Laser control of an excited-state vibrational wave packet in neutral H_2

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We observe and control a molecular vibrational wave packet in an electronically excited state of neutral H_2. Using extreme-ultraviolet (XUV) light in the spectral range between 13 eV and 17 eV we drive multiple transitions between the ground state and excited vibronic states in the neutral hydrogen molecule, Fig.1(a). Through spectral filtering of the rich absorption spectrum we select only the transitions to the electronically excited $D\,^1\Pi_u\,3p\pi$ state, Fig.1(b), and use the technique of real-time reconstruction of the dipole response [1] to obtain the time-dependent coherent dipole emission associated to the D vibrational wave packet, with the wave-packet revival at 270 fs imprinted on it, Fig.1(c). A subsequent interaction with a 5-fs short near-infrared (NIR) pulse arriving 7 fs after the fast (attosecond-scale) XUV excitation modifies the vibrational wave packet and changes the revival signature in the reconstructed dipole, Fig.1(d). An experimental study of the intensity dependence of the wave-packet revival and a supporting multi-level simulation solving the time-dependent Schrödinger equation show: the higher the NIR intensity, the earlier the revival time. We further extract the state-specific NIR-induced phase-shifts, which are a key contributor to the observed time shifts. In this general approach for wave-packet modification, transferable also to complex molecules, the molecular ground state acts as the probe for the visualisation of the modified wave packet and only a pump and a control laser pulse are required for both the time-dependent reconstruction and the control of the molecular vibrational wave-packet [2].

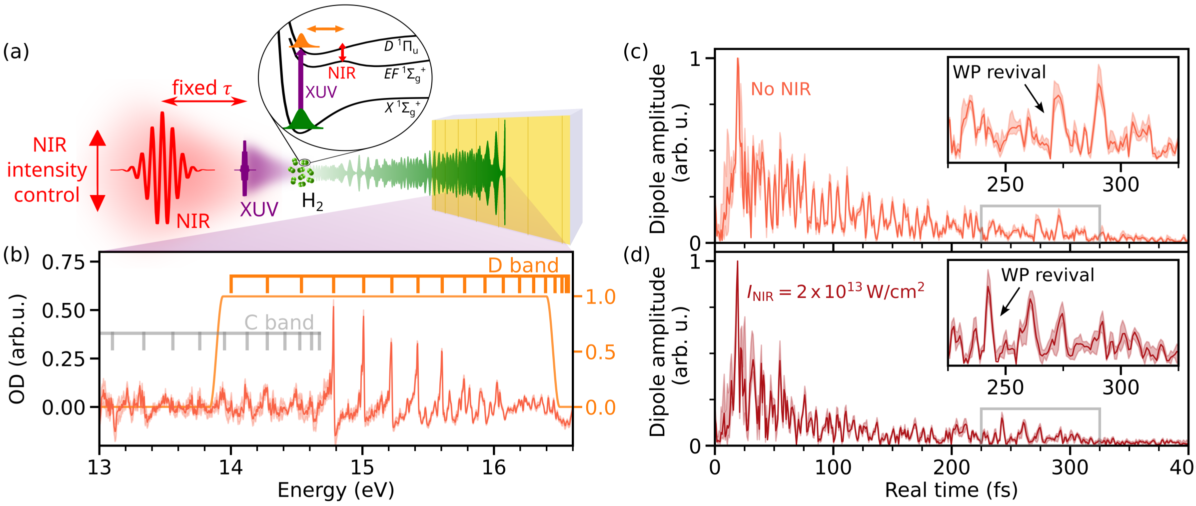

Figure 1: **(a)** Schematic of the experimental setup for XUV time-domain absorption spectroscopy in H_2. **(b)** Optical density (OD) spectrum as measured after transmission through a 10 mbar 3 mm H_2 sample. **(c)** and **(d)** Reconstructed time-dependent dipole amplitude of the D-state vibrational wave packet in an XUV-only configuration and for an NIR control field with $I_{\mathrm{NIR}} \approx 2 \times 10^{13}\,\mathrm{W/cm^2}$, respectively. The insets show the wave-packet revival region.

References

[1] Stooß V *et al.* 2018 *Phys. Rev. Lett.* **121** 173005
[2] Borisova G D *et al.* 2023 arXiv:2301.03908