Giant nonreciprocal second harmonic generation from layered antiferromagnetism in bilayer CrI_3

With the support by the National Natural Science Foundation of China and the Ministry of Science and Technology of China, the research team led by Prof. Wu ShiWei (吴施伟) at Fudan University and Prof. Xiaodong Xu at the University of Washington Seattle discovered a giant and nonreciprocal second harmonic generation from layered antiferromagnetism in bilayer CrI_3, which was published in *Nature* (2019, 572: 497—501). This work may lead to new nonlinear and nonreciprocal optical devices based on 2D magnets and highlight SHG as a highly sensitive probe for 2D antiferromagnetic order.

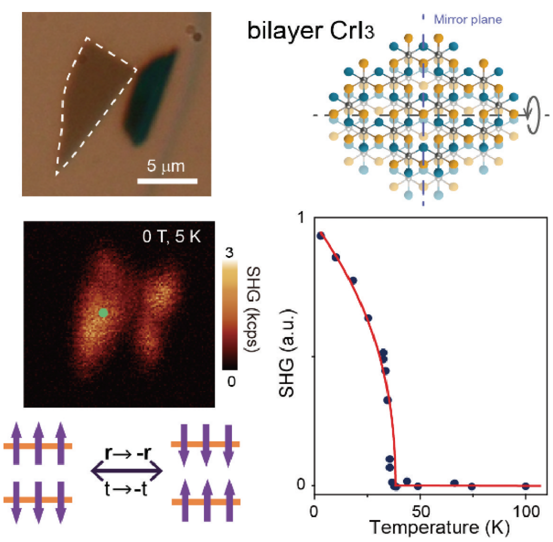

Figure Second harmonic generation from layered antiferromagnetism in bilayer CrI3.

Second harmonic generation (SHG) is a ubiquitous nonlinear optical process, which is not only important for technological applications, but also indispensable for investigating symmetry-related fundamental phenomena that are otherwise challenging to probe. To date, SHG phenomena are usually dominated by the systems with broken lattice inversion symmetry. In crystals with a centrosymmetric lattice structure, SHG can exist too, for example, due to the underlying magnetic order, but is generally believed to be a very weak effect.

Now the team uncovered a giant and nonreciprocal SHG in the newly discovered, antiferromagnetic CrI_3 bilayers, which possess a centrosymmetric lattice structure. They demonstrated that the SHG solely originates from the unique layered antiferromagnetism, where individual monolayers are ferromagnetic and their interlayer magnetic coupling is antiferromagnetic. Such a magnetic structure breaks both spatial inversion and time reversal symmetries, necessary for the electric dipole-allowed nonreciprocal SHG. For a direct comparison, they showed that the value of $|\chi^{(2)}|$, a measure of the strength of SHG, is ~3 orders of magnitude stronger than that in the Cr_2O_3 bulk (a model material for studying antiferromagnetism-induced SHG), and ~10 orders of magnitude stronger than the SHG induced by surface ferromagnetism in transition metal thin films (the first material system for observing the magnetization-induced SHG).

The ability to access antiferromagnetic states then enabled them to reveal the symmetry of spin-lattice structure in CrI_3 for the first time by polarization-resolved SHG. This is crucial for understanding the microscopic origin of interlayer antiferromagnetism in CrI_3, which was elusive due to lack of structural information. It was reported that bulk CrI_3 crystal possesses rhombohedral structure at low temperatures (< 200 K). However, theoretical calculations suggest that such a crystal structure should lead to ferromagnetic interlayer coupling, rather than the experimentally established antiferromagnetic coupling. With the extreme sensitivity of SHG to symmetry, they ruled out the rhombohedral structure by observing the broken three-fold rotational symmetry in the CrI_3 bilayer below the Neél temperature. Rather, they uncovered the C_{2h} symmetry, which establishes that the monoclinic structure is responsible for the layered antiferromagnetic ground states. This understanding should be the first step for developing a new theory for understanding the magnetic properties of CrI_3.