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Few-Body Systems

Fragmentation in Intense Time-Dependent Fields*

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Abstract. The mechanism of enhanced ionization or charge-assisted tunneling, first discovered for small linear molecules under an intense laser pulse, is briefly reviewed. Then it is shown that this mechanism also applies to rare gas clusters in strong laser fields. A suitable observable which can be accessed experimentally, is the degree of ionization of the cluster as a function of the laser pulse length. Examples are given for Ne_{16} and Ar_{16}.

1 Introduction

Critical stability, understood as a delicate balance between stabilizing and destabilizing factors in a dynamical system, plays an important role for weakly bound states in few-body systems, and of course, to understand and interpret resonances. Both situations have in common that the Hamiltonian of the respective few-body system is time-independent, i.e., energy-conserving.

What happens, if an explicit time-dependent interaction is added, e.g., an intense laser pulse? How does the irradiated system fragment? Clearly, conventional fragmentation thresholds at a certain energy will not be important in this situation. Yet, are there still certain structural properties which have a distinct influence on the fragmentation or excitation of the system?

In this contribution we will describe such a property, *enhanced ionization*. It is a source of critical *instability* for a few-body system which decays into a number of charged fragments under the influence of an intense laser pulse. Enhanced ionization was first discovered for H_2^+ in Corkum's and Bandrauk's group [1]. It has been proven to play also a role for more complicated diatoms and triatoms and recently for long linear molecules composed of a number of atoms [2].

Here, we will briefly review the basic mechanism of energy transfer from laser light to bound electrons which goes back to Keldysh [3]. Then we will summarize the principle of enhanced ionization or, as we call it, charge-assisted

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tunneling. Finally, in the main part of this contribution, we will show that this principle of ionization and fragmentation enhancement due to a local (in time and space) instability of the system is also at work in rare gas atom clusters of moderate size of the order of 10 atoms. We will use atomic units unless stated otherwise.

2 Energy Transfer from a Laser Field to Bound Electrons

2.1 Tunneling Ionization for an Atom

Adding a time-dependent interaction via dipole coupling

$$V(\mathbf{R}, t) = \mathbf{R}\hat{e}_p F_0 f(t) \cos(\omega t) \tag{1}$$

to the dynamics of a bound electron with a Hamiltonian $H = p^2/2 - Z/R$ increases the space of parameters on which the dynamics depends by the envelope function $f(t)$ with a maximum field strength of F_0, by the pulse length T, the frequency of the field ω and by the polarization direction $\hat{e}_p$ (we assume linearly polarized light only). The fluency or energy content of the pulse is then $E_T = F_0^2 \int_0^T f(t)^2 dt$. Most experiments have been performed at $\omega = 0.057$ a.u. which is the frequency of the Titan-Sapphire laser. Typical pulse lengths are of the order of $10–10^3$ fs or longer, depending on the peak intensity F_0^2 one needs.

For the theoretical approach the ratio $\gamma = \omega/F_0(2\omega_B)^{1/2}$ is important and decides if the light field can be treated classically ($\gamma < 1$) but non-perturbatively or if one deals with multiphoton dynamics where each absorbed photon can be traced and increases the energy by $\hbar\omega$. The regime $\gamma < 1$ is sometimes called tunneling regime since the ionization proceeds by tunneling through an instantaneous barrier formed by the time-dependent potential $-Z/R + \mathbf{R}\hat{e}_p F_0 f(t) \cos \omega t$ seen by the electron. Keldysh gave an analytical expression of the ionization rate under such conditions which is dominated by the tunneling through the barrier (see Fig. 1a; for simplicity we discuss a one-dimensional case here where the electron has only one degree of freedom along the polarization axis of the laser, $\mathbf{R}\hat{e}_p = R$). For our purpose, also in the context of clusters as explained later, we use a semiclassical

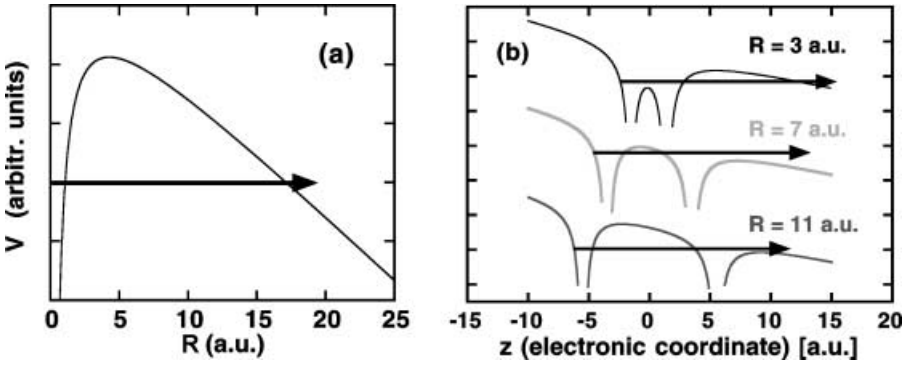

Fig. 1. Schematic representation of tunneling ionization at maximum field strength (**a**) for an atom, and (**b**) for H_2^+ at different internuclear distances R

approximation of the tunneling rate

$$\Gamma(t) = 1/\tau \int (V(R) - RF_0 f(t) \cos \omega t - E_B)^{1/2} dR, \tag{2}$$

where $1/\tau$ is the frequency $\omega_B/2\pi$ with which the particle bound with energy $\hbar\omega_B$ tries to tunnel through the barrier. For a hydrogen atom with binding energy $E_B = \hbar\omega_B = 0.5$ a.u. and the laser frequency of $\omega = 0.057$ a.u. at a field strength of $F = 0.15$ a.u. we have $\gamma \approx 1/10$ as well as $\omega/\omega_B \approx 1/10$ which implies a separation of time scales for the electron dynamics and the periodic change of the laser field: The electron in its hydrogenic ground state returns roughly ten times before one optical cycle is completed. This allows one to formulate a time-dependent rate as in Eq. (2) and to integrate over time afterwards. The envelope function $f(t)$, of $\sin^2$-shape in our case, controls the pulse length and varies even more slowly for a pulse of a typical length of 10 to 100 optical cycles.

2.2 Enhanced Ionization for a Diatomic Molecule

Looking at the simplest molecule, H_2^+, exposed to a strong laser field, one notes that an additional time scale enters the dynamics, namely that of the nuclear motion. It is typically (depending on the mass of the nuclei) a factor 10^3 to 10^5 slower than the electronic time scale (1 a.u.) and can be of the same order as the pulse length. The interesting dynamical effects happen when the internuclear axis is parallel to the polarization axis of the laser light. The simplest representation of the system still capturing the essential dynamical features is therefore one-dimensional for the electronic and nuclear degree of freedom. Since the latter can be treated as an adiabatic variable to a first approximation we have now an ionization (or tunneling) rate in the strong laser field which depends on time t and internuclear distance R, as shown in Fig. 1b. Since the energy absorption is non-linear a larger part of the wavefunction is accumulated in the higher-lying well at each instant of time [1]. The internuclear distance which produces the lowest barriers will allow for the highest tunneling rate. At small internuclear distance, the electron lives still in one common well and has to tunnel through the outer barrier. For large internuclear separation the electron has to tunnel through the inner barrier. Hence, an intermediate internuclear distance R_c must exist where both barriers are of roughly equal height and easy to tunnel through for the electron. For H_2^+ this is the case at about 7 a.u. for which distance also in the experiment a strong enhancement of ionization has been observed. The distance of the nuclei before the Coulomb explosion can be determined from the measured kinetic energy of the ions [4]. In Fig. 2 we show the one-dimensional result for enhanced ionization for fixed nuclei. This means that the laser pulse is applied to the system of the bound electron and the two nuclei at a distance R and the ionization yield is recorded. One clearly sees a maximum of ionization around 7 a.u. Note that the effect of charge-assisted tunneling does not depend on the laser frequency as long as the dynamical time scales are separated well. The quasiclassical curve compared to the quantum result in Fig. 2 has been obtained with our mixed quantum-classical formulation of few-body dynamics and represents a test for the following application to rare gas atom clusters.

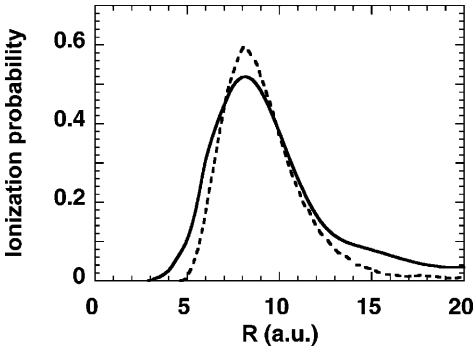

Fig. 2. Ionization probability for H_2^+ in one dimension as a function of the internuclear distance R for a pulse of 10 optical cycles, peak intensity of $5 \times 10^{13}\,\mathrm{W/cm^2}$ and frequency of $\omega = 0.055$ a.u.; quantum result solid and quasi-classical model dashed

3 Rare Gas Atom Clusters in Strong Laser Fields

3.1 Theoretical Approach

The description of clusters of 10–50 atoms with more than 10 electrons (Ne, Ar, etc.) in an intense laser field requires some severe approximations since more than a hundred particles cannot be propagated quantum-mechanically in time. The laser pulse fragments and ionizes the cluster very efficiently. Hence, it is reasonable to model the Coulomb explosion of charged fragments classically by solving Newton's equations for all the fragments. However, as described in the previous section, barriers and tunneling play a significant role for the absorption of energy from the laser pulse by bound electrons. An adequate treatment of these processes requires some quantum elements in the description which have to be compatible with the classical formulation for the motion of the particles. We model the ionization probability $P(t) = \Gamma(t)\,\Delta t$ for each atom in the cluster with the time-dependent tunneling rate of Eq. (2) in a small time interval Δt, where the potential in the tunneling rate is now not only the atomic potential but in addition the instantaneous effective potential generated by all other surrounding charged particles. We decide by comparison of the probability with a random number if the atom is ionized. If the answer is yes, a new "classical" electron is created and the charge of the atom/ion increases by one. Simultaneously the next bound electron of the ion (with correspondingly higher binding energy) is subject to the possibility of ionization by the effective tunneling rate. This procedure is carried through simultaneously for all atoms/ions of the cluster. More details of the theoretical formulation and numerical implementation will be reported elsewhere. However, similar approaches have been formulated in refs. [5–7]. Our results agree quantitatively with ref. [7], while the results in ref. [6] for a given peak intensity indicate in general a higher degree of ionization than in our case.

3.2 The Traces of Charge-Assisted Tunneling in Rare Gas Atom Clusters

It is not obvious beforehand if charge-assisted tunneling is relevant for clusters since they do not have the preferred orientation of a linear molecule which gets aligned along the polarization axis. Another practical problem is the detection of the mechanism since the kinetic energy of all ions (even if it could be measured)

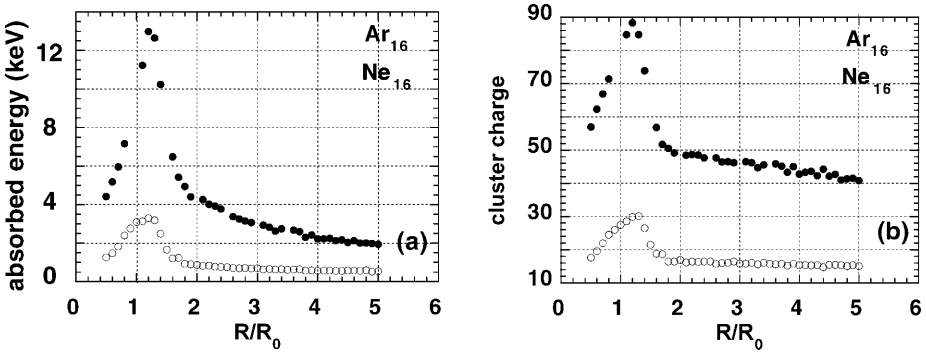

Fig. 3. Absorbed energy (**a**) and charge of the cluster (**b**) after application of the laser pulse of Fig. 2 but with an intensity of $10^{15}\,\mathrm{W/cm^2}$ and a pulse length of 20 optical cycles. Shown are results for Ne_{16} and Ar_{16} clusters for fixed mean next-neighbour distance of the ions $R = \left(N^{-1}\sum_{i=1}^{N}\min_{i>j}\{|\mathbf{R}_i - \mathbf{R}_j|^2\}\right)^{1/2}$ given in terms of the equilibrium distance R_0. The results are smoothed and averaged over 20 runs for each R

would not directly allow one to determine the ionic distances where the electron was ionized. However, we can tune the pulse length through an optimum for enhanced ionization: If the pulse is too short, the ions will not reach the critical distance for enhanced ionization during the laser pulse. If the pulse is too long, the intensity of the pulse is too weak when the critical distance is reached. In between, there should be an optimum pulse length putting enhanced ionization most effectively to work, provided the mechanism exists.

We proceed as in the case of H_2^+ and calculate the ionization yield for fixed cluster size measured in terms of the averaged next-neighbour ion distance in the cluster first (Fig. 3). We find a clear signature for charge-assisted tunneling for the absorbed energy as well as for the number of ionized electrons in both of our clusters considered, namely Ne_{16} and Ar_{16}, see Fig. 3. However, as one can see in Fig. 3, the critical distance is very close to the equilibrium distance since rare gas atom clusters are loosely bound compared to covalent molecules. Hence, it is not clear a priori if the absorbed energy and created charge of the cluster will also reflect charge-assisted tunneling in a full dynamical calculation as a function of laser pulse length. As can be seen from Fig. 4 the energy is monotonically decreasing while the charge shows a maximum for an optimum pulse length. In contrast to Fig. 3 we have now different laser pulses with constant fluency, i.e. $F_0^2 \propto 1/T$. Hence, the shortest pulses have the highest intensity. In this situation, ionization happens fast and almost at R_0 leading to highly energetic ions, as well as to energetic electrons (through the high intensity of the field). This effect masks tunneling ionization since the energy rises for short pulses.

Note also that the maximum in the charge for the Neon cluster is weaker than for the Argon cluster and that it occurs at longer pulses. This result is somewhat unexpected since the Argon atoms are heavier than the Neon atoms which leads to a slower Coulomb explosion for Argon. Based on this argument one would expect that the critical distance is reached at longer pulses for Argon than for Neon while the opposite is the case. The reason lies probably in the level structure of the Ne and the Ar atom: The latter has more weakly bound electrons than the Ne atom.

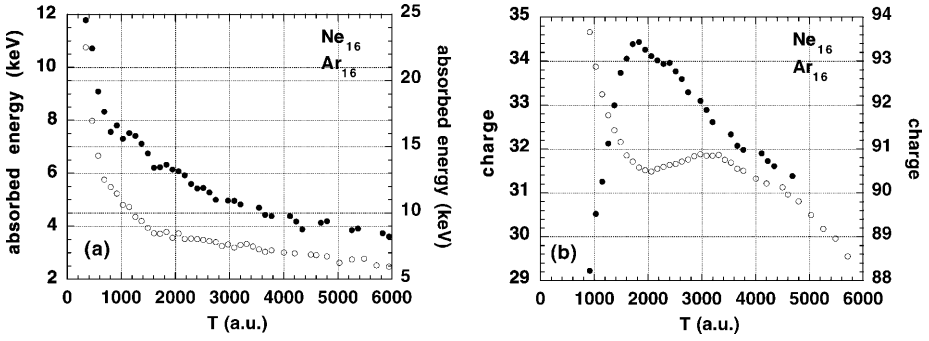

Fig. 4. Absorbed energy (**a**) and charge of the cluster (**b**) for laser pulses of different length with the same fluency, normalized to the pulse of Fig. 3 for Ne_{16} and Ar_{16}

Hence, during the same laser pulse, ions are created faster for an Ar cluster and therefore the Coulomb explosion sets in earlier and also progresses faster, due to the higher degree of ionization, compared to Neon. This may explain why the maximum occurs for shorter pulses in Ar_{16} compared to Ne_{16}. If this interpretation is correct, which has to be validated with more detailed analysis in the future, then it is feasible that clusters from heavy rare gas atoms such as Xenon show a pronounced effect of charge-assisted tunneling.

4 Summary

We have shown that the mechanism of enhanced ionization or charge-assisted tunneling, first discovered for diatomic molecules, also occurs in the fragmentation of rare gas clusters of moderate size (of the order of 10 atoms) in a strong laser pulse. As an experimentally accessible signature of charge-assisted tunneling in clusters we propose the degree of ionization of the cluster as a function of laser pulse length while keeping the fluency of the laser pulse constant.

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