# Ultrafast Scanning Tunnelling Spectroscopy of a Phonon-driven Atomic Single-Photon Emitter in a Monolayer Crystal

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Atomic-defect-based quantum systems in monolayers and moiré heterostructures of 2D materials have attracted huge interest for their qubit and single-photon emission functionalities. Whereas time-resolved ARPES provides critical insights into the dynamics of electronic energies in momentum space, its counterpart with atomic spatial instead of momentum resolution is necessary to directly observe the interplay of electronic structure of a single defect with the microscopic excitations of the environment on the intrinsic atomic and femtosecond scales.

Here we directly resolve in space, time and energy how spin-orbit-split bound states of an individual Se vacancy – an atomic single-photon emitter – evolve under coherent lattice vibrations in moiré-distorted $WSe_2$ using lightwave-driven scanning tunnelling spectroscopy (LWSTS) [1] (Fig. 1a, b). We selectively launch a drum phonon mode (Fig. 1c) with a THz pulse coupled to the tip and take ultrafast snapshots of electronic spectrum on atomic scales faster than a vibration period. Such ultrafast tunnelling spectra reaching ~300 fs temporal resolution reveal transient energy shifts of the lower bound vacancy state by up to 40 meV, depending on the amplitude and phase of the coherent lattice vibration (Fig. 1d). We discuss how THz fields can couple via the Coulomb interactions to the drum mode, and how the interplay of Se-W bonds distortion and image charge renormalization due to vertical motion of the vacancy, induced by the drum mode, affect the energy levels of the vacancy.

The combination of atomic spatial, sub-picosecond temporal, and meV energy resolution marks a disruptive development towards a comprehensive understanding of complex quantum matter, paving the way to disentangling microscopic interactions one by one and tuning many-body states by transiently shifting their energetic position.

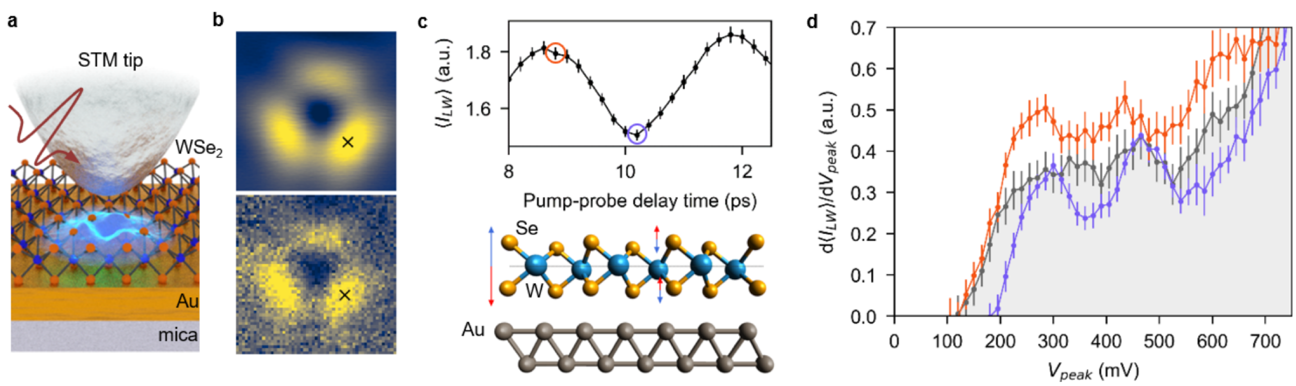


Fig.1. **a,** Schematic of LWSTS: an ultrashort THz pulse (red) of variable intensity coupled to the tip transiently modulates the bias between tip and Se vacancy (blue orbital) in monolayer $WSe_2$. **b,** Constant-current images ($1.5 \times 1.5$ nm$^2$) of the vacancy measured with DC (top, $V_{DC} = 720$ mV) and THz-driven currents (bottom, $V_{DC} = 0$, peak THz bias $V_{peak} \sim 720$ mV). **c,** Tunneling current driven by a THz probe pulse as a function of the delay time after local excitation by a THz pump pulse (top). Ball-and-stick model of the induced drum phonon mode (bottom), which involves a center-of-mass motion of the $WSe_2$ monolayer as well as out-of-phase intra-cell distortion (red and blue arrows mark opposite oscillation phases). **d,** Transient tunneling spectra measured at the defect lobe (cross in (b)) for delay times corresponding to maximum (red, 8.8 ps) and minimum (blue, 10.2 ps) distance between $WSe_2$ and Au in (c), grey – unpumped spectrum. The rising edge shows the onset of tunnelling into the lower bound state of the vacancy.

References

[1] C. Roelcke, L. Z. Kastner *et al.*, Nature Photonics (2024) DOI: 10.1038/s41566-024-01390-6