# Internal structure and formation dynamics of interlayer excitons

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Interlayer excitons in hetero-bilayers of transition metal dichalcogenides [1] feature unique properties for optoelectronics and valleytronics [2]. All future applications hinge on a precise understanding of the binding energy and formation dynamics of these bound states. Yet, their weak coupling to light brings interband absorption spectroscopy to its limits. Here, we study interlayer excitons in a $WSe_2/WS_2$ heterostructure (Fig. 1a) by directly interrogating their internal structure with phase-locked mid-infrared probes [3]. We reveal a novel transition between 1*s* and 2*p* orbitals of interlayer excitons (Fig. 1b, upper panel) at an energy of 67 ± 6 meV. This value coincides with our numerical solution of the Wannier equation (Fig. 1c) and allows us to determine the binding energy of itinerant interlayer excitons as 126 ± 7 meV. Since the complex-valued spectral response functions of inter- and intralayer excitons differ strongly (Fig. 1b) [4], we can sensitively track the ultrafast evolution of the respective exciton densities as a function of the pump-probe delay time $t_{pp}$ (Fig. 1d). Interestingly, intralayer excitons photoinjected into the $WSe_2$ monolayer transform into interlayer species by direct ultrafast electron tunnelling without the occurrence of a transient conductive phase. Depending on the stacking angle, intra- and interlayer species coexist on picosecond scales and the 1*s*-2*p* resonance becomes renormalized. Our work provides the first direct measurement of the binding energy of interlayer excitons and opens the possibility to trace and control correlations in novel artificial materials.

## Figures

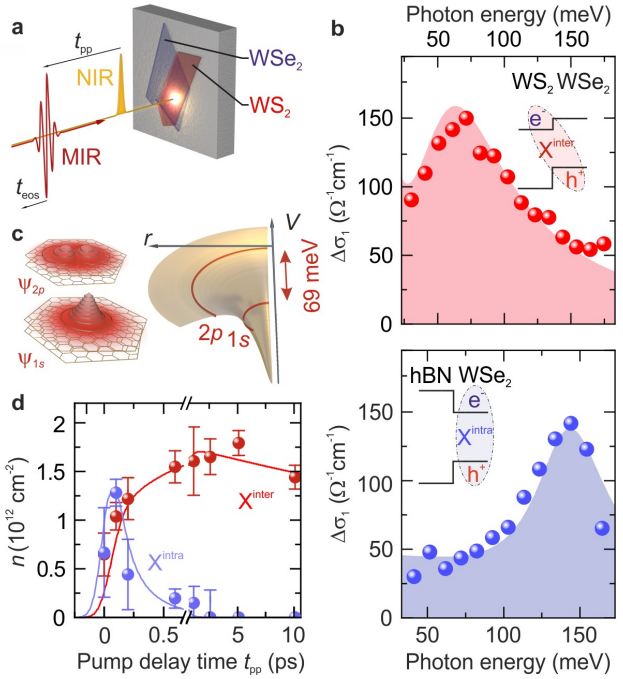


**Figure 1: a** Time-resolved near-infrared (NIR) pump – mid-infrared (MIR) probe spectroscopy of a $WSe_2/WS_2$ heterostructure on a diamond substrate. The NIR pump pulse resonantly injects 1*s* A excitons in the $WSe_2$ monolayer, whereas the MIR probe pulse samples the dielectric response of the heterostructure. **b** Pump-induced change of the real part of the optical conductivity $\Delta\sigma_1$ revealing 1*s*-2*p* transitions of interlayer excitons (upper panel) and intralayer excitons (lower panel). Insets: band alignments of the heterostructures. **c** Interlayer Coulomb potential and wavefunctions of the 1*s* and 2*p* interlayer excitons in real space. **d** Intra- and interlayer exciton densities as a function of pump-probe delay time $t_{pp}$ for a $WSe_2/WS_2$ heterostructure with a twist angle of $\theta = 5°$. Solid lines: results of the microscopic theory.

## References

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