Highlights

RESEARCH AREA 2 - **Functional and Complex Materials for Innovative Electronics and Sensing** - 2022

"Evidence for a single-layer van der Waals multiferroic"

Qian Song [1,2,9], Connor A. Occhialini [1,9], Emre Ergeçen [1,9], Batyr Ilyas [1,9], Danila Amoroso [3,4], Paolo Barone[5], Jesse Kapeghian [6], Kenji Watanabe[7], Takashi Taniguchi[8], Antia S. Botana[6], Silvia Picozzi[3], Nuh Gedik[1], Riccardo Comin[1]

[1] Department of Physics, Massachusetts Institute of Technology, Cambridge, MA, USA.
[2] Department of Material Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA, USA.
[3] Consiglio Nazionale delle Ricerche CNR-SPIN, c/o Università degli Studi 'G. D'Annunzio', Chieti, Italy.
[4] NanoMat/Q-mat/CESAM, Université de Liège, Liège, Belgium.
[5] Consiglio Nazionale delle Ricerche CNR-SPIN, Area della Ricerca di Tor Vergata, Rome, Italy.
[6] Department of Physics, Arizona State University, Tempe, AZ, USA.
[7] Research Center for Functional Materials, National Institute for Materials Science, Tsukuba, Japan.
[8] International Center for Materials Nanoarchitectonics, National Institute for Materials Science, Tsukuba, Japan.

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Multiferroic materials have attracted wide interest because of their exceptional static and dynamical magnetoelectric properties. In particular, type-II multiferroics exhibit an inversion-symmetry-breaking magnetic order that directly induces 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 high-strength magnetoelectric effects. Two-dimensional materials possessing such intrinsic multiferroic properties have been long sought for to enable the harnessing of magnetoelectric coupling in nanoelectronic devices. Here we report the discovery 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 a proper-screw spin helix with given handedness, which couples to the charge degrees of freedom to produce a chirality-controlled electrical polarization. We use circular dichroic Raman measurements to directly probe the magneto-chiral ground state and its electromagnon modes originating from dynamic magnetoelectric coupling. Combining birefringence and second-harmonic-generation measurements with theoretical modelling and simulations, we detect a highly anisotropic electronic state that simultaneously breaks three-fold rotational and inversion symmetry, and supports polar order. The evolution of the optical signatures as a function of temperature and layer number surprisingly reveals an ordered magnetic polar state that persists down to the ultrathin limit of monolayer NiI2. These observations establish NiI2 and transition metal dihalides as a new platform for studying emergent multiferroic phenomena, chiral magnetic textures and ferroelectricity in the two-dimensional limit.

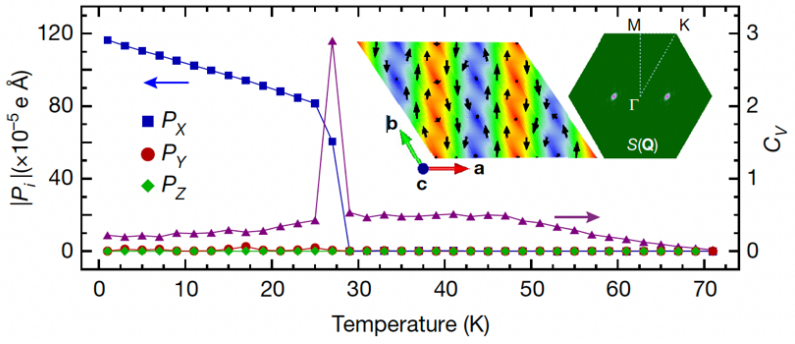

Fig. 1 - Electrical polarization components |Pi| in units of 10−5 e . (absolute value, closed squares) and specific heat (CV, triangles) as a function of temperature, relative to the proper-screw spiral order represented in the insets (for the spin texture, the black arrows represent in-plane components of the spins and the colour map indicates the out-of-plane spin component with red (blue) denoting sz = +(–)1, S(Q) represents the spin structure factor, corresponding to the single-Q helical spin configuration Q).