Giant chiral magnetoelectric oscillations in a van der Waals multiferroic

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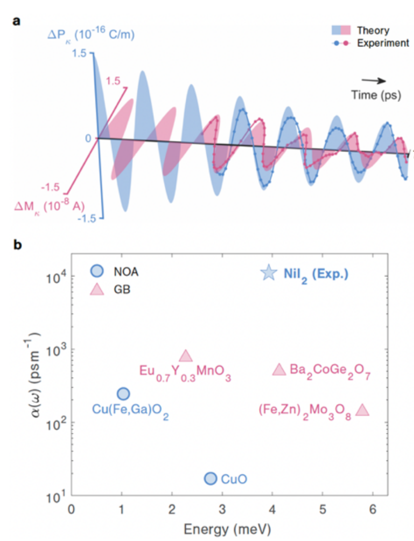
Helical spin structures are expressions of magnetically-induced chirality, entangling the dipolar and magnetic orders within materials[1–4]. The recent discovery of helical van der Waals multiferroics down to the ultrathin limit raises prospects of large chiral magnetoelectric correlations in two dimensions[5, 6]. However, the exact nature and magnitude of these couplings have remained unknown to date. Here, we perform a precision measurement of the dynamical magnetoelectric coupling for an enantiopure domain in an exfoliated van der Waals multiferroic. We evaluate this interaction in resonance with a collective electromagnon mode, capturing the impact of its oscillations on the material's dipolar and magnetic orders with a suite of ultrafast optical probes. Our data reveals a giant natural optical activity at terahertz frequencies, characterized by quadrature modulations between the electric polarization and magnetization components. First-principles calculations further reveal that these chiral couplings originate from the synergy between the non-collinear spin texture and relativistic spin-orbit interactions, resulting in substantial enhancements over lattice-mediated effects. Our findings highlight the potential for intertwined orders to enable exotic functionalities in the two-dimensional limit and pave the way for the development of van der Waals magnetoelectric devices operating at terahertz speeds.



Recently, a concerted effort has been made to extend type-II multiferroics into the atomically thin regime by exploiting van der Waals materials that crystallize in centrosymmetric structures[5, 6]. These quasi-two-dimensional multiferroics have the potential to achieve enhanced magnetoelectric properties with a level of tunability that is beyond the reach of traditional three-dimensional helimagnets. At finite frequency, such properties are expected to be controlled by different manifestations of the dynamical magnetoelectric coupling, expressed via the first-order magnetoelectric response[7] $\Delta P_i(\omega) = \alpha_{ij}(\omega)\Delta H_j(\omega)$. In this relation, $\alpha_{ij}(\omega)$ is the complex dynamical magnetoelectric susceptibility tensor, which represents the cross-coupling between the modulation of the material's electric polarization $\Delta P_i(\omega)$ and magnetizing field $\Delta H_j(\omega)$. Among the various phenomena governed by this tensor, the natural optical activity stands out as the effect most intimately connected to the spontaneous breaking of chiral symmetry[8, 9]. This property describes the rotation of light polarization that is symmetric under time reversal and antisymmetric under spatial inversion, and its strength can be enhanced in resonance with the collective modes of the magnetochiral order[9]. Unveiling the presence and magnitude of this elusive effect in the realm of exfoliated van der Waals multiferroics is pivotal for realizing high-speed chiral applications based on artificially stacked layered structures. However, the lack of suitable probing methods has so far hindered the identification of this phenomenon at the microscopic length scales of individual chiral domains within a van der Waals flake.

To address this challenge, here we perform a precision measurement of the dynamical magnetoelectric coupling for a native enantiopure domain of the van der Waals multiferroic nickel iodide (NiI_2)[10]. We use a tailored laser pulse to coherently drive an electrically active magnon mode and directly track the resulting modulation of the dipolar and spin-spiral orders in real time via optical second harmonic generation (SHG) and Kerr rotation microscopy. Our protocol uncovers the existence of natural optical activity at terahertz frequencies, signaled by the emergence of a $\pi/2$ phase shift between the electric polarization and magnetization oscillations. Extraction of the corresponding magnetoelectric tensor element reveals that the strength of natural optical activity greatly surpasses that of any other known helimagnet. Our comprehensive analysis, which incorporates model Hamiltonian, density-functional theory (DFT), and group theory calculations, fully rationalizes these findings, underscoring the role of the relativistic spin-orbit interaction in the origin of such giant natural optical activity.

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