A type-II multiferroic in two dimensions

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Multiferroic materials have garnered wide interest for their exceptional static and dynamical magnetoelectric properties. In particular, type-II multiferroics exhibit an inversion-symmetry-breaking magnetic order which directly induces a ferroelectric polarization through various mechanisms, such as the spin-current or the inverse Dzyaloshinskii-Moriya effect. This intrinsic coupling between the magnetic and dipolar order parameters results in record-strength magnetoelectric effects. In this context, there has been a recent surge of interest in 2D materials possessing such intrinsic multiferroic properties, enabling the integration and control of magnetoelectric effects in artificial heterostructures and nanoelectronic devices.
In this talk, I will present our recent study and realization of type-II multiferroic order in a single atomic layer of the transition metal-based van der Waals material NiI2. The multiferroic state of NiI2 is characterized by an inversion-symmetry-breaking helimagnetic order which induces a chirality-dependent electrical polarization. Using circular dichroic Raman measurements, we directly probed the magneto-chiral ground state and its electromagnon modes originating from dynamic magnetoelectric coupling. Using birefringence and second-harmonic generation measurements, we observed a highly anisotropic electronic state simultaneously breaking three-fold rotational and inversion symmetry to support polar order. The evolution of the optical signatures as a function of temperature and layer number surprisingly revealed an ordered magnetic, polar state that persists down to the ultrathin limit of monolayer NiI2.