Strong magnetic second harmonic generation effect discovered in two-dimensional $CrPS_4$ monolayer

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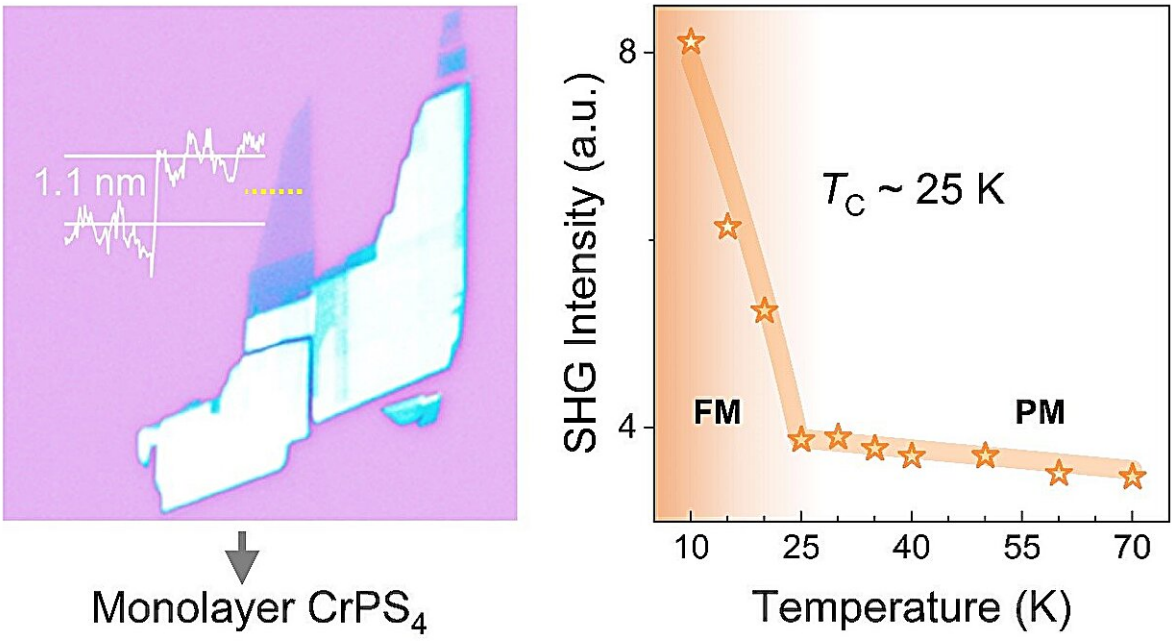

$CrPS_4$ exhibits a monolayer ferromagnetic order induced second-harmonic generation effect. Left: optical images of monolayer $CrPS_4$; Right: atomic force microscopy characterization. Credit: Hou De

A research team led by Professor Sheng Zhigao at the Hefei Institutes of Physical Science of the Chinese Academy of Sciences has, for the first

time, observed the strong nonlinear magnetic second harmonic generation (MSHG) induced by the ferromagnetic order in monolayer $CrPS_4$, with the help of the Steady High Magnetic Field Facility.

The results have been published in *Advanced Optical Materials*.

Second harmonic generation (SHG) is a nonlinear optical effect sensitive to symmetry breaking in materials. SHG was first seen in crystals with broken symmetry (i type), and also occurs in magnetic systems (c type), though it is much weaker. This limits its use in optical devices.

Magnetic two-dimensional van der Waals materials, gaining attention for their unique properties, still have unexplored nonlinear optical properties, particularly the link between SHG and magnetic order, which limits their potential in optoelectronics.

In this study, Sheng's team delved deeply into the magnetic order-related SHG effects in the two-dimensional antiferromagnetic material $CrPS_4$. They found that in bulk and even-layered $CrPS_4$, the antiferromagnetic order did not produce any c type SHG effects, whereas a substantial c type SHG effect induced by monolayer ferromagnetic order was observed in odd-layered $CrPS_4$.

This is the first observation of ferromagnetic order induced c type SHG effects in a 2D magnet under the electric-dipole approximation, stemming from the dual breaking of spatial and time inversion symmetries.

More importantly, the research team found that the ferromagnetic order induced c type SHG has a signal strength comparable to that of i type SHG, which arises from the breaking of crystal structural symmetry. This is extremely rare among all known magnetic materials.

"Our discovery revealed a new mechanism for SHG in two-dimensional magnetic materials," said Hou De, first author of the paper, "and greatly enriched the category of SHG generated by magnetic order."

More information: De Hou et al, Extraordinary Magnetic Second Harmonic Generation in Monolayer $CrPS_4$, *Advanced Optical Materials* (2024). DOI: 10.1002/adom.202400943

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