Tailoring Coulomb correlations in WSe_2 homobilayers via interlayer twist

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Recently novel phase transitions in atomically thin layers were discovered by stacking them at magic twist angles, opening a new paradigm in solid state physics. Topological phases [1] and shear solitons [2] have been discussed for twisted bilayers of transition metal dichalcogenides. Yet, a precise understanding of the underlying Coulomb correlations has remained challenging, even though it is the key to tailor novel functionalities of matter.

Here, we isolate how the internal structure and mutual interaction of Coulomb bound electron-hole pairs – called excitons – in WSe_2 bilayers are shaped by the twist angle alone [3]. To this end, we employ near-infrared (NIR) pump – mid-infrared (MIR) probe spectroscopy (Fig. 1a) and interrogate Lyman-like 1*s*–2*p* transitions, which dominate the characteristic MIR response function of excitons [4] (Fig. 1b). Remarkably, the 1*s*–2*p* transition energy – a direct measure for the exciton binding energy – is renormalized by a factor of two by varying the twist angle (Fig. 1c). These findings are explained by a microscopic theory, which identifies the underlying mechanism as hybridization between different excitonic species (Fig 1d). The interlayer hybridization of excitons also allows us to tune their lifetime and many-body interactions. By changing the stacking angle, the recombination lifetime exhibits a giant enhancement by two orders of magnitude (Fig. 1e) while the exciton-exciton interactions become highly tuneable (Fig. 1f). Our work opens the possibility for tailoring quasiparticles in search of new phases of matter in a broad range of van der Waals heterostructures.

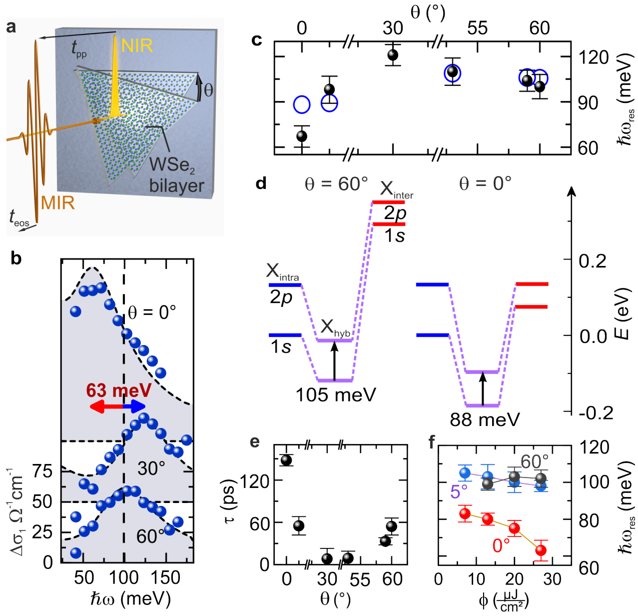

Figure 1: a Sketch of the experiment: A (NIR) pump pulse (yellow) injects 1*s* A excitons in the WSe_2 BLs. The excited sample is probed by a MIR waveform (orange). **b** Pump-induced change of the real parts of the optical conductivity $\Delta\sigma_1$ for a pump-probe delay time of t_{pp} = 5.1 ps for samples with different twist angles θ. Blue spheres: experimental data (pump fluence Φ = 27 µJ/cm²; sample temperature, 5 K). Grey shaded areas: phenomenological model. The dashed line and the blue/red arrow indicate the blue/red shift of the 1*s*–2*p* resonance energy $\hbar\omega_{res}$. **c** $\hbar\omega_{res}$ (black spheres) extracted from the data in b and derived from the microscopic theory (blue circles) as function of θ. **d** Hybridization of 1*s* and 2*p* excitons X_{hyb} lowers the energy levels (magenta lines) with respect to X_{intra} and X_{inter} (blue/red lines). Zero energy is set to 1*s* X_{intra} at θ = 60°. The vertical black arrows mark respective 1*s*–2*p* transition energies. **e** Exciton decay time τ as a function of θ. **f** $\hbar\omega_{res}$ for different pump fluences Φ at t_{pp} = 5.1 ps for different θ.

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[3] P. Merkl *et al.*, *Nat. Commun.*, under review.
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