Ultrafast Transient Absorption Spectroscopy of the charge-transfer insulator NiO: Beyond the Dynamical Franz-Keldysh Effect

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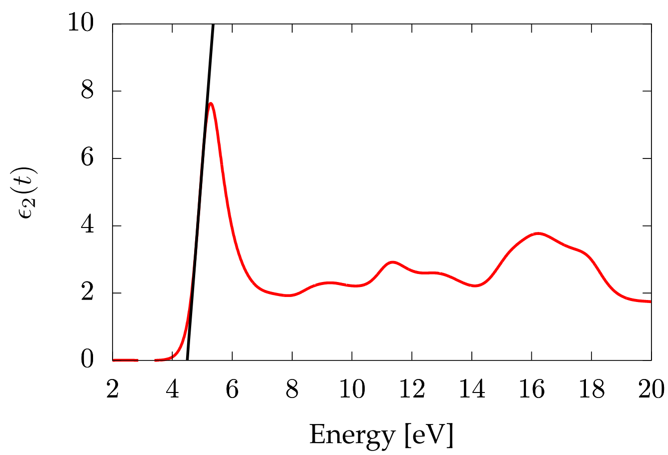
EXTRACTION OF THE OPTICAL GAP

In this section, we present how we extracted the optical gap from the calculated optical spectra.

In order to get the optical gap, we first find the values of the imaginary part of the dielectric function at 40% and 90% of the maximum peak value. Then we fit this portion of the dielectric function in between these two points by a straight line.

The value of the optical gap is taken as the point for which this tangent crosses the y=0 axis. This is exemplified in Supplementary Fig. 1 for the equilibrium spectrum of the NiO.

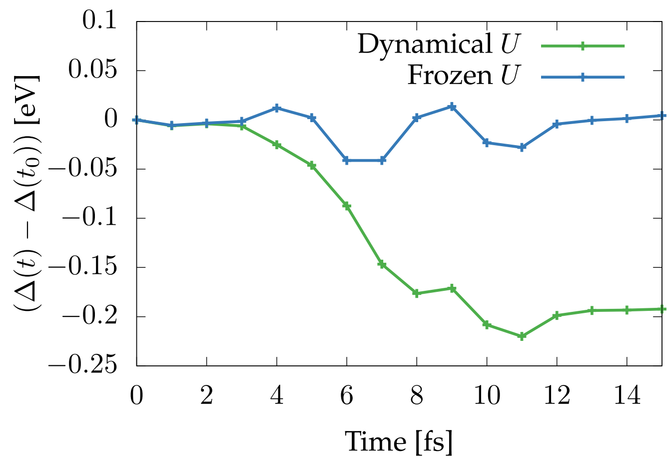


Supplementary Fig. 1. Extraction of the optical gap of NiO. See the main text for details.

Note that small shoulder at the absorption edge is a numerical artifact, that can be removed by a tighter convergence in **k**-points.

COMPARISON OF THE OPTICAL GAP DYNAMICS FOR FROZEN AND DYNAMICAL U

In this section, we compare the dynamics of the optical gap in the case of the dynamical U (as shown in the main text), and the one obtained for a frozen U. As shown in Supplementary Fig. 2, while the optical gap shows a clear decrease during the laser pulse when the U decreases, the optical gap for the case of a frozen U (blue curve) display mainly a modulation around an average value, as expected from the dynamical Franz-Keldysh effect.

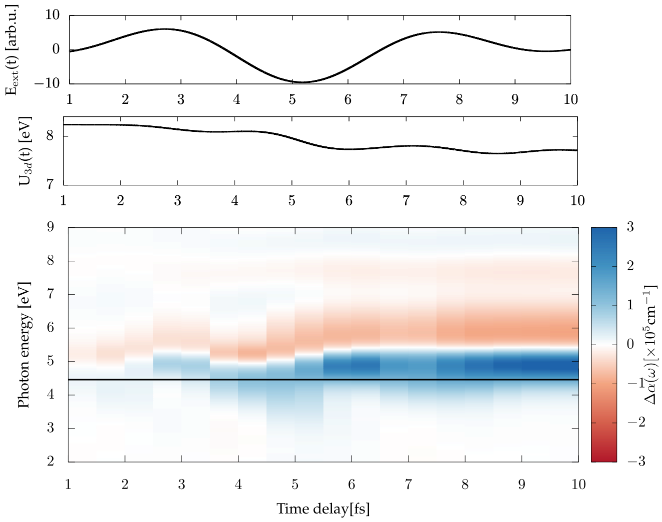


Supplementary Fig. 2. Variation of the optical gap, extracted from the calculated transient absorption, for both dynamical and frozen U cases.

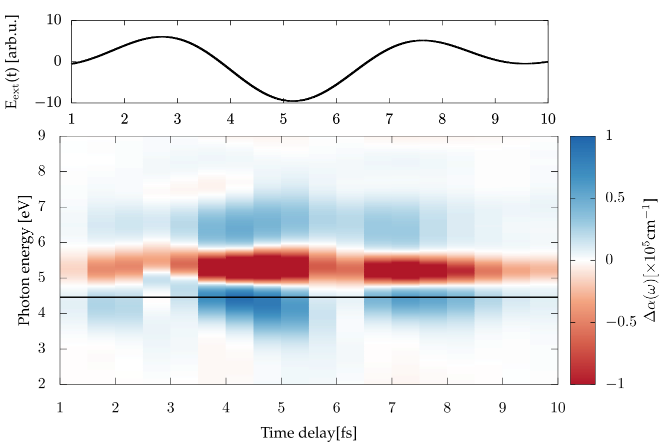
TRANSIENT ABSORPTION MAP FOR A 2100NM WAVELENGTH DRIVER

In order to verify if the that the reported effects are general, we also performed simulation for a driver of 2100nm, for a shorter pulse of 5 fs FWHM and for an intensity of the external field in matter of $I_0 = 5 \times 10^{11}$W.cm^{-2}. Due a shorter time propagation after each kick, we used here a Gaussian broadening of 0.5 eV. The results are shown in Supplemental Fig. 3 and Fig. 4. Qualitatively, the findings are very similar. For the dynamical U, (Supplemental Fig. 3), the dominant feature is the band-gap renormalization, whereas for the frozen U, the band-gap renormalization is closer to the DFKE, even if for a one cycle pulse, the transient absorption map is hard to interpret.

The change in the transient absorption map is much more important in the dynamical U case, consistent with the findings presented in the main text for a driver of 800 nm driving wavelength. For the frozen U case, we observe a



Supplementary Fig. 3. Calculated transient absorption of NiO for a driving field of wavelength 2100 nm. The top panel shows the time profile of the external driving electric field $\vec{E}_{\text{ext}}(t)$. The middle panel shows the corresponding time-evolution of the on-site U for the $3d$ orbitals of Ni. A similar variation is obtained for the $2p$ orbitals of oxygen atoms. The bottom panel shows the calculated transient absorption of NiO. The dashed line indicates the calculated ground-state gap of NiO.



Supplementary Fig. 4. The same as Fig. 3, but keeping the Hubbard U fixed during the time evolution.

clear increase of the absorption below the bandgap oscillating at twice the laser frequency. We note that as the photon energy is much smaller here (0.58 eV), the area in which the absorption decreases at the bandgap edge is expected to be much smaller, as found here in Fig. 4.

Supplementary References

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