Optical markers of magnetic phase transition in layered CrSBr.

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Two-dimensional (2D) magnetic semiconductors give the opportunity to take advantage of the electron charge and the electron spin at the same time, which gives the possibility of broadening modern semiconductor technology and spintronic devices. It can greatly expand the applications of ferromagnets (FM) in other devices such as transformers, electromagnets, high-density storage, and magnetic random access memory [1].

Here, we investigate the role of the interlayer magnetic ordering of CrSBr: antiferromagnetic van der Waals crystal whose single monolayer is intrinsic ferromagnetic (AFM:A), in the framework of <u>ab initio calculations</u>, and by using <u>optical spectroscopy [2]</u>**.** We have demonstrated that optically induced band-to-band transitions visible in optical measurement are remarkably well assigned to the band structure by the momentum matrix elements and energy differences for magnetic ground state (A-AFM). In addition, our study reveals significant differences in electronic properties for two different interlayer magnetic phases. When the magnetic ordering of A-AFM to FM is changed, the crucial modification of the band structure reflected in the direct-to-indirect band gap transition and the significant splitting of the conduction bands are obtained.

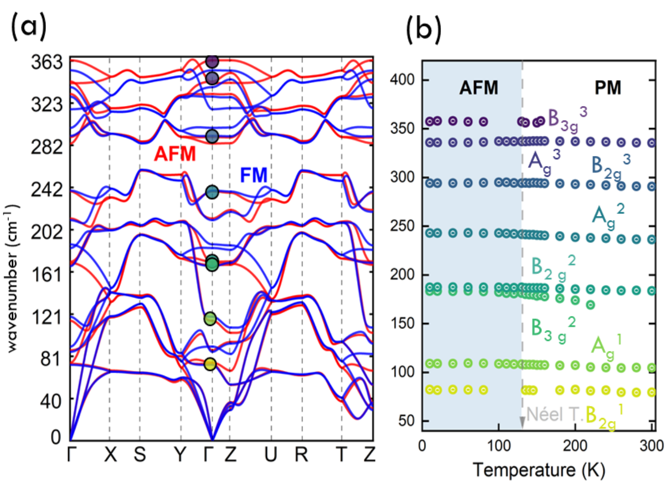

Fig. 1 (a) Phonon dispersion for AFM, FM magnetic states (b) Experimentally measured raman peaks positions in function of temperature.

In addition, Raman measurements (Fig.(b)) demonstrate a splitting between the in-plane modes B_{2g}^2/B_{3g}^2, which is temperature dependent and can be assigned to different interlayer magnetic states, corroborated by the DFT+U study (Fig.(a)). Finally, our results point out the origin of interlayer magnetism, which has its origin in electronic rather than structural properties. Our results reveal a new approach for tuning the optical and electronic properties of van der Waals magnets by controlling the interlayer magnetic ordering in adjacent layers.

[1] S. Chen,C. Huang, H. Sun, J. Ding, P. E. Jena, J. Kan, J. Phys. Chem. C 2019, 123, 17987–17993
[2]W. Linhart, M. Rybak, M. Birowska, K. Mosina, V. Mazanek,P. Scharoch, D. Kaczorowski, Z. Sofer, and R. Kudrawiec, Optical markers of magnetic phase transition in CrSBr, under review in ACS Nano