Attosecond resolved charging of clusters

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Attosecond laser pulses open the door to resolve microscopic electron dynamics in time. Experiments performed include the decay of a core hole [1], the time-resolved measurement of photo ionization [2] and electron tunneling [3]. The processes investigated share the coherent character of the dynamics involving very few, ideally one active electron. Here, we introduce a scheme to probe dissipative multi-electron motion in time. In this context attosecond probing enables one to obtain information which is lost at later times and cannot be retrieved by conventional methods in the energy domain due to the incoherent nature of the dynamics. As a specific example we will discuss the charging of a rare-gas cluster during a strong femtosecond pulse with attosecond pulses. The example illustrates the proposed use of attosecond pulses and suggests an experimental resolution of a controversy about the mechanism of energy absorption by rare-gas clusters in strong vacuum-ultraviolet (VUV) pulses [4, 5, 6].

Using attosecond pulses for making microscopic time measurements involves a unique pump-probe scheme. In recent experiments [1, 2, 3] the system is pumped by an attosecond extreme-ultraviolet (XUV) pulse and probed by a few-cycle infra-red (IR) pulse. In one case [1, 2], referred to as attosecond streaking, the final electron momenta \( p_{\text{final}} = p_{\text{excited}} + \vec{A}(\tau) \), in which the excitation time \( \tau \) is encoded by the instantaneous vector potential \( \vec{A}(\tau) \) of the IR pulse, is measured. In the other case [3], the ion yield \( F_{\text{final}} = \int_{-\infty}^{\infty} dt' P(t') \) is recorded, whereby the nonlinear dependence of the tunnel ionization \( P \) on the instantaneous electric field \( \vec{E}(t) \) of the IR pulse allows one to determine the initial time \( \tau \) with subfemtosecond resolution.

An alternative imaging method with attosecond resolution which does not even need ultrashort pulses has been proposed and demonstrated [7]: Using high harmonics generated by illuminating an aligned molecule with an intense femtosecond laser pulse, the Fourier transform of an electronic amplitude could be imaged and the corresponding spatial amplitude tomographically reconstructed. Although the measured amplitude looks similar to the highest occupied molecular orbital of the illuminated molecule this is certainly not the case since the experiment probes the multi-electron ground state of \( \text{N}_2 \) which, moreover, is entangled due to the Pauli principle. The details are by now well understood [8, 9].

Since the new attosecond technique leads into unparalleled territory the first experiments have addressed relatively simple coherent dynamics: one-electron, two-electron, and in the third case a seemingly simple almost separable case of multi-electron motion. One may argue that, at least regarding the pump-probe experiments, the time-resolved quantities could in principle also be retrieved from an experiment which exhausts the full parameter space in electron energy. This is the case since the electron distribution of interest (directly after the attosecond pulse) remains essentially coherent until it can be measured (time-resolved or more conventionally, energy-resolved). Time- and energy-resolved measurements are to a large extent Fourier related in this context.

Here, we would like to draw attention to another possibly fruitful use of attosecond pulses in the context of complex systems, exemplified with rare-gas clusters. The difference to the simpler electron dynamics as discussed above lies in the fact that due to dissipation and other many-particle interaction effects one can by the very nature of the process not recover time-resolved information from the (energy resolved) traditional observables. Hence, time-resolved information from ultrashort pulses will provide far unobtainable insight into the dynamics of those larger, dissipative systems. Clearly, such information is dominantly incoherent due to the dissipation processes.

To be specific we will propose a pump-probe scenario which is inverse to the one mentioned above: We expose a small rare-gas cluster to a femtosecond VUV laser pulse and probe the time-dependent excitation and ionization dynamics in the cluster, i.e., its charging, by time-delayed attosecond XUV pulses. The latter simply kick out electrons, still bound to an ion in the cluster at that time, and provide in this way information on the (transient) charging status of the cluster ions, as will be detailed below and is schematically shown in Fig. 1.

We will use for the pump pulse a 100 fs pulse with a frequency of 20 eV similar to the one applied at FLASH in Hamburg in 2002 [4]. The interpretation of the experimental results is still somewhat controversial [8, 10, 11] and a future attosecond resolved observation of the charging of the cluster could discriminate between the different theoretical scenarios which predict quite different degrees of transient charging of the cluster.

In our specific example we want to trace the ionization stage of the cluster ions through the kinetic energy spectra of the photo electrons from the attosecond pulse. Three possible problems come immediately to mind. They all have to do with the unique relation of the time of flight spectra of the electrons to the time dependent charging of the target:

(i) The attosecond pulse will act perturbatively on the electrons bound to the ions, but how does one make
FIG. 1: Scheme for the slow-pump (VUV pulse, blue line) fast-probe (XUV pulse, red line) time-resolved measurement of the transient charging in a multi-ion and multi-electron system. Before the pulse (a) the electrons are localized in atomic orbitals (thick red lines) and the cluster is held together by van-der-Waals forces. The initial ionization will lead to a cluster potential (b), allowing for further excitation of bound electrons into the cluster; these quasi-free electrons (blue dots) are eventually ionized with low kinetic energies (blue arrows). The attosecond XUV pulse pushes electrons from the upper bound states, directly into the continuum by one-photon absorption (dashed red arrow). The measured energy of these electrons traces their instantaneous binding energy and thus the transient charge state of the ion and the overall charge of the cluster as a function of the time delay $\Delta t$ between both pulses. Finally, after the pulse, the charged cluster fragments (c).

sure that the least bound electron is kicked out and therefore identifies uniquely the present charge state of the ion?

(ii) How can one avoid that the atto-pulse photo electron looses its energy characteristic of the bound state it came from through inelastic collisions while leaving the cluster?

(iii) How does one distinguish between “normal” ionized electrons and the atto-pulse photo electrons?

In order to answer these questions and to see if the idea of slow-pump fast-probe is feasible one has to do a realistic calculation. We base our calculation on previous experience in modeling cluster dynamics [5, 11, 12, 13, 14]. As a new element we add the atto-probe pulse which acts perturbatively (via photo ionization rates) onto individual ions in their respective charge state. The photo-ionization rates for the argon atom in the XUV region are easily accessible [15] since 3s and 3p electrons from the outer shell can be ionized by single-photon absorption.

Electrons in other states are not affected by the attosecond XUV pulse. Quasi-free electrons, on one hand, have due to their delocalization in the cluster volume much lower cross sections for ionization. Core-shell electrons (1s, 2s, 2p), on the other hand, would require multiphoton processes which are negligible at the XUV intensity applied.

Altogether, the well defined coupling of the $n = 3$ shell electrons to the attosecond pulse solves problem (i), although in the case of Argon, indeed, 3s and 3p electrons are ionized, as we will see. Problem (ii) is solved by minimizing the probability of collisions for the photo electron using a small Ar$_{13}$ cluster and a high frequency for the atto pulse (fast photo electron). The latter also solves problem (iii) since the quasi-free electrons form a well characterized electron plasma whose Maxwellian yields electrons in the continuum but with energies of only a few eV [16], which should be very small compared to the energies of the photo electrons. One may think that a slower probe than with attosecond pulses may be sufficient for the charging dynamics. However, the simulation (without atto-pulse) suggests that the interesting charging happens within some 10 fs, see Fig. 2. Hence, probe pulses of at least subfemtosecond length are convenient. Moreover, the probe pulse must have considerably higher photon energy to generate photo-electrons, clearly separated in energy from the cluster electrons, see problem (iii). We have used 500 as pulses with 150 eV photon energy and a peak intensity of $1 \times 10^{15}$ W/cm$^2$.

Figure 3 presents a series of electron spectra with different delays $\Delta t$ between the femto- and the atto-pulse. One can clearly follow the charging of the cluster with increasing delay of the atto pulse. For a time delay
that the low-energy part of the electrons from the plasma (blue wings in Fig. 3) is not influenced by the XUV pulse.

Since for the high-energy part the kinetic energy is given by $E_{\text{kin}} = \hbar \omega_{\text{xuv}} - E_{\text{ip}}(\text{Ar}^{q+})$, with the latter the ionization potential of a $q$-fold charged argon ion, one can measure the average charge from the centre of mass of the obtained energy distribution. The measured charge (red in Fig. 2) leads the real one (blue). Within the standard deviation the measured charge is systematically larger due to the cluster’s overall charge and the excited states of the ions.

We have proposed a scheme to probe the transient charging of a rare-gas cluster during exposure to a strong VUV pulse with attosecond pulses. This specific example conveys the general idea for a so far not suggested use of attosecond pulses: Initiate a relatively slow excitation process in an extended system through a femtosecond laser pulse, and probe the non-stationary, most likely dissipative relaxation dynamics by time delayed attosecond pulses.

**Methods**

A hybrid quantum-classical approach is used to simulate the time-dependent dynamics of the cluster ions and electrons under external light pulses [11]. Bound electrons are not explicitly treated, but can be ionized with probabilities according to their quantum ionization rates. Special attention has been paid to adopt the photo ionization rates to the cluster environment containing electrons which screen the ion and neighboring ions which lower the threshold for ionization into the cluster. Once an electron is ionized into the cluster, it becomes “quasi-free” and is propagated together with all other charged particles (ions and electrons) classically under the full Coulomb interaction and the dipole coupled electric field $E(t) = \tilde{e}_{\mathbf{z}} \sqrt{I_{\text{xuv}}/I_0} \sin^2 \left( \frac{2\pi}{\lambda_{\text{xuv}}} t \right) \sin (\omega_{\text{xuv}} t)$ of the VUV pulse which has a peak intensity of $I_{\text{xuv}} = 7 \times 10^{13} \text{W/cm}^2$ at $\hbar \omega_{\text{xuv}} = 20 \text{eV}$ photon energy and $T_{\text{xuv}} = 100 \text{fs}$ pulse length. The effect of the attosecond pulse is treated in perturbation theory. The intensity $I_{\text{xuv}} = 1.4 \times 10^{15} \text{W/cm}^2$ of the atto pulse (photon energy $\hbar \omega_{\text{xuv}} = 90 \text{eV}$, duration $T_{\text{xuv}} = 500 \text{as}$) is chosen such that on average 0.5 electrons/cluster are photo ionized by the atto pulse to ensure that the atto electrons do not perturb the charging situation of the cluster themselves.

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