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Surface-plasma resonance in small rare-gas clusters by mixing ir and vuv laser pulses

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The ionization dynamics of a xenon cluster with 40 atoms is analyzed under a pump-probe scenario of laser pulses where an infrared laser pulse of 50-fs length follows with a well-defined time delay a vuv pulse of the same length and peak intensity. The mechanism of resonant energy absorption due to the coincidence of the ir laser frequency with the frequency of collective motion of quasifree electrons in the cluster is mapped out by varying the time delay between the pulses.

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In recent years, much work has been devoted to the ionization mechanisms of clusters in few-cycle, intense laser fields (i.e., pulse lengths of the order of 100 fs and intensities $I=10^{13}$–10^{16} W/cm^2): from the case of plasmon excitation, when exposing metal clusters to relatively weak fields [1] over *enhanced ionization* akin of molecular ionization for small rare-gas clusters in intense fields [3], to collective excitation of a plasma resonance in clusters of intermediate [4] to large sizes [5], ultimately leading to ionic charge states of 40+ and higher [6], thus potentially providing a unique source for the generation of x rays, energetic ions or electrons, and, via nuclear fusion, even neutrons [7]. A new parameter regime for laser-cluster interaction has been proven to become accessible with the first experiment using vuv—free electron laser (FEL) light of 98-nm wavelength for the ionization of rare-gas clusters [8], soon followed by proposals for an explanation of the unexpectedly high charge states seen in this experiment [9,10].

While xuv-cluster interaction is still the subject of an ongoing debate, there seems to be a more or less common understanding regarding the qualitative picture of ir laser-cluster interaction: during the rising part of the laser pulse, a few electrons are ionized [16] leaving the cluster with a net positive charge which leads to an expansion typically on the same time scale as the duration of the laser pulse. Hence, effects which depend on the internuclear distances can be resolved by varying the pulse length [11] and/or applying pump-probe techniques [12,13]. The resolution of an optimum time delay Δt and the contrast of the signal in a pump-probe experiment increases if $\Delta t \gg T$, the length of each pulse. Since $\Delta t \approx t_c$, the critical expansion time of the cluster at which maximum absorption of energy from the cluster pulse is possible, long times t_c are desirable, which implies large clusters consisting of heavy atoms (slower Coulomb explosion). Also, the large number of quasifree electrons temporarily trapped in the cluster lead to a good contrast for the optimized versus nonoptimized signal [13].

The critical time t_c originates from a critical radius $R_c = R(t_c)$ of the cluster, usually larger than the equilibrium radius R_0, where energy absorption is most efficient. For larger clusters (resonant mechanism) this radius is determined by the surface-plasma frequency which is (in atomic units, which will be used throughout the paper) approximately given by

$$\Omega_t = \sqrt{\frac{N_t Z_t}{R_t^3}} = \frac{\omega_{pl}}{\sqrt{3}}, \tag{1}$$

where N_t is the number of atoms or ions in the cluster, Z_t is their average charge, R_t is the cluster radius, and ω_{pl} the bulk plasma frequency. The indices t indicate a slow dependence on time (at this point we want to emphasize that, at least as long as the cluster is neutral, the surface-*plasma* resonance is mathematically completely equivalent to the surface-*plasmon* resonance and can be derived along the same pathway. Nevertheless, we prefer to call it a plasma resonance since there is in principle a physical difference between a metal cluster being excited perturbatively and a rare-gas cluster turned into a nanoplasma by a nonperturbative laser field). If $\Omega_t \approx \omega$, the laser frequency, then the cloud of electrons which are trapped inside the cluster behaves like a (damped) harmonic oscillator driven to resonance [4], leading to efficient energy absorption and ionization.

The most important prerequisite for this mechanism is a significant amount of trapped (quasifree) electrons before R_c is reached, which can be achieved in two ways: (1) the laser-field strength is small enough to leave enough electrons inside the cluster before $R=R_c$; this possibility is limited, however, by the fact that the inner ionization process will eventually not start if the field strength is too small, so that no quasifree electrons will be created in the first place. For metal clusters, this problem obviously does not occur; on the other hand, one has to ionize the cluster to a certain degree in order to start the expansion process, and the range of intensities and pulse lengths which can start the Coulomb explosion while at the same time keeping the valence electron cloud intact is quite small [14]. (2) On the other hand, the force that keeps the electrons inside the cluster is generated by the space charge of the ions. Hence, going to larger clusters while leaving the average ion charge constant will make it more and more difficult for electrons to leave the cluster, so that the number of trapped electrons will increase with the

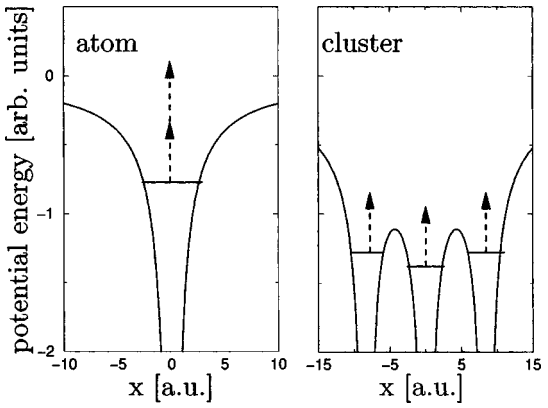

FIG. 1. Comparison of the inner ionization process for a single atom and a model cluster of three atoms. The effective barrier is lowered in a cluster due to the charged environment.

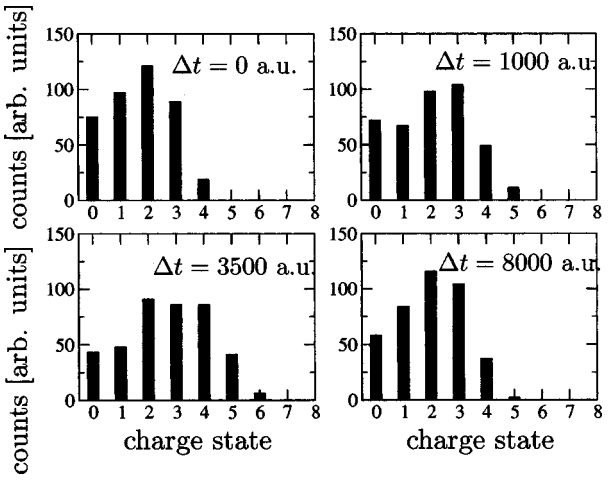

FIG. 2. Single-ion charge spectra after the interaction of Xe_{40} with a vuv pump pulse followed by an ir probe pulse as a function of the delay Δt between the two pulses. The highest (maximum and average) charge states are created for $\Delta t = 3500$ a.u. The scale of the ordinate axis is identical in all four cases.

cluster size. This is the reason why the resonance absorption is much clearer when seen with ir pulses for large clusters (compare Ref. [12] with Ref. [13]).

To summarize, resonant absorption in intense ir fields occurs (i) for metal clusters in weak fields ($I \lesssim 10^{13}$ W/cm^2); the size of the clusters then only plays a role insofar as it will change the expansion speed, which has to be accounted for by changing the pulse lengths accordingly; or (ii) for clusters with $N \gtrsim 10^2$ and intensities of $I \lesssim 10^{14}$ W/cm^2, where a substantial fraction of the quasifree electrons which are created by the laser is kept inside the cluster, so that a collective oscillation can develop.

In the case of small rare-gas clusters in strong ir fields, none of the above scenarios applies. Rather, ionization is dominated by *charge enhanced ionization*, known already from diatomic molecules, where the shape of the interatomic barrier leads to an optimal distance between the cluster nuclei which results in an efficient interplay between inner and outer ionization [2]: the neighboring charges must be close enough to start an *ionization avalanche* [3] once the first electrons are created; on the other hand, they must not be too close in order to decrease the space charge which prevents the electrons from escaping the cluster. Obviously, the whole process relies on the ionization of the first electrons relatively early in the pulse; once the field strength drops significantly below the field strength F_{th} required to field ionize a single cluster atom ($F_{th} = E_b^2/4$, where E_b is the first atomic ionization energy), the avalanche will not be started and the cluster will survive the radiation relatively undamaged. For Xe clusters, for example, $F_{th} = 0.0493$ a.u., which corresponds to an intensity of $I_{th} = 8.53 \times 10^{13}$ W/cm^2.

As has been shown in the Hamburg experiment, the threshold for an ionization avalanche is considerably lower when using vuv instead of ir light. With intensities of the order of 10^{12} W/cm^2 and a photon energy of 12.7 eV, complete breakup of Xe clusters and unexpectedly high ionic charges have been observed [8]. These findings can be explained [10] by using standard atomic photoabsorption rates but taking into account the effective (inner) ionization threshold which is lowered by the surrounding charges in a cluster (see Fig. 1). Due to this mechanism, and due to the fact that the quiver amplitude ir is two orders of magnitude smaller than for ir radiation, a vuv pulse is much more efficient in creating quasifree electrons than an pulse of the same peak intensity.

This opens up an elegant way to study the dynamics of collectively excited electrons and hence the resonance absorption mechanism in a *small* cluster by combining a vuv pump pulse with a time-delayed ir probe pulse: The vuv pulse generates a large number of quasifree electrons. At the same time the cluster gets only moderately charged and a slow expansion sets in, mainly driven by the hydrodynamic pressure of the quasifree electrons. Hence, one can observe with a time-delayed probe pulse very cleanly the optimum condition for energy absorption by the quasifree electrons as a function of cluster size starting at a size as small as $N = 40$ as we will demonstrate with the following pump-probe scenario: a Xe_{40} cluster is first irradiated by a 50-fs vuv pulse ($\omega = 12.7$ eV, $I = 7.9 \times 10^{12}$ W/cm^2). Then, after a variable time delay Δt, we apply a second pulse of the same length and intensity, but now with a wavelength of 780 nm. The simulation has been done using the quasiclassical model introduced in Ref. [3].

The charge spectra after the interaction of the cluster with the two pulses is shown in Fig. 2 for various times delay Δt. Two features stand out: First, ions with charges of 5+ or 6+ are produced already with these comparatively low intensities. In fact, the ionization efficiency is comparable to the results from our calculation in Ref. [10], where only a single vuv pulse was applied, but with an intensity one order of magnitude higher than in the present case (note that, with an average charge per atom of 2.5 for $\Delta t = 3500$ a.u., the space charge of the Xe_{40} cluster from the present work is approximately equal to the space charge of the Xe_{80} cluster from Ref. [10], where an average charge per atom of only 1.5 was achieved, so that the two cases can really be compared). This shows that by combining vuv and ir pulses, significantly higher charge states can be achieved than by applying an ir or a vuv pulse alone. Second, we see that the ionization is

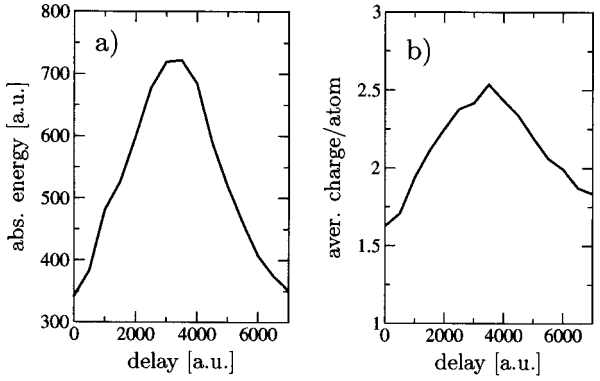

FIG. 3. Absorbed energy (a) and average charge per atom (b) as a function of time delay between the vuv and the ir pulse.

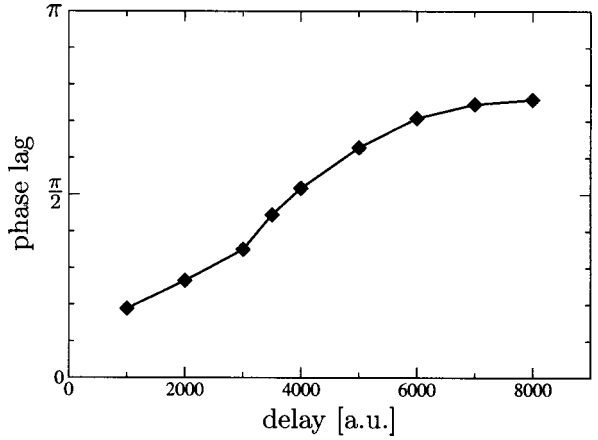

FIG. 4. Phase lag between the center-of-mass oscillation of the electron cloud and the driving laser field as a function of time delay between the vuv and the ir pulse.

most efficient for a time delay of $\Delta t = 3500$ a.u.

The absorbed energy and the average charge state per atom after the cluster has disintegrated are shown in Fig. 3 as a function of the time delay Δt. The curve for the energy absorption as well as the one for the charge shows a maximum $\Delta t \approx 3500$ a.u. between the pump vuv and the probe ir pulse. If our physical picture is correct, this maximum should be due to the existence of a collective resonance with frequency Ω_t of the quasifree electrons that were created by the vuv pulse. This resonance is most efficiently excited by the ir pulse when the laser frequency $\omega = \Omega_t$. We will now proceed to give evidence for this hypothesis.

In principle there are two ways to check numerically whether the electron cloud is at resonance with the laser field: First, treating the electrons and ions of the cluster as a homogenic positively and negatively charged sphere, respectively, one ends up with the resonance condition $\Omega_t = \omega$. However, the definition Eq. (1) of Ω_t is by no means unique, since one has to define a cluster "volume" which itself is time dependent (through the increase of the cluster radius) and so are the charge of the ions and the number of electrons. The resonance condition can be determined more reliably by calculating the phase difference between the oscillation of the electronic center of mass (ECM) and the driving laser field $F(t) = F_t \cos \omega t$ [4], where F_t is the envelope of the laser field: if one assumes a collective oscillation with a damping constant Γ, the time-dependent dipole amplitude for the ECM reads

$$X(t) = A_t \cos(\omega t - \phi_t) \tag{2}$$

with

$$A_t = F_t / \sqrt{(\Omega_t^2 - \omega^2)^2 + (2\Gamma_t \omega)^2}, \tag{3}$$

$$\phi_t = \arctan[2\Gamma_t \omega / (\Omega_t^2 - \omega^2)]. \tag{4}$$

For $\phi_t = \pi/2$ the system is at resonance and the laser cycle averaged energy absorption

$$\langle dE/dt \rangle = \frac{1}{T} \int_0^T \frac{dX_t}{dt} F(t) dt \propto \sin \phi_t \tag{5}$$

is at its maximum. Note, however, that the amplitude X_t does not necessarily increase considerably at resonance due to strong damping [see Eq. (3)]. Hence, we take the condition $\phi_t = \pi/2$ as the *definition* for the plasmon resonance and calculate ϕ_t from the phase lag between the driving field $F(t)$ and the dipole oscillation dX_t/dt by extracting the maximum of the time correlation $c(s)$ between the two signals, where

$$c(s) = \int_{t_1}^{t_2} F(t) X(t+s) dt. \tag{6}$$

We chose the limits of integration in Eq. (6) to be 10 ir cycles before and after the maximum of the probe laser; the radial evolution R_t of the cluster is sufficiently slow so that the phase lag only changes by a small amount during that time. The outcome of this calculation is shown in Fig. 4. Indeed the phase lag is equal to $\pi/2$ for $\Delta t \approx 3800$ a.u. This proves that the maxima in Fig. 3 are due to a resonance of the collective electron oscillation with the driving laser field.

Hence, the vuv pump combined with a ir probe pulse can map out the internal collective cluster dynamics very clearly and may be the only possibility to resolve this dynamics for small clusters. The reason is simply that the vuv pulse produces a large number of quasifree electrons which can participate in collective electron motion. On the other hand, the vuv pulse itself does not couple to this collective motion. Hence, only the probe pulse probes literally the collective electron dyamics. Using two ir pulses for pump and probe make this distinction difficult: The pump pulse must be very short in order not to "probe" itself the collective electron dynamics. On the other hand, if it is very short it will not produce a significant number of quasifree electrons which could move collectively since they are produced most efficiently through resonant coupling. In the next phase of the vuv-FEL at Hamburg, a pump-probe facility as "used" in this

theoretical investigation will be available for experiments. Furthermore, a similar experiment, but much simpler than the one at DESY as far as the experimental set is concerned, is planned in Saclay [15]; it is planned to use an 800-nm femtosecond laser, together with its ninth harmonic (delivering photons with $E \approx 14$ eV) as the ir and vuv pulse, respectively; the intensities will be a bit lower than the ones considered here, but this should only result in a quantitative difference. The present paper shows that indeed, interesting and unique experiments can be done with such a time-delayed combination of pulses.

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[1] E. Suraud and P. G. Reinhard, Phys. Rev. Lett. **85**, 2296 (2000).
[2] T. Seideman, M. Y. Ivanov, and P. B. Corkum, Phys. Rev. Lett. **75**, 2819 (1995).
[3] Ch. Siedschlag and J. M. Rost, Phys. Rev. Lett. **89**, 173401 (2002); Phys. Rev. A **67**, 013404 (2003).
[4] U. Saalmann and J. M. Rost, Phys. Rev. Lett. **91**, 223401 (2003).
[5] C. Jungreuthmayer, M. Geissler, J. Zanghellini, and T. Brabec, Phys. Rev. Lett. **92**, 133401 (2004).
[6] T. Ditmire, T. Donnelly, A. M. Rubenchik, R. W. Falcone, and M. D. Perry, Phys. Rev. A **53**, 3379 (1996).
[7] I. Last and J. Jortner, J. Phys. Chem. A **106**, 10872 (2002).
[8] H. Wabnitz *et al.*, Nature (London) **420**, 482 (2002).
[9] R. Santra and C. H. Greene, Phys. Rev. Lett. **91**, 233401 (2003).
[10] Ch. Siedschlag and J. M. Rost, Phys. Rev. Lett. **93**, 043402 (2004).
[11] L. Köller, M. Schumacher, J. Köhn, S. Teuber, J. Tiggesbäumker, and K. H. Meiwes-Broer, Phys. Rev. Lett. **82**, 3783 (1999).
[12] T. Döppner, Th. Diederich, J. Tiggesbäumker, and K. H. Meiwes-Broer, Eur. Phys. J. D **16**, 13 (2001).
[13] S. Zamith, T. Martchenko, Y. Ni, S. A. Aseyev, H. G. Muller, and M. J. J. Vrakking, Phys. Rev. A **70**, 011201(R) (2004).
[14] F. Calvayrac, P.-G. Reinhard, E. Suraud, and C. A. Ullrich, Phys. Rep. **337**, 493 (2000).
[15] H. Wabnitz (private communication).
[16] For rare-gas clusters, the concepts of *inner* and *outer* ionization, the first being the ionization of an electron out of an atomic orbital into the cluster environments, the latter the process of an electron leaving the cluster as a whole, have proven to be very useful. If one is dealing with the valence electrons of a metal cluster, the inner ionization step is skipped, since these electrons can already move freely throughhout the cluster. From the second shell onwards, metal clusters should not behave differently from rare-gas clusters.