Observation of anisotropic dispersive dark exciton dynamics in CrSBr

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CONTENTS

I. CRYSTAL GROWTH OF CrSBr

Single crystals of CrSBr were synthesized via chemical vapor transport [1]. A slightly off-stoichiometric amount of chromium powder, chromium(III) bromide chunks, and sulfur pieces were loaded into a 12.7 mm outer diameter and 10.5 mm inner diameter fused silica tube and sealed to a length of 20 cm under vacuum ($\sim$ 30 mtorr). The ampule was subjected to a temperature gradient of 950°C (source side) to 850°C (sink side) for 72 hours in a computer-controlled two-zone tube furnace. The crystals were washed using a Cr^{2+} aqueous solution, deionized water, toluene, and acetone, resulting in shiny black plate-like single crystals with the surface normal along the crystallographic c axis. The crystal morphology tends to be rectangular, with the larger dimension parallel to the a axis. The single crystal used for this measurement had dimensions 1 mm along the b direction and 4 mm along the a direction. The crystal orientation was verified by Laue and single crystal diffraction prior to the resonant inelastic x-ray scattering (RIXS) experiment.

II. EXCITON DISPERSION FITTING

The spectra in Fig. 1 of the main text, were fit with either 5 or 6 Gaussian functions, depending on whether the low-energy exciton was visible. In the $0KL$ plane, the position and width of the excitation at $\sim$1.6 eV were fixed to the fitted value from the scan at low K. The full width of the exciton at $\sim$1.37 eV was fixed to the experimental resolution of 30 meV.

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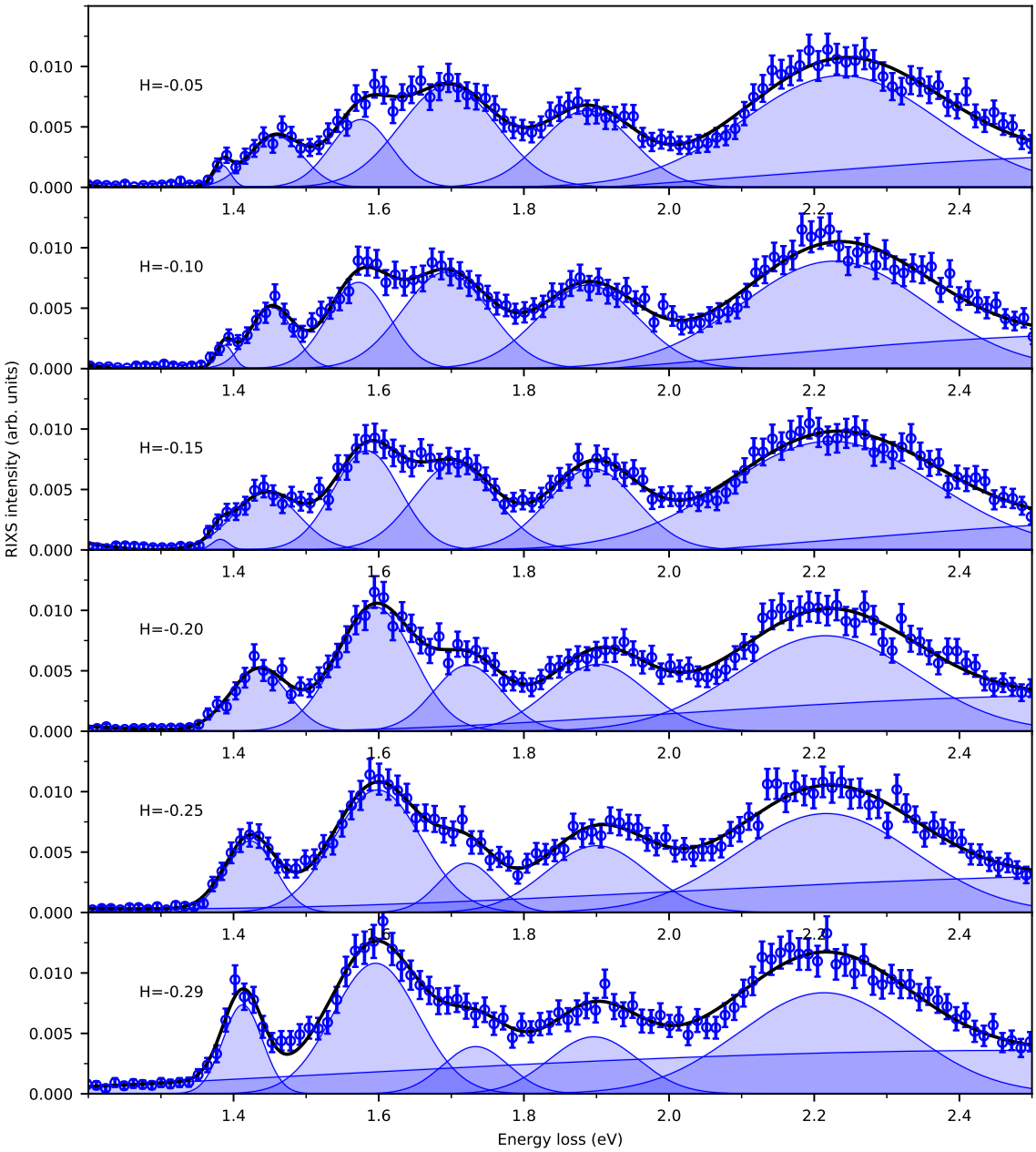

FIG. S1. Fits to the spectra in the $H0L$ plane.

[1] A. Scheie, M. Ziebel, D. G. Chica, Y. J. Bae, X. Wang, A. I. Kolesnikov, X. Zhu, and X. Roy, Spin waves and magnetic exchange Hamiltonian in CrSBr, Advanced Science **9**, 2202467 (2022).

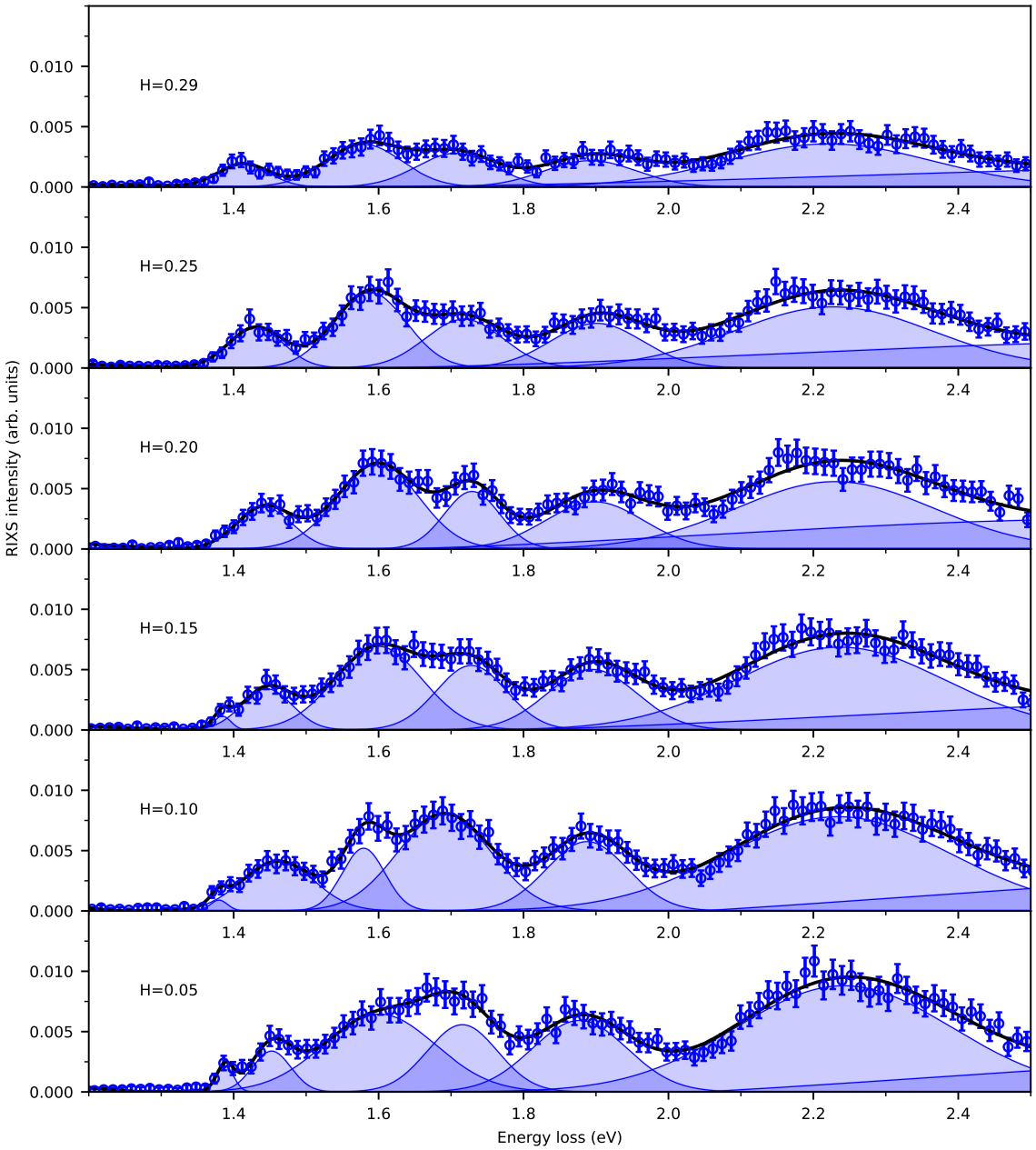

FIG. S2. Fits to the spectra in the $H0L$ plane.

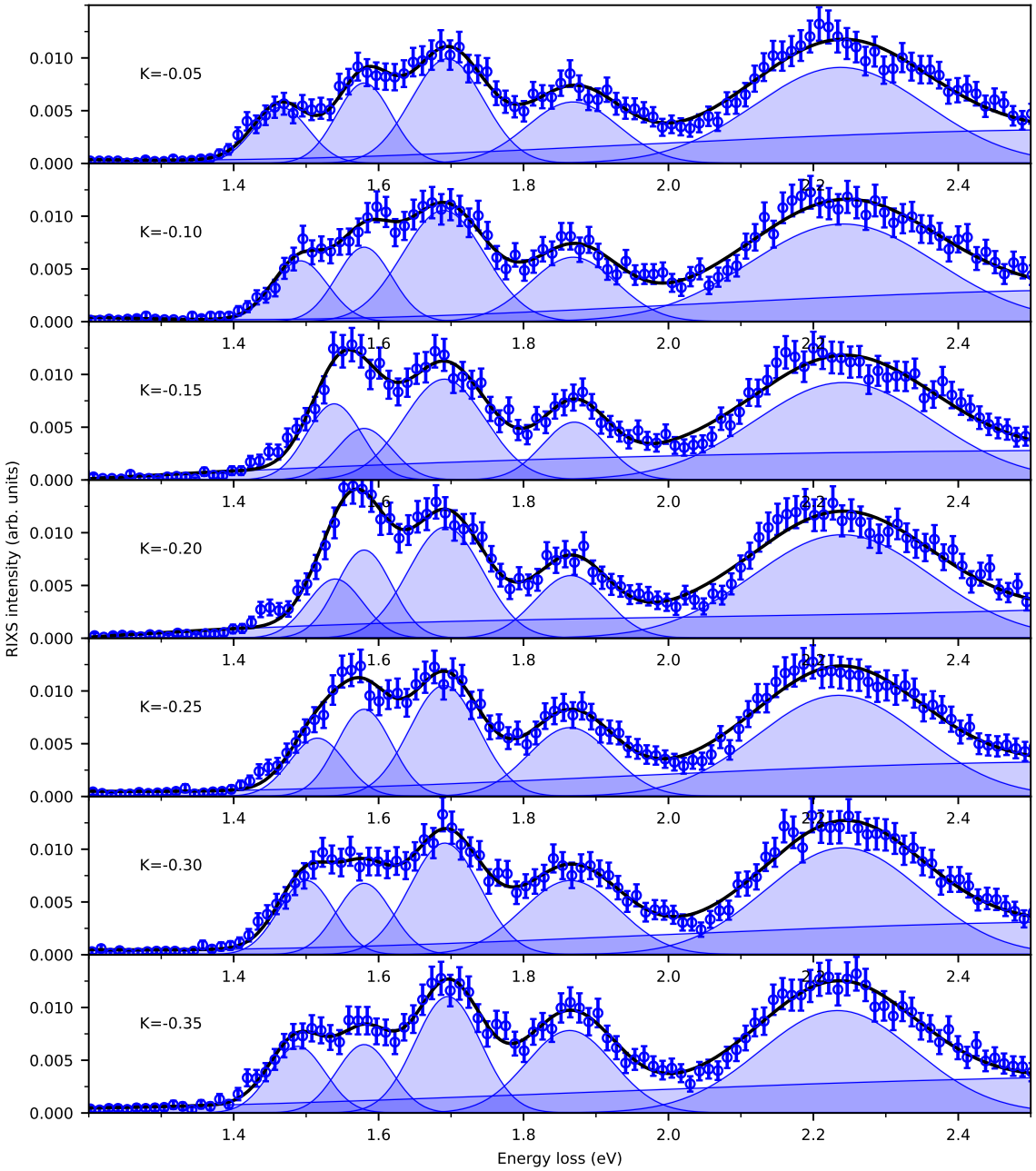

FIG. S3. Fits to the spectra in the $0KL$ plane.

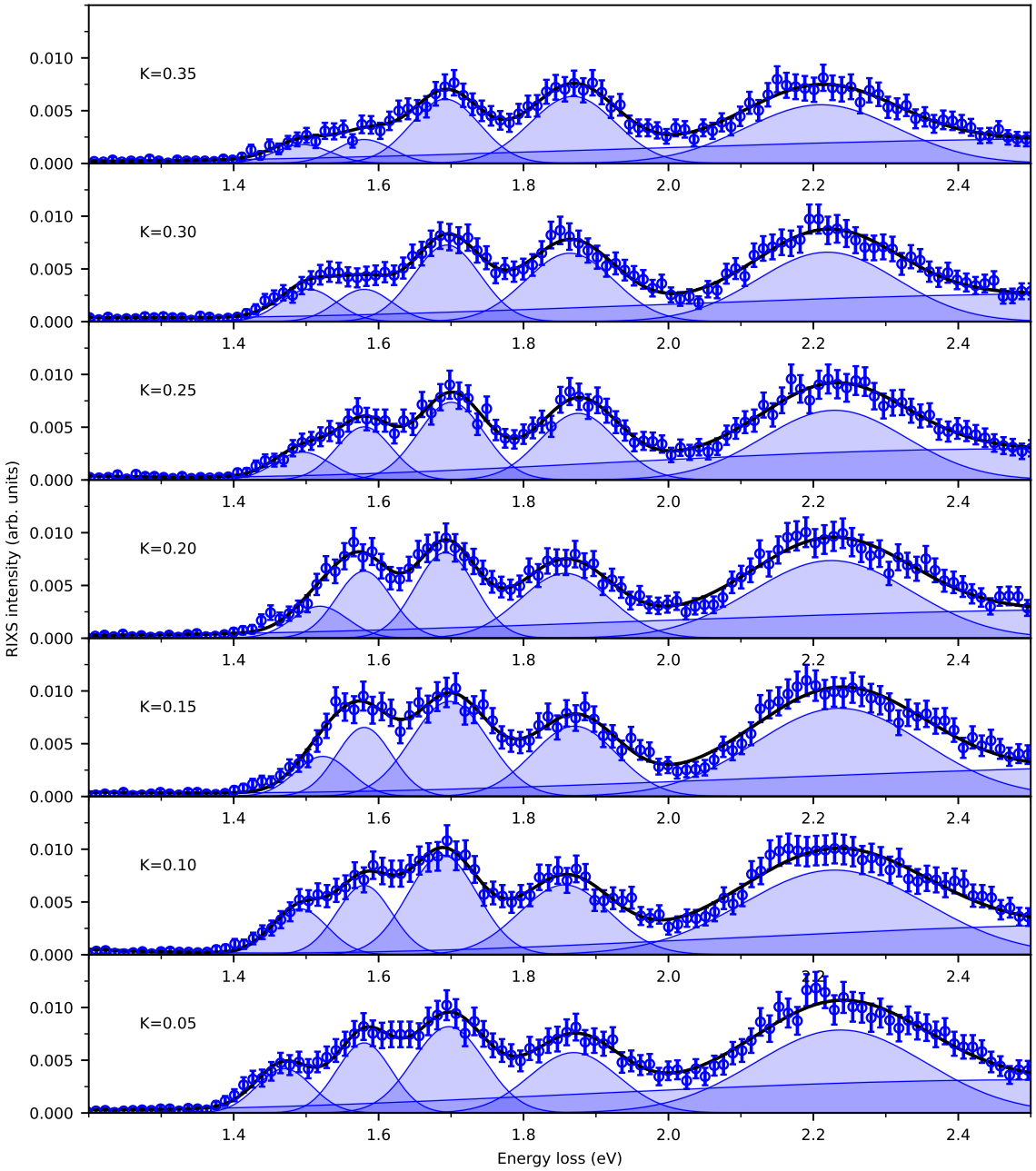

FIG. S4. Fits to the spectra in the $0KL$ plane.