see figures A1(a), (b) and A2).

Figure A2 shows a non-zero SMR signal in the helical phase with maxima around 200 nV which is close to the value observed at 20 mT, emphasizing the fact that the SMR signal does not vanish at low-fields in the helical phase. At $B < 5$ mT,

Figure A1. (a) and (b) shows the angular dependence of the SMR signal (V_{SMR}) at different field and temperatures. The black and red curves represent the trace and retrace curves, respectively.

Figure A2. Angular dependence of the SMR signal (V_{SMR}) at 50 K at $B = 5$ mT. The black and red curves represent the trace and retrace curves, respectively.

the magnetization does not follow the applied field which can be related to some pinning effect and can be seen in the trace and retrace measurements which look very different from each other (the data is not included here). Therefore, to check the SMR signals below 5 mT we measured the magnetic field dependence of the SMR signal at fixed values of α . We performed both trace and retrace curves for the field dependent measurements. Both trace and retrace agree with each other but for simplicity we only included trace curves for both values of angle α (45° , 135°) in figure A3. For a $\sin(2\alpha)$ dependence of the SMR in the transverse configuration, we expect no change in the SMR signal at $\alpha = 90^\circ$, 180° and maximum change at $\alpha = 45^\circ$, 135° . Therefore to check the maximum variation in the field dependence and to relate it with the observed angular dependence of the SMR signals, we performed the field dependent scans at $\alpha = 90^\circ$, 135° . Such a measurement is shown in figure A3, after subtraction of the background signal V_0 from the measured first harmonic response V_1 .

Figure A3 shows that the change in SMR signal at $B = 20$ mT is around 150 nV which is consistent with the $V_{\text{SMR}}^{\text{ampl}}$ observed at 20 mT (see figure 2(e)). At $B < 20$ mT the SMR signal increases by decreasing the applied field for $\alpha = 90^\circ$. This observation can be explained from the fact that at $B < 20$ mT the angular dependence of the SMR signal deviates strongly from the $\sin(2\alpha)$ dependence (see figure 2(f)). The SMR signal at $\alpha = 90^\circ$ increases by decreasing magnetic field and saturates around 7 mT. For $\alpha = 135^\circ$, the SMR signal

Figure A3. Magnetic field dependence of the SMR signal ($V - V_0$) at 50 K measured at fixed values of angle α (90° and 135°). Here V_1 and V_0 represents the observed first harmonic and the background signals, respectively.

changes slightly by decreasing field till 7 mT and below 7 mT the SMR signal decreases till 60 nV at $B = 0$ mT. The field dependent measurement shown in figure 2(e) clarifies that the SMR signal does not vanish in the helical phase. Although to understand the change in the SMR signals in the helical phase, a detailed theoretical modelling is needed which is beyond the scope of this paper. The observations at 50 K agree qualitatively with the model described in the main text and in figure 4(d).

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