On the role of resonances in photoionization of metal clusters

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Abstract. We analyze electron emission from irradiated clusters by means of time-dependent density-functional theory (TDDFT) in real time. We focus on photo-electron spectra (PES) which deliver an invaluable tool to explore static and dynamical properties of irradiated species. We discuss, in particular, the role of resonances in the PES once the laser frequency is below the emission threshold which implies multiphoton processes. We show that the resonances in the electronic spectrum lead to the occurrence of several peaks in the PES and also strongly affect the standard scaling relations between ionization and the number of required photons for electronic emission.

1. Introduction

The analysis of electron emission from irradiated atoms, molecules and clusters is a key tool to explore both static and dynamical properties of these systems. This brings an invaluable complement to optical absorption measurements, for an overview on clusters see [1, 2, 3, 4]. Short laser pulses offer here a most versatile tool to consider various irradiation scenarios. A first useful indicator is the mere net ionization yield while photo-electron spectroscopy (PES) which measures the distribution of the kinetic energy of emitted electrons, appears as an especially valuable method for delivering information on the energies of occupied single-electron states [5, 6, 7, 8, 9]. Photo-electron angular distribution (PAD) offer an even more involved signal often combined with PES, then providing double-differential cross-sections (energy- and angular-resolved) [10, 11, 12]. The PES nevertheless remains a quite valuable technique by itself. In the one-photon regime, PES provides an image of the sequence of occupied single-particle levels, an aspect which was explored in the early days of cluster physics in cluster anions [8]. Resolving deeper levels requires higher frequencies than visible ones, and hence one finds also UV [7] and X-ray PES [13]. The great variety of modern coherent light sources in large ranges of frequency, intensity and pulse length, nowadays even allow for time-resolved studies in molecules [14, 15, 16] or for deeply lying core states of atoms embedded in a material [17]. Studies of PES are thus now found in all areas of molecular physics, from atoms over simple molecules [18] to complex systems such as clusters [19] and organic molecules [20] and this allows the investigation of both structural and dynamical properties of these various atomic systems.

The general need for a robust theoretical tool of analysis of PES in various dynamical regimes is thus obvious. The traditional approaches relying on (multi-)photon perturbation theory...
[21] are naturally limited to low laser intensities. Moreover, they require a good knowledge of the continuum states for the outgoing electrons thus becoming extremely inconvenient for systems with low or without any symmetry. In clusters, time-dependent density-functional theory (TDDFT) at the level of the local-density approximation (TDLDA) directly propagated in the time domain has proven to be a robust, reliable, and efficient method, see e.g. [22, 23, 24]. When using absorbing boundary conditions [25, 26] and when augmented by a self-interaction correction (SIC) [27], TDLDA provides a convenient framework for analyzing electronic emission properties. A simple but efficient procedure for calculating PES based on TDLDA, was thus proposed a few years ago in [28, 29] and has been taken up recently in [30] for a combined analysis of PES and PAD. The technique of [28, 29] has been applied to a variety of systems with possibly large electron emission induced by still moderate laser intensities such as free metal clusters [31, 32, 33], dimer molecules [30], and deposited clusters [34]. It has been recently extended to larger laser intensities [33].

The aim of this paper is to discuss some aspects of PES dynamics in the simple case of small sodium clusters but exploring in detail the dynamical mechanisms at work, and how they affect the actual PES. The paper is organized as follows. Section 2 provides a short summary of the formal framework discussing, in particular, the evaluation of PES. Some illustrative results are discussed in section 3 focusing on the role of resonances in PES.

2. Theoretical framework
Cluster dynamics is described by real-time TDDFT at the level of the time-dependent local-density approximation (TDLDA) using the exchange-correlation functional of [35]. In order to properly analyze electron emission, especially at low energy (namely close to threshold emission), one has to augment TDLDA with a self-interaction correction (SIC) [27] which is chosen here at the level of average-density self-interaction correction (ADSIC), following [36]. The coupling to the ions is mediated by soft local pseudo-potentials for Na [37]. The ionic background is kept frozen which is a legitimate approximation as we consider short laser pulses and direct electron emission proceeding rather fast. From a practical point of view, real-time TDLDA is realized with standard coordinate-space techniques. Emission is basically recorded by means of absorbing boundary conditions. Laser excitation is described by an external classical dipole laser field (as the used wavelengths are much larger than the system size) with a finite temporal envelope \( \propto \sin(\pi t/T_{\text{pulse}})^2 \). For formal and technical details, we refer the reader to [26, 38].

Observables from electron emission deserve somewhat more detail. As already pointed out, the description of electron emission requires absorbing boundary conditions. The absorption is performed after each time step according to the following scheme. We first perform a standard time step from \( t \) to \( t + \delta t \), \( \phi(r,t) \rightarrow \tilde{\phi}(r) \). We then apply a mask function to the preliminary wavefunction \( \tilde{\phi} \) to obtain \( \phi(r,t+\delta t) = M(r)\tilde{\phi}(r) \). This allows to gradually remove any flow towards the bounds. In the following, we have used a spherically symmetric mask profile

\[
M = \cos\left(\frac{|r| - R_{\text{in}}}{R_{\text{out}} - R_{\text{in}}} \frac{\pi}{2}\right)^{1/8}
\]  

which is active in an absorbing margin \( R_{\text{in}} < |r| < R_{\text{out}} \). The spherical profile turns out to minimize gridding artifacts when computing angular distributions [39]. For a detailed discussion of absorbing bounds and optimal choice of absorption profiles, see [40].

The absorbing bounds practically reduce in a gradual manner the norm of the wavefunctions, which mimics the dynamical ionization of the system. The net ionization, namely the number of escaping electrons is directly accessible from the single-particle norms as

\[
N_{\text{esc}} = N_{\text{el}}(t=0) - \sum_\alpha (\phi_\alpha | \phi_\alpha).
\]
The PES require a bit more effort. They are evaluated for each state separately from the phase oscillations of the single-electron wavefunction at an analyzing point \( r_{\text{meas}} \) located near the absorbing bounds, just a bit before the absorbing layer [28]. The single-electron wavefunctions \( \phi_\alpha(r_{\text{meas}}, t) \) are thus recorded in time at measuring point(s), and then time-frequency Fourier transformed to \( \tilde{\phi}_\alpha(r_{\text{meas}}, \omega) \). This delivers the total PES spectrum as

\[
P(\omega) \propto \sum_\alpha |\tilde{\phi}_\alpha(r_{\text{meas}}, \omega)|^2
\]

which is translated to a kinetic-energy spectrum of emitted electrons by identifying \( \hbar \omega = \epsilon_{\text{kin}} \). This simple identification is possible thanks to the absorbing bounds which leave only outgoing waves in its vicinity (thus establishing a unique relation between frequency and momentum). High laser intensities require slight modifications of this recipe [33]. An energy- (or state-)resolved PES is, of course, immediately attained by skipping the summation on states in Eq. (3).

3. Results

3.1. A simple example in the one-photon regime

Basic computed features of PES for clusters have been collected in a series of papers [28, 29, 31, 32]. We briefly summarize here the major results. PES for one-photon processes provide a direct map of the ground-state single-particle (s.p.) spectra as used since long in experiments [8, 9]. This regime is easily attained once the laser frequency is larger than the span of the s.p. spectra to be measured and once the excitation is sufficiently gentle to avoid perturbation of the ground-state of the system. Larger intensities blur the signal from PES and we shall not consider them here. Lower frequencies quickly lead into the regime of multiphoton ionization (MPI). In that case, one can spot multiple copies of PES peaks, each of them standing for a certain photon number \( \nu \). The envelope of this multiple peak structure exponentially decreases with kinetic energy \( \epsilon_{\text{kin}} \) of the outgoing electrons. Finally for even higher intensities in the MPI regime, the signal turns to pure exponential decrease which can easily be misinterpreted as thermal emission [32].

We illustrate the simple one-photon case (laser frequency above ionization threshold) in Figure 1. The system Na\(_8\) is treated in the soft jellium approximation [41, 42] which is fully justified for such a simple metal. The jellium parameters have been chosen as: Wigner-Seitz radius \( r_s = 3.65 \, \text{a}_0 \), jellium surface width \( \sigma = 1 \, \text{a}_0 \). The laser frequency has been chosen as \( \omega_{\text{las}} = 0.9 \, \text{Ry} \) namely well above the ionization potential (IP) of the system (0.3 Ry), thus safely into the one-photon ionization regime. The figure nicely shows how the PES directly maps the single-electron spectrum. It provides a straightforward proof of the capability of the PES to access structural properties of a system. The case is nevertheless especially forgiving. Indeed, an anomalous behaviour has been noticed for frequencies below the IP and we shall now discuss that point in more detail. The following considerations are restricted to the Na\(_8\) cluster (with explicit ionic structure). However, the effect was also observed for other Na clusters. For the sake of simplicity, the discussion is focused on the (0, 0, 1)-orientation of the cluster, where the ionic rings are located in the \((x, y)\)-plane (see Figure 2 for details). The laser will be always polarized along the \(z\)-axis.

3.2. Ionization dynamics

The cluster is excited by a laser pulse of \( \omega_{\text{las}} = 0.08 \, \text{Ry} \), \( T_{\text{pulse}} = 120 \, \text{fs} \), and \( I = 3.1 \times 10^{11} \, \text{W/cm}^2 \). We recall that with explicit ionic background the Na\(_8\) clusters displays three occupied levels (each furthermore doubly degenerate because of spin). The associated energies are \( \epsilon_{1s} = -0.42 \), \( \epsilon_{1px,y} = -0.33 \) and \( \epsilon_{1pz} = -0.31 \, \text{Ry} \). For \( \omega_{\text{las}} = 0.08 \, \text{Ry} \) (see right panel of Figure 1). The
Figure 1: PES (left panel) computed for the Na\textsubscript{8} cluster in jellium approximation and in the one-photon regime (\(\omega_{\text{laser}} > \text{IP}\)). The laser frequency is \(\omega_{\text{laser}} = 0.9\ \text{Ry}\), the intensity \(I = 6.9 \times 10^{13}\ \text{W/cm}^2\) and the pulse duration \(T_{\text{pulse}} = 60\ \text{fs}\). The net ionization is \(N_{\text{esc}} = 0.003\). The right panel recalls the ionic and electronic spectrum of Na\textsubscript{8} as computed in jellium approximation as well as with full ionic structure.

1s state needs six, the 1p\textsubscript{x,y} states five, and the 1p\textsubscript{z} state four (which is very close to the threshold) photons to be ionized. The total number of emitted electrons is plotted in Figure 2 as a function of time. Although the laser stops at \(t = 120\ \text{fs}\), the cluster surprisingly continues to emit electrons. This is a clear non-perturbative behaviour.

A closer look at the dipole signal \(D_z(t)\) in the direction of the laser polarization, see the left panel in Figure 3, reveals what happens. For \(t < 120\ \text{fs}\), the dipole follows the electric field as expected. For \(t > 120\ \text{fs}\), however, the electronic cloud continues to oscillate, but faster than before. Furthermore, the post-pulse oscillations seem to be very “clean”, i.e., with one single

Figure 2: Typical emission profile for a laser frequency below the ionization threshold. The laser pulse begins at \(t = 0\) and ends at \(t = 120\ \text{fs}\). Significant electron emission starts not until \(t \approx 50\ \text{fs}\).
Figure 3: Left: Dipole signal $D_z$ in the direction of the laser polarization as a function of time. Right: Fourier transformation $|\tilde{D}_z(\omega)|$ of the dipole during ($t < 120$ fs, thin curve) and after the pulse ($t > 120$ fs, thick line).

frequency only. The spectral analysis of the signal from $t = 120 - 250$ fs in the right panel confirms that. The electronic cloud oscillates with $\omega_r = 0.237$ Ry after the pulse is over. During the pulse, the spectrum is mixed: the main frequency is the laser frequency, but $\omega_r$ also appears.

This analysis provides the following possible explanation to the phenomenon: the laser pulse has excited a dipole resonance of the cluster. The resonance is triggered by absorption of three photons, since $\omega_r \approx 3\omega_{\text{las}}$. Figure 4 shows the dipole strength $S_D(\omega) \sim \Im \{\tilde{D}_z(\omega)\}$ of the dipole signal. The dipole response has been obtained by an initial boost of all s.p. wavefunctions $|24, 26|$, $\varphi_\alpha(r, 0) \rightarrow e^{i\vec{p}_{\text{boost}} \cdot \vec{r}} \varphi_\alpha(r, 0)$.

The strength function is then directly related to the photoabsorption cross-section [26]. The predominating excitation mode is, of course, the plasmon at $\omega_{\text{pl}} = 0.187$ Ry. However, besides the plasmon, one can observe several 1ph excitations and resonances, all of them below the IP. Nevertheless, close to the laser frequency, there is only one stronger mode at 0.106 Ry related to 1ph excitations from the 1s state to the 1p states. This mode is too far away for the laser

Figure 4: Dipole strength function $S_D \sim \Im \{\tilde{D}_z(\omega)\}$ of Na$_8$ with explicit ionic background, in the direction of the laser polarization (denoted here by $z$). The plasmon frequency is $\omega_{\text{pl}} = 0.187$ Ry, the ionization potential at IP = 0.315 Ry.
frequency. But the system can still be excited by multiphoton absorption. For laser frequencies below the IP, the laser can therefore very likely hit a resonance. However, it has to be noted that a dipole resonance can only be induced by an odd number of photons. The present case corresponds to three photons. Note that it is also sufficient that the energy $3\omega_{\text{las}}$ lies just in the close vicinity of the resonance energy $\omega_r$, an exact matching is not compulsory.

3.3. Photo-electron spectra

It is now interesting to analyze the photoemission observables in more detail. Figure 5 shows the PES (in logarithmic scale) for different time spans: during and after the pulse, and for the full calculated time. The full PES (line at the top of the panel) exhibits pronounced peaks with certain distance to each other. At first glance, these peaks just resemble MPI peaks. The four-photon peak of the $1p_z$ state should appear closely above the IP, but has not fully passed the threshold. This is due to the positive charge of the residual cluster which shifts the whole spectrum.

More interesting is the shape of the PES after the pulse (line in the middle of the panel). Because of numerical artifacts of the Fourier transformation, it shows more noise. Again, the PES exhibits sharp peaks, but many less. These peaks could finally be identified as multi-resonance peaks. Their energy exactly corresponds to the s.p. energy plus multiples of the resonance frequency, here $\epsilon_i + \mu \omega_r$, with $\mu = 2, 3$. The last relations have been marked with solid vertical lines. Due to the residual cluster charge, the s.p. spectrum has been shifted by about $\delta\epsilon = -0.012 \text{Ry}$ in order to achieve better match. The first visible peak at $\epsilon_{\text{kin}} = 0.035 \text{Ry}$ corresponds to the doubly excited ($\mu = 2$) $1s$ state, the following peaks to the $1p_{x,y}$ and $1p_z$ states. The triply excited $1s$ peak disappears behind the noise, but the $1p$ states are still present. No MPI peaks appear in the spectrum after the pulse. Hence, the important energy for the post-pulse emission is the resonance energy $\omega_r = 0.237 \text{Ry}$ and not $\omega_{\text{las}} = 0.08 \text{Ry}$. The full PES,
finally, is a superposition of MPI peaks, $\epsilon_i + \nu \omega_{\text{las}}$ (also shifted by $\delta \epsilon$ and marked with black dashed lines), and the multi-resonance peaks. The resonance-enhanced emission already starts during the pulse (line at the bottom of the panel) and continues after the laser excitation has stopped.

### 3.4. Impact of laser intensity

We now turn to the impact of laser intensity on actual electron emission in Na$_8$. Figure 6 shows the total number of emitted electrons $N_{\text{esc}}$ as a function of the laser intensity in the range $I \sim 10^{10} - 10^{12}$ W/cm$^2$. The frequency $\omega_{\text{las}} = 0.08$ Ry and the pulse length $T_{\text{pulse}} = 120$ fs have been kept constant. All curves are normalized to unity. The results indicate that the post-pulse emission profile sensitively depends on the intensity: for intensities below $3.1 \times 10^{11}$ W/cm$^2$, all profiles look very similar, typical post-pulse emission occurs. In contrast, for intensities $I \gtrsim 5 \times 10^{11}$ W/cm$^2$ the post-pulse emission becomes weaker and finally disappears. It is worth noting that the ionization is very large for such intensities, $N_{\text{esc}} = 0.2 - 1.4$ for $I \geq 5.6 \times 10^{11}$ W/cm$^2$. The dipole spectrum of Figure 4 is then blue-shifted during the emission process and resonances are broadened. Consequently, several resonances can be excited at once, as can be seen from the right panel of Figure 6 for $I = 5.6 \times 10^{11}$ W/cm$^2$. In this case, two close frequencies are interfering. However, at large intensities one could also imagine that the blue shift of the spectrum has the consequence that no resonance is induced. All in all, the conclusion stemming from these various calculations is that the emission behaviour is hard to predict in this intensity range.

The number of emitted electrons $N_{\text{esc}}$ for the different intensities is shown in Figure 7 in double-logarithmic scale, during (left panel) and after the pulse (right). For a pure multiphoton process, one expects that $N_{\text{esc}} \sim I^n$, where $n$ is at least the minimum number of photons required for ionization, derived from perturbation theory. Actually, the $1p_z$ state needs four photons to be ionized. However, according to the PES in Figure 5 the four-photon peak has not fully passed the threshold. Moreover, the $1s$ state needs six photons while for the $1p_{x,y}$ states already five photons are sufficient. A value between $6 > n > 4$ and close to five should thus be reasonable if emission is dominated by multiphoton processes. According to Figure 5, this is not the case.
Instead, the number of emitted electrons is proportional $\sim I^p$, with $p \approx 3$. This applies for the emission during and after the pulse, as long as the laser intensity is weak. For higher intensities, the post-pulse emission disappears and $p \approx 1$.

The consequence is that multiphoton processes play only a minor role at weak intensities and the emission is dominated by the resonance at $\omega_r$. Otherwise, one would have expected a much higher power $p$. Instead, the power corresponds to the number of photons required to induce the resonance. It is interesting to note that the number $\mu$ of resonance energies $\omega_r$ needed for ionization does not seem to be important, either.

As already mentioned, the presented phenomenon is very sensitive to intensity variation, since for too large intensities, the emission could be on- or off-resonant, or several resonances could be hit due to dynamical shifts of the dipole spectrum. Finally, it should be noted, however, that multiple resonances can also be induced by a variation of the laser frequency. The laser frequency could lie next to two or more resonances which may be excited by one photon. One could also imagine a situation where $\omega_{\text{las}}$ is close to an excitation mode and a multiple of the photon energy as well. In all these cases, several excitation modes interfere, the dipole signal appears “dirtier” and the PES should be more puzzling. Moreover, it becomes difficult to distinguish between multiphoton and multi-resonance ionization. In this respect, the chosen frequency $\omega_{\text{las}} = 0.08\text{ Ry}$ is a kind of exceptional case. The first photon energy is below any excitation, the second photon is in the gap between the first 1ph excitation and the plasmon, and the third photon hits almost exactly the resonance whose energy is, in turn, different from the photon energy.

4. Conclusions and perspectives

We have briefly reviewed the theoretical description of photo-electron spectra (PES) focusing on the special case of simple metal clusters. The numerical evaluation relies on a Cartesian coordinate-space grid with absorbing boundary conditions cutting a dynamically active sphere into the 3D grid. The PES are obtained from recording the time evolution of each single-electron wavefunction at a few selected measuring point near the absorbing bounds. This technique adds only a small overhead to the underlying TDDFT propagation thus allowing large scale studies.

We have discussed the basic features of electron emission in the simple case of Na$_8$. The PES outcome depends very much on the dynamical conditions mainly set by laser frequency and intensity. The one-photon regime of ionization (low intensity, frequency larger than the lowest bound state) is particularly clean allowing a detailed analysis of the single-particle states in the cluster ground-state. It thus provides a direct and detailed structural information on the...
system as known since long. Higher intensities and/or lower frequencies lead into the regime of multiphoton ionization which turns out to be much more involved. We have focused here on the lower end of intensities, where one still stays in the frequency dominated regime and where one can count photons and sees them acting. This, in particular, shows up in the PES as repeated peaks separated by the photon frequency. A detailed discussion of that case further shows that resonances which can be directly identified from the PES, are also coming into play. Nevertheless, the resulting picture becomes very involved, basically blurring the original simple idea of the single-photon regime. No clear systematics emerges and each case is likely to be considered separately and in detail.

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