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Twisting for tunable nonlinear optics

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Yang et al. recently introduces the twisting degree of freedom into nonlinear physics and demonstrates tunable second-order nonlinear optical responses in twisted bilayer graphene. The demonstration opens a new route to future nonlinear optical applications.

The ability to stack two-dimensional (2D) layered materials and control the twist angle between them offers an exotic structure degree of freedom for van der Waals homo- and hetero-structures: the interlayer twist angle (Figure 1a)[1-3]. Such new degree of freedom presents new possibilities to tailor their electrical, optical, magnetic and topological properties and hence results in a wide variety of fascinating physical phenomena[4-8], such as Hofstadter's butterfly, correlated insulator behaviour, unconventional superconductivity, non-trivial topological Chern number, Hubbard model simulator and moiré excitons. This initiates the age of twistronics and twistoptics to transform the landscape of various fields (e.g., material science and condensed-matter physics).

Further, the interlayer twisting fundamentally determines the crystal space group and thus opens up exciting strategies for engineering the inversion symmetry breaking. More specifically, the inversion symmetry is explicitly broken for twisted van der Waals bilayers with a relative twist angle other than $\frac{2m\pi}{N}$, where N denotes the N-fold rotation symmetry of the monolayer and m is an integer (Figure 1b). Since the inversion symmetry breaking provides the necessary ingredient for a large portfolio of fascinating physical effects (e.g., even-order nonlinear effects, unconventional Ising superconductivity, Rashba spin-orbit coupling, non-vanishing Berry curvature/dipole, valley-contrasting light-valley interactions, piezoelectricity and ferroelectricity), the interlayer twisting provides a novel degree of freedom for numerous potential nanoelectronic and nanophotonic applications.

For instance, second-harmonic generation (SHG) where two photons with the same frequency are converted into a new photon with twice the frequency of the incident photons is the most widely studied nonlinear optical process[9]. According to the generally accepted nonlinear optics principles, SHG under the electric-dipole approximation is strongly inhibited by crystalline symmetries and thus can manifest only in materials with broken inversion symmetry. Now, writing in *Matter*, Fuyi Yang and colleagues from the Yao group at University of California Berkeley have reported that the interlayer twisting offers an exotic approach to demonstrate efficient SHG in artificially twisted bilayer graphene (tBLG) created from centrosymmetric monolayer constituents. The achieved second-order nonlinear susceptibility is up to $\sim 2.8 \times 10^5\ pm^2/V$, comparable to these of the most well-known 2D nonlinear optical materials studied (e.g., MoS_2). This twisting approach can significantly expand the scope of suitable materials for SHG and be applicable to other physical phenomena that are closely linked with broken inversion symmetry, such as nonlinear Hall effect, circular photogalvanic effect, piezoelectricity, ferroelectricity, quantum spin Hall effect and Majorana zero modes.

Yang and colleagues have also demonstrated tunable SHG in tBLG. Owing to the strong interlayer coupling and hybridized electronic states, interlayer twisting can enable the formation of flat bands with high density of states in the conduction and valance bands of tBLG, dubbed as van Hove singularities. The transition energies (E_{vHs}) bettwen the van Hove singularities of conduction and valance bands in tBLG are strongly dependent on the twist angle. Therefore, for a fixed twist angle (incident wavelength), one- and two-photon resonances could be selectively turned on or off via changing the incident wavelength (twist angle), giving rise to highly tunable SHG (Figures 1c-1e). Yang and colleagues have shown that SHG intensity is significantly enhanced when the incident wavelength of 785 nm is resonant to $E_{vHs} = 1.58$ eV (3.16 eV) with a twist angle of ~ 8° (20°), corresponding to the one-photon (two-photon) resonance. Here we point out that other nonlinear optical processes, such as third-harmonic generation, difference frequency generation, four-wave mixing, and high-harmonic generation, can also be resonantly enhanced and widely tunable by controlling the interlayer twisting.

The seminal work of Yang et al. has introduced the interlayer twisting degree of freedom into nonlinear optics, providing an exotic strategy for the engineering of inversion symmetry breaking and hence the realization of widely tunable nonlinear optical responses. The twist-tunable resonance in tBLG, in principle, can range from zero at θ = 0° to ultraviolet (~ 4 eV) at θ = 30°, covering the most common spectral range for silicon based photonics and optical fiber communications (e.g., O band around 1300 nm and the S and C bands around 1550 nm). Therefore, tBLG can be fully integrated with the current optical material, waveguide and resonator platforms for compact, efficient and fast nonlinear optical nanodevices. Moreover, there is no doubt that the strategy of twisting can be applied to other 2D materials and hold substantial future growth potential to construct next-generation nanophotonics and nanophysics for new and emerging applications in metrology, quantum technology, communications and sensing. The age of nonlinear twistoptics is coming!

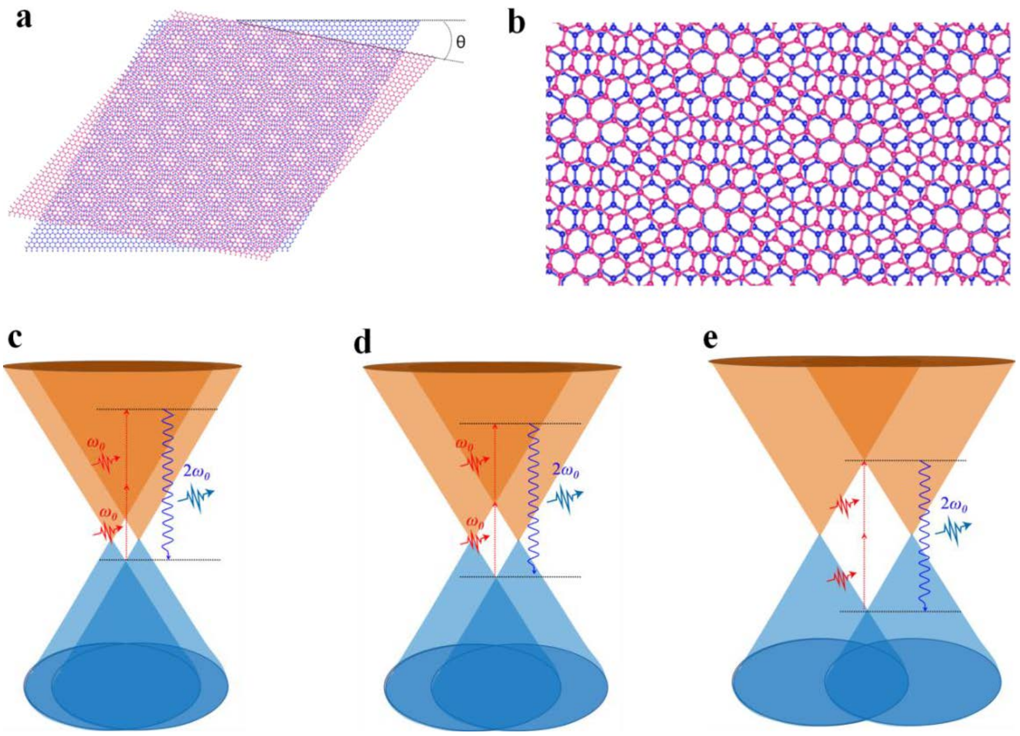

Figure 1. Twisting for nonlinear physics. (a) Schematic diagram of moiré pattern in tBLG with twist angle θ. (b) Schematic crystal structure of tBLG, indicating the the inversion symmetry breaking. (c-e) Illustrations of the SHG with off-resonance (c) one-photon ($\omega_0 = E_{\mathrm{vHs}}$) (d) and two-photon resonances ($2\omega_0 = E_{\mathrm{vHs}}$) (e) by controlling the interlayer twist angle of tBLG.

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