Unveiling the Complex Nonlinear Susceptibility in Atomically Thin WSe_2 through Valley-Polarization-Electric-Dipole Interference

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All-optical methods to probe the valley degree of freedom are fundamental not only to move forward the field of valleytronics, but also as a tool to study the interplay between space inversion and time-reversal (TRS) symmetries. Since second harmonic generation (SHG) is an ultrafast process and additionally highly sensitive to the symmetry of crystals, it was proposed as an alternative probe of VP [1], namely by detecting the rotation of the polarization-resolved SHG pattern induced by an elliptically polarized pulse [1]. This method suffers from limited valley addressability and cannot probe the complex nature of the valley polarization (VP). In this work, we demonstrate a novel method to study VP and thus broken TRS in a monolayer TMD by probing the interference between the intrinsic electric-dipole (ED) and VP-induced SHG and overcome aforementioned limitations of the approach proposed so far [2]. We compare the SH intensity generated by linearly and circularly polarized fundamental beams (FB) (Fig.1). If TRS is preserved, the ratio should be exactly 2. Instead, we measure a clear deviation from 2, with values ranging from 1.6 to 2.4. We analytically show that this result is captured by interference between the VP-induced and electric-dipole (ED) susceptibility, analogous to the magnetic-dipole-electric-dipole interference observed in bulk magnets [3]. Our work provides an advanced method to probe the valley degree of freedom or more generally the breaking of TR symmetry in TMD monolayers based on SHG, highlighting the unique capabilities of nonlinear optical processes for valleytronics and, possibly, as an all-optical probe of non-trivial topologies.

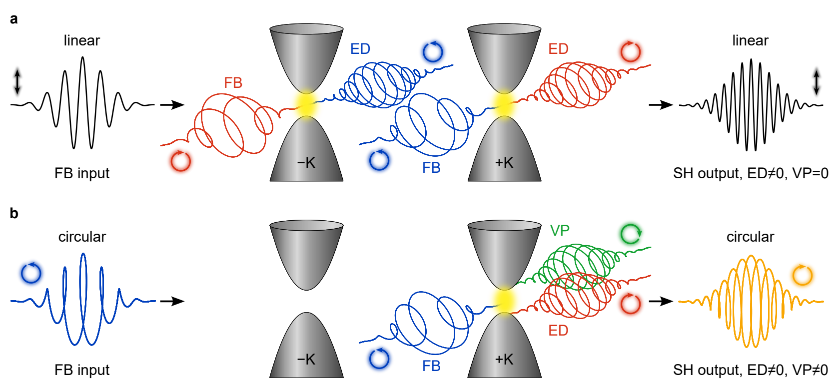

Fig. 1. **a**, A linearly polarized FB (left, black) can be decomposed into right (red) and left (blue) circular components interacting with the $\mp K$ valleys, respectively. As no VP is induced, only the ED contributes to the counter-rotating SH. Coherent superposition of the SH contributions from the $\mp K$ valleys results in linearly polarized SH (black, right). **b**, A left circularly polarized FB (left, blue) interacts only with the $+K$ valley. Simultaneously the fundamental induces a VP response. Therefore also the VP (green) contributes to the counter-rotating SH. Coherent superposition of the ED and VP contributions from the $+K$ valley results in circularly polarized SH (orange, right).

References

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