Inducing out-of-plane magnetocrystalline anisotropy by substitutional doping in the multiferroic $CuCrP_2S_6$ monolayer

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Magnetocrystalline anisotropy (MCA) is a crucial property of lowdimensional magnetic materials for both its fundamental role in stabilizing long-range magnetic order in two-dimensional (2D) materials [1] and for its pivotal influence on spintronic devices based on those materials. Specifically, perpendicular magnetic anisotropy (PMA), where the magnetization is out of plane, has emerged as a key focus in spintronics due to its inherent stability against thermal fluctuations, scalability, endurance, and low switching current density, causing an ongoing search for new ways to control PMA.

MCA is of particular interest for thin-film and monolayer multiferroics, which have attracted significant interest for spintronic applications due to their coexistence of multiple ferroic phases, suitable for tuning and control of magnetic properties. In recent years, large progress has been made in understanding room temperature multiferroicity in the 2D metal thiophosphates (MTP) family of the $ABP_2S(Se)_6$ type, where A and B are transition metals which are responsible for ferroelectric (FE) and ferromagnetic (FM) properties, respectively. However, only a few 2D multiferroics have been experimentally confirmed so far, with $CuCrP_2S_6$ (CCPS) being the only one from the MTP family [2].

In this study, we predict a transition in magnetocrystalline anisotropy (MCA) orientation from in-plane to out-of-plane in 2D multiferroic material $CuCrP_2S_6$ (CCPS) by chemically doping it with Indium, where a linear increase in MCA with In concentration in the mixed $CuCr_{1-x}In_xP_2S_6$ is observed based on first-principles calculations. We attribute this shift in MCA to structural and chemical effects. The chemical effect comprises all the changes caused by substitution of Cr by In while preserving the pristine CCPS structure. We find it to be related to the depletion of the Cr d in-plane orbitals consequent to a reduction in the Cr magnetic moment upon its partial substitution by a non-magnetic element. The structural effect is analyzed in terms of the changes in the in-plane lattice parameter and the monolayer thickness upon the addition of In. Finally, we extend our findings relative to the tuning of MCA towards more positive values to other non-magnetic dopants, isovalent with Cr^{3+} and of atomic radius larger than that of Cr^{3+} in 2D CCPS and related metal thiophosphates.

References

[1] S. Hope, B.-Ch. Choi, P. J. Bode, and J. A. C. Bland. Direct observation of the stabilization of ferromagnetic order by magnetic anisotropy. *Phys. Rev. B*, 2000, *61*, 5876–5879.

[2] Y. Lai, Z. Song, Y. Wan, M. Xue, C. Wang, Y. Ye, L. Dai, Z. Zhang, W. Yang, H. Du, and J. Yang, *Two-dimensional ferromagnetism and driven ferroelectricity in van der waals $CuCrP_2S_6$*, Nanoscale, (2019), *11*, 5163.