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Far out-of-equilibrium spin populations trigger giant spin injection into atomically thin MoS₂

Liang Cheng^{1,†}, Xinbo Wang^{1,2†}, Weifeng Yang³, Jianwei Chai³, Ming Yang³, Mengji Chen⁴, Yang Wu⁴, Xiaoxuan Chen¹, Dongzhi Chi³, Kuan Eng Johnson Goh³, Jian-Xin Zhu⁵, Handong Sun¹, Shijie Wang², Justin C. W. Song^{1,6*}, Marco Battiato^{1,7,*}, Hyunsoo Yang^{4,*}, and Elbert E. M. Chia^{1,*}

¹Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, 21 Nanyang Link, 637371, Singapore.

²Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

³Institute of Materials Research and Engineering, A*STAR (Agency for Science, Technology and Research), 3 Research Link, 117602, Singapore.

⁴Department of Electrical and Computer Engineering, National University of Singapore, 117576, Singapore.

⁵Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos 87545, USA.

⁶Institute of High Performance Computing, Agency for Science, Technology and Research, Singapore 138632

⁷Institute of Solid State Physics, Vienna University of Technology, 1040 Vienna, Austria.

[†]These authors contributed equally to this work.

*Correspondence to: Marco Battiato (marco.battiato@ntu.edu.sg), Justin Song (justinsong@ntu.edu.sg), Hyunsoo Yang (eleyang@nus.edu.sg), Elbert Chia (elbertchia@ntu.edu.sg)

Injecting spins from ferromagnetic metals into semiconductors efficiently is a crucial step towards the seamless integration of charge- and spin-information processing in a single device^{1,2}. However, efficient spin injection into semiconductors has remained an elusive challenge even after almost three decades of major scientific effort³⁻⁵, due to, for example, the extremely low injection efficiencies originating from impedance

mismatch^{1,2,5,6}, or technological challenges originating from stability and costs of the approaches⁷⁻¹². We show here that, by utilizing the strongly out-of-equilibrium nature of sub-picosecond spin current pulses, we can obtain a massive spin transfer even across a bare ferromagnet/semiconductor interface. We demonstrate this by producing ultrashort spin-polarized current pulses in cobalt and injecting them into monolayer MoS₂, a two-dimensional semiconductor. The MoS₂ layer acts both as the receiver of spin injection and as a selective converter of the spin current into a charge current, whose THz emission is then measured. Strikingly, we measure a giant spin current, orders of magnitude larger than typical injected spin current densities using currently available techniques. Our result demonstrates that technologically-relevant spin currents do not require the very strong excitations typically associated with fs lasers. Rather, they can be driven by ultralow-intensity laser pulses, finally enabling ultrashort spin current pulses to be a technologically-viable information carrier for THz spintronics.

Recently the discovery of ultrashort spin currents brought to the fore a very promising carrier of information¹³⁻¹⁷. Sub-picosecond spin-polarized current pulses (which, from now on, we will call simply spin currents) can be created in a FM after femtosecond (fs) laser excitation¹³. Their appeal comes from their intrinsic extreme time compression of signal, with a maximal overlap-free information processing and transfer frequency up to several terahertz (THz). They have already been successfully injected into other metals^{18,19}. However, it is an open question whether they could provide any advantage in tackling the issue of spin injection into semiconductors, as it has been recently proposed²⁰. We here set out to experimentally demonstrate that such

out-of-equilibrium spin current pulses possess the remarkable characteristic of an extremely highly-efficient injection in semiconductors across the simplest of contacts — the bare metal/semiconductor interface. We will also illustrate how this arises from strong out-of-equilibrium effects, empirically demonstrating how far-from-equilibrium distributions, with their increased number of degrees of freedom allow for the appearance of a number of exciting emergent phenomena.

The strategy is as follows: a strongly out-of-equilibrium carrier distribution is generated in the FM layer by a fs laser excitation (Fig. 1b). As the out-of-equilibrium excited carriers diffuse, they relax in energy¹³ (schematically depicted in Fig. 1a). Due to the strong spin asymmetry of the high energy electron-electron scattering lifetimes²¹, minority spin electrons are expected to thermalize very quickly towards a hot Fermi-Dirac distribution, while a significant number of majority spin electrons will persist for a longer time at high energies (Fig. 1c). As theoretically proposed in Ref. 20, of those electrons that reach the FM/SC interface, only those with excitation energy higher than the SC conduction band minimum can cross into the SC layer. The SC's bandgap will hence filter only the high-energy carriers where the population is almost fully spin polarized²⁰; this is expected to lead to an extremely-high spin polarization of the current injected into the SC, as depicted in Fig. 1d.

However, realizing the above spin-injection concept experimentally is a formidable challenge. While measuring spin currents is already in general problematic, it is in this case even more complicated since such spin currents exists for just few hundreds of fs. Even more importantly, it is imperative to prove that the measurement implies a spin current flowing within the SC, and not a spurious signal generated within the metallic regions of the sample.

To address the first problem, we focus on semiconductors with high spin-orbit (SO) interaction. In these, the spin current flowing through the SC can be converted, via the SO-mediated effects^{3,12}, into a charge current flowing in a direction orthogonal to both the current propagation direction and the spin axis. Such charge current pulses (flowing parallel to the surface) will produce THz radiation that can be collected at a detector and used to reconstruct the temporal shape of the current itself¹⁴⁻¹⁷.

Unfortunately, not all high-SO SCs are suitable. First, the SC needs to possess a finite bandgap everywhere. Moreover, the FM/SC interface has to remain chemically and structurally stable. MoS₂ (in a Co/MoS₂ heterostructure) proved to be an ideal venue. Its high SO interaction allows for a high spin to charge conversion (SCC) efficiency²², which is essential to selectively pinpoint the SC layer as the source of the THz radiation. Furthermore, the 2D semiconducting character of MoS₂ — the exposed gapped surface states together with the smooth and atomically thin Co/MoS₂ interface — guarantees that no metallic character is expected anywhere on the MoS₂ layer, and therefore that any signal coming from MoS₂ implies that spin has been injected across its bandgap.

To this end, we fabricated Co/MoS₂ heterostructures, with care to ensure that the MoS₂ film is (1) pure, (2) continuous, (3) truly monolayer, and (4) contains no intrinsic free carriers. Large-area monolayer MoS₂ films (> 7 mm diameter) were grown by physical vapour deposition (PVD)²³ on c-plane sapphire substrates; Raman and photoluminescence data confirm its good quality and monolayer thickness²³. A ferromagnetic (5-nm cobalt) and protection layer against oxidation (3-nm SiO₂) were then grown by magnetron sputtering, giving rise to a SiO₂/Co/MoS₂/sapphire heterostructure, and the Raman data of this heterostructure shows that the damage of

sputtering on MoS₂ is insignificant. X-ray photoelectron spectroscopy (XPS) data shows that the MoS₂ film contains only Mo and S atoms, while atomic force microscopy (AFM) data shows the MoS₂ film to be contiguous, and the roughness of both the Co/MoS₂ and pure MoS₂ are small (0.3–0.4 nm; for a full account of the sample growth and characterization, see Supplementary Information). The quality of the sample ensures that the MoS₂ layer is indeed a 2D SC. This is confirmed by THz transmittance measurements (Fig. 2a) which show that the MoS₂ film has zero THz absorption within our sensitivity. Indeed, the THz absorption of Co/MoS₂ heterostructure and pure Co film are virtually identical. This indicates that MoS₂ acquires no Drude weight after Co deposition, hence possessing no intrinsic free carriers or metallic character.

We irradiated the sample (Fig. 2b) with 50-fs linearly-polarized optical laser pulses (800 nm wavelength, 1 kHz repetition rate, diameter 7 mm, 0.78 mJ/cm²). The magnetization direction of Co is controlled by an external magnetic field (~800 Oe). The emitted THz radiation impinges on a THz polarizer and is then detected by electro-optic sampling using a ZnTe detector crystal. A strong THz emission is detected from the Co/MoS₂ (Fig. 2c). Interestingly, the THz emission has amplitude ~2% of that generated by the commonly-used 1-mm thick THz generating crystal ZnTe at the same pump fluence.

In the following we pinpoint the origin of our emitted THz signal, by providing a full parameter-space characterization of the phenomenon. In particular we will show that the emitted THz amplitude (1) originates from the heterostructure itself, rather than from the Co or MoS₂ layers independently; (2) rotates with the magnetic field direction, indicating its *spin*-based origin, and (3) switches sign upon sample flip, indicating a *spin-current*-based origin²⁴.

Previous research reported that pure Co thin film can emit THz radiation originating from a fs-laser induced demagnetization process^{25,26}. Fig. 2c shows that the emitted THz amplitude from the Co/MoS₂ heterostructure is ~ 10 times that from pure Co film of the same thickness. While some THz emission is expected to originate in MoS₂ due to nonlinear optical effects²⁷⁻²⁹, which depend on the relative orientation between lattice and light polarization, we measure a negligible THz emission from pure monolayer MoS₂ (magenta line, Fig. 2c). This is consistent with the polycrystalline nature of our PVD-grown MoS₂ thin films. Further, we find that the emitted THz amplitude is independent of pump-polarization θ_P (the angle between the laser polarization and the THz polarizer axis, see red points in Fig. 3a). These, together, establish that the THz generation in the Co/MoS₂ heterostructure does not depend on the nonlinear optical response that is related to the crystalline structure of the samples and its relative orientation with respect to the laser polarization axis.

Second, to confirm the spin origin of our signal, the THz amplitude was measured as a function of the direction of the applied magnetic field (θ_M), as shown in Fig. 3a and 3b. These show the strong Co magnetization tuning of the THz peak. In particular, three features of the θ_M -dependence stand out: (1) THz peak amplitude exhibits a 360° symmetry which can be fitted to a sinusoidal function (Fig. 3a), (2) the maximum (minimum) of the THz peak is at $\theta_M = 180^\circ$ (0°), establishing that the polarization of emitted THz pulses is always perpendicular to the magnetic field, and (3) the THz time-domain normalized waveforms (and their corresponding frequency-domain spectra after Fourier transform) at different θ_M are similar, (excluding important contributions from nonlinear effects which should rotate following crystal symmetry axis²⁷⁻²⁹). These [1-3] are fully consistent with the SCC mechanism in MoS₂ that obeys $\vec{j}_C \propto \vec{j}_S \times \vec{M} / |\vec{M}|$.

Lastly, to confirm the spin-*current*-based origin of our signal, we flip the sample (to change the sign of the spin injection current $\vec{j}_S$ in the lab frame), Fig. 3b shows that the polarity of the emitted THz pulses from Co/MoS₂ with front- and back-incident pump are opposite. This eliminates Co-demagnetization as a source of the measured THz radiation (as that would give the same polarity for both configurations²⁶). Note that the observed time difference between THz signal from front and back pump is due to the different refractive index of substrate (c-cut sapphire) for 800-nm and THz pulses.

All the three above experimental observations, as illustrated by Fig. 3a and 3b, collectively confirm the spin-current origin of the giant THz emission from the Co/MoS₂ interface. Indeed, this picture finds further corroboration in our photocurrent measurements, which directly measures the charge current induced by the spin injection. Fig. 3a (magenta triangles) shows the simultaneous measurements of photocurrent and THz emission on Co/MoS₂ (see Supplementary Information). The same θ_M -dependence of the THz peak and photocurrent indicate that the two signals arise from the same origin: the SCC of large spin-currents injected from the Co/MoS₂ interface discussed above.

The above results establish that we have generated and directly probed ultrafast spin injection across a FM/SC interface. Even more striking, however, is the strong dependence of the emitted THz amplitude on laser wavelength in Fig. 4a and 4d at constant absorbed fluence (the absorbed fluence is obtained by measuring the incident, reflected and transmitted power of the device and beam diameter). As we explain below, these provide clear empirical evidence/demonstration of the strongly-out-of-equilibrium nature of spin injection in our devices, and its dependence on MoS₂ material parameters.

We first clarify that these results (Fig. 4a and 4d) are incompatible with a close to equilibrium (hot) electron distribution. If the spin diffusion after a fs laser excitation

were to happen when the electrons have already attained internal thermalization^{30,31} as shown in Fig. 4b, the spin polarization and the amount of injection would depend *only* on the total deposited energy (number of absorbed pump photons $\times$ pump photon energy) displaying insensitivity to photon energy/wavelength; the total deposited energy, in turn, defines the temperature of the thermalized hot Fermi-Dirac distribution of carriers. Remarkably, our observation in Fig. 4a and 4d show that the THz peak amplitude increases with increasing pump photon energy for the same amount of absorbed energy. As a result, we conclude that a non-thermalized and far out-of-equilibrium spin population dominates our THz signal (Fig. 4c).

The detailed photon-energy resolved map (from 0.5 – 2.5 eV) in Fig. 4d, enables us to sensitively track the energy dependence of the out-of-equilibrium spin-current-injection process. While the THz emission of Co by itself is photon-energy independent (grey region Fig. 4d), THz emission from the Co/MoS₂ heterostructure rapidly rises for photon energies above 0.5 eV, achieving signals ~ 10 times larger when photons of 2.5 eV are incident. To understand this rapid increase, we note that the out-of-equilibrium distribution of laser excited electrons (holes) in the FM is continuous and extends from the Fermi energy to energies that are up to (dip below) the laser frequency above (below) the Fermi energy. At very low excitation energies (< 0.55 eV, see red region of Fig. 4d, and Fig 4e) none of the excited electrons or holes have sufficient energy to overcome the Co/MoS₂ Schottky barrier. Indeed, in this regime, our measured Co/MoS₂ THz signal coincides with that of Co by itself – see grey shaded region in Fig. 4d. As a result, we conclude that the dominant THz emission observed in Co/MoS₂ is dominated by processes within the Co layer (e.g. via ultrafast demagnetization).

However, above the ~ 0.55 eV threshold (see white region of Fig. 4d, and Fig. 4f), a fraction of laser-excited (spin-polarized) electrons in the Co has enough energy to enter the semiconductor above the bottom of the conduction band of MoS₂ and produces spin current within it (for a more detailed discussion, see Supplementary Information and Ref. 20). This fraction of electrons increases (roughly linearly) with the photon energy. Under higher energy photon pump (> 1.20 eV, see purple region of Fig. 4d, and Fig. 4g), the hole population (whose currents' spin polarization is opposite and smaller than that from the electrons) reaches the top of the valance band of MoS₂, adding a further contribution to the net spin current, and causes a kink in Fig. 4d, and a slight decrease in the slope.

The two thresholds correspond to the electron and hole Schottky barrier heights in the Co/MoS₂ heterostructure; from our data, we can estimate these to be (0.55 ± 0.06) eV and (1.20 ± 0.11) eV respectively³². Their sum (1.75 ± 0.17) eV should be equal to the bandgap of MoS₂. This value agrees well the expected value for MoS₂ bandgap (~ 1.9 eV) found in the literature (see e.g. Ref. 33).

Finally, we estimate³⁴ the magnitude of the spin current density directly from the measured THz electric field: $J_S^{peak} = J_{\square}^{peak} - J_{\square}^{peak} \approx 4 \times (10^6 - 10^8)$ A/cm² (the detailed calculations and the discussion of spin-to-charge conversion in MoS₂ are in Supplementary Information). Even in the most conservative estimation this enormous injected spin current density is orders of magnitude larger than typical spin current injection amplitudes into semiconductors [e.g. $10^2 - 10^3$ A/cm² using spin-pumping³⁵, $10^{-1} - 10^0$ A/cm² via tunnel junction^{8,36}]. Its efficiency is a direct result of the far-out-of-equilibrium nature of spin current injection in our devices. It moreover works across a bare metal/semiconductor interface, circumventing the problem of impedance

mismatch, which, instead close to equilibrium spin currents face. Interestingly we obtain our record spin injection without any attempt at optimizing the device. Our work sets the bedrock for ultrafast spin currents as a technologically-viable carriers of information, and provides fundamental science with a simple method to inject massive spin currents into gapped materials.

Methods

Sample fabrication. High-quality monolayer MoS₂ films were grown by physical vapor deposition, on c-plane sapphire substrates at 800 °C by reactively sputtering of Mo target in a sulphur vapor ambient. During the deposition process, the chamber pressure was kept at 3.2×10^{-4} mbar and the sputtering power was kept at 10 W. Prior to the growth, the Mo target was pre-sputtered for 15 min to remove the contaminants on its surface. A ferromagnetic (5-nm cobalt) and protection layer (3-nm SiO₂) was then grown by magnetron sputtering. The protection layer is to prevent oxidation of the magnetic layer. The monolayer nature of MoS₂ was confirmed by Raman spectroscopy and the thickness of Co/MoS₂ device is measured by atomic force microscopy (see the sample characterization in Supplementary Information for details).

THz emission setup. As shown in Fig. S1 in Supplementary Information, THz emission spectroscopy is built in a typical time-resolved THz spectroscopy setup³⁷. Femtosecond laser pulses (Coherent Legend-Elite, 50-fs pulse duration, 1 kHz repetition rate, 800 nm wavelength, 7 mm diameter) are incident on the device (or ZnTe THz emitter), then the generated THz pulses are projected onto the x-axis (vertical) by using a wire-grid THz polarizer and then collected and focused by two parabolic mirrors onto a 1-mm-thick <110> ZnTe crystal for electro-optic sampling. The magnetic field at the sample position is ~800 Oe, which is provided by permanent magnets. The polarization angle

and magnetic field direction are controlled by motorized rotational stages (Thorlabs PRM1/MZ8). Low-temperature measurements were performed in a Cryo Industries cryostat.

Photocurrent measurement was performed on a Co/MoS₂ heterostructure with THz emission spectroscopy simultaneously. Two electrodes at opposite ends on the device were made, and the distance between them is larger than the laser diameter (~7 mm) to make sure the electrodes are not excited by the pump pulses. In the measurement, the sample with electrodes was rotated to make sure the direction of measured photocurrent (the direction from one electrode to another) is the same as THz polarizer, which lead to the similar phase of the two measurements (Fig. 3b in main text).

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

E.E.M.C. and H.Y. conceived the experiments. W.Y., Y.W. and M.C. fabricated the heterostructures. L.C. and X.W. carried out the THz measurements and data analysis with the help and guidance of E.E.M.C. and H.Y. M.B., J.C.W.S., and J.X.Z. provided theoretical inputs. W.Y. and S.W. performed and analysed the XPS and Raman measurements. L.C., X.W., J.C.W.S, M.B., and E.E.M.C wrote the manuscript together. All authors discussed the results and commented on the manuscript.

Additional information

Supplementary information accompanies this paper is available.

Competing financial interests

The authors declare no competing financial interests.

Figure 1. Ultrafast spin injection process. (a) Schematic of spin injection from Co to monolayer MoS₂. High energy electrons, generated by the fs laser, have spin dependent scattering rates. Minority electrons quickly decay to low energy and are consequently prevented by the bandgap from entering into MoS₂. **(bottom panels)** Electron population during spin injection process: **(b)** initial out-of-equilibrium electronic distribution generated by the laser absorption; **(c)** due to the comparatively smaller scattering rates, the majority-spin channel (red) retains an out-of-equilibrium distribution, while the minority-spin electrons (blue) have already decayed to a Fermi-Dirac distribution; **(d)** the MoS₂ bandgap acts as a filter and allows only high

energy electrons from Co to cross the interface, leading to a high spin polarisation of the injection.

Figure 2. Characterization of MoS₂ and Co/MoS₂ samples. (a) THz transmittance of Co/MoS₂ (blue triangle), pure Co (red circle) and pure MoS₂ (black square) film, the magenta line represents the vacuum-to-vacuum transmittance that indicates the accuracy of the THz spectrometer; **(b)** Schematic of Co/MoS₂ heterostructure and experimental configuration. **(c)** THz emission spectrum of Co/MoS₂, pure Co, pure monolayer MoS₂ and 1-mm-thick <110> ZnTe crystal ($\times 0.01$) in time and frequency domain (inset).

Figure 3. THz emission of Co/MoS₂ under different experimental conditions. (a) THz peak of Co/MoS₂ as a function of pump polarization (θ_P , red squares) and magnetic field direction (θ_M , blue balls), and photocurrent as a function of magnetic field direction (θ_M , magenta triangles), the curves indicate the sinusoidal fitting (the error bars represent standard deviations). **(b)** THz emission signal with front and back pump (the inset is the signal in frequency domain).

Figure 4. Strongly out of equilibrium distribution of spin injection (a) THz peak amplitude as a function of the absorbed power under different pump photon energy (the error bars represent standard deviations). (b-c) Comparison of thermalized (b) and non-thermalized (c) spin-dependent injection into semiconductors. The injection is insensitive to the pump photon wavelength in the thermalized case (b), while it is strongly affected by pump photon wavelength when the electrons have a non-thermalized far out-of-equilibrium distribution (c). Dot-dashed lines indicate the energy level E_c for the conduction band minima in MoS₂. (d) (THz-peak)/(absorbed fluence) of our devices under different pump wavelength. The pink circles are emitted THz signal from Co/MoS₂ device, and the gray diamonds are signal from pure Co thin film. The dotted lines (dark yellow) are guides to the eye, and the shaded gray area

represents for the level of THz emission from pure Co (the error bars represent for standard deviations). **(e-g)** Scheme of spin polarized electrons injection under different pump photon energy.

Supplementary Figure S1

Supplementary Figure S1. Schematic of THz emission and electro-optic detection setup.

Supplementary Figure S2

Supplementary Figure S2. Sample characterization: Data of XPS (a), Raman (b) and PL (c) measurement on monolayer MoS_2 . (d) is the data of thickness measurement of Co/MoS_2 measured by AFM. (e) is the topography of monolayer- MoS_2 and Co/MoS_2 heterostructure measured by AFM.

Supplementary Figure S3

Supplementary Figure S3. Scheme of spin conserving electron decay.

Supplementary Figure S4

Supplementary Figure S4. Temperature dependence of THz emission amplitude of Co/MoS₂. The signal is almost temperature-independent.

Supplementary Figure S5

Supplementary Figure S5. Ratio $|R[\omega]|$ as a function of frequency. The peaks in the frequency domain originate from multiple reflections in the substrate. The red line represents the frequency-averaged $|R[\omega]|$.

Supplementary Figure S6

Supplementary Figure S6. Two geometries of spin-charge conversion. (a) Charge to spin conversion in spin torque measurement; **(b)** Spin to charge conversion in our experiment.

Supplementary note 1

Sample characterization. The quality of MoS₂ thin film has been confirmed by XPS and Raman measurements. The thickness of MoS₂/Co devices has been measured by Atomic Force Microscopy (AFM) and confirmed by THz Time-Domain Spectroscopy (THz-TDS).

(1) XPS measurement

The surface chemistry of monolayer MoS₂ was examined with Thermo Scientific Theta Probe XPS with monochromatic Al K _{α} source ($h\nu = 1486.7$ eV) at the pass energy of 40 eV for high resolution measurement. The result (Fig. S2a) is consistent with previous reports (Ref. 23), which indicates good quality of our samples.

(2) Raman measurement

Raman measurements were performed to characterize the layer number of MoS₂ thin films using a Thermo Scientific DXR Raman microscope with a 514-nm excitation. The frequency difference between A_{1g} and E_{2g}^1 modes are 19.4 cm^{-1} (Fig. S2b), which shows good agreement with previous research (Ref. 23 and Ref. S1), and confirms the monolayer nature of MoS₂ in our devices.

(3) Photoluminescence measurement

Photoluminescence measurements were performed to characterize MoS₂ thin film. The A- and B-exciton peaks in Fig. S2c show good agreement with other research (Ref. 33), which confirms the good quality and monolayer nature of MoS₂ in our devices.

(4) AFM measurement

AFM measurements on MoS₂ and Co/MoS₂ (Fig. 2e) show the smooth surface of the devices, and their roughness is 0.3–0.4 nm. The thickness measurement (bottom of Fig. 2d) shows that the total thickness of Co/MoS₂ device is ~ 8.7 nm, which is exactly

the sum of the thicknesses of MoS₂ thin film (~0.7 nm) and capping layer (Co 5 nm+ SiO₂ 3 nm), which indicates the correct thickness of each constituent layer in the device.

(5) THz-TDS measurement

THz-TDS measurements were performed with THz spectrometer (TeraView TPS 3000) at room temperature. As shown in Fig. 2a in main text, there is no THz absorption of pure MoS₂ thin film observed from our measurement. The same THz transmittance of all the Co and Co/MoS₂ device indicates that MoS₂ has no Drude weight after Co deposition, hence possessing no intrinsic free carriers or metallic character. The vacuum-to-vacuum measurement is $(100 \pm 1\%)$, which shows that the THz spectrometer is well calibrated.

Supplementary note 2

Wavelength dependent THz emission. THz emission at parity of absorbed power by tuning the laser wavelength (see Fig. 4 in main text) which as we now discuss provide clear *empirical* confirmation of our claims in the main text.

As shown in Fig. 4a and 4d in the main text, our new measurements show large THz signals when photon energies are far above the band-edge of MoS₂, but quenched THz signals when the photon energy is tuned below the band-edge of MoS₂. This measurement clearly shows that injection of spin-polarized currents into MoS₂ is the key to the large THz amplitude that we observe. Together with a detailed analysis of the features in our new measurement (e.g., two-threshold behavior – see discussion below), our new measurements empirically establishes

- 1) the strongly-out-of-equilibrium nature of the spin-polarized-current injection process, and
- 2) that the THz generation happens *within* the MoS₂ semiconductor.

To further understand the detailed structure of the energy dependence of the spin-polarized-current-injection processes in the Co/MoS₂ heterostructure, we first recall that a wide range of transitions can be activated in Co by the laser's photons, even when they are assumed perfectly monochromatic (the time compression of the pump laser pulse will induce an energy spread). For generic metallic bands, electrons can be expected to be excited from just below the Fermi energy to well above it (generating high-energy excited electrons and shallow holes), or from well below the Fermi level to just above it (generating high-energy holes and low-energy electrons), as well as intermediate configurations. Therefore, after the pump excitation, a distribution of excited electrons exists from zero energy all the way to the photon frequency above the Fermi energy. Similarly, for holes we expect a distribution down to the photon frequency below the Fermi energy.

Tuning incident photon energy is, therefore, a natural means of adjusting the energy distribution of the out-of-equilibrium distribution function in the ferromagnet. Indeed, depending on the photon energy and the position of the metal's Fermi level compared to the semiconductor's band-edge (either conduction or valence) at the interface, parts of the excited electron or hole out-of-equilibrium distribution in the ferromagnetic metal may have enough energy to be injected into the semiconductor. For example, for photon energies smaller than the Schottky barrier in Co/MoS₂ (Fig. 4e in main text), no electrons or holes have enough energy to be injected into the semiconductor, and the THz signal emitted is small; THz emitted in this regime of photon energy (energies below ~ 0.55 eV) arise from processes within the Co itself (e.g. ultrafast demagnetization), see shaded band in Fig. 4d of the main text. We remark that THz

emission from the Co by itself does not exhibit any photon energy dependence (gray shaded band).

However, for photon energies above this threshold, electrons excited in the Co (see below for justification of carriers as electron type) have enough energy to reach the bottom of the semiconductor's conduction band (see Fig. 4f in main text). Their spin polarization (as described in Ref. 20) induces the spin-polarized current in the semiconductor. The higher the photon energy, the larger is the fraction of the excited spin-polarized electrons that can travel into the semiconductor, and the larger the THz emission. This is clearly seen in the linear increase of the THz emission all the way up to ~ 1.20 eV in Fig. 4d in main text.

Above the ~ 1.20 eV threshold, holes have enough energy below the Fermi level to enter the semiconductor's valence band (see Fig. 4g in main text). Holes are expected to have a spin polarization that is smaller and with opposite sign compared to the electrons (see below, for a full discussion) We clearly observe a kink in the pump-energy dependence (see Fig. 4d in main text). This kink and slope change fully agrees with theoretical expectations, see Fig. 4e-g in main text.

The two thresholds correspond to the electron and hole Schottky barrier heights in the Co/MoS₂ heterostructure. From our data, we can estimate these as (0.55 ± 0.06) eV and (1.20 ± 0.11) eV respectively. The error in our extractions for the Schottky barrier heights can be estimated by considering the bandwidth of the pulse from our OPA (bandwidth ~ 80 nm at center wavelength 1000 nm and bandwidth ~ 250 nm at 2300 nm) that broadens the excitation profile. Their sum (1.75 ± 0.17) eV should be equal the bandgap of monolayer MoS₂. This value agrees well the expected value for monolayer MoS₂ bandgap (~ 1.9 eV) found in the literature (see e.g., Ref. 33.). This shows the

excellent agreement between the measured THz phenomena and the MoS₂ material parameters.

We note parenthetically that one might be tempted to expect some kink at a photon energy equal the MoS₂ bandgap. That however is not the case for the following reasons:

- (1) We have pumped the pure MoS₂ film (with 800 nm and 400 nm) in the absence of the Co layer, and no THz emission was observed.
- (2) The direct excitation in the MoS₂ monolayer is only a very small fraction of that in the Co layer, due to the very limited thickness of MoS₂, and the fact that the presence of the bandgap generates (even when the laser has sufficient energy to overcome the bandgap) a much smaller joint density of states for absorption compared to the metallic layer.
- (3) Even for very low-energy photon excitation (where there should be no direct excitation of carriers in MoS₂), we could still detect an emitted THz signal (see Fig. 4d in main text). This supports the model in our work.

Therefore, the direct excitation of MoS₂ does not induce any sizeable spin-polarized current, and does not influence or contribute to the generation of THz radiation.

This photon-energy-resolved measurement, therefore, empirically demonstrates that the injected spin-polarized currents are (1) a strongly-out-of-equilibrium effect, and (2) that the THz is indeed generated within the MoS₂ layer. Further, it provides vivid frame-by-frame (energy selective) picture of the injection process.

Further details about the above discussion. Here we provide further details into the discussion above. First, we recall that the spin polarization of the current generated by carriers within a certain energy range depends on the spin asymmetry in their group velocity and lifetime (see Ref. 13 and 20). Below we provide an intuitive argument to

estimate this asymmetry; precise values can be obtained from *ab initio* calculations (see Ref. 21, Ref. S2 and S3).

We proceed with a rough but meaningful estimation of the overall behavior of the (spin-conserving) electron scattering lifetimes for carriers at a given energy and spin, by integrating the states in the same spin channel that have energies between the considered energy and the Fermi level (see Fig S3). The more the states available for decay, the shorter the lifetime. By considering the Co density of states, we can see how majority-spin electrons tend to have longer lifetimes compared to the minority-spin electrons. The situation is similar for holes — majority-spin holes tend to have longer lifetimes compared to minority-spin ones (yet with a smaller spin asymmetry compared to electrons). However, since majority-spin holes carry minority spin, the spin polarization of the hole current is smaller, and is reversed in sign, compared to that from electrons.

The arguments above allow us to identify that the first threshold is due to electrons, while the second is due to holes.

Supplementary note 3

Temperature dependent THz emission. We also study the temperature dependence of THz emission from Co/MoS₂ device. Fig. S4 shows insensitivity of the THz emission strength on the temperature. This is consistent with a previous report (Ref. S4), where such insensitivity was ascribed to temperature-insensitive Rashba strength and Fermi level in MoS₂.

Supplementary note 4

Photocurrent measurement. Two electrodes are fabricated at the opposite edges of the sample. And the photocurrent data shows in Fig. 3a was taken simultaneously with THz

emission measurement. Due to the fact that (1) our electrical contacts are deposited on the SiO₂ surface, (2) there is a large conductivity difference between Co and MoS₂ thin films, and (3) the current meter has a slow response, the measured value of the photocurrent will not be indicative of the actual *transient* photocurrent that flows through MoS₂. The aim of our photocurrent measurement is to show that it has the same magnetic-field-direction symmetry as that of the THz emission signal. That is why we do not display the actual measured photocurrent in our manuscript.

Supplementary note 5

Electric field strength of emitted THz pulse from ZnTe and Co/MoS₂. Fig. S1 gives a schematic of the EO-detection setup. The 800 nm pump beam comes in from the right. To obtain the absolute amplitude of the electric field, we follow the standard method in Ref. S5. For completeness we detail this below. First, we calculate the electric field strength of the THz radiation detected inside the ZnTe detector crystal (Position A in Fig. S1)⁵:

$$\frac{\Delta I}{I} = \frac{\omega n^3 E_{THz}^{Det, ZnTe} r_{41} L}{2c} (\cos \alpha \sin 2\phi + 2 \sin \alpha \cos 2\phi), \quad (S1)$$

where c is speed of light in vacuum, $\omega = 2\pi \times c/(800 \text{ nm})$ is angular frequency of detection/gating laser (see Fig. S1), $n = 2.85$ is the refractive index of ZnTe crystal at ω (Ref. S6), $r_{41} = 3.9 \text{ pm/V}$ is the non-linear coefficient of ZnTe (Ref. S7), $L = 1 \text{ mm}$ is the thickness of our ZnTe detection crystal. In our setup, $\alpha = \pi/2$ is the azimuthal angle of detection ZnTe crystal, $\phi = \alpha + \pi/2 = \pi$ is the angle of the probe/gating beam polarization with respect to the crystal (001) axis, and $\Delta I/I$ is the normalized intensity difference of the polarization components of the gate pulse (see Fig. S1), which we measured as $\frac{\Delta I}{I} = \frac{0.16 \pm 0.01}{2.00 \pm 0.01} = 0.080 \pm 0.005$ at the THz peak.

Using Equation S1, we find the electric field strength detected by the EO sampling inside ZnTe crystal to be $E_{THz}^{Det,ZnTe} = 1.13 \pm 0.07$ kV/cm.

The THz electric field immediately after the ZnTe emitter crystal (Position B in Fig. S1) can be obtained from $E_{THz}^{Det,ZnTe}$ [electric field strength of the THz radiation detected inside the ZnTe detector crystal (Position A in Fig. S1)] by considering the transmittance of various optics in our setup: (1) high-density polyethylene (HDPE) $t_{HDPE} = 0.93 \pm 0.01$, (2) THz polarizer $t_{THzP} = 1.00 \pm 0.01$ (from our measurement), (3) beamsplitter $t_{BS} = 1.00 \pm 0.01$ (from our measurement), and (4) surface of the detection ZnTe crystal $t_{ZnTe} = \frac{2 \times n_{air}}{n_{air} + n_{ZnTe}^{THz}} = \frac{2 \times 1}{1 + 3.3} = 0.465$ [from $n_{ZnTe}^{THz} \approx 3.3$, taken from Ref. S6]. Taking these into account, the electric field strength of the emitted THz pulse from the ZnTe emitter is:

$$E_{THz}^{Emit,ZnTe} = \frac{E_{THz}^{Det,ZnTe}}{t_{HDPE} t_{THzP} t_{BS} t_{ZnTe} M} \approx 2.61 \pm 0.08 \text{ kV/cm},$$

where M is the ratio of the spot sizes on the detector and emitter. In our setup, we used a confocal arrangement to ensure $M = 1$ — the focal lengths of two off-axis parabolic mirrors are the same, and the THz emitter and detection crystal are placed at the focal point, so the area of the THz emission spot on the ZnTe emission crystal, and THz detection spot on the ZnTe detection crystal, are the same.

Since the ratio of the THz peaks between Co/MoS₂ and ZnTe is $\sim 2.1\%$, the THz peak electric field strength of Co/MoS₂ is

$$E_{peak}^{Co/MoS_2} = E_{THz}^{Emit,ZnTe} \times 2.1\% \approx 54.9 \text{ V/cm}.$$

Conversion from electric field to surface charge current. We report below how to reconstruct the shape of the charge current from the experimental values of the electric field. We assume the MoS₂ monolayer as a plane, and the charge current running

through it as a 2D surface current. We solve the Maxwell's equation for a layered sample (air/5-nm Co/surface current/0.5-mm sapphire/air). For completeness and the reader's convenience we report the full derivation, even if it is a straightforward application of electromagnetism.

In the considered geometry the Maxwell's equations hold separately in each layer (we will address the boundary conditions later on) and reduce to

$$\begin{aligned}\frac{\partial E_y}{\partial x} &= -\frac{\partial B_z}{\partial t}, \\ \frac{\partial H_z}{\partial x} &= -\frac{\partial D_y}{\partial t},\end{aligned}$$

For convenience we focus on a single frequency component and drop the direction indices

$$\begin{aligned}E[x, t] &= \Re[E[x]e^{i\omega t}], \\ B[x, t] &= \Re[B[x]e^{i\omega t}],\end{aligned}$$

where $\Re$ represents the real part. We now explicitly write the material properties (in the frequency domain)

$$\begin{aligned}H &= B/\mu_0, \\ D &= (\epsilon - i\sigma/\omega)E,\end{aligned}$$

where ϵ and σ are the refractive index and the conductivity of the material. We obtain

$$\begin{aligned}E' &= -i\omega B, \\ B' &= -i\omega\mu_0(\epsilon - i\sigma/\omega)E,\end{aligned}$$

where the prime represents the spatial derivative. We rewrite as

$$\begin{aligned}E'' &= k^2 E, \\ B &= iE'/\omega,\end{aligned}$$

where

$$k = \sqrt{\omega\mu_0(i\sigma - \omega\epsilon)},$$

is a generalized complex momentum and incorporates the response of the material.

The solution to the equations above is

$$E[x] = \alpha e^{kx} + \beta e^{-kx},$$

$$B[x] = \frac{i\alpha k}{\omega} e^{kx} + \frac{i\beta k}{\omega} e^{-kx},$$

leading to the fields

$$E[x] = \Re[\alpha e^{i\omega t + kx} + \beta e^{i\omega t - kx}],$$

$$B[x] = \Re\left[\frac{i\alpha k}{\omega} e^{i\omega t + kx} + \frac{i\beta k}{\omega} e^{i\omega t - kx}\right].$$

The component of the electromagnetic radiation with amplitude α (β) propagates from right to left (left to right).

The boundary conditions are continuity of the electric fields at each surface, and continuity of the magnetic field at every surface except the one where the surface current is flowing, where instead the magnetic field has a jump equal to $\mu_0 \Re[J_{surf} e^{i\omega t}]$. Furthermore, we need to impose no radiation is incoming in the sample.

After some math and keeping into account that the boundary conditions have to hold at any time, we can write the set of equations

$$(\alpha_{airLeft} e^{k_{air} x} - \alpha_{Co} e^{k_{Co} x} - \beta_{Co} e^{-k_{Co} x})|_{x=x_{air-Co}} = 0,$$

$$(\alpha_{airLeft} k_{air} e^{k_{air} x} - \alpha_{Co} k_{Co} e^{k_{Co} x} + \beta_{Co} k_{Co} e^{-k_{Co} x})|_{x=x_{air-Co}} = 0,$$

$$(\alpha_{Co} e^{k_{Co} x} + \beta_{Co} e^{-k_{Co} x} - \alpha_{AlO} e^{k_{AlO} x} - \beta_{AlO} e^{-k_{AlO} x})|_{x=x_{Co-AlO}} = 0,$$

$$(\alpha_{Co} k_{Co} e^{k_{Co} x} - \beta_{Co} k_{Co} e^{-k_{Co} x} - \alpha_{AlO} k_{AlO} e^{k_{AlO} x} + \beta_{AlO} k_{AlO} e^{-k_{AlO} x})|_{x=x_{Co-AlO}}$$

$$= \mu_0 \omega J_{surf},$$

$$(\alpha_{AlO} e^{k_{AlO} x} + \beta_{AlO} e^{-k_{AlO} x} - \beta_{airRight} e^{-k_{air} x})|_{x=x_{AlO-air}} = 0,$$

$$(\alpha_{AlO} k_{AlO} e^{k_{AlO} x} - \beta_{AlO} k_{AlO} e^{-k_{AlO} x} + \beta_{airRight} k_{air} e^{-k_{air} x})|_{x=x_{AlO-air}} = 0,$$

The equations above are a simple linear system, which we solve to obtain the ratio

$$R[\omega] = \frac{\beta_{airRight}[\omega]}{J_{surf}[\omega]},$$

between the electric field amplitude emitted on the sapphire side and the driving current at a certain frequency ω . We do not explicitly report the expression since it is lengthy; we however remind that all the generalized momenta are explicitly frequency dependent.

As materials' properties we use $\epsilon = 3.1\epsilon_0$ and $\sigma = 0$ for sapphire (thickness 0.5 mm). In the case of Co (thickness 5 nm) we use $\sigma = 1.7 \times 10^7$ S/m. Given the good metallic nature of Co, we use $\epsilon = \epsilon_0$, de facto ignoring the induced dipole moment generated by its dielectric response, since the one generated by induced currents is importantly larger.

The ratio $R[\omega]$ is found to be oscillating with frequency (due to interference within the sapphire layer, see Fig. S5) around an average value within the THz frequency range of

$$\langle |R[\omega]| \rangle \approx 14.0 \text{ V/A}.$$

For simplicity we will use the average to estimate the charge current. We can thus estimate the peak surface THz current as

$$J_{surf,peak} = \frac{54.9 \text{ V/cm}}{14.0 \text{ V/A}} = 3.92 \text{ A/cm}.$$

We stress that the charge current density obtained above is an underestimation. In reality, carriers are photogenerated by the femtosecond laser pulses, leading to greater

THz absorption by these carriers and increased conductivity of the Co and MoS₂ layers. In our derivation above we have used equilibrium properties — this is a conservative choice, where the underestimated charge current density leads also to an underestimated spin current density (shown below).

Supplementary note 6

Conversion from charge current to spin current. To estimate the spin current density, we need to know the conversion efficiency from spin current to charge current. In MoS₂, the spin to charge conversion is based on the inverse Rashba-Edelstein effect (Ref. S4) which can be described by (Ref. S8):

$$J_C(\text{A/m}) = \lambda_{IREE} j_S(\text{A/m}^2),$$

where λ_{IREE} is the measure of spin-to-charge conversion efficiency. Conventionally, the spin-to-charge conversion efficiency in the (inverse) Rashba-Edelstein effect has been expressed in the language of the (inverse) spin Hall effect (Ref S4, S8 and S9) via the "spin torque ratio" or "effective spin Hall angle" θ

$$J_C(\text{A/m}) = \lambda_{IREE} j_S(\text{A/m}^2) = \theta t_{\text{MoS}_2} j_S(\text{A/m}^2),$$

where t_{MoS_2} is the thickness of the MoS₂ layer (0.7 nm).

Literature provides data for θ ranging over 2 orders of magnitude (Ref. S4). For illustrative purpose only, in the next section we show an argument that hints that such large discrepancy of experimental values is only apparent and might be explained by differences in the geometry and a proper application of the Onsager relations.

Nonetheless, it is not our intention to contribute to that particular research field with the present work: we therefore simply provide here an estimation of the spin current using the highest $\theta = 12.7$ (Ref. S10), and lowest $\theta = 0.14$ (Ref. S4) values found in literature. We obtain the range

$$j_{S,peak} \approx 4 \times (10^6 - 10^8) \text{ A/cm}^2.$$

Summarizing, we have estimated the spin current by reconstructing in turn the electric field within the detector, the electric field at the sample surface, the charge current generating the THz, and finally the spin current. In these three steps we have always made conservative approximations, which result in an underestimation of the current. In the last step, we have opted for taking the full range of experimentally measured spin to charge conversions. Taking the lower bound in the range of $j_{S,peak}$ above provides a conservative estimation of the spin current.

Supplementary note 7

Onsager relations for charge current to spin current and spin current to charge current. For illustrative purpose only, we provide here a qualitative indication as to why the large discrepancy between the experimental values provided in literature for spin/charge conversion might stem from the fact that the conversion factor has been measured on the direct effect in one case and the inverse in the other. In Ref. S⁴, where a charge current in the y direction (Fig. S6a) is used to drive a spin current in the x direction, the authors obtain $\theta_{C \rightarrow S} = 0.14$. Conversely a spin current in the x direction is used to drive a charge current in the y direction (Fig. S6b) in Ref. S10, and the value $\theta_{S \rightarrow C} = 12.7$ is obtained. Naively one might expect the two conversion factors to have the same value due to Onsager relations. However, we show below that in more complex geometries, those symmetries need to be applied with some more care.

To derive the relation between $\theta_{S \rightarrow C}$ and $\theta_{C \rightarrow S}$, we explicitly account for the strong (in-plane vs out-of-plane) anisotropy of the MoS₂ layer. For simplicity we provide equations for bulk transport, since our only aim is showing the effect of the system anisotropy on the considered effects, and not providing a precise description.

Using a simple two-spin channel model, we can write

$$\begin{bmatrix} J_{x\uparrow} \\ J_{x\downarrow} \\ J_{y\uparrow} \\ J_{y\downarrow} \end{bmatrix} = \frac{1}{2e} \begin{bmatrix} (1-\delta)\sigma_x & \delta\sigma_x & (1-\delta)\sigma_{xy} & -\delta\sigma_{xy} \\ \delta\sigma_x & (1-\delta)\sigma_x & \delta\sigma_{xy} & -(1-\delta)\sigma_{xy} \\ -(1-\delta)\sigma_{xy} & \delta\sigma_{xy} & (1-\delta)\sigma_y & \delta\sigma_y \\ -\delta\sigma_{xy} & (1-\delta)\sigma_{xy} & \delta\sigma_y & (1-\delta)\sigma_y \end{bmatrix} \begin{bmatrix} d\mu_{\uparrow}/dx \\ d\mu_{\downarrow}/dx \\ d\mu_{\uparrow}/dy \\ d\mu_{\downarrow}/dy \end{bmatrix},$$

where $J_{x\uparrow}$ ($J_{x\downarrow}$, $J_{y\uparrow}$, $J_{y\downarrow}$) is the current of spin up (down, up, down) carriers in the x (x, y, y) direction, $e^{-1}d\mu_{\uparrow}/dx$ ($e^{-1}d\mu_{\downarrow}/dx$, $e^{-1}d\mu_{\uparrow}/dy$, $e^{-1}d\mu_{\downarrow}/dy$) is the effective electromotive force in the x (x, y, y) direction for spin up (down, up, down) carriers; σ_x (σ_y) is the conductivity in the x direction, σ_{xy} is the off-diagonal element of the conductivity tensor which can be non-zero for a specific spin component only in the presence of finite spin orbit coupling; δ comes from spin-flip scatterings and can be estimated from Drude model as $\delta = \frac{\tau}{\tau + \tau_{sf}}$, where τ is the Drude lifetime, and $\tau_{sf} > \tau$ the spin-flip lifetime (we highlight that the presence of spin-flip scatterings will not be necessary to our argument, but is added for completeness). The conductivities σ_x and σ_y are spin independent, but direction dependent. The symmetries of the terms including σ_{xy} are dictated by the Onsager relations. The other symmetries can be obtained by the symmetry of spin-orbit coupling. As we will see, the origin of the various electromotive forces will not matter since we will be interested only in the ratio between currents flowing. It is easy to verify that the conductivity tensor is, as it should be,

$$\begin{bmatrix} J_x \\ J_{y\uparrow} \end{bmatrix} = \begin{bmatrix} J_{x\uparrow} + J_{x\downarrow} \\ J_{y\uparrow} + J_{y\downarrow} \end{bmatrix} = \frac{1}{e} \begin{bmatrix} \sigma_x & 0 \\ 0 & \sigma_y \end{bmatrix} \begin{bmatrix} d\mu/dx \\ d\mu/dy \end{bmatrix}$$

We first consider the case of Ref. S4, where a charge current is used to drive a spin current. According to the geometry of Ref. S4 (Fig. S6a), the electromotive forces are

$$\frac{1}{e} \begin{bmatrix} d\mu_{\uparrow}/dx \\ d\mu_{\downarrow}/dx \\ d\mu_{\uparrow}/dy \\ d\mu_{\downarrow}/dy \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ \nabla V \\ \nabla V \end{bmatrix},$$

leading to a charge current along y and spin current along x as

$$J_{Cx} = J_{x\uparrow} + J_{x\downarrow} = 0 = J_{Sy} = J_{y\uparrow} - J_{y\downarrow},$$

$$J_{Cy} = J_{y\uparrow} + J_{y\downarrow} = \sigma_y \nabla V,$$

$$J_{Sx} = J_{x\uparrow} - J_{x\downarrow} = \sigma_{xy}(1 - 2\delta) \nabla V.$$

As a result, the ratio between the *induced* spin current and a *driving* charge current is

$$\theta_{C \rightarrow S} = \frac{J_{Sx}}{J_{Cy}} = \frac{\sigma_{xy}(1 - 2\delta)}{\sigma_y}.$$

The geometry in Ref. S11 (similar to our experiment), however, is different (Fig. S6b): we directly inject a spin current. As described in Ref. S11, this injected spin current is largely charge neutral. The nature of the driving force is not relevant for the current discussion. Further, given that the driving force will disappear in taking the ratio of spin to charge currents, for simplicity we use the same notation as above.

Using the notation above, the injected spin current can be modeled as

$$\frac{1}{e} \begin{bmatrix} d\mu_{\uparrow}/dx \\ d\mu_{\downarrow}/dx \\ d\mu_{\uparrow}/dy \\ d\mu_{\downarrow}/dy \end{bmatrix} = \begin{bmatrix} \nabla V \\ -\nabla V \\ 0 \\ 0 \end{bmatrix}.$$

It is easily verified that the form of the driving forces above induces a pure spin current along x. For this configuration, the spin current along x and the charge current along y are

$$J_{Cx} = J_{x\uparrow} + J_{x\downarrow} = 0 = J_{Sy} = J_{y\uparrow} - J_{y\downarrow},$$

$$J_{Sx} = J_{x\uparrow} - J_{x\downarrow} = \sigma_x(1 - 2\delta) \nabla V,$$

$$J_{Cy} = J_{y\uparrow} + J_{y\downarrow} = -\sigma_{xy}\nabla V.$$

Taking the ratio between the induced charge current and the inducing spin current above (for this configuration) we obtain

$$\theta_{S\rightarrow C} = \frac{J_{Cy}}{J_{Sx}} = -\frac{\sigma_{xy}}{(1-2\delta)\sigma_x}.$$

Interestingly, the ratios between the currents are *not* the same in the two geometries, but are related by the anisotropy of the device geometry:

$$|\theta_{S\rightarrow C}| = |\theta_{C\rightarrow S}| \frac{\sigma_y}{(1-2\delta)^2\sigma_x} \cong |\theta_{C\rightarrow S}| \frac{\sigma_y}{\sigma_x}$$

where the approximate equality holds because δ is a small number.

We can therefore estimate $\theta_{S\rightarrow C}$ from $\theta_{C\rightarrow S} = 0.14$ [provided in Ref. S4] and the ratio between the in-plane (σ_y) and out-of-plane (σ_x) conductivities in MoS₂. As an illustration of the strong effect anisotropy can have, we note that the literature provides a number of experimental measurements of the ratio σ_y/σ_x in *bulk* MoS₂ of order 100 to 170 (Ref. S12, S13). These yield $\theta_{S\rightarrow C} = 14.0 - 23.8$. While we expect that for heterostructures [such as the Co/monolayer-MoS₂ system we consider here], values of the anisotropy ratio σ_y/σ_x may be different, we remark that the values of $\theta_{S\rightarrow C}$ inferred above turn out to coincide with the value of $\theta_{S\rightarrow C} = 12.7$ obtained in Ref. S10.

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