Multielectron Ionization of Large Rare Gas Clusters

Isidore Last and Joshua Jortner*

School of Chemistry, Tel-Aviv University, Ramat Aviv, Tel Aviv 69978, Israel

Received: June 10, 1998; In Final Form: August 28, 1998

In this paper we describe high-order multielectron ionization of large rare-gas clusters in strong laser fields in terms of initial ionization due to the suppression of electrostatic barriers and a subsequent dynamic ignition process in the Coulomb exploding cluster. The mechanism of a Xe_{1100} cluster ionization in a strong laser field of 10^{16} W/cm^2 is studied in a one-electron, one-dimensional approximation. The initial ionization is realized up to the total charge of $Q_c \approx 1200$. In the interval $2600 < Q_c < 6600$, where electrons are moving almost freely inside the cluster due to their high kinetic energy, the mechanisms of a direct energy enhancement are responsible for ionization. The $Q_c > 6600$ ionization becomes possible when the cluster spatial expansion due to Coulomb explosion is taken into account.

I. Introduction

Multielectron ionization of molecules and clusters can be realized by photoionization in a strong laser field.[1−11] The multielectron ionization leads to a Coulomb explosion and to the production of multicharged atomic ions. The kinetic energy of the product ions formed by Coulomb explosion is of the order of several or tens eV in diatomics,[1,2] hundreds of eV in small van der Waals (vdW) clusters,[3,4] and 100 KeV−1 MeV in large ($n > 1000$) vdW clusters.[5−11] The Coulomb explosion of large rare-gas clusters is of special interest due to the extremely large energy release, which approaches the energy of nuclear processes.[5] Another new feature of the Coulomb explosion of large clusters containing more than a few hundred constituents is the generation of highly charged (up to Xe^{40+}) product ions.[6] The electronic and nuclear dynamics of molecules and clusters in strong laser radiation fields (intensity $I > 10^{14}$ W/cm^2) are novel and very interesting.

The energetics and dynamics of ultrafast Coulomb explosion on the time scale of nuclear motion can be adequately accounted for by Coulomb repulsion between multicharged ions.[1,12,13] A much more difficult and challenging task pertains to the understanding of the ionization process initiated by the laser field. In diatomic molecules and in small clusters multielectron ionization is most probably caused by the quasiresonance energy enhancement of electrons, which becomes possible due to the presence of inner potential barriers.[14−16] While treating the ionization process in multicharged large clusters, it is imperative to bear in mind that in these systems the electrons are extremely strongly bound. For example, the ionization potential (IP) of uniformly charged $(Xe^{8+})_{1000}$ clusters is estimated to be as large as ∼4.8 KeV. Electrons can only be removed from such systems provided that their energy is highly enhanced.

The plasma-heating mechanism[8,10] provides a possible route for the ionization of large clusters. However, as will be discussed subsequently, the existence of a quasistable plasma in multicharged large clusters of the size of a few thousand atoms is questionable (see section III). Furthermore, the ionization mechanism may be distinct for different levels of ionization. In this paper we describe high-order multielectron ionization of large rare-gas clusters in terms of two sequential processes: (1) initial ionization due to the electrostatic barrier suppression mechanism,[17,18] and (2) the dynamic ignition mechanism. Subsequent high-order ionization processes are induced by the inner electric field of the initially ionized cluster[19−20] which undergoes Coulomb explosion.

II. Cluster Initial Ionization

In order to treat the problem of large cluster ionization, we will consider here electron dynamics in the multicharged $(Xe^{q+})_{1100}$ cluster, which was studied experimentally by Hutchinson and his colleagues.[9] The cluster is supposed to be initially in the neutral equilibrium geometry described by a sphere with radius $R_c = 25$ Å. In our study we shall use the laser power $S = 10^{16}$ W/cm^2 (the force amplitude $eF_0 = 27.4$ eV/Å) and the frequency $\nu_F = 0.386$ fs^{-1} employed in the experiments of Hutchinson et al.[9] The laser field $F = F_0 \cos(\omega t + \varphi_0)$ is described as a sinusoidal field (frequency $\omega = 2\pi\nu_F$, initial phase φ_0) with a constant amplitude F_0. In real-life experiments, the laser pulse envelope is not rectangular and the field amplitude increases at the beginning up to the maximal value $eF_0 =$ 27.4eV. Our treatment does not account for the turn on of the laser field, considering the electron dynamics when the field amplitude reaches its maximal value.

By the ionization of a cluster we will refer to the removal of electrons to infinity, whereas by the inner ionization we will imply the process when electrons are removed from their host atoms but remain bound to the cluster. One of the possible mechanisms of the ionization by an outer electric field is the suppression of the electrostatic barriers.[16−18] Following our previous analysis[16] it is possible to show that the field of $eF_0 =$ 27.4 eV/Å suppresses the electrostatic barrier in neutral Xe atoms leading to the inner ionization of the neutral Xe cluster. Some of the electrons, which are removed from their host atoms, can also leave the cluster, so that the cluster becomes ionized with a charge Q_c. This charge attracts the electrons enhancing their binding to the cluster. Outside the strongly charged (with a total charge $Q_c \gg 1$) cluster, considered as a continuous medium, the electron potential along the x axis (Figure 1) is

$$U(x,t) = -\frac{BQ_c}{x} - eF_0(x-x_0)\cos(\omega t+\varphi_0); \quad x > Rc, \quad B = 14.385 \text{ eV Å} \quad (1)$$

10.1021/jp9825821 CCC: $15.00
Published on Web 11/19/1998

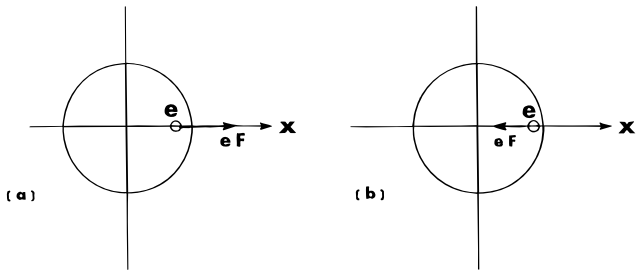

Figure 1. Two different orientations of the outer field force eF acting on an electron e located at the cluster periphery. (a) eF is directed outside the cluster, (b) eF is directed inside the cluster.

where x_0 is the initial ($t = 0$) electron location. We will assume that the electron is initially located at the cluster periphery ($x_0 = R_c$). The potential (eq 1) may present an electrostatic barrier which prevents the cluster ionization. This outer electrostatic barrier is suppressed when the force generated by the potential (eq 1) is directed outside the cluster (Figure 1a). This condition is fulfilled for the cluster charge

$$Q_c \leq \frac{1}{B} R_c^2 eF_0 \tag{2}$$

For the accepted Xe_{1100} cluster and the laser field parameters used herein this criterion provides a total charge $Q_c \leq 1190$ ($q \leq 1.08$ per atom). It follows that the cluster loses its first ~1000 electrons by direct electron removal. Most of these electrons are removed as soon as the field strength becomes close to the amplitude value F_0 so that the process of initial cluster ionization is expected to be accomplished during one laser light oscillation (2.6 fs), but after the laser amplitude becomes close to its maximal value F_0.

III. Electron Dynamics in the Ionized Cluster

After the cluster becomes initially ionized, due to direct electron removal, to the level of $Q_c \approx 1000$, another ionization mechanism has to be responsible for the further loss of electrons. The electron behavior in a charged cluster is different from that in a neutral one because of the inner Coulomb field which depends on the cluster charge distribution. In the absence of the outer field and in the state of equilibrium the cluster can be considered as a conductive sphere whose charge is located at the surface. However, the ionization process in the presence of a strong laser field is far from equilibrium so that the determination of a real charge distribution is not feasible and we are compelled to use some model charge distributions. We will apply here the spherically symmetric charge distribution with the charge density

$$\rho(r) = \frac{1.67 Q_c}{V_c} (r/R_c)^2, \quad Q_c = 0.6\, n q_c \tag{3}$$

where r is the distance from the cluster center, V_c is the cluster volume, n is the number of the cluster atoms, and q_c is the charge of the outer shell ions (at the $r = R_c$ periphery). The charge distribution given by eq 3 reflects the tendency of the charge concentration at the cluster periphery. In order to estimate the charge distribution effect on the electron dynamics, we also performed some calculations for a uniform charge distribution. According to these calculations both models provide similar results so the choice of the charge distribution model is not of great importance.

The inner Coulomb field, which is pointed toward the cluster center contributes, in particular, to the electron removal from their host atoms (the so-called ignition mechanism[19,20]). More electrons can be removed when the outer field is also pointed to the center (Figure 1b). The level of the inner ionization in the charged cluster can be estimated by the analysis of the inner electrostatic barriers, which can be suppressed by the inner Coulomb field or by the total effect of both the inner and the outer (laser) fields. According to our analysis, in the initially ionized cluster of $Q_c \approx$ l000 and in the absence of the laser field most atoms are deprived from their 5p electrons. When the total charge is higher, e.g., $Q_c = 2640$ (periphery charge $q_c = 4$ per atom) the periphery ions are deprived from all eight outer shell (5s,5p) electrons, and most inner ions from all six 5p electrons. With laser field assisted ionization more electrons are deprived from their host ions including some of the 4d electrons. As a result of the inner ionization about 2000−2500 of unbound (delocalized) electrons are moving inside the $Q_c = 2640$ cluster forming some kind of plasma.

TABLE 1: Electron Kinetic Energy (in eV) in Three Cluster Points x (in angstroms) for the Initial Field Phase $\varphi_0 = 150°$ (Figure 1b) as Depending on the Periphery q_c Charge. The Total Charge Q_c (see eq 3) Is Also Presented

x	$q_c = 2$ $Q_c = 1320$	$q_c = 4$ $Q_c = 2640$	$q_c = 6$ $Q_c = 3960$	$q_c = 8$ $Q_c = 5280$	$q_c = 10$ $Q_c = 6600$	$q_c = 12$ $Q_c = 7920$
12.5	470	690	860	960	1170	1380
0	820	1050	1230	1410	1600	1790
−25	1290	1340	1360	1310	1280	1320

The dynamics of the delocalized electrons inside the cluster can be significantly affected by electron−electron and electron−ion collisions. In order to find the effect of these collisions we particularly need to know the electrons' kinetic energy which determines to a large degree the collision probability. We will estimate the electron's kinetic energy by treating first the electron dynamics in the collision-free approximation. The role of this approximation will be discussed later using the results of the collision-free calculation.

Let us consider an electron initially located on a periphery ($r = -R_c$) with zero energy (the results have been shown not to depend much on the initial energy). The electron motion is restricted to the one-dimensional trajectory located along the cluster diameter (axis x) which is arranged in parallel to the outer field (Figure 1b). In the framework of the charge density model given by eq 3 the equation of electron motion is

$$\frac{d^2x}{dt^2} = C[-BQ_c g(x) + eF_0 \cos(\omega t + \varphi_o)];$$
$$C = 17.604 \text{ Å (fs)}^{-2} \text{ (eV)}^{-1} \tag{4}$$

where

$$g(x) = \begin{cases} x^3/R_c^5 & -R_c \leq x \leq R_c \\ 1/x^2 & x > R_c \\ -1/x^2 & x < R_c \end{cases} \tag{4'}$$

with R_c and x given in angstroms. According to the results of the calculation the electron crosses the multicharged cluster with a high kinetic energy when the initial field phase φ_0 roughly lies in the interval $120° \leq \varphi_0 \leq 180°$. The electron kinetic energy K at the three cluster points $x = 12.5$, 0, and −25 Å, is presented in Table 1 for the initial phase of $\varphi_0 = 150°$ (corresponding to the field direction of Figure 1b). The kinetic energy increases with Q_c at the points $x = 12.5$ Å and $x = 0$ but does not vary much at $x = -25$ Å, where the electron leaves the cluster. At the initial fragment of the trajectory ($25 > x > 12.5$ Å) the kinetic energy increases from zero to $K = 470$ eV

for $Q_c = 1320$ and to $K = 690$ eV for $Q_c = 2640$. Most of its way through the cluster ($12.5 > x > -25$ Å) the electron moves with a high kinetic energy of $K > 700$ eV for $Q_c > 2640$. The electron passes the central region with an energy of about $K = 1000$ eV for $Q_c = 2640$ and $K = 1800$ eV for $Q_c = 7920$.

We will now try to estimate the importance of the electron-atom (ion) collisions for the electron dynamics. Considering the collisions with atoms (ions), we will take into account the ionization (inelastic) and the elastic scattering channels. The ionization cross section can be estimated approximately by the expression[21]

$$\sigma = a\sum_i \ln(K/I_i)\frac{1}{KI_i},\ a = 450\ (\text{eV})^2\ \text{Å}^2 \qquad (5)$$

where K is the kinetic energy of the impact electron and I_i is the ionization potential of the bound electrons. In the case of electron motion through a neutral cluster, eq 5 presents $\sigma = 0.49$, 0.32 Å^2 for kinetic energies $K = 700$ and 1000 eV, respectively. The atomic density of the Xe_n clusters is $N_a = 0.017$ Å^{-3} which provides the free path of ionization $l = (\sigma N_a)^{-1} = 120-190$ Å. The elastic scattering cross section can be determined by using the results of the approximate Thomas−Fermi calculation.[22] For the scattering angles $\theta > 30°$ and the kinetic energy $K = 1000$ eV one obtains $\sigma \approx 3.6$ Å^2 which provides $l \approx 16$ Å. Since the ionization free path is not much larger than the cluster size ($2R_c = 50$ Å) and the elastic scattering free path is even smaller than the cluster size, both these processes are of importance when the neutral cluster is considered.

The situation is, however, quite different in multicharged clusters. In a cluster with $Q_c > 2640$, as considered before, most of the outer shell (5s,5p) electrons are removed from the ions, so that the $4d^{10}$ electrons can be considered as the bound ones. Using eq 5 we obtain the cross section of the $4d^{10}$ electrons ionization by $K = 700$ eV electrons being equal to $c = 0.026$ Å^2 which provides a free path as large as $l = 2600$ Å. We will also roughly estimate the cross section of the elastic scattering on the bound $4d^{10}$ electrons, taking it to be proportional to the mean square radius $\langle r^2\rangle$ of their orbitals. Such an estimate provides us with the cross section $\sigma = 0.29$ Å^2 and the free path of $l \sim 200$ Å which is noticeably larger than the cluster size. In addition to the elastic scattering with the bound electrons of the ions one also has to take the elastic scattering with the ionic Coulomb potential into account. This potential is proportional to the ionic charge q_i in the case of an isolated ion. Inside the cluster, however, the inner Coulomb potential is smoothed by the common effect of all surrounding ions. According to simple estimates, when the electron passes an ion with $q_i = 8$ at the impact parameter of about $(\sigma/\pi)^{1/2} \approx 0.3$ Å, the maximal variation of the inner potential is about 300 eV. This potential variation is smaller than the electron kinetic energy of $K > 700$ eV. We infer that the mean free path for the elastic scattering with the inner Coulomb potential arising from multicharged ions is comparable to the elastic scattering with the bound $4d^{10}$ electrons.

Since in a charged cluster there is a large number of free electrons one also has to take the electron−electron collisions into account. The contribution of these collisions can be estimated by using the expression for the free path of the plasma electrons[23]

$$l = 1.35 \times 10^3\, K^2/[n_e N_a \ln(0.03K)/(n_e N_a)^{-1/3}] \qquad (6)$$

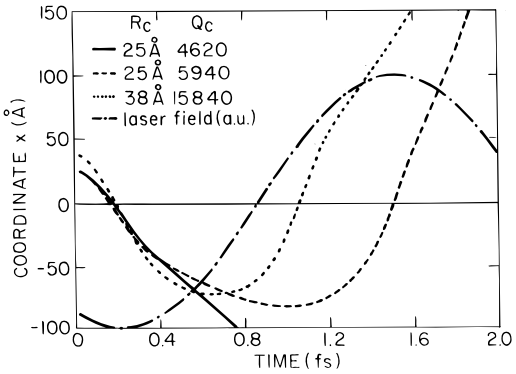

Figure 2. Electron trajectories for different cluster charges and radii. The outer field force eF is also shown. The initial ($t = 0$) eF phase is $\varphi_0 = 150°$ which corresponds to the eF orientation shown in Figure 1b.

where l is in angstroms, n_e is the number of free electrons per atom, and N_a is the atomic density (in Å^{-3}). Taking $n_e = 6$ and $K = 700$ eV, one obtains the free path to be as large as $l = 1650$ Å. Our estimates show that the free paths of the collision processes are considerably larger than the cluster size. It follows that, in the multicharged $Q_c \geq 2640$ clusters, the fast ($K > 700$ eV) electrons can be considered as free moving electrons whose collisions with cluster ions and other electrons are of minor effect. This finding confirms the possibility to perform the dynamical simulation of the ionization process in these clusters by using the collision-free approximation. This approximation may have to be modified for relatively weakly charged clusters with $Q_c < 2640$ where the collisions may be of importance. In spite of this constraint we have also utilized the collision-free approximation for $Q_c < 2640$, Xe_{1100} cluster.

IV. Ionization of Charged Clusters

In order to study the ionization process in charged clusters we performed a dynamic simulation of the electron motion in Xe_{1100} clusters using the collision-free approximation. The potential energy for the (classical) electron motion is characterized by eq 1. The dynamic simulation was performed for the most favourable initial conditions, namely for the initial electron location at $x = 25$ Å and the initial phase $\varphi_0 = 150°$. It was found that in the charge interval $1320 \leq Q_c \leq 5280$ (the periphery charge is $2 \leq q_c \leq 8$ per atom) the electron, which moves from the initial point $x = 25$ Å to the left (Figure 1b), gets enough kinetic energy to leave the cluster and to go to $x = -\infty$ (Figure 2). Such a one-way motion through the cluster takes an extremely short time of 0.4−0.5 fs. This ionization mechanism is realized when the characteristic frequency of the electron motion inside the cluster is much larger than the laser light frequency. In the charge interval $5280 < Q_c \leq 6600$ (the periphery charge being $8 \leq q_c \leq 10$) the ionization process is more complicated. The electron first moves left to some turning point located outside the cluster. When the electron goes back to the cluster, the field changes its direction enhancing the electron energy once more. After crossing the cluster once more the electron leaves it and goes to $x = +\infty$ (Figure 2). Such a two-way ionization process, which can be called a quasiresonance process, takes about 1 fs. The electron can leave the cluster also in the charge interval $6600 < Q_c \leq 7920$ (with the periphery charge $10 < q_c \leq 12$) but only after performing more than two oscillations. For higher charges the ionization process becomes precluded.

Our treatment shows that in a strong laser field electrons are very quickly removed from a large charged cluster, after crossing

it once or twice. In very strongly charged clusters ($10 < q_c \leq 12$) electrons are removed after crossing the cluster a few times. The characteristic times of the electron removal for the one- and two-way ionization processes are smaller than the laser light period of $T = 2.6$ fs, so that these ionization processes are expected to be mostly completed during one light oscillation (on condition that the field strength is already close to its maximal value F_0). Since the electrons do not have enough time to exchange their kinetic energy with each other, their dynamics drastically differs from that of the electrons in an equilibrium plasma. Such a conclusion contradicts the consideration of the ionization of large clusters as the process of the equilibrium plasma heating, proposed by Ditmire et al.[8] and by Hutchinson et al.[10]

V. Ionization During Coulomb Explosion

According to the results presented above a fast (~2.6 fs or less) ionization of the cluster at its neutral geometry ($R_c = 25$ Å) can be realized, resulting in the charge of $Q_c = 6600$. Charged clusters are, however, expanding, due to the Coulomb repulsion effect. The increase in the cluster size, even if moderately small, amplifies the ionization process. Thus increasing the cluster radius by 8% only makes the fast two-way ionization of the $Q_c = 7920$ ($q_c = 12$) cluster possible. When the cluster radius exceeds $R_c = 38$ Å, the two-way ionization becomes possible for the $Q_c = 15840$ ($q_c = 24$) cluster (Figure 2). A further increase in the cluster size is not considered here since in larger clusters of roughly $R_c > 40$ Å, the assumption of the continuous charge distribution, implemented in eq 3, is not well grounded.

Since the formation of highly charged ions with $q_c > 12$ depends on the cluster expansion, it is of interest to elucidate the dynamics of this expansion. The time of the cluster expansion can be estimated by using the expression for systems with fixed charges.[13] The Coulomb explosion time τ is

$$T = t_0 Z(\xi) \tag{7}$$

where

$$Z(\xi) = \eta/\xi + 0.5 \ln[(1 + \eta)/(1 - \eta)] \tag{8}$$

Here R_0 is the initial radius, R is the cluster radius at time τ, $\xi = R_0/R$, $\eta = (1 - \xi)^{1/2}$, and t_0 is the time parameter that depends on the system structure. In a large spherically symmetric A_n cluster with the charge distribution given by eq 3, the dynamics of the periphery ($r = R_c$) ion expansion was found to be determined by the following time parameter t_0 (in femtoseconds)

$$t_0 = \frac{1.04}{q_c}\left[\frac{md^3}{1 - 3.25n^{-1/3}}\right]^{1/2} \tag{9}$$

where m is the atom mass (in AM) and d is the initial interatomic distance (in angstroms). According to eqs 7−9 the increase in the cluster size from the initial radius $R_0 = 25$ Å to $R = 33$ Å for $q_c = 14$ takes about 11 fs. The expansion to $R = 38$ Å, the size where the ions with the charge as large as $q_c = 24$ can be generated, occurs on a slightly longer time scale of 14−16 fs. Accordingly, the rate determining time scales of ~10 −20 fs for the multielectron ionization at a high level of cluster ionization ($q_c = 14-24$) involves the Coulomb explosion.

The one-electron approximation used by us ignores the screening effect which is expected to attenuate the electron energy enhancement and, consequently, the ionization process. Rough estimates indicate that the screening effect reduces the laser field inside the charged cluster with the initial radius R_0 by only 20−30%. The contribution of this effect sharply decreases while the cluster size increases due to the Coulomb explosion. It follows that neglecting the screening effect is not expected to affect the ionization process in any significant way.

The simplifications pertaining to the one-dimensional model and the treatment of scattering effects overestimate, most probably, the ionization efficiency. In order to take the scattering effects into account, one needs to perform a rigorous simulation of the ionization process. Such a simulation was performed by Ditmire[24] for small Ar_n clusters ($n < 55$), where the energetics differ from that in large clusters. According to Ditmire's results, the electron impact ionization is roughly of the same importance as the ignition mechanism. These findings contradict our results. In our treatment of large clusters the inner Coulomb field is considerably stronger and the kinetic energy of electrons is much larger than in the small clusters studied by Ditmire.[24] Accordingly, our treatment for large clusters results in a higher level of the ignition mechanism ionization and in a lower efficiency of the impact electron ionization. Our conclusions are supported by recent three-dimensional many-electron dynamics simulation of the ionization process in large clusters.[25]

VI. Concluding Remarks

The interaction of high-intensity lasers ($I = 10^{16}$ W/cm^2) with large atomic clusters containing more than a few hundred constituents, gives rise to highly energetic electrons and a very high level of atomic core ionization. These novel phenomena can be traced to the manifestation of both electronic and nuclear motion, i.e., initial ionization due to barrier suppression, and subsequent ionization in conjunction with nuclear motion, that is dynamic ignition during cluster Coulomb explosion. Our study rests on the admittedly oversimplified classical treatment of the electron motion in a one-dimensional approximation. Since the multielectron ionization of clusters or molecules in a strong laser field involves tens or even hundreds of photons, the quantum nature of the light absorption is practically lost and a classical treatment of the light field becomes appropriate.[26] Accordingly, one may attempt to treat the ionization in a strong laser field in the same way as in an electrostatic field.

The one-dimensional description of the electron motion in the cluster provides a temporal lower limit for the characteristic time scales for the intracluster passage time of the electron and the ionization dynamics. A more detailed treatment of this interesting problem will involve a three-dimensional simulation of the (classical) electron dynamics. The application of classical dynamics for the electron both for the one-dimensional model utilized herein and for three-dimensional simulations is a-posteriori justified in view of the high electron kinetic energy, resulting in a low de Broglie particle wavelength, which is considerably smaller than the internuclear spacing in the cluster.

From the point of view of general methodology, the exploration of the multielectron ionization dynamics induced by intense laser−large cluster interaction, constitutes a borderline between laser−matter interaction for molecules and for condensed matter systems, with the dynamics being dominated by both electronic and nuclear processes. These constitute a novel set of phenomena of electronic and nuclear control of ultrafast femtosecond and attosecond dynamics in large finite systems.

Acknowledgment. This research was supported by the Binational German-Israeli James Franck program on Laser-Matter Interaction.

References and Notes

(1) Boyer, K.; Luk, T. S.; Solem, J. C.; Rhodes, C. K. *Phys. Rev. A* **1989**, *39*, 1186.

(2) Armstrong, D. P.; Harkins, D. A.; Compton, R. N.; Ding, D. *J. Chem. Phys.* **1994**, *100*, 28.

(3) Purnell, J.; Snyder, E. M.; Wei, S.; Castleman, A. W., Jr. *Chem. Phys. Lett.* **1994**, *229*, 333.

(4) Snyder, E. M.; Buzza, S. A.; Castleman, A. W., Jr. *Phys. Rev. Lett.* **1996**, *77*, 3347.

(5) . Ditmire, T.; Tisch, J. W. G.; Springate, E.; Mason, M. B.; Hay, N.; Smith, R. A.; Marangos, J.; Hutchinson, M. H. R. *Nature* **1997**, *386*, 54.

(6) Ditmire, T.; Tisch, J. W. G.; Springate, E.; Mason, M. B.; Hay, N.; Marangos, J.P.; Hutchinson, M. H. R. *Phys. Rev. Lett.* **1997**, *78*, 2732.

(7) Ditmire, T.; Smith, R. A.; Tisch, J. W. G.; Hutchinson, M. H. R. *Phys. Rev. Lett.* **1997**, *78*, 3121.

(8) Ditmire, T.; Donnelly, T.; Rubenchik, A. M.; Falcone, R. W.; Perry, M. D. *Phys. Rev. A* **1996**, *53*, 3379.

(9) Shao, Y. L.; Ditmire, T.; Tisch, J. W. G.; Springate, E.; Marangos, J. P.; Hutchinson, M. H. R. *Phys. Rev. Lett.* **1996**, *77*, 3343.

(10) Hutchinson, M. H. R.; Ditmire, T.; Springate, E.; Tisch, J. W. G.; Shao, Y. L.; Mason, M. B.; Hay, N.; Marangos, J. P. *Phil. Trans. R. Soc. London A* **1998**, *356*, 297.

(11) Lezius, M.; Dobosh, S.; Normand, D.; Schmidt, M. *Phys. Rev. Lett.* **1998**, *80*, 261.

(12) Jortner, J.; Levine, R. D. *Isr. J. Chem.* **1990**, *30*, 207.

(13) Last, I.; Schek, I.; Jortner, J. *J. Chem. Phys.* **1997**, *107*, 6685.

(14) Seideman, T.; Ivanov, M. Yu.; Corkurn, P. B. *Phys. Rev. Lett.* **1995**, *75*, 2819.

(15) . Chelkowski, S.; Bandrauk, A. D. *J. Phys. B* **1995**, *28*, L723.

(16) Last, I.; Jortner, J. *Phys. Rev. A.* **1998**, *58*, 4.

(17) Augst, S.; Strickland, D.; Meyerhofer, D.; Chin, S. L.; Eberly, J. H. *Phys. Rev. Lett.* **1989**, *63*, 2212.

(18) Yu, H.; Zuo T.; Bandrauk, A. D. *J. Phys. B* **1998**, *31*, 1533.

(19) (a) Rose-Petruck, C.; Schafer, K. J.; Barty, C. P. J. *Proc. SPIE Int. Soc. Opt. Eng.* **1995**, *2523*, 272. (b) Rose-Petruck, C.; Schafer, K. J.; Wilson, K. R.; Barty, C. P. J. *Phys. Rev. A* **1997**, *55*, 1182.

(20) Snyder, E. M.; Buzza, S. A.; Castleman, A. W., Jr. *Phys. Rev. Lett.* **1996**, *77*, 3347.

(21) Lotz, W. Z. *Phys.* **1968**, *216*, 241.

(22) Mott N. F.; Massey, H. S. W. *The Theory of Atomic Collisions*; Clarendon Press: Oxford, 1965.

(23) Ginzburg, V. *The Propagation of Electromagnetic Waves in Plasmas*; Pergamon Press: Oxford, 1970.

(24) Ditmire, T. *Phys. Rev. A* **1998**, *57*, R4094.

(25) Last, I.; Jortner J. To be published.

(26) Keldysh, L. V. *Sov. Phys. JETP* **1965**, *20*, 1307.