Spin-flip Determination in CrX_3 (X = Cl, Br, and I) 2D Magnets using High-resolution X-ray Scattering

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Abstract:

Chromium tri-halides CrX_3 (X = Cl, Br, and I) exhibit low-temperature, layer-dependent magnetism that can be manipulated by an electric field, which makes them important candidates for spintronics applications. Their magnetic ground states depend keenly on electronic parameters such as spin-orbit coupling (SOC), Hund's coupling (J_H), $p-d$ covalency, and interorbital Coulomb interactions. Therefore, accurately determining these parameters is paramount for understanding the CrX_3 physics. We have used resonant inelastic x-ray scattering (RIXS) spectroscopy facilitated by ligand field multiplet calculations in C_3 symmetry to simulate experimental RIXS spectra. Tanabe-Sugano-like energy level diagram calculations facilitated the determination of detailed electronic structure parameters in CrX_3. These methods provide the most detailed description of CrX_3 magneto-optical and electronic energetic (terms) to date. The determined $10Dq$ values are in good agreement with the spectrochemical series, the Racah B parameter follows the expected Nephelauxetic effect, and the crystal field distortion parameters $D\sigma$ and $D\tau$ are calculated for the first time. Moreover, high-resolution RIXS spectra reveal a clear energy separation between spin-allowed quartet and spin-forbidden doublet states in CrX_3, showcasing the potential of this technique in determining materials' electronic parameters. This study validates the role of SOC in Cr *2p* spin-flip excitations. Such precise measurements offer insights into the energy design of spintronic devices that utilize quantum state tuning and the effect of halides in determining spin-flip excitation energies in 2D magnetss.