Poster-2-10

Spin-orbital excitations encoding the magnetic phase transition in the van der Waals antiferromagnet $FePS_3$

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Magnetic van der Waals (vdW) materials offer new avenues for exploring exotic magnetic phases and collective behavior. [1] Recent research, mainly using optical spectroscopy, reveals the sensitivity of the acoustic phonon response to magnetic states even in few-layer structures. [2] $FePS_3$, whose interlayer exchange interaction is very small, is an $S = 2$ zig-zag quasi-two-dimensional antiferromagnetic insulator with a honeycomb lattice. [3] Here we performed resonant inelastic X-ray scattering (RIXS) at Fe L_3-edge as a function of temperature across the magnetic transition to elaborate the spin-orbital excitations of $FePS_3$ and their relation to magnetism. The multiplets probed by Fe L3 RIXS have a large sensitivity to the spin state. By comparing with ligand-field-theory calculations, we identified the essential role of the trigonal lattice distortion and negative metal-ligand charge transfer to account for the low-energy spin-orbital excitations. We reveal the robustness of these elementary excitations down to the few atomic layer limits, in accordance with the persistent magneto-optical excitations and phonon modes in monolayers measured by Raman scattering. [4] This provides crucial insight into how the spontaneous magnetic symmetry-breaking stabilizes in the quasi-two-dimensional limit of the vdW magnet $FePS_3$. Our work highlights RIXS as an ideal generalized approach for studying 2D magnetic materials, featuring sensitivities to all degrees of freedom.

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