Uncovering colossal magnetooptic properties of a two-dimensional magnet CrSBr

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The discovery of 2D layered materials paved the way for investigating new quantum phenomena emerging from lower dimensionality. With the first exfoliation the these materials, they gave rise to the concept of "nano-lego" with a number of classes of 2D materials to be combined into heterostructures. In particular, in recent years there has been a considerable focus on such a special class exhibiting a magnetic ordering.

A prominent example of such a material is chromium sulfide bromide – CrSBr, which is particularly interesting due to combination of semiconducting and magnetic properties. This semiconductor exhibits strongly anisotropic direct-band gap excitations in the near-infrared energy range[1]. Showcasing the intriguing coupling of electronic and magnetic properties, the energy of an exciton is notably influenced by the magnetic interlayer ordering.

The standard exciton states with energy around 1.32 eV have been thoroughly examined for their prominent tunability in magnetic field of around 15 meV. In this work we present observation of higher-bands exciton states in the material. With identical anisotropy and continuous field dependence, reported states offer over 7 times wider span of possible magnetic field-tunable exciton energies. The phenomenological model of magnetic field-assisted coupling between two higher energy states fits to the extracted energies of the states, explaining the enormous shift in magnetic field.

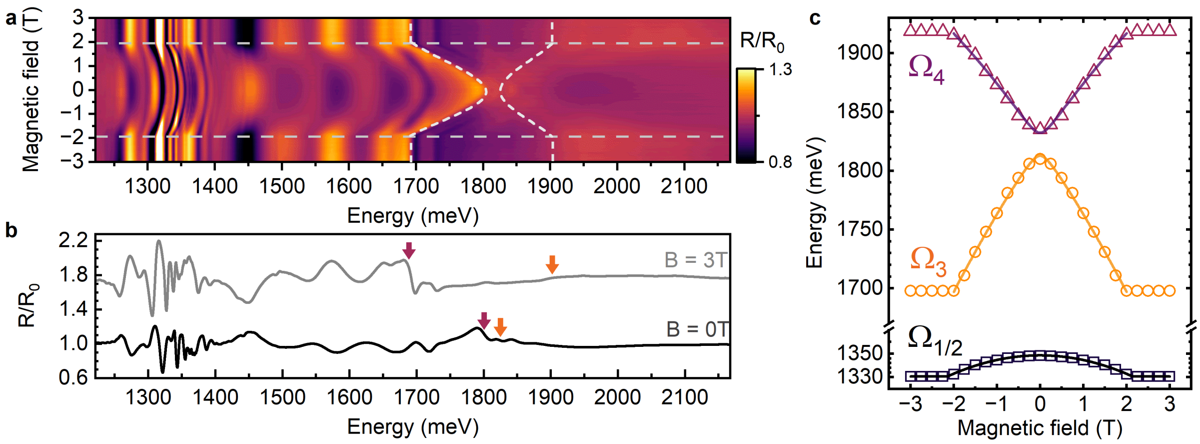

Figure 1: **a**, Magnetoreflectance map with reference on substrate in wide range of energies showcasing absorption of both fundamental (1.3 eV) and higher-bands (1.8 eV) exciton states. **b**, Comparison of extracted reflectance profiles in magnetic fields of 0T and 3T with two chosen higher-bands states highlighted with arrows. **c**, Extracted energies of earlier denoted states with model fit to all states in function of magnetic field.

[1] Wilson, N.P., Lee, K., Cenker, J. et al. *Nat. Mater.* **20**, 1657–1662 (2021).