2D materials

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Transforming into fully commensurate bilayers

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Heat treatment can transform some moiré superlattices into fully commensurate bilayers, where atoms in opposite layers align perfectly with each other. This structural transformation gives rise to markedly brighter interlayer excitons.

Monolayer transition metal dichalcogenides (TMDs), such as MoS_2, $MoSe_2$ and WSe_2, constitute an outstanding family of two-dimensional (2D) semiconductors with remarkable electronic and optical properties. These materials exhibit conduction-band and valence-band valleys at the corners of the hexagonal Brillouin zone. In a typical optical process, the absorption of a photon generates an electron in the conduction-band valley and a hole in the valence-band valley. Subsequently, the electron–hole pair relaxes to form a bound state, known as an exciton, which may recombine to emit a photon. Because TMD exciton energies span from the infrared to the visible range, TMD materials hold promise for optical and optoelectronic applications.

An important challenge in TMD research is controlling the properties of excitons, including their energy, brightness and lifetime. A clever strategy involves stacking two different TMD monolayers to create a heterogeneous bilayer, often referred to as a heterobilayer. Some heterobilayers exhibit staggered band alignment, such as the one depicted in Fig. 1a for a $MoSe_2/WSe_2$ heterobilayer. In this case, the conduction-band minimum resides in the $MoSe_2$ layer, while the valence-band maximum is found in the WSe_2 layer. Consequently, electrons tend to relax to the $MoSe_2$ layer, while holes migrate to the WSe_2 layer, forming interlayer excitons (Fig. 1a). These interlayer excitons possess lower energy than the monolayer excitons and exhibit longer lifetimes due to the separation of electrons and holes[1]. This behaviour paves the way for expanding the functionality of TMD materials.

Although the above approach seems straightforward in theory, practical implementation shows complexities, primarily arising from the different lattice constants and/or orientations of the two stacked TMD layers. The interlayer lattice and orientation mismatch yield two notable consequences. First, the electron and hole within the interlayer exciton exhibit different momenta because the Brillouin zones of the two TMD layers differ in size and orientation (Fig. 1a). The mismatch in the electron and hole momenta strongly suppresses exciton light emission because visible photons have negligible momentum. Second, lattice mismatch and/or interlayer twisting give rise to a moiré pattern in the heterobilayer (Fig. 1c,e). The periodic moiré landscape can confine excitons and modulate their properties. Although the moiré superlattice can lead to novel excitonic phenomena[2–4], it complicates the materials' characteristics and, in many cases, makes it difficult to control the exciton properties.

Now, writing in *Nature Materials*, Ji-Hwan Baek and colleagues[5] report a straightforward yet effective method to address these challenges. Their method is applicable to $MoSe_2/WSe_2$ heterobilayers

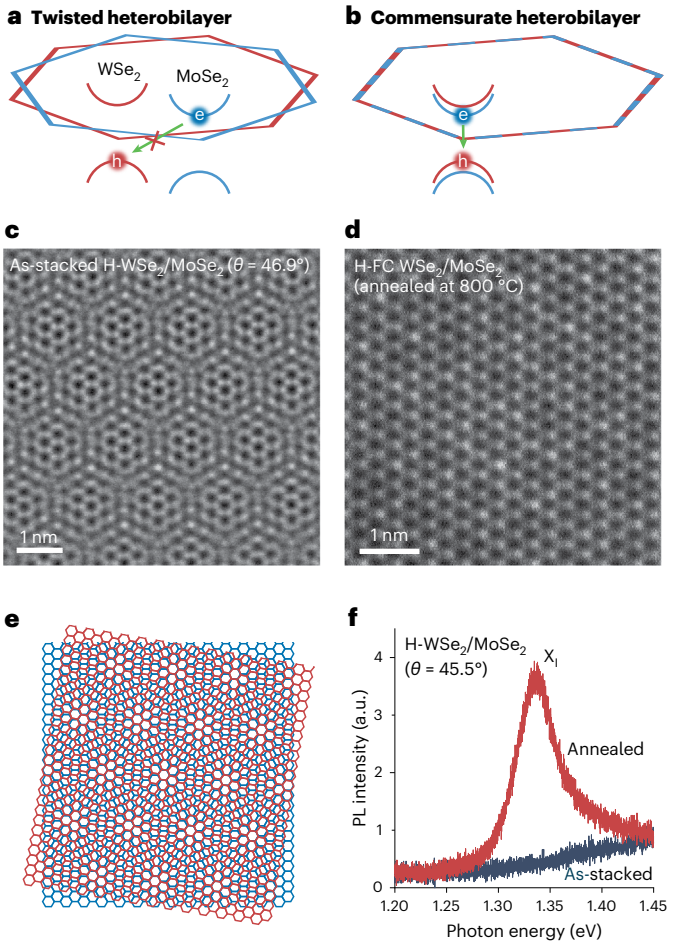

Fig. 1 | Turning $WSe_2/MoSe_2$ moiré superlattices into fully commensurate heterobilayers by annealing. a, Illustrative band alignment in a twisted $WSe_2/MoSe_2$ heterobilayer. The red (blue) hexagon represents the Brillouin zone of the WSe_2 ($MoSe_2$) layer. The arcs with corresponding colour represent their conduction-band and valence-band valleys at the zone corners. Owing to their staggered band alignment, the electrons (holes) relax to the $MoSe_2$ (WSe_2) layer and form interlayer excitons. **b**, Band alignment of a fully commensurate $WSe_2/MoSe_2$ heterobilayer. **c**,**d**, Scanning transmission electron microscopy images of a H-type $WSe_2/MoSe_2$ heterobilayer with twist angle $\theta = 46.9°$ before (**c**) and after (**d**) annealing at $T = 800$ °C. **e**, Illustration of the moiré pattern in **c**. **f**, The photoluminescence (PL) spectra of interlayer excitons in a twisted $MoSe_2/WSe_2$ heterobilayer before (black) and after (red) annealing. Initially, the interlayer excitons cannot recombine optically because the electrons and holes have different momenta (**a**). But annealing transforms the twisted heterobilayer into a fully commensurate heterobilayer, removing the electron–hole momentum mismatch (**b**), thus enabling the interlayer excitons to recombine to emit light (**f**). FC, fully commensurate; e, electron; h, hole. Figure adapted with permission from ref. 5, Springer Nature Ltd.

with minimal (approximately 0.1%) lattice mismatch and also to some twisted TMD homogeneous bilayers. The effectiveness of the method crucially hinges on the utilization of boron nitride to encapsulate the bilayers. The authors discovered that by heating the encapsulated bilayer to a high temperature (T = 800 °C) and then cooling it down (a process called annealing), the moiré pattern disappears, and that the moiré superlattice spontaneously transforms into a heterobilayer with perfect interlayer atomic alignment (Fig. 1c,d). Remarkably, their method works even when the two TMD monolayers initially have a large interlayer twist angle. Baek and colleagues find that in the annealing process the two twisted layers do not rotate macroscopically as a whole to align with each other; instead, the atoms in one or both layers undergo microscopic movement to achieve interlayer atomic registry. Such atomic reconstruction eventually produces a fully commensurate heterobilayer, where the atoms in opposite layers align perfectly with each other (Fig. 1c,d). In this new phase, the $MoSe_2$ and WSe_2 layers have exactly the same lattice constant and orientation.

One remarkable consequence of this structural transition is that the interlayer excitons are markedly brightened. In contrast to the twisted heterobilayer with misaligned Brillouin zones (Fig. 1a), the fully commensurate heterobilayer features two Brillouin zones with identical size and orientation for the $MoSe_2$ and WSe_2 layers (Fig. 1b). As a result, the electrons and holes exhibit matching momenta. The initially momentum-indirect interlayer excitons now become momentum-direct, leading to much brighter luminescence (Fig. 1a,b,f).

Besides brightening the interlayer excitons, achieving full commensuration is expected to change interlayer electronic coupling, leading to modification of the band structure and distinct optical properties[6]. In this context, Baek and colleagues demonstrate the controlled formation of commensurate heterobilayers with both R-type (0° interlayer rotation) and H-type (180° interlayer rotation) stacking configurations. These two phases are expected to have different band structures and optical properties due to their distinct interlayer electronic coupling[7–9]. Consequently, the authors' research holds the potential to reveal novel materials properties not attainable in incommensurate heterobilayers.

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Competing interests

The author declares no competing interests.