Transient and remnant electronic states

June 7, 2019

1 Transient and remnant electronic states (Dynamical localization)

In this section we consider behavior of the correlated system during and shortly after ultrashort pump pulse, and analyze creation of the dynamic Mott insulating state. We choose $\lambda = 3000$ nm pulse in order to maintain high enough vector potential, keep field strength reasonable for experimental applications (low enough not to burn a sample), and fit a few-cycle pulse into calculation tilmestep window. We apply a short linearly polarized gaussian pulse with central frequency $\omega = 0.413$ eV (wavelength $\lambda = 3000$ nm), pulse duration (Full Width at half-maximum, FWHM) d =7.7 fs, total simulation time is 33 fs. The pulse bandwidth (FWHM) is 0.24 eV.

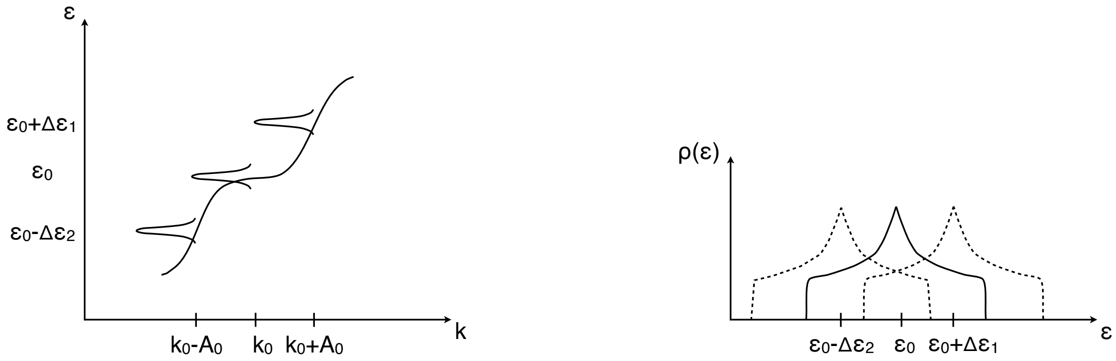
The peak value of the vector potential is varied in a range $A_{max} = 0.46 - 3.68$, that corresponds to electric fields of $5 \times 10^8 - 4 \times 10^9$ V/m for lattice constant 3.78 Åfor CuO_2 plane of La cuprate. These values correspond to $0.23 - 1.8$ mJ/cm^2 pulse fluence for $150 \times 200\mu$m focus size. The pulse wave vector is parallel to [001] direction, so electric field vector is in the CuO_2 plane.

@HA As I have emphasized when we discussed in Hamburg, you have to characterize the field strength in terms of the Bessel function for the renormalized band width as a function of @. "Weak" and "strong" fields can be characterized only in terms of that. Then you can talk about the renormalized U/W, where W is the renormalized bandwidth. @EG, VV we are renormalizing the bandwidth, and the effective U is being changed in the units of renormalized bandwidth, keeping it's "bare" value constant (i.e. it is not changing w.r.t. other characteristic energies of the model, e.g. pulse frequency).

In this section we choose laser's linear polarization to be parallel to the lattice diagonal ([110] direction), that bring us to the situation similar to hypercube-lattice, studied e.g. in (cite !! Tsuji et al), but here with the 2D dispersion that includes van-hove singularity and sharp band edges. We have also considered the case of electric field along one of the lattice axes ([100]). Then the model becomes quasi-one-dimensional, giving results similar to ones discussed in Ref. [**?**] with no population inversion, no sign-change of the U_{eff}. The benchmark of our method with the exact diagonalization of a one-dimensional finite chain used in Ref. [**?**] is shown in Appendix. We also considered the case of circularly polarized short pulse, and no qualitative difference in dynamical localization processes with respect to [110] polarization have been found. @HA Really? Hard to understand. Anyway, you have said that you'll have another paper for the circularly polarized light, haven't you? @VV All circularly polarized pulses are out of scope of the current paper.

The shape of the pulse vector potential is given by Eq. **??** with $t_0 = 16.4$ fs, the profile is shown in Fig. 2. Figure 3 displays the time-resolved photoemission intensity, Eq.(**??**). For low field strengths [Fig.3(a,b)] we can see the spectral weight transfers from van Hove singularity to lower Hubbard band (LHB), situated at $-U/2 = -1.25$ eV.

@HA I don't understand this. Take t (nearest-neighbor hopping) as the unit of energy. Then indicate the energy position of the vH singularity. @HA As the initial state, where is the peak



(a) (b)

Figure 1: Schematic band structure of square lattice. Pulse with vector potential amplitude A_0 drive the electrons with initial momentum $\mathbf{k_0}$ (zero?) from high-density region ε_0 to regions with momentum $\mathbf{k_0} \pm \mathbf{A_0}$ and energy $\varepsilon_0 - \Delta\varepsilon_1$ and $\varepsilon_0 + \Delta\varepsilon_2$. @HA I don't understand this figure AT ALL! @VV Maximal intensity of Hubbard bands appears at certain values of the vector potential, namely at zeros of the zero order Bessel function ($J_0(A)$). This effect could also be called dynamical localization, because renormalizing the hoppings we increase effective Coulomb U; and at $J_0(A) = 0$ we have the perfect Mott insulator.

of the population density located? @HA The color code around t (time) = 0 is no clear. Another quite important factor is, you are dealing with the half-filled case, right? @HA Then, initially, the density should be electron-hole symmetric about E=0, but this is not clear, either. @EG we changed the figures (that actually suffered from imperfectness of the Fourier transform applied, leading e.g. to particle-hole asymmetry at t=0) from population density (not used anymore) to simulated photoemission spectra (with probe FWHM = 2.5 fs)

Above a certain field strength the upper Hubbard band (UHB) starts to be populated (Fig.3(d)). At even higher fields we can see the Mott gap opening after some time, accompanied by an oscillatory behavior of the population of LHB and UHB (Fig.3(e)).

Before 6 fs the system stays metallic, demonstrating van Hove singularity at the Fermi energy and emitting at $\omega = 0.5$ eV, see Fig. 4, lower panel. Please note, that despite the pulse central frequency is 0.413 eV, and the current's fundamental frequency is 0.5 eV, and the third harmonic of current is at 1.5 eV (1.2 times higher).

Around 7 fs a gap opens due to dynamical localization. The details of the transient processes for responses for pulses can be analyzed in terms of the current (Eq. **??**), for high field strengths, e.g. $E_{max} = 8 \times 10^9$ V/m. If we plot the Gabor transform (a window Fourier transform) of the current in Fig. 4, lower panel, we can see that shows dynamics of higher harmonic generation (HHG) process. The corresponding spectrum is shown in Fig. 4, middle panel. @HA Is the Gabor transform really meaningful for such a short time interval (1.2 fs, which is much shorter than the pulse period as displayed @EG the width of Gabor window was 3 fs in FWHM, actual time window is wider due to it's Gaussian nature,so it makes sense. Also, we checked ordinary FT over all time, it shows the same peak positions of current in frequency space.

We can then see in Fig. 4 that the HHG occurs concomitantly with the gap opening in the spectrum. Since this occurs when the field is high enough (for $E_{max} = 8 \times 10^9$ V/m in the present case), the tunneling process between Hubbard bands is considered to start, leading to the generation of higher harmonics. Around 7 fs the third harmonic starting to be generated,

In general, as it has been checked for longer pulses, after few periods of the driving field (e.g. two periods, or 10 fs for the $\omega = 0.827$ eV pulse) the double occupancy saturates to some constant value (see Fig. **??** in Sec. **??**), then no more current is induced in the system, arrived at the dynamical Mott insulating state (cite Martin), at least for reasonable field strengths.

The total energy and its time derivative for various pulse fluencies are shown in Fig. 5. Comparing peak values of dE_{tot}/dt for different pulse fluencies we can conclude (!! to Olga

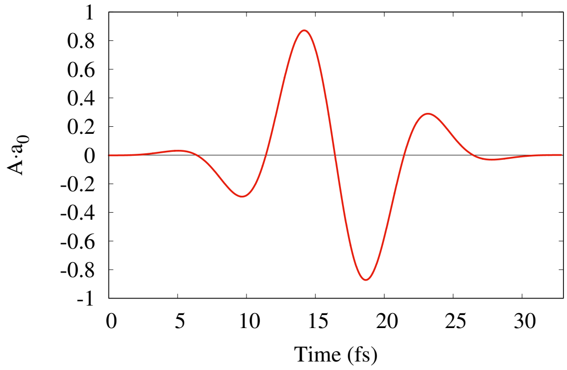


Figure 2: Shape of the vector potential for a gaussian pulse with a central frequency ω = 0.413 eV, duration (FWHM) d = 7.7 fs, and intensity $A_{max} \times a_0 = 1$.

and Misha: I_p analysis !!). Even before the phase transition to the HHG regime the maximal derivative $dE_{\rm tot}/dt$ is being reached earlier for stronger fields. @I don't see this. For the field 2×10^{10}, the peak seem to appear exactly when you have the HHG.

@HA Another question is what is the physical meaning of the derivative?

Some notes on $dE_{\rm tot}/dt$. $dE_{\rm tot}/dt|_{t=t_i} = \alpha e^{-\frac{2}{3}\frac{(2I_p)^{3/2}}{E(t_i)}}$, where α is unknown coefficient, I_p is energy gap (and should be proportional to Hubbard U?), $E(t_i)$ is the electric field strength at time t_i. @HA How have you derived this expression? Choosing the first and the second maxima of $dE_{\rm tot}/dt$ at times $t = t_1$ and $t = t_2$ we can get rid of α and find I_p. @HA I don't understand this sentence at all. Let's note $\dot{W}_i = dE_{\rm tot}/dt|_{t=t_i}$ and $E_i = E(t_i)$. Then $\frac{\dot{W}_1}{\dot{W}_2} = e^{-\frac{2}{3}\left(\frac{(2I_p)^{3/2}}{E_1} - \frac{(2I_p)^{3/2}}{E_2}\right)}$, and $I_p = \frac{1}{2}\left(\frac{-\frac{3}{2} ln\frac{\dot{W}_1}{\dot{W}_2}}{E_1^{-1} - E_2^{-1}}\right)^{2/3}$. @HA So what you wanted to say with this?

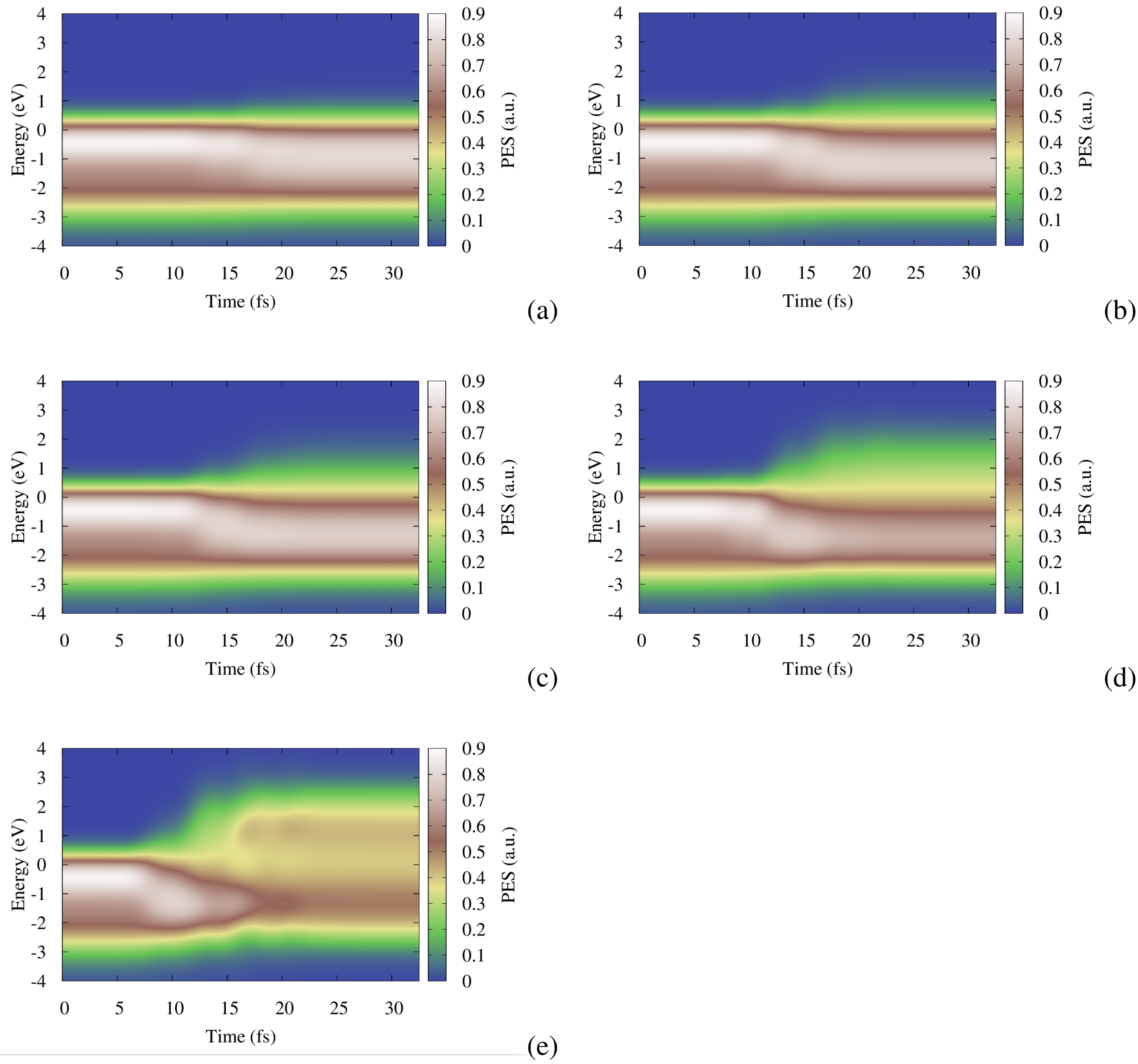


Figure 3: Time-dependent PES for gaussian pump pulse with central frequency $\omega = 0.413$ eV, $FWHM = 7.7$ fs, and $E_{max} = 5 * 10^8$V/m (a), $8 * 10^8$V/m (b), $1 * 10^9$V/m (c), $2 * 10^9$V/m (d), $5 * 10^9$V/m (e) with polarization along [110] direction. The probe pulse duration (FWHM) is 2.53 fs.

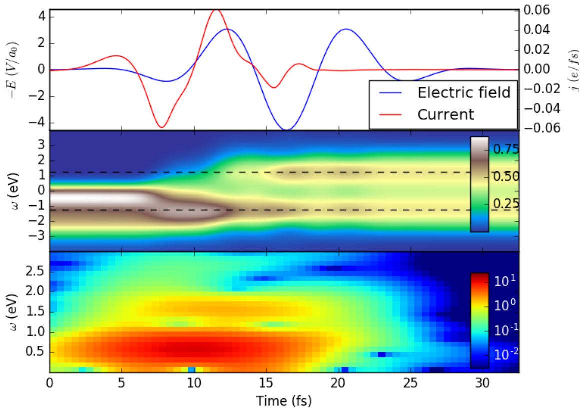


Figure 4: Upper panel: A gaussian pulse (with the electric field strength displayed in blue) at a central frequency $\omega = 0.413$ eV, width $d = 7.7$ fs, and intensity $E_{\max} = 8 \times 10^9$ V/m, polarization along [110]. The generated current is shown in red. Middle panel: Time-dependent PES, dash lines denotes positions of Hubbard bands $\pm U/2$. Lower panel: Gabor transform of the current.

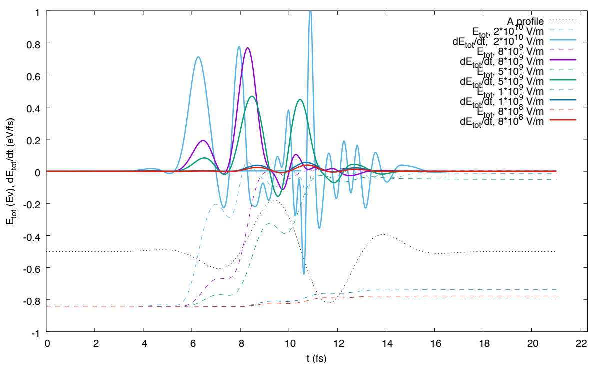


Figure 5: Total energy, E_{tot} (dashed curves), and its time derivative (solid curves) for a gaussian pulse with central frequency $\omega = 0.827$ eV, width $d = 3.8$ fs, and polarization along [110] direction, for various intensities $E_{max} = 8 \times 10^8, ..., 2 \times 10^{10}$ V/m. Black dotted line represents the pulse shape.