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Twist-Tailoring Hybrid Excitons In Van Der Waals Homobilayers

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In homobilayer systems, the twist angle θ has emerged as a powerful tuning knob for tailoring novel phase transitions in atomically thin layers stacked at magic twist angles [1] – a paradigm shift for condensed-matter physics. For twisted bilayers of transition metal dichalcogenides, topological phases [2] and even potential signatures of a superconducting state [3] have been discussed. Yet, a precise understanding of the underlying Coulomb correlations has remained challenging.

Here, we introduce a direct, ultrafast access to Coulomb correlations in van der Waals heterostructures out of equilibrium [4,5]. Phase-locked mid-infrared pulses interrogate Lyman-like 1*s*–2*p* transitions of optically bright and dark excitonic species on subcycle timescales. These transitions manifest as novel resonances in the extracted, complex-valued dielectric response (Fig. 1a,b) and reveal how the twist angle alone affects the properties of hybrid excitons in WSe_2 bilayers [5]. Remarkably, the exciton binding energy is renormalized by as much as a factor of two (Fig. 1c), their lifetime exhibits a giant enhancement by more than an order of magnitude (Fig. 1d), and the exciton-exciton interaction is widely tunable. Density functional theory and density matrix theory identifies variable degrees of exciton hybridization between the layers as the microscopic origin for all these observations.

Our work significantly advances the concept of tailoring quasiparticles in search of new phases of matter and is applicable to a broad range of man-made materials. Furthermore, combining sub-cycle time-domain spectroscopy with near-field microscopy [6] represents a promising route towards resolving ultrafast charge carrier dynamics in van der Waals heterostructures on the nanoscale.

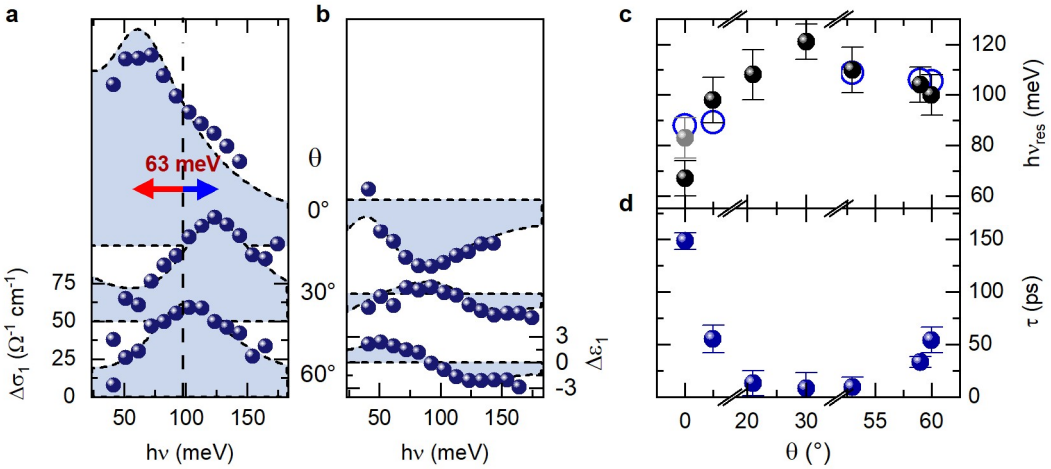

Fig. 1 a,b, Pump-induced changes of the real parts of the optical conductivity $\Delta\sigma_1$ (**a**) and the dielectric function $\Delta\varepsilon_1$ (**b**) for a fixed pump-probe delay time t_{pp} = 5.1 ps as a function of the photon energy hν for WSe_2 bilayers with different twist angles θ. Blue spheres: dielectric response measured for the photoexcited samples (pump fluence, Φ = 27 μJcm^{-2}; temperature, T = 5 K). Blue shaded areas: fits to the experimental data by a phenomenological model. Dashed line and the blue/red arrow: blue/red shift of the 1*s*–2*p* resonance energy with respect to the 2H stacking. **c**, Intraexcitonic 1*s*–2*p* transition energy $h\nu_{res}$ (black spheres) extracted from the data in **a**, **b** and derived from the microscopic theory (blue circles) as function of θ. In the case of θ = 0°, the gray sphere indicates $h\nu_{res}$ as obtained for a pump fluence of Φ = 7 μJcm^{-2}, yielding the best agreement with the microscopic theory. **d,** Exciton decay time τ as a function of the twist angle θ, determined by fitting exponential decays to the pump-induced changes to the mid-infrared electric field Δ*E* (recorded at fixed electro-optic sampling time t_{eos} = 0 fs) as a function of t_{pp}.

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