



Far Off Equilibrium Dynamics in Clusters and Molecules

Phuong Mai Dinh^{1*}, Marc Vincendon¹, Jordan Heraud¹, Eric Surraud¹ and Paul-Gerhard Reinhard²

¹ Laboratoire de Physique Théorique, Université de Toulouse, CNRS, UPS, Toulouse, France, ² Institut für Theoretische Physik, Universität Erlangen, Erlangen, Germany

This brief review illustrates on a few typical applications fully fledged dynamical simulations of finite electronic systems (atoms, molecules, clusters) using time-dependent density-functional theory (TDDFT). It concentrates on aspects which are different from nuclear applications. These are: the correct handling of electron emission, the self-interaction correction, the enormous versatility of laser excitation to probe systems properties, and with it the exploitation of detailed observables of electron emission as photo-electron angular distributions and photo-electron spectra (PES). Finally, we demonstrate the impact of electronic dissipation putting question marks on the reliability of TDDFT simulations over long times.

Keywords: time-dependent density-functional theory, molecules, electron emission, photo-electron distributions, dissipation

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Artur Polls,
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*Correspondence:

Phuong Mai Dinh
dinh@irsamc.ups-tlse.fr

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1. INTRODUCTION

Far-off equilibrium dynamics in quantum many-body systems is since numerous decades a challenging task much studied in experimental and theoretical investigations. In the realm of finite electronic systems, the availability of versatile laser pulses has given way to detailed analysis of the response of clusters and molecules [1–11]. In particular it allows one to explore the properties of electrons emitted after irradiation in correlation with the laser pulse. This has led to detailed analysis of angular- and energy-resolved electron spectra, e.g., their angular distributions and photoelectron spectra (PES) or both simultaneously as angular resolved PES (ARPES) recorded via Velocity Map Imaging techniques [12]. The theoretical description of such far-off equilibrium situations is also demanding. It requires large phase spaces and must cover vastly different time scales ranging from basic processes of excitation over collisional redistribution with subsequent relaxation to ionic motion and possible coupling to environment. They thus require dedicated fully time-dependent approaches, such as the widely used time-dependent density functional theory (TDDFT), often combined with molecular dynamics for the description of ionic motion (see e.g., [13]). This approach is both versatile and robust and allows one to simulate numerous dynamical scenarios with high degree of accuracy. The electronic part, TDDFT, is formally exactly the same as time-dependent Hartree-Fock (TDHF) in the nuclear domain [14]. The difference lies mainly in energy functionals which are constructed by very different strategies, see Dreizler and Gross [15] for electronic systems and Bender et al. [16] for nuclei.

In a typical laser irradiation, electrons immediately react to the external electro-magnetic field driving the system quickly far-off equilibrium long before ionic motion and thermal relaxation processes set on [17]. Thus, the system fully remains in the quantum regime for quite a while, as seen in recent experiments, exhibiting clear quantum and thermal pattern in photoelectron spectra [18–20]. The crucial first stage, i.e., the doorway to any photo-reaction, is predominantly

and particularly well for metal clusters. It is not applicable to situations with fragmented electron density and to bulk systems (the latter because the particle numbers N_σ grow infinite).

2.3. Observables

The KS equations are solved in real time on a 3D grid with standard techniques [13, 17, 37]. The predominantly local structure of the KS Hamiltonian allows one to use the very efficient time-splitting scheme for time propagation [38]. The Coulomb field is computed with successive over-relaxation [39] or Fast Fourier Transform techniques [37, 40]. To describe ionization dynamics, we use absorbing boundary conditions [17], which, when properly optimized [41], gently remove outgoing electron flow at the boundaries of the numerical grid. This is a crucial ingredient which is so far not much used in nuclear dynamics. But it is essential in atoms/molecules where observables from electron emission (explained below) are very important.

The analysis of electron dynamics is performed through a set of well-established observables [10, 17, 42, 43] that we briefly recall here. As the laser field couples to the electronic dipole, dipole response is the most prominent observable in many studies [10, 13, 42, 44]. We compute the dipole moment of electrons (with respect to ionic background) from the electronic density as:

$$\mathbf{D}(t) = \int d^3\mathbf{r} \mathbf{r} \varrho(\mathbf{r}, t) \quad . \quad (5)$$

Spectral properties are then obtained from the time-frequency Fourier transform $\mathbf{D}(t) \rightarrow \tilde{\mathbf{D}}(\omega)$ after unfolding the spectrum of the exciting pulse [44]. Spectral analysis becomes particularly simple for excitation by an instantaneous dipole boost. The dipole strength becomes then $\Im\{\tilde{\mathbf{D}}(\omega)\}$ and the dipole power spectrum $|\tilde{\mathbf{D}}(\omega)|^2$.

For sufficiently strong excitation, electron emission becomes important. It can be analyzed at various levels of sophistication thanks to the use of absorbing boundary conditions mentioned above. Simplest is the global measure, the total ionization which can be computed from the given density as

$$N_{\text{esc}}(t) = N - \int d^3\mathbf{r} \varrho(\mathbf{r}, t) \quad (6)$$

where N is the initial electron number.

More detailed observables are Photo-Electron Spectra (PES), obtained from measuring the distribution of kinetic energies of the emitted electrons, or their angular distribution. Measuring both simultaneously yields Angular-Resolved PES (ARPES) for which often valuable experimental data exist to compare with [43]. The strategy to compute PES consists in defining a set of “measuring points” \mathbf{r}_M near the absorbing boundaries, and to record the time evolution of s.p. wave functions $\varphi_n(\mathbf{r}_M, t)$ at these points. The information thus gathered can be post-processed in two ways. Simple time integration of $|\varphi_n(\mathbf{r}_M, t)|^2$ yields the amount of ionization at this measuring point. This together with properly accounting for the solid angle $\Omega_{\mathbf{r}_M}$ associated with the vicinity of the measuring point direction yields the angular

distribution of emitted electrons [42, 45]. The PES is obtained by Fourier transforming the s.p. wave functions at the measuring point from time to frequency $\varphi_n(\mathbf{r}_M, t) \rightarrow \tilde{\varphi}_i(\mathbf{r}_M, E_{\text{kin}})$ and identifying the kinetic energy as $E_{\text{kin}} = k^2/(2m) = \hbar\omega$. This delivers the PES from s.p. state i for emission in direction of the measuring point \mathbf{r}_M . The total PES is then obtained by summing up the contributions from each s.p. state

$$\mathcal{Y}_{\Omega_{\mathbf{r}_M}}(E_{\text{kin}}) \propto \sum_{n=1}^N |\tilde{\varphi}_i(\mathbf{r}_M, E_{\text{kin}})|^2 \quad . \quad (7)$$

This simple Fourier transform applies to weak and moderate fields. For stronger field one still needs a phase correction which is explained in detail in [46].

3. RESULTS

3.1. Impact of SIC

We illustrate the importance of SIC for TDDFT on the example of the Na_5 cluster irradiated by a femtosecond (fs) laser pulse with FWHM of 10 fs, intensity $I = 2.2 \times 10^{11}$ W/cm², and frequency $\omega_{\text{las}} = 10.9$ eV, well above ionization threshold (ionization potential IP= 4.1 eV) and far away from dominant eigenfrequencies of the system. We use the simple ADSIC approximation (which is found to be appropriate for Na systems [35, 36]) and compare it to LDA. **Figure 1** collects the results. The top right part of the figure shows the (planar) ionic configuration. The lower right panel compares the sequence of s.p. energies for LDA and ADSIC. The latter produces a global down-shift of the s.p. levels as compared to those from LDA (by 1.2 eV for the HOMO) due to the enhanced Coulomb attraction (self-interaction neutralizes the asymptotic Coulomb field). The IP can be checked independently by comparing with the difference of energies of Na_5 and Na_5^+ (Koopman’s theorem) and ADSIC matches this energy difference perfectly while LDA fails badly. As a consequence, the s.p. spectrum from ADSIC carries the correct information on the ionization threshold.

The left panels show the dynamical response of the system to the laser pulse. The left lower panel displays the time evolution of the electronic dipole moment along laser polarization (x axis). There is almost no difference between LDA and ADSIC for the dipole signal. This is also found for the optical absorption strength (not shown here) which is no surprise because the optical absorption is another way to look at the dipole response. There is no urgent need to employ SIC when computing these quantities. This is plausible because the dipole signal is a global signal deduced from the density and density is, by construction, well-described in DFT.

The left upper panel shows the time evolution of the total ionization N_{esc} (see Equation 6). This signal reveals a dramatic difference between ADSIC and LDA. This is no surprise as ADSIC provides a larger IP than LDA (and altogether, more deeply bound levels, see bottom right panel). The reason is that, in TDDFT, ionization is mediated via the actual s.p. energies such that it is crucial to put the s.p. energies right within DFT.

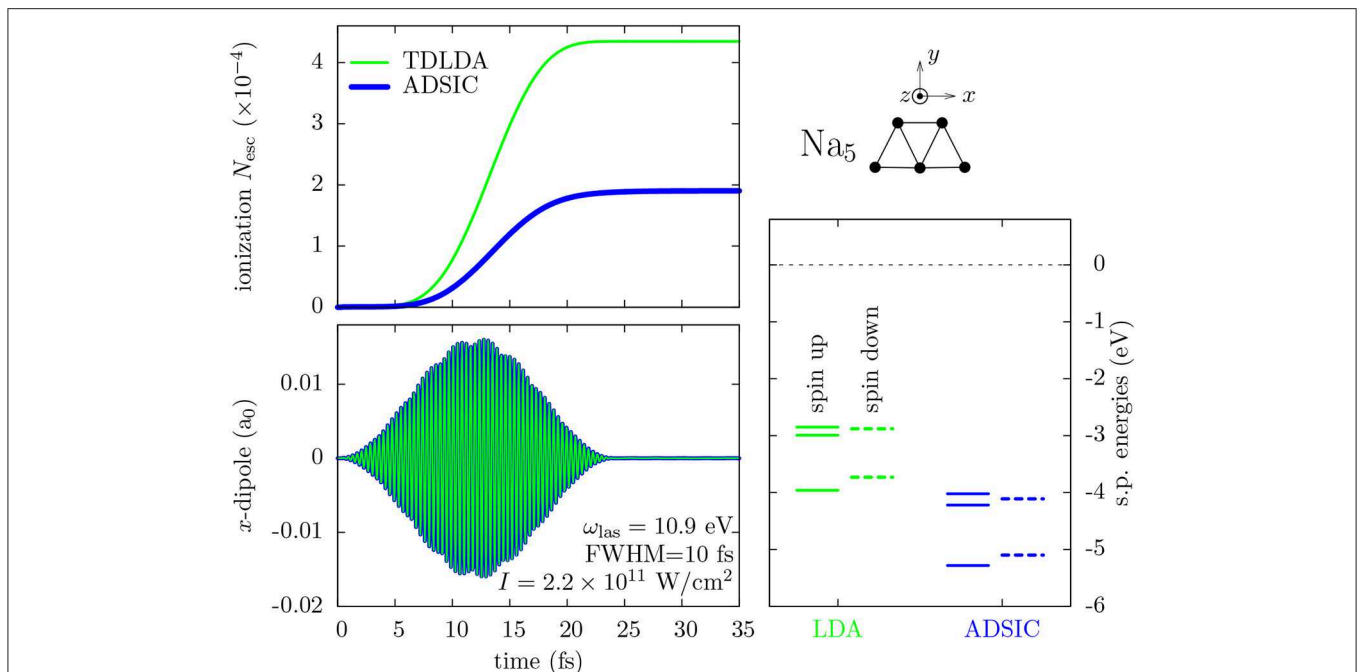


FIGURE 1 | Comparison of (TD)LDA and ADSIC for the Na_5 cluster. (Top right) Sketch of ionic configuration of Na_5 . (Bottom right) Single particle (s.p.) energies for spins up (full lines) and down (dashes). (Left) Time evolution of total ionization N_{esc} (top) and of the electronic dipole in the x direction (bottom) after irradiation by a laser pulse polarized along x with $\omega_{\text{las}} = 10.9$ eV, $T_{\text{pulse}} = 24$ fs, and $I = 2.2 \times 10^{11}$ W/cm 2 .

3.2. Optical Response

Optical response is the most prominent observable for dynamical properties of a system. Its role in nuclear physics and in molecular or cluster physics is similarly important. The conceptually simplest way is to compute it by spectral analysis of a time-dependent simulation (see section 2.3). Traditionally, it has mostly been computed by linearized TDDFT which complicates coding, but simplifies calculations, particularly if a system has certain symmetries (see e.g., [47–49]). The pathway through spectral analysis is competitive in fully 3D calculations and it is more flexible as it also allows one to explore the transition to the non-linear regime [50]. We use that here.

Figure 2 shows optical absorption strengths for a nucleus, ^{208}Pb , and three different electronic systems of different bond types, a metal cluster Na_{40} , a covalent molecule H_2O , and the noble gas Ar atom. The three electronic systems have significantly different spectra. For Ar and H_2O , the strength is heavily fragmented over many dipole states, most of them having predominantly the structure of one-particle-one-hole excitations. Quite different is the metallic Na_{40} , with one dominating and almost exhaustive peak, the highly collective Mie plasmon [54, 55]. The nuclear spectrum looks very similar to the cluster case. It is dominated by the a strong collective mode, the giant dipole resonance [56]. The similarity is not surprising as both, the metallic electron cloud and the nucleons, have the same bulk limit, namely a Fermi liquid [57].

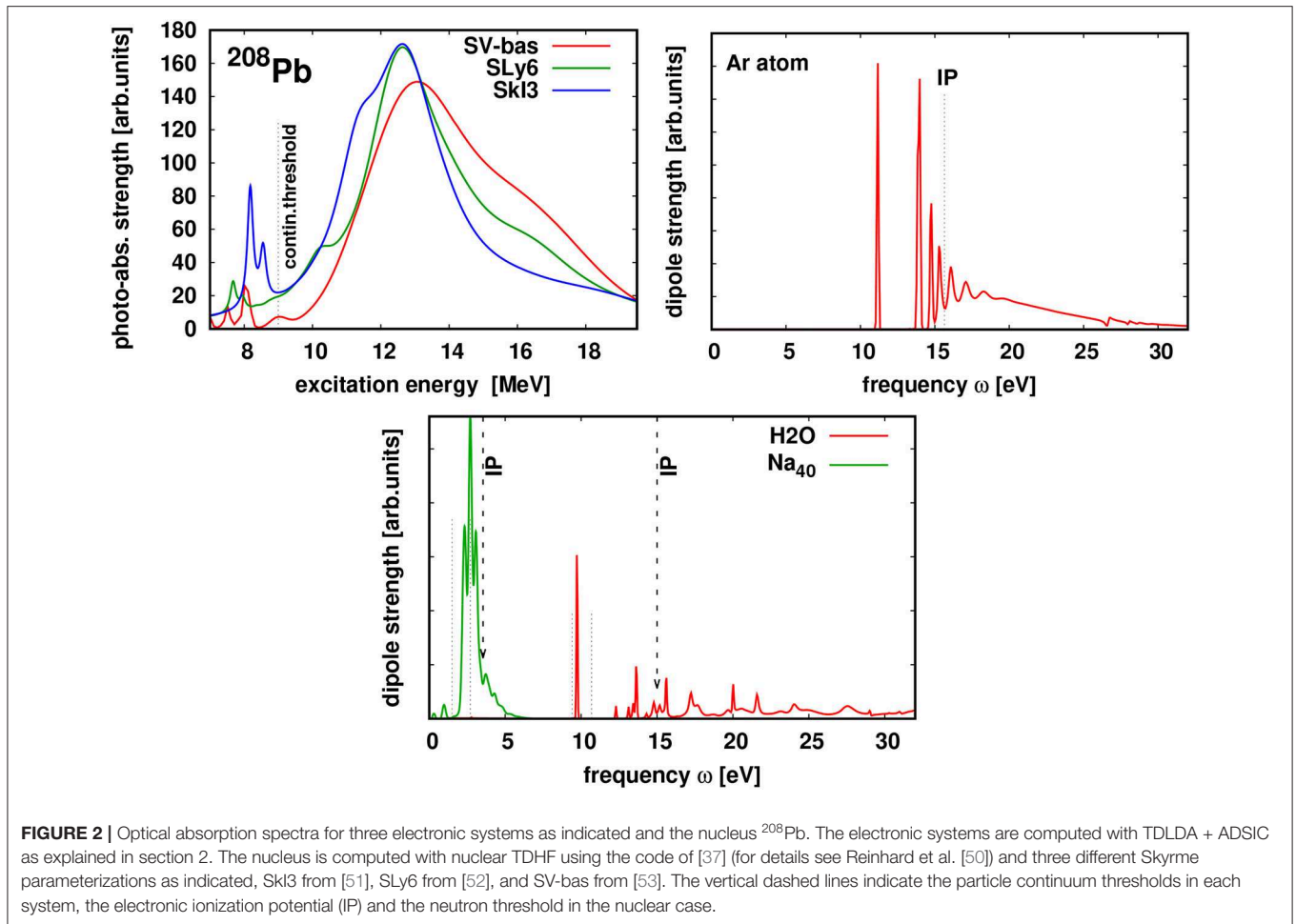
The nuclear dipole resonance and the cluster's plasmon resonance both have some width. However, the mechanisms producing these widths is different. The nuclear dipole resonance

lies in the nucleon continuum and a large part of the width stems from limited lifetime due to nucleon emission. The remaining part is due to dissipative processes from nucleon-nucleon collisions. Both processes also play some role in metal clusters. But here the dominant broadening mechanism is given by thermal fluctuations of the underlying ionic configuration [58, 59].

3.3. Laser Excitation

Laser pulses offer a unique and extremely versatile tool for dedicated probing and switching of electronic systems. Coherent pulses in keV and MeV regime, as required for nuclear experiments, appear at the horizon [60–62], but deliver not yet sufficient field strengths to attain the most interesting multiphoton regime. This day will come and thus it is worth having a look at electronic applications and see what experiments with coherent pulses can reveal.

A great deal of information is gained when combining laser excitations with detailed analysis of the emitted electrons. The top panels of **Figure 3** show as an example ARPES produced from the Ar atom (left) and the Na_9^+ cluster (right) after irradiation by a few-cycle laser pulse. Cuts along a fixed angle represent the PES and cuts along a fixed kinetic energy the angular distribution. The PES decrease with increasing E_{kin} because more photons have to cooperate to supply the higher emitted energy. And yet, one finds a plateau around 20 eV which is typical for above-threshold ionization spectra [63, 64]. The two different systems deliver different patterns, demonstrating that system properties



form the signal which, in turn, can be used to analyze a system by ARPES studies.

One of the interesting features in ARPES from very short pulses is the asymmetry of the angular distributions which appears in various energy bins for both systems. Indeed, in such laser pulses, the carrier-envelope phase ϕ_{CEP} , see Equation (2), becomes a decisive laser parameter because the CEP controls the net momentum exerted on the electron cloud and so impacts the pattern of the pulse dramatically which, in turn, can have a strong impact on laser-induced electron dynamics. For example, photo-electron emission induced by few-cycle laser fields can be controlled by the CEP, leading to a pronounced forward-backward (also called “right-left”) asymmetry in the PES. We can quantify this asymmetry by a simple number as

$$\eta(E_{\text{kin}}) = \frac{\int_0^\Theta d\theta \mathcal{Y}(E_{\text{kin}}, \theta) - \int_{180^\circ-\Theta}^{180^\circ} d\theta \mathcal{Y}(E_{\text{kin}}, \theta)}{\int_0^\Theta d\theta \mathcal{Y}(E_{\text{kin}}, \theta) + \int_{180^\circ-\Theta}^{180^\circ} d\theta \mathcal{Y}(E_{\text{kin}}, \theta)} \quad (8)$$

where $\mathcal{Y}(E_{\text{kin}}, \theta)$ is the ARPES strength at given kinetic energy and emission angle. For the opening angle in the integration, we take $\Theta = 15^\circ$ in accordance with many experiments in that field (see e.g., [65]). A value of $\eta = +1$ indicates prevailing forward emission and $\eta = -1$

backward dominance. Condensing the angular distribution into one compact number η allows one to visualize trends with laser parameter. The lower panel of **Figure 3** shows an energy-integrated asymmetry η as a function of ϕ_{CEP} . The effects are impressive. Tuning ϕ_{CEP} allows one to switch emission from forward to backward and vice versa. And we also see a significant system dependence of the trend. The marked difference between both systems can be explained by the difference in their optical response displayed in **Figure 2**. The 1.8 eV laser frequency lies far below any considerable dipole strength for Ar while it comes close to the strong Mie plasmon resonance for the Na cluster. For a detailed discussion with more material see Reinhard et al. [29].

3.4. Impact of Dissipation

So far, we have demonstrated the use of TDDFT in electronic systems on a few selected examples. TDDFT has shown to be a robust and versatile tool for simulating dynamical processes. However, the more energy comes into a system, the more likely come dynamical correlations beyond TDDFT into play. This is also well-known in nuclear physics and a variety of methods has been developed to deal with those correlations. They employ all semi-classical concepts and most of them

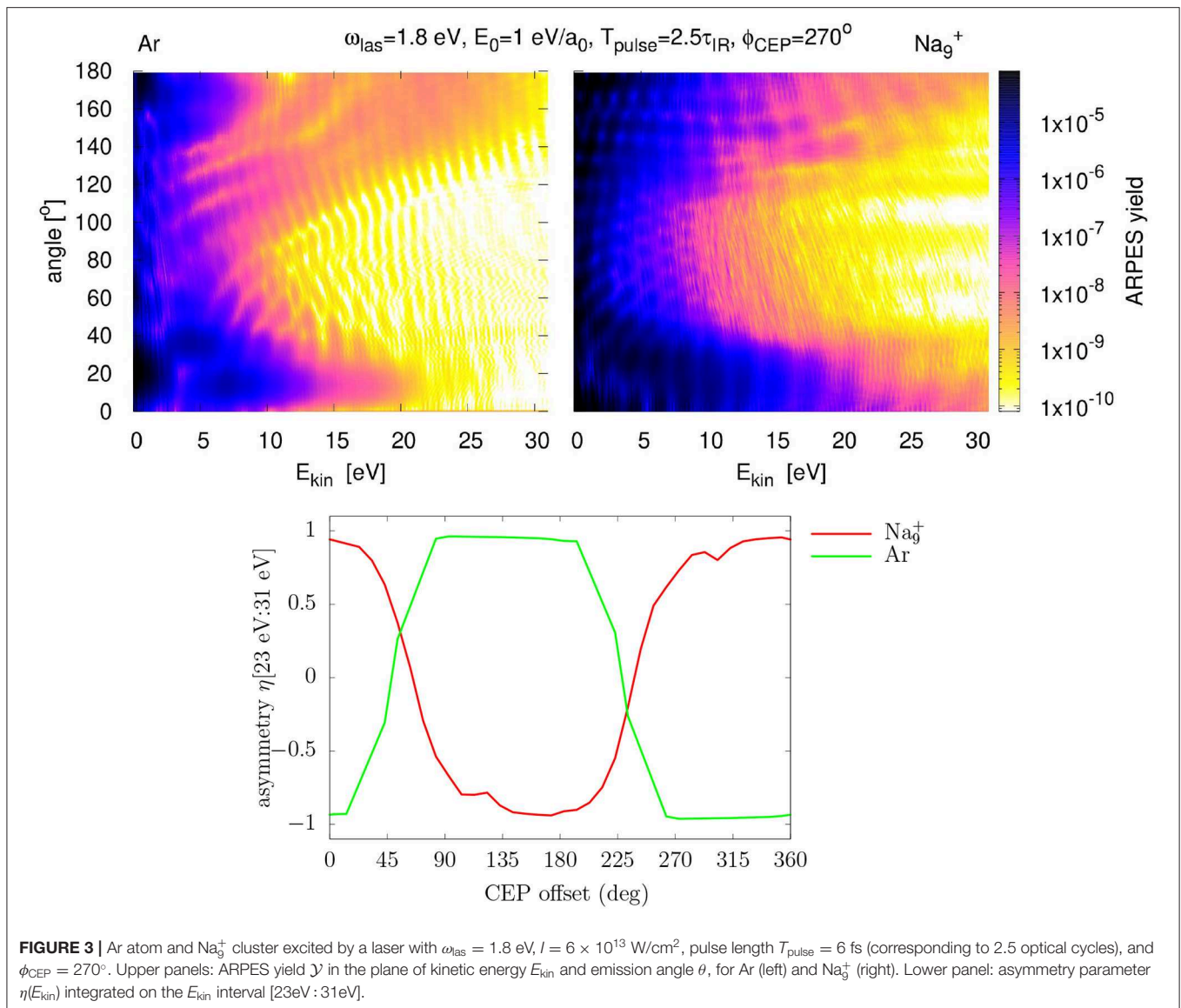


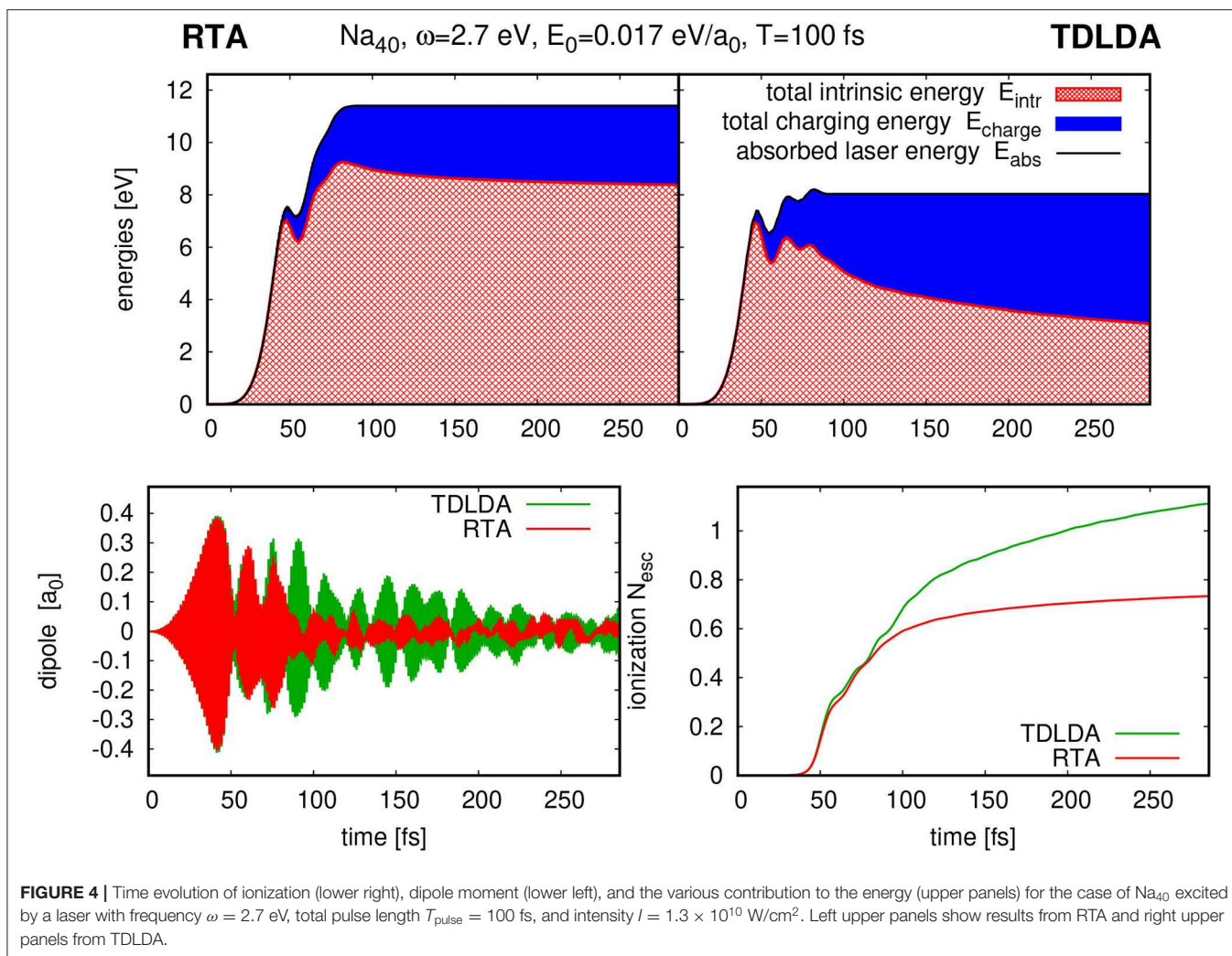
FIGURE 3 | Ar atom and Na_9^+ cluster excited by a laser with $\omega_{las} = 1.8 \text{ eV}$, $I = 6 \times 10^{13} \text{ W/cm}^2$, pulse length $T_{pulse} = 6 \text{ fs}$ (corresponding to 2.5 optical cycles), and $\phi_{CEP} = 270^\circ$. Upper panels: ARPES yield \mathcal{Y} in the plane of kinetic energy E_{kin} and emission angle θ , for Ar (left) and Na_9^+ (right). Lower panel: asymmetry parameter $\eta(E_{kin})$ integrated on the E_{kin} interval [23eV : 31eV].

are based on the Vlasov-Ühling-Uhlenbeck collision term [66] (for reviews see [67, 68]). This has also been applied to clusters [10, 69, 70]. But laser excitation processes sail often for a long initial time span through the quantum regime. There is thus a need for a quantum description of dynamical correlations from electron-electron collisions and associated dissipation. Just recently, there were promising moves to develop such approaches, one along the line of stochastic jumps (comparable to the collisions in semi-classical methods) [71–73] and another one with the more empirical relaxation-time approximation (RTA) [74, 75]. Both approaches are based on two-body collisions implying the in-medium electron-electron interaction and take care of the available phase space of final states. RTA, for which we will show an example here, maps cross-section and phase space into a collision rate with which the system relaxes toward local-instantaneous equilibrium, the latter being evaluated at each relaxation step by density- and current-constrained minimization of the free energy. By

construction, RTA maintains the continuity equation and energy conservation.

Figure 4 exemplifies the consequences of the dissipation thus introduced with an application of RTA to resonant laser excitation of a Na_{40} cluster. The lower left panel shows the effect on the dipole signal. The additional damping introduced by RTA is clearly visible. It is interesting to note that the TDLDA signal is also damped, although at lower rate. This damping stems from electron emission which is the dominant cooling mechanism in TDLDA. This is corroborated by the time evolution of ionization shown in the lower right panel. The trends agree up to about 100 fs, the same span where the dipole signals agree. But then the RTA curve levels off whereas TDLDA continues to emit. That happens because RTA has meanwhile converted much of the initial excitation energy into internal heat and so distracted it from moving into direct electron emission.

A comparison of energy balances is detailed in the upper panel of **Figure 4**. It shows the contributions from intrinsic



energy and energy invested in electron emission for TDLDA and RTA (for details of the definition see [75]). It confirms what we have inferred from the ionization trends in the lower right panel. Most interesting is the large difference in the total energy absorbed from the laser pulse (black line). In pure TDLDA, the entrance channel for energy absorption, the dipole channel, has limited capacity as one can see from the onset of oscillations in the absorption signal. Dissipation in RTA clears the entrance channel and thus allows more energy to come in. Altogether, we see that dissipation can make a difference. Note, however, that this example deals with a considerable excitation energy and that deviations from TDLDA develop only slowly. In turn, we can conclude that TDDFT provides a pertinent description of dynamical processes in its early stages. The time scale of validity of TDDFT depends on excitation energy: the lower the excitation is, the longer TDDFT holds.

4. CONCLUSIONS

We have presented a couple of typical applications of time-dependent density functional theory (TDDFT) to electronic

dynamics in clusters and molecules, as triggered by laser pulses. Basis of our description is TDDFT propagated in real time which delivers a versatile and robust tool for a great variety of dynamical situations. At the numerical side it is important to implement absorbing boundary conditions which then allow a detailed description of electron emission. We have addressed in detail two known defects of TDDFT in the local density approximation, namely the self-interaction error and the lack of dynamical correlations. The first problem is less important in nuclear physics, but highly relevant in electronic systems. It is solved by a self-interaction correction (SIC) which still can be formulated within a mean field theory (thus affordable) and which is particularly crucial if electron emission plays a role. The second problem, also important in nuclear dynamics, requires extensions beyond TDDFT for which manageable quantum mechanical approximations are presently being developed.

We have considered typical examples of applications in clusters and molecules excited by a laser pulses of different strengths. In all cases, the early response of the systems addresses predominantly the electrons and remains fully in the quantum mechanical regime (as properly described by TDDFT). In

particular, we have focused on an analyzing tool which is not yet available in nuclear dynamics, namely widely tunable laser excitation in combination with detailed resolution of the distributions of the emitted electrons with respect to angles and kinetic energies. We have demonstrated that such a setup delivers valuable information on the system and its dynamical response.

Finally, the inclusion of dissipative features is a key step to address the long-time evolution of dynamics far off-equilibrium in which one observes, in addition to ionization, a thermalization of the electron cloud. Formerly, the problem was attacked with semi-classical approaches much similar as done in nuclear dynamics, particularly in heavy-ion collisions. This approach is less justified in connection with laser excitations for which the (electronic) system stays much longer in the quantum regime. Promising and manageable solutions to deal with dissipation in the quantum regime are just presently coming up. First results from treating dynamical correlations in the relaxation-time approximation (RTA) show that dissipation has a crucial impact on the energy balance in the system (internal energy vs. electron emission) and on the energy intake from the laser pulse.

Proper treatment of dynamical correlations is still in its infancy and will attract much future development.

DATA AVAILABILITY STATEMENT

All datasets generated for this study are included in the article/supplementary material.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct and intellectual contribution to the work, and approved it for publication.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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