Control and Enhancement of Optical Nonlinearity in Twisted Bilayer Graphene

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1. Introduction

Nonlinear optic processes have been extensively used in diverse fields, including ultrafast optical signal control, wavelength conversion, and generation of entangled light sources in quantum technology. Graphene, a single sheet of carbon atoms forming a hexagonal structure, has received increasing attention as a promising material for nonlinear optic applications. Since monolayer graphene (MLG) possesses inversion symmetry, second-order nonlinearity is not allowed. Thus, the third-order optical nonlinearity, called Kerr nonlinearity, becomes the primary nonlinear response. Graphene is known to have a large third-order optical nonlinearity over broad bandwidth because of its strong optical coupling and linear energy dispersion of the Dirac fermions. Recent studies have shown that the efficiency of the nonlinear optical signal could be electrically controlled and enhanced in MLG [1, 2].

2. Nonlinear optic properties in tBLG

Twisted bilayer graphene (tBLG) is a pair of MLG stacked with a specific rotation angle, forming Moiré superlattices of carbon atoms. The interlayer interaction in tBLG notably reconstructs the electronic band structure of graphene, leading to very intriguing physical properties such as superconductivity, Mott insulator, and Moiré excitons. Here we investigated the nonlinear optical properties in tBLG according to the twist angle θ. The generated third harmonic signal intensity is found to change with the twist angle in tBLG considerably. Figure 1 shows the results according to the Raman 2D/G ratio when tBLG interacts with a femtosecond light source with a center wavelength of 1560 nm. We observed that the Raman 2D/G ratio is inversely proportional to the THG intensity generated in tBLG. Particularly tBLG in θ ~ θc group shows more enhanced THG intensity. Here θc is a critical angle where the energy gap at van Hove singularity matches the three-photon resonance of incident light. Moreover, we examined THG characteristics by electrically controlling third-order nonlinear optic responses. The maximum value of THG intensity in tBLG was about 60 times larger than that in neutral MLG [3].

3. Conclusions

In summary, we demonstrate THG in bilayer graphene, which shows a strong dependence on stacked angle between graphene layers. Enhanced THG was observed when the rotation-induced nonlinear coupling is matched with the photon energy of the incident laser. Moreover, we investigated the correlation between the twist angle and THG intensity by Fermi-level tuning through electrical gating. Our result provides a basic understanding of third-order nonlinearity possessing a strong relationship with twist angle in tBLG, which provides a novel way to design and enhance the optical nonlinearity in a two-dimensional material system.

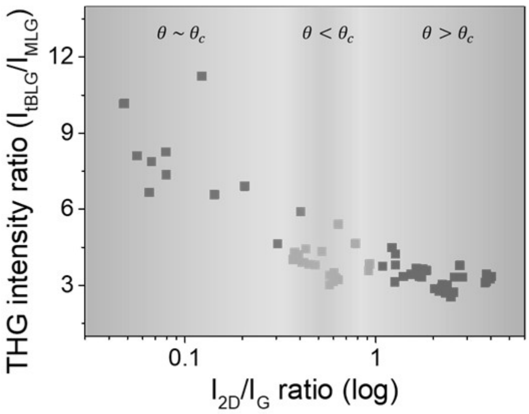

Fig.1 The measured THG intensity as a function of the I_{2D}/I_G ratio of tBLG samples.

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