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Photo-induced ultrafast phase transition in twisted bilayer graphene

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Twisted bilayer graphene (TBG) has recently attracted immense attention as they exhibit several intriguing phenomena at magic angles such as superconductivity[1], correlated insulating behavior[2] and ferromagnetism[3]. It is widely recognized that the moiré pattern plays an important role in these new properties by modulating the electronic band structure of the material. In our recent work[4], we found the moiré pattern of TBG plays another important role in the formation of transient 2D diamond through naturally providing stacking configurations like AA and AB' which are favorable for the preferential formation of sp^3 bonds, providing a new methodology to utilize moiré pattern for developing next-generation optoelectronic devices.

In graphite (aligned graphene), carbon atoms are arranged in planar honeycomb sheets with alternating carbon atoms in adjacent honeycomb centers, so called AB stacking. This AB stacking makes graphite thermodynamically stable but difficult to directly convert into diamond. Therefore, theorists have predicted that before transforming into diamond, graphite sheets must go through a sliding process forming high energy intermediate phase with matching AA stacking or close matching AB' stacking. By simply twisting two graphene sheets, we can arrive to this kind of intermediate stacking, which provides local areas close to AA and AB' stacking in moiré pattern facilitating transient 2D diamond formation. Using SLAC mega-electron-volt ultrafast electron diffraction[5] (MeV UED), we show diamond-like transient states in TBG under fs laser irradiation. The UED pattern of TBG is shown in Fig. 1a. The time-dependent intensity changes of {100} and {110} diffraction rings at pump fluence of ~9.7 mJ/cm^2 clearly show a rapid intensity rise in the {100} ring and a drop in the {110} ring (Fig. 1b), indicating this process is not a thermal effect. This can be further supported by the following Q shift as illustrated in Fig. 1c. The time-dependent differential pair distribution function (ΔPDF) was calculated to further extract the structural dynamics of TBG. The experimental ΔPDF is shown in Fig. 2a, with blue indicating loss and red indicating gain in the ΔPDF at various atom pair distances compared with unexcited TBG. Ultrafast changes in interatomic distances were observed from the ΔPDF. By choosing the appropriate time delays, we obtained a series of one-dimensional ΔPDFs (Fig. 2b). The positive peaks (II, Ⅳ) represent a significant increase in the ΔPDF at interatomic distances r of 1.94 Å and 3.14 Å, while the negative peaks (I, III) correspond to a reduction of the ΔPDF at interatomic distances at 1.42 Å and 2.46 Å. The appearance of new bond lengths of 1.94 Å and 3.14 Å other than 1.42 Å, 2.46 Å and 2.84 Å (bond lengths in graphene) in PDF reflects the interlayer distance is reached to the smallest valve of new bond lengths, indicating the ultrafast formation of interlayer sp^3bonds after the fs laser irradiation. To understand the detailed structural evolution during compression, we carried out molecular dynamics (MD) simulations for TBG with different twist angles. All TBGs with twist angles other than 0° with AB stacking showed similar structural dynamics. Taking a typical angle of 12° as an example, one can see that after excitation, sp^3bonds labeled with red and orange (warm colors) are preferentially formed in local areas with stacking close to AA and AB', but almost no sp^3 bonds are formed (shown in blue) in the area with AB stacking from the top-view snapshots (Fig. 3). These simulation results support our experimental observations of ultrafast phase transitions in TBG but not in aligned graphene sheets that is composed entirely of AB stacking configuration. Using the simulated transient structures, we calculated their ΔPDFs by selecting several representative moments. These calculated ΔPDFs match well with the experimental ones (Fig. 2b).

This new discovery using TBG offers a promising way to open and precisely tune a bandgap in graphene system, enabling the exploration of innovative applications for the future. In addition, this work sheds light on

the direct graphite-to-diamond transformation mechanism, which has not been fully understood for more than 60 years.

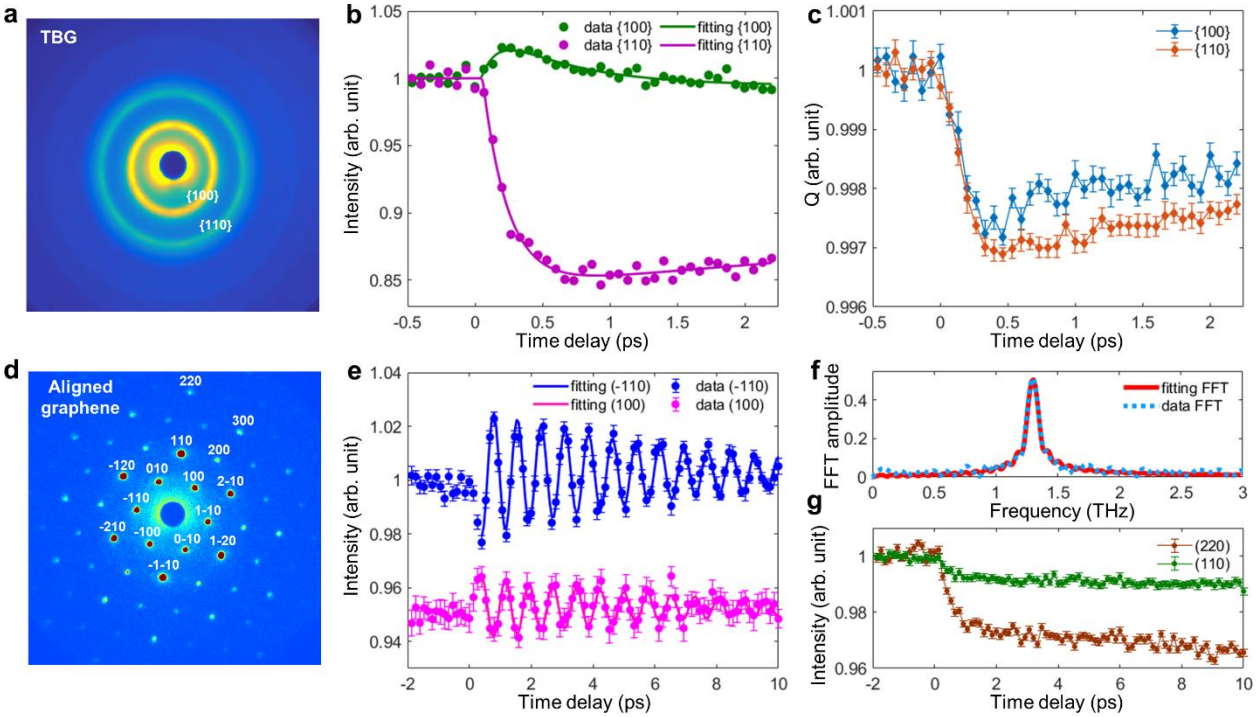

Figure 1. Figure 1. Ultrafast structural dynamics in TBG. a, Typical diffraction pattern of TBG. b, Diffraction intensity changes as a function of time delays. c, Time-dependent Q shift. d-f, for aligned graphene.

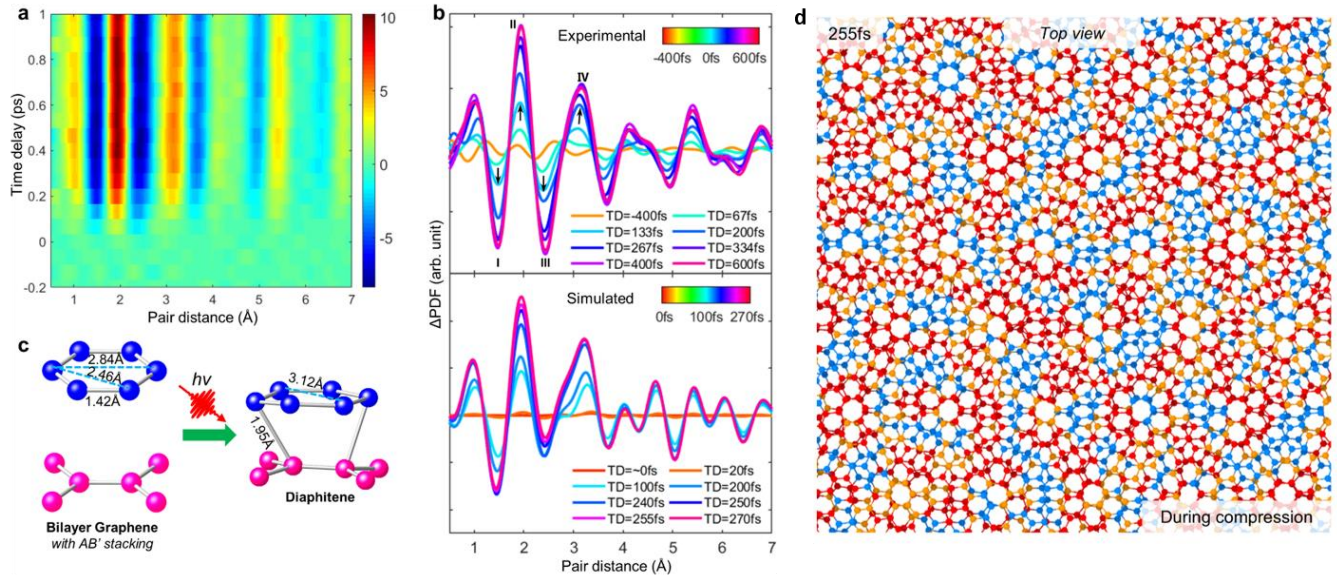

Figure 2. Figure 2. a, Experimental 2D ΔPDF of TBG. b, 1D experimental and simulated ΔPDF. c, transient structures extracted from simulation. d, Snapshot of the top view during compression from a MD simulation.

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