Observation of nonreciprocal magnon Hanle effect

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The precession of magnon pseudospin about the equilibrium pseudofield, the latter capturing the nature of magnonic eigen-excitations in an antiferromagnet, gives rise to the magnon Hanle effect. Its realization via electrically injected and detected spin transport in an antiferromagnetic insulator demonstrates its high potential for devices and as a convenient probe for magnon eigenmodes and the underlying spin interactions in the antiferromagnet. Here, we observe a nonreciprocity in the Hanle signal measured in hematite using two spatially separated platinum electrodes as spin injector/detector. Interchanging their roles was found to alter the detected magnon spin signal. The recorded difference depends on the applied magnetic field and reverses sign when the signal passes its nominal maximum at the so-called compensation field. We explain these observations in terms of a spin transport direction-dependent pseudofield. The latter leads to a nonreciprocity, which is found to be controllable via the applied magnetic field. The observed nonreciprocal response in the readily available hematite films opens interesting opportunities for realizing exotic physics predicted so far only for antiferromagnets with special crystal structures.

The quantized excitations of the spin system in ordered magnets - magnons - offer a unique platform for intriguing science and technology. Their solid state host and associated quantized spin make them promising as information carriers [1–4], comparable to electrons. At the same time, their bosonic nature allows for phenomena typically exploited in, e.g., optics and optomechanics [5]. Combining these features, antiferromagnetic magnons with their high frequencies [6–8] and tunable spin [9, 10] offer fast operation and robustness against thermal fluctuations, among several advantages [7, 8]. Numerous theoretical proposals exploit the diversity and engineerability of antiferromagnetic magnons for unprecedented phenomena [11–19]. However, their high frequencies also pose challenges making conventional GHz spectroscopies, although showing rapid experimental progress with ferromagnetic magnons, not suitable for probing antiferromagnetic magnons.

Electrically injected and detected magnonic spin transport overcomes this limit to a large extent and is mediated by magnons in the full frequency range [20–29]. In this respect, the recent observation of the magnon Hanle effect in an antiferromagnetic insulator has opened new opportunities [30–33]. On the one hand, it shows that the antiferromagnetic magnon pseudospin can be manipulated and used in devices, similar to the electronic spin in spintronics [34, 35]. On the other hand, it offers a new powerful tool for studying the rich nature of antiferromagnetic magnons, parameterized via a pseudospin, thereby providing crucial information about the underlying spin interactions [14, 18, 19, 36, 37].

Several of the exciting theoretical predictions in quantum matter, such as topological antiferromagnetic magnons [13, 15, 16, 38], essentially exploit inversion symmetry-breaking in the spin system. This, in turn, is intricately related to spin-orbit interaction and nonreciprocity [39]. These have been the basis of exciting chiral or rectification phenomena observed across platforms [39], from supercurrents [40] to magnetoacoustic waves [41, 42]. Moreover, nonreciprocal antiferromagnetic magnons have been observed in α-Cu2V2O7 using neutron scattering experiments [43, 44]. However, antiferromagnetic materials with broken inversion symmetry are still scarce and mostly do not offer high Néel temperatures. This is in contrast to ferromagnets, where nonreciprocal magnonic responses have been reported in various widely used magnetic hybrids employing easily accessible detection schemes at room temperature, thereby triggering rapid advancements [29, 39, 45–50]. Hence, several undiscovered nonreciprocity-based phenomena with antiferromagnetic magnons await finding suitable widely available materials and detection methods.

Here, we report a nonreciprocal response in the widely available antiferromagnetic insulator (AFI) hematite, observed as direction-dependent electrically-induced magnon spin transport and Hanle effect [30]. The degree of nonreciprocity is found to vary with the equilibrium Néel vector direction and thus, is tunable. Our observations are understood in terms of the different pseudofields, and thus pseudospin precession rates, experienced by magnons propagating in the forward and backward directions. Since the pseudofield is directly related to the magnon eigenmodes and the underlying
Let us consider the upper panel of Fig. 1(a). The left injection and detection of magnon spin currents [51–53]. Consequently, the magnon spin signal, given by the pseudospin z-component, detected by the right NM depends on the pseudofield \( \omega_z \). The latter can further be controlled via an applied magnetic field [30] and vanishes at a specific value denoted as \( H_c \).

If we interchange the two NM electrodes roles’ [lower panel, Fig. 1(a)] injecting spin with the right and detecting it using the left, magnons may experience a slightly different pseudofield \( \omega_z \) due to inversion symmetry breaking. Consequently, the magnon spin signal detected in this configuration is slightly different. This difference \( (\propto \delta \omega \equiv (\omega_+ - \omega_-)/2 \) allows us to quantify the pseudofield nonreciprocity in the system. Furthermore, as depicted in Fig. 1(b), this difference changes sign together with the average pseudofield \( \omega \equiv (\omega_+ + \omega_-)/2 \) due to a corresponding reversal of the precession sense. Moreover, this difference vanishes with the pseudofield, as depicted schematically in Fig. 1(c). These key features, validated by our experimental data reported below, allow us to confirm the nonreciprocal pseudofield as the origin of the observed nonreciprocity in the magnon Hanle effect.

The nonequilibrium magnons and their transport in the AFI can be described in terms of the pseudospin chemical potential \( \mu_s \), which is a vector [31]. Its magnitude, z-component, and direction respectively capture the densities of nonequilibrium magnons, spin, and their nature. For the system of interest, it suffices to consider that \( \mu_s \) varies only along the z-coordinate between the injector and detector (Fig. 1). It is thus described by a one-dimensional diffusion equation [31, 32]:

\[
\frac{\partial \mu_s}{\partial t} = D_m \frac{\partial^2 \mu_s}{\partial z^2} - \frac{\mu_s}{\tau_m} + \mu_s \times \omega \vec{x} - l \frac{\partial \mu_s}{\partial z} \times \delta \omega \vec{x},
\]

where \( D_m \) is the diffusion coefficient, \( \tau_m \) is the spin relaxation time, and \( l \) is the mean free path, all quantities pertaining to the AFI magnons. The last term on the right hand side of Eq. (1) is the new contribution here as compared to the previous inversion-symmetric considerations [31, 32]. It is obtained by allowing different pseudofields in the forward (+) and backward (−) directions within the random walk model describing the diffusive pseudospin transport with precession [31, 54, 55].

The magnon spin injection by the NM electrode injects a magnonic spin current into the AFI, which corresponds to injecting a z-polarized pseudospin current. The AFI under consideration bears an easy-plane anisotropy, which harbors z-directed pseudofield and, correspondingly, spin-0 magnon eigenmodes. Thus, the spin-1 magnons injected by the NM are not the eigenmodes and start to transmute into other kinds of magnons with varying spin. This process is represented by pseudospin precession about the pseudofield \( \omega_z \).
ity that relates the pseudospin density with its chemical potential, and \( \mu_{el} \) is the magnitude of the magnon spin current density driven by the injector NM. Its direction is positive (negative) for transport along \( \hat{z} (-\hat{z}) \), which further leads to the \(+ (-)\) sign in the boundary condition above. Finally, imposing the stability requirement \( \mu_{el}(z \rightarrow \infty) = 0 \) \( \mu_{el}(z \rightarrow -\infty) = 0 \) for the forward [backward] case, we obtain the desired solutions to Eq. (1) for \( \mu_{el}(z) \) for both cases: forward \( z > 0 \) and backward \( z < 0 \).

The detected magnon spin signal is directly proportional to \( \mu_{el} \) at the detector location [31, 32]. Hence, \( \mu_{el}(+d) [\mu_{el}(-d)] \) represents the detected magnon spin signal in the forward [backward] transport configuration (see upper [lower] panels in Fig. 1), where \( d \) is the injector-detector distance [Fig. 2(a)]. We further define \( \mu_{sym} \equiv [\mu_{el}(+d) + \mu_{el}(-d)]/2 \) and \( \mu_{asym} \equiv [\mu_{el}(+d) - \mu_{el}(-d)]/2 \), which are evaluated as

\[
\mu_{sym} = m_{el} a e^{-a m_{el}} \left( a \cos \left( \frac{b d}{l_m} \right) - b \sin \left( \frac{b d}{l_m} \right) \right),
\]

\[
\mu_{asym} = \frac{\omega m_{el} \partial \mu_{sym}}{2 l_m},
\]

where \( a \equiv \sqrt{\left(1 + \sqrt{1 + \omega^2 \tau_m^2} \right) / 2} \), \( b \equiv \sqrt{\left(-1 + \sqrt{1 + \omega^2 \tau_m^2} \right) / 2} \), and \( l_m \equiv \sqrt{D_m \tau_m} \) is the magnon diffusion length. Here, we have retained terms up to the first order in \( \delta \omega \), assuming \( |\delta \omega/(l_m \omega)| \ll 1 \).

Equation (2) constitutes the desired and our main theoretical result, which is employed to analyze the experimental data below. The expression thus obtained for \( \mu_{sym} \) is the same as that in the previous inversion-symmetric analysis [31]. In contrast, \( \mu_{asym} \) captures the nonreciprocity and is finite only when the pseudofield is nonreciprocal, i.e., when \( \delta \omega \neq 0 \). Furthermore, Eq. (2) shows that \( \mu_{asym} \) manifests the odd-in-\( \omega \) behavior motivated and discussed schematically in Fig. 1. This feature allows us to distinguish a nonreciprocal pseudofield contribution from other potential sources of nonreciprocity.

In our experiments, we use a \( t_m = 89 \) nm thick film of hematite \( (\alpha - Fe_2O_3) \) as the AFM. The hematite film undergoes a transition from an easy-axis to an easy-plane AFI above the Morin transition \( T_M \approx 200 \) K [56] (see Supplemental Material (SM) [57] for details). All measurements are conducted in the easy-plane phase at \( T = 250 \) K, where the hematite film features an out-of-plane Dzyaloshinskii-Moriya interaction (DMI), in agreement with previous works [30, 32]. As depicted in the left panel of Fig. 2(a), the equilibrium Néel vector \( \mathbf{n} \), the two sublattice magnetizations \( \mathbf{M}_1 \) and \( \mathbf{M}_2 \) as well as the induced net magnetization \( \mathbf{M}_{net} = \mathbf{M}_1 + \mathbf{M}_2 \) due to DMI lie in the \( xz \) or \( (0001) \) Fe\(_2\)O\(_3\)-plane. Both, \( \mathbf{n} \) and \( \mathbf{M}_{net} \) can be controlled by the orientation and magnitude of the applied magnetic field \( \mathbf{H} \), where \( \mathbf{M}_{net} \) encodes the canting angle [30]. To investigate the magnon spin transport by all-electrical means, we employ two spatially separated narrow Pt strips on top of the film (see SM [57] for fabrication details).

To characterize the sample we perform angle-dependent electrical transport measurements by changing the orientation of the external magnetic field \( \mathbf{H} \) within the \( xz \)-plane [see Fig. 2(a)]. A dc charge current \( I_{inj} = 500 \) mA is applied first to the left electrode leading to spin injection into the hematite film via the spin Hall effect (SHE) [51–53]. The resulting diffusive pseudospin magnon current is detected electrically as a voltage sig-
nal $V_{\text{det}}$ at the right electrode [left panel, Fig. 2(a)] (see SM [57] for experimental details). In a second step, we interchange the injector and detector electrode, i.e., $I_{\text{inj}}$ is injected at the right electrode and the voltage $V_{\text{det}}$ is detected at the left Pt strip [right panel, Fig. 2(a)]. The measured magnon spin signal $R_{\text{el}} = V_{\text{det}}/I_{\text{inj}}$ is plotted in Fig. 2(b) versus the angle $\varphi$ of the applied in-plane magnetic field for three different magnitudes $\mu_0 H$ for both configurations. The full circles correspond to the forward transport direction $[+d$, left panel of Fig. 2(a)], while open circles represent the backward direction $[-d$, right panel of Fig. 2(a)]. Evidently, all curves appear to exhibit the $\sin^2(\varphi)$ angular dependence characteristic of a factor $\sin(\varphi)$ contributed by both of the injection and detection processes [20, 21]. However, a careful examination shows that there are differences between the two propagation directions for $\mu_0 H = 5$ T and 7 T, predominantly at $\varphi = 90°, 270°$, where $R_{\text{el}}$ is largest. This corresponds to $\mathbf{n} \parallel \hat{z}$ or $H \parallel \mathbf{n}$ as $H \perp \mathbf{n}$.

To quantify this observation, we plot the symmetric $R_{\text{sym}} = [R_{\text{el}}(+d) + R_{\text{el}}(-d)]/2$ and antisymmetric $R_{\text{asym}} = [R_{\text{el}}(+d) - R_{\text{el}}(-d)]/2$ components of the magnon spin signal for the two measurement configurations in Fig. 2(c) and (d), respectively [29]. The angle dependence in Fig. 2(c) follows a simple $\Delta R_{\text{sym}} \sin^2(\varphi)$ behavior, where $\Delta R_{\text{sym}}$ is the amplitude of the symmetric magnon spin signal. As discussed below (cf. Fig. 2), this amplitude also exhibits the expected Hanle curve consistent set of parameters. Here, we have restricted the fit range for $\Delta R_{\text{sym}}$, and $\Delta R_{\text{asym}}$ to $\mu_0 H = 4.5 - 7$ T, as the simplified considerations resulting in Eq. (2) do not account for low-energy magnons, which contribute to the spin transport at smaller magnetic fields [32] (see SM [57] for fit procedure and parameters).

Thus, employing our theoretical analysis leading to Eq. (2), we deduce the origin of our experimentally observed nonreciprocity to be an antisymmetric pseudospin field $\delta \omega$, finding it to be angle dependent as $\sin(\varphi)$. This angle dependence is reminiscent of a related, but distinct, nonreciprocity of the magnon dispersion found in yttrium iron garnet/gadolinium gallium garnet heterostructures [29], which has been attributed to the interfacial DMI. Considering that the antisymmetric signal in our experiments is smaller in thicker samples (see SM), our observed nonreciprocity also likely stems from the interface between hematite and its substrate. This has not been observed before, to the best of our knowledge, and opens novel perspectives for engineering nonreciprocal effects in a widely employed AFI. At the same time, the spin Hamiltonians used to describe hematite in the literature [30], for example in the original article by Moriya [58], could be oversimplified and might have missed such a nonreciprocity stemming from the crystal structure. To examine this potential origin, atomistic spin modeling of hematite taking into account its exact crystal structure is desirable and, hopefully, will be motivated by our findings [59, 60].

In summary, we demonstrate nonreciprocal magnon spin transport in the widely used antiferromagnetic insulator - hematite - employing electrical spin injection and detection. Our theoretical modeling allows us to understand this observation as due to an antisymmetric pseudospin field along the spin transport direction. It further enables extraction of its dependence on the applied
magnetic field. This antisymmetric pseudofield, in turn, directly translates to magnon dispersion [31] and constitutes an observation of emergent pseudospin-orbit interaction. Hence, our work establishes nonlocal magnon transport as a powerful probe for underlying spin interactions in antiferromagnetic insulators. It also demonstrates the widely available hematite as a promising material for searching topological and nonreciprocal phenomena.

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