Attenuation of ultraintense laser radiation in an assembly of molecular clusters

Isidore Last and Joshua Jortner
School of Chemistry, Tel Aviv University, Ramat Aviv, Tel Aviv 69978, Israel
(Received 8 November 2005; published 7 June 2006)

We present a theoretical and computational study of the effects of the laser light intensity attenuation, due to absorption by an assembly of $(D_2)_{n/2}$ ($n=250-3\times10^4$) clusters in ultraintense laser fields (peak intensity $I_M=10^{16}-10^{18}$ W cm^{-2}). The laser intensity attenuation was described by a relation between the local, time-integrated energy flow of the laser pulse and the energy deposited per ion in the Coulomb explosion products. In the cluster vertical ionization (CVI) domain, the local laser intensity manifests a linear decrease along the plasma filament, so that under the conditions of partial light absorption the energy absorbed by the plasma filament is independent of the incident laser intensity. In the non-CVI domain, the local laser intensity decreases exponentially across the plasma filament and the energy absorbed is proportional to the incident intensity. Cluster sizes for total light absorption were established on the basis of numerical solutions of our first-order differential equation for the local laser intensity. The effect of strong light absorption on the kinetic energy distribution of the ions from Coulomb explosion in an assembly of clusters results in a marked deviation from the energy distribution under CVI conditions. This exhibits a sharp rise in the range of low energies with the location of a low-energy maximum in the energy distribution providing a benchmark for the assessment of the contributions of laser absorption effects. Finally, we consider the effects of laser light absorption in a single cluster, demonstrating that both in the CVI and in the non-CVI domains the attenuation of the laser intensity is small for the cluster sizes considered in this work, while in the non-CVI domain laser light absorption by a single large ($R_0=330$ Å and $n\sim5\times10^6$) deuterium cluster can be realized.

DOI: 10.1103/PhysRevA.73.063201 PACS number(s): 36.40.Vz, 36.40.Wa, 52.38.Dx

I. INTRODUCTION

Elemental [1–8] and molecular [6,9–16] clusters subjected to ultraintense and ultrashort laser fields, $I \geqslant 10^{15}$ W cm^{-2}, manifest sequential-parallel electron dynamics processes involving inner ionization, nanoplasma formation, and outer ionization [17–29]. As a result of outer ionization, the cluster becomes charged, which leads to Coulomb explosion and to the formation of atomic ions or nuclei. The energetics of the product ions (nuclei) depends on the cluster size and on the laser intensity, as well as on other laser pulse characteristics, e.g., light frequency, pulse shape, and width [27]. Most of the theoretical studies addressed electron and nuclear dynamics of a single cluster [17–19,21,22,24–29], while experiments focused on an assembly of clusters coupled to a laser field [1–16]. A meaningful interpretation of the experimental data has to account for the spatial inhomogeneity inside the focus volume (the plasma filament [10,16,23]), and for the absorption of laser radiation by the assembly of clusters in a beam [6,10,15,20]. The attenuation of the light intensity $I(t;x,y,z)$ along the light propagation direction (denoted here as the z axis) at time t, will markedly affect dynamic data in ultraintense laser fields. These dynamic effects will be manifested in the cluster size dependence of the ion energy spectrum as well as in the yields of dd nuclear fusion driven by Coulomb explosion (NFDCE) of an assembly of deuterium containing clusters [10,12,14–16]. In this paper we address the attenuation of the light intensity from an ultraintense laser in an assembly of clusters, and the manifestation of this process in the energetics, i.e., ion average kinetic energy, ion energy distribution, and the final energy of the Coulomb explosion products of an assembly of deuterium containing clusters. As a physical system we shall treat laser-cluster interactions and Coulomb explosion of deuterium $(D_2)_{n/2}$ ($n=250-33\,000$) homonuclear clusters [27,30]. We shall describe the laser intensity attenuation by a relation between the local, time integrated energy flow of the laser pulse and the energy deposited per ion in the Coulomb explosion products. For this analysis we utilize an empirical parametrization scheme for the cluster size dependence and the laser intensity dependence of the energy deposited in a cluster, which was obtained in our previous work [30].

II. ATTENUATION OF ENERGY FLOW AND LOCAL INTENSITY

Light absorption by clusters constitutes a complicated nonlinear and time-dependent process [6,20,24,31–34]. We shall advance an equation, which describes the attenuation of the local z-dependent time-integrated energy flow of the laser pulse

$$S(z) = \int_{-\infty}^{\infty} I(t,z)\cos^2(2\pi v t)dt, \tag{1}$$

where $I(t,z)$ is the local intensity envelope at the light propagation z axis at time t and v is the laser frequency. Here, and in what follows, we consider the decrease of the light intensity along the z axis and ignore the inhomogeneity of the light intensity in the xy plane perpendicular to z. The decrease of the energy flow along the propagation direction will be described by a global energy balance equation, with the attenuation of $S(z)$ being due to energy absorption by the cluster constituents. We take a uniform cluster spatial distribution within the cluster beam. Ignoring the cluster size dispersion, the assembly of clusters of n constituents will be characterized by a number density ρ_c. Concurrent Coulomb explosion of the multicharged clusters will result in a nearly homogeneous plasma filament with an average ion density $\rho^{(p)}=n\rho_c$. The energy deposited via absorption by a cluster located at the coordinate z is $nE^{(p)}(z)$, where $E^{(p)}(z)$ is the energy deposited per ion. The total energy deposition at z is $nE^{(p)}(z)\rho_c=E^{(p)}(z)\rho^{(p)}$. Accordingly, the energy balance equation is

$$\frac{dS(z)}{dz}=-E^{(p)}(z)\rho^{(p)}. \tag{2}$$

For elemental and molecular clusters the neutral cluster binding energy can be neglected and the energy $E^{(p)}$ absorbed by the clusters becomes equal to the sum of the ionization energy of cluster atoms $E^{(i)}$, the final kinetic energy of atomic particles (ions or nuclei) $E^{(a)}$, and the final kinetic energy of the ionized electrons $E^{(e)}$ [20,34], so that the energy balance relation is

$$E^{(p)}=E^{(i)}+E^{(a)}+E^{(e)}. \tag{3}$$

The ionization energy $E^{(i)}$ of deuterium clusters $(D_2)_{n/2}$ is $E^{(i)}=22.6$ eV per D atom [27]. This energy is considerably smaller than the deuteron energy $E^{(a)}$, which exceeds 150 eV for $n>100$ [27]. Neglecting $E^{(i)}$ one presents $E^{(p)}$ as the final energy of the Coulomb explosion products

$$E^{(p)}=E^{(a)}+E^{(e)}. \tag{4}$$

In what follows the total average energy $E^{(p)}$ will be represented in terms of the average deuteron energy $E^{(a)}$ (see Sec. III), with the energy balance for the $(D_2)_{n/2}$ cluster being given by Eq. (4). For extreme ionization of atomic or molecular clusters consisting of many-electron atoms, e.g., $(\mathrm{Xe})_n$ (which produces Xe^{q+} ions with (averaged) charge $q\simeq 7$ for $I=10^{16}$ W cm^{-2} and $q\simeq 25$ for $I=10^{18}$ W cm^{-2} [35]) or $(\mathrm{CD}_4)_n$ (which produces C^{q+} ions with $q=4$ at $I=10^{17}-10^{18}$ W cm^{-2}, and $q=6$ at $I\geqslant 10^{19}$ W cm^{-2} [28]), the ionization energy has to be included in the energy balance equation.

The energies $E^{(a)}$ (ions) and $E^{(e)}$ (electrons) of the Coulomb explosion products and, consequently, the energy $E^{(p)}$, Eq. (4), depend on the local laser intensity $I(t,z)$. In view of the two-dimensional character of the function $E^{(p)}$ $(I(t,z))$, Eqs. (1) and (2) cannot provide the general $S(z)$ versus z dependence. However, in case the product energy $E^{(p)}$ can be presented as a function of some time-integrated physical property, such as the energy flow $S(z)$ [Eq. (1)], a solution for the absorption process [Eq. (2)] can be obtained. The energy flow, Eq. (1), will then be described by the equation

$$\frac{dS(z)}{dz}=-E^{(p)}(S(z))\rho^{(p)}. \tag{2$'$}$$

Other laser and cluster parameters which determine the energy $E^{(p)}$, i.e., the laser temporal width, the light frequency and the cluster size, and also determine the energy $E^{(p)}$, are not explicitly shown in Eq. (2′).

We shall consider a Gaussian $I(t,z)$ pulse which at the spatial onset $(z=0)$ of the plasma filament is given in the form [27]

$$I(t,0)=I_M(0)\exp[-5.55(t/\tau)^2], \tag{5}$$

where $I_M(0)$ is the peak intensity at $z=0$ and τ is the temporal pulse width at the field half maximum of $0.5eF_M$. The peak electric field is $eF_M=2.74\times 10^{-7}(I_M)^{1/2}$ eV Å^{-1}, with I_M being expressed in W cm^{-2}. For a fixed laser frequency and temporal pulse width, the product energy $E^{(p)}\times(I_M(z),R_0)$ is a function of the local laser peak intensity $I_M(z)$ and the radius R_0 of the neutral cluster. This representation is valid provided that the pulse maintains its Gaussian shape along the propagation coordinate. In this case the relation between the energy flow $S(z)$ and the Gaussian peak intensity $I_M(z)$ is obtained from Eqs. (1) and (5) in the form

$$S(z)=2360\tau I_M(z). \tag{6}$$

In the derivation of Eq. (6) we took the average $\langle\cos^2 2\pi\nu t\rangle=1/2$ over the fast laser oscillations. In Eq. (6) S is expressed in eV cm^{-2}, $I_M(z)$ in W cm^{-2}, and τ in femtoseconds. Substituting Eq. (6) into Eq. (2′) we obtain an ordinary first-order differential equation

$$\frac{dI_M(z)}{dz}=-E^{(p)}(I_M(z),R_0)\rho^{(p)}/2360\tau, \tag{7}$$

where z is expressed in cm, $E^{(p)}$ in eV, and $\rho^{(p)}$ in cm^{-3}.

Equation (7) rests on the use of a Gaussian pulse, while the attenuation process deforms the pulse shape. Since the laser light energy transfer to the cluster electrons takes place mainly during the laser pulse intensity rise [18,25,31,32], i.e., at $t<0$, Eq. (5), a stronger intensity attenuation is expected at $t<0$ than at $t>0$. This contributes to the shift of the pulse peak to the $t>0$ domain. To describe such a deformation of the pulse profile, we chose the asymmetric pulse of the form

$$\tilde{I}(t)=(I_M/\Phi)\exp[-2X^2]\{1-\gamma\exp[-\theta(X-X_a)^2]\}^2, \tag{8}$$

where $X=1.665t/\tau$ and $X_a=1.665t_a/\tau$, with t_a being the parameter of the $\tilde{I}(t)$ temporal asymmetry. Φ is a normalization factor, which establishes the equality between the energy flow of the deformed pulse, Eq. (8), and the energy flow of the Gaussian pulse, Eq. (6). This normalization factor is given in the form

$$\Phi=1-2c_1\gamma\exp[-\theta c_1^2X_a^2]+c_2\gamma^2\exp[-2\theta c_2^2X_a^2], \tag{8a}$$

where $c_1=[2/(2+\theta)]^{1/2}$ and $c_2=(1+\theta)^{-1/2}$. For a pulse width of $\tau=25$ fs, we chose the following numerical parameters in Eq. (8): $t_a=-7$ fs, $\theta=2$ and $\gamma=0.7$, providing a strongly deformed pulse, with the pulse peak shifted to the $t>0$ domain (inset to Fig. 1). In order to explore the effect of the pulse shape deformation on the energy $E^{(p)}$, Eq. (4), we performed simulations [27,35] of the energetics of electrons and ions in Coulomb explosion for both the Gaussian pulse, Eq. (5), and for the deformed pulse, Eq. (8). An initially truncated pulse was used in all the simulations [27,35] with the initial laser field being located at a finite time $(t=t_s)$, where the initial

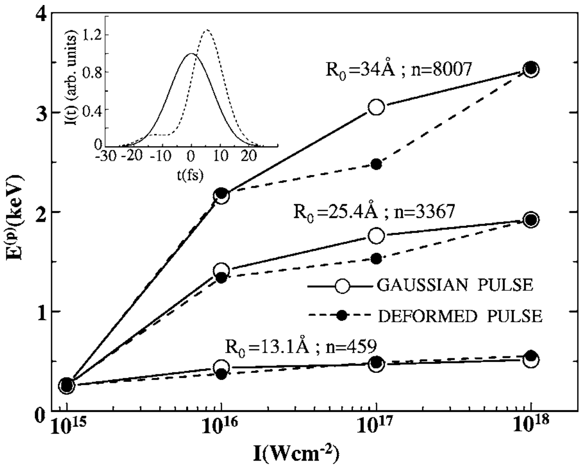

FIG. 1. Effects of the deformation of the laser pulse shape on the energetics of Coulomb explosion. The simulation results (see text) show the cluster size dependence and the intensity dependence of the energies per ion of the Coulomb explosion products $E^{(p)} = E^{(a)} + E^{(e)}$, Eq. (4), produced by the Gaussian pulse with $\tau = 25$ fs (open circles) and by the deformed pulse (black circles) with the same energy flux. The points were connected by curves (solid lines for the Gaussian pulse and dashed lines for the deformed pulse) to guide the eye. R_0(Å) denotes the initial cluster radius and n denotes the number of atoms in the $(D_2)_{n/2}$ cluster. The inset portrays the Gaussian laser pulse, Eq. (5) (solid line) and the deformed asymmetric laser pulse, Eqs. (8) and (8a), with the same energy flux, Eq. (1) (dashed line).

laser field corresponds to $(F^{\text{th}} + F^{\text{co}})/2$, where F^{th} is the threshold field for the first (single electron) ionization of D_2, and F^{co} is the threshold field for the two-electron ionization of the D_2 molecule [27,35]. The simulation results (Fig. 1) reveal that the energies $E^{(p)}$ induced by the deformed pulse differ by less than 20% from those induced by the Gaussian pulse. These small differences in the energies $E^{(p)}$ induced by the deformation of the pulse shape will be neglected. Accordingly, the energy $\tilde{E}^{(p)}(S(z))$ induced by the deformed pulse with intensity $\tilde{I}(t,z)$ and energy flow $S(z)$ is taken to be equal to the energy $E^{(p)}(S(z))$ induced by the Gaussian with the same energy flow, i.e., $\tilde{E}^{(p)}(S(z)) = E^{(p)}(I_M(z))$. Here $I_M(z)$ is related to $S(z)$ by Eq. (6). We infer that for a pulse, which is characterized by a Gaussian spatial onset [Eq. (5)], Eq. (7) is applicable for the description of the intensity variation across the propagation direction.

III. CLUSTER SIZE AND INTENSITY DEPENDENCE OF LOCAL PLASMA ENERGIES

In order to solve the differential equation for the local intensity, Eq. (7), we have to establish the cluster size and intensity dependence of $E^{(p)}(I_M(z), R_0)$, Eq. (4). The average atomic kinetic energy $E^{(a)}$ of D^+ (ions) nuclei from the Coulomb explosion of $(D_2)_{n/2}$ clusters was determined from molecular dynamics simulations for a cluster coupled to a Gaussian-shaped laser pulse, Eq. (5), with a pulse width of $\tau = 25-50$ fs and a laser frequency of $\nu = 0.35$ fs^{-1} [30]. We have shown [30] that over the entire intensity and cluster radius domains the (average) $E^{(a)}$ energies provided by the simulations are well described by the empirical relation [30]

$$E^{(a)}(I_M(z), R_0) = \alpha(4\pi/5)\bar{B}q^2\rho R_0^2[1 + (\beta R_0/R_0^{(I)})^4]^{-1/2}, \tag{9}$$

where $E^{(a)}$ is expressed in electron volts, R_0 and $R_0^{(I)}$ in angstroms, the ion charge $q=1$, the cluster atomic density $\rho = 0.05$ Å^{-3} and $\bar{B} = 14.4$ eV Å. The border radius $R_0^{(I)}$ is represented by the empirical relation [27,30]

$$R_0^{(I)} = 2.65 \times 10^{-8} \tau^{0.62} I_M^{1/2}, \tag{10}$$

where $R_0^{(I)}$ is expressed in angstroms, τ in femtoseconds and I_M in W cm^{-2}. Using Eq. (6), $R_0^{(I)}$ can be expressed in the alternative form

$$R_0^{(I)} = 5.45 \times 10^{-10} \tau^{0.12} S^{1/2} \tag{10a}$$

with the same units as in Eq. (6). The radius $R_0^{(I)}$ [the border radius for cluster vertical ionization (CVI)] separates the CVI domain $R_0 < R_0^{(I)}$ (where $E^{(a)}$ does not depend on I_M being scaled as $E^{(a)} \propto R_0^2$) from the non-CVI domain, $R_0 > R_0^{(I)}$ [27,30]. In the non-CVI domain $E^{(a)}$ depends on I_M and its increase with R_0 is slowed down, becoming saturated at $R_0 \gg R_0^{(I)}$ [27,30]. In the $R_0 \gg R_0^{(I)}$ domain, usually realized for $I_M \leqslant 10^{16}$ W cm^{-2}, the cluster outer ionization is qualitatively described by a "cold" nanoplasma model [30]. The parameters α and β of Eq. (9) depend on the pulse width τ [30]. For the parameters α and β we shall use a linear representation, which fits the α and β values of Ref. [30] ($\alpha = 0.93$ and 0.87 and $\beta = 0.52$ and 0.41 for $\tau = 25$ and 50 fs, respectively) and satisfies the condition $\alpha(\tau = 0) = 1$ (i.e., leading to the exact CVI energy for very short pulses [27])

$$\alpha(\tau) = 1.00 - 0.0027\tau, \tag{11a}$$

$$\beta(\tau) = 0.63 - 0.0044\tau. \tag{11b}$$

The simulations of the final energy $E^{(p)}$, Eq. (4), are much more time consuming than the calculations of $E^{(a)}$, and were therefore performed only for small and moderately large $(D_2)_{n/2}$ clusters, $n \leqslant 8007$. The relation between the $E^{(p)}$ and $E^{(a)}$ energies obtained herein from the simulations was fit by the empirical expression

$$E^{(p)} = [1.15 + 0.68/(1 + [R_0/R_0^{(I)}]^2)]E^{(a)}, \tag{12}$$

with $E^{(a)}$ being presented [30] by Eq. (9). The accuracy of the fitting of the simulation data by Eq. (12) is 10% (Table I). In the CVI domain ($R_0^{(I)} > R_0$) the electron energy $E^{(e)}$, in contrast to the ionic energy $E^{(a)}$, depends on the $R_0^{(I)}$ and, consequently, on the laser intensity I_M. This dependence of $E^{(e)}$ on $R_0^{(I)}$ contributes to the decrease of $E^{(p)}$, Eq. (4), with decreasing I_M for a fixed cluster size R_0. Only at high intensities I_M, in the $R_0 \ll R_0^{(I)}$ domain, $E^{(p)}$ becomes independent on I_M with $E^{(p)}/E^{(a)} = 1.83$. This ratio is close to the experimental ratio $E^{(p)}/E^{(a)} = 1.8$ obtained by Eloy *et al.* [36], and is somewhat higher than the ratio $E^{(p)}/E^{(a)} = 1.5$ reported by Parks *et al.* [20]. When the intensity I_M decreases due to the

TABLE I. The ratio $(E^{(p)})_{SIM}/(E^{(p)})_{FIT}$ between the final total energies (of nuclei and electrons) of the products of Coulomb explosion of $(D_2)_{n/2}$ clusters provided by simulations, $(E^{(p)})_{SIM}$, and by the fitting to Eq. (12), $(E^{(p)})_{FIT}$. I_M(W cm^{-2}) is the pulse peak intensity, R_0 (Å) is the cluster initial radius, and n is the number of cluster atoms.

		I_M(W cm^{-2})			
		10^{15}	10^{16}	10^{17}	10^{18}
R_0 (Å)	n		$(E^{(p)})_{SIM}/(E^{(p)})_{FIT}$		
8.15	135	1.152	1.052	1.039	1.127
13.1	459	0.931	0.921	0.905	0.973
21.1	1961	1.020	0.981	0.943	1.007
25.4	3367	0.988	0.975	0.958	0.973
34.0	8007	—	1.008	0.969	0.980

light absorption, and $R_0 \lesssim R_0^{(I)}$, the ratio $E^{(p)}/E^{(a)}$ begins to decrease, reaching the value of $E^{(p)}/E^{(a)}=1.49$ at $R_0^{(I)}=R_0$. The $E^{(p)}/E^{(a)}$ ratio continues to decrease in the non-CVI domain, $R_0^{(I)}<R_0$. In the "cold" nanoplasma domain, $R_0^{(I)} \ll R_0$, $E^{(p)}/E^{(a)}$ is saturated at the $E^{(p)}/E^{(a)}=1.15$ level, Eq. (12).

IV. NUMERICAL RESULTS FOR THE ATTENUATION OF THE LASER INTENSITY

The plasma filament will be described by a cylinder of radius r and length h (with $r \ll h$). The attenuation of the laser intensity and the absorbed energy in an assembly of clusters can be expressed analytically for two cases of physical interest: in the CVI domain $R_0^{(I)} \gg R_0$, and in the "cold" nanoplasma domain $R_0^{(I)} \ll R_0$. In the CVI domain $R_0^{(I)} \gg R_0$, Eq. (12) results in $E^{(p)}=1.83E^{(a)}$ with $E^{(a)}=(4\pi/5)\alpha\bar{B}\rho R_0^2$, Eq. (9), being independent of I_M [16,22–24,37]. The solution of Eq. (7) becomes

$$I_M(z) = I_M(0)(1 - z/Z_A); \quad z < Z_A; \quad z \leq h \tag{13}$$

with

$$Z_A = 1290\tau I_M(0)/\rho^{(p)}E^{(a)}. \tag{13a}$$

Z_A is the attenuation parameter (in cm), with $\rho^{(p)}$ in cm^{-3} and $E^{(a)}$ in eV. Thus, in the CVI domain $I_M(z)$ manifests a linear decrease with increasing z. According to Eq. (6) the energy absorbed by the plasma filament is

$$W_{abs} = 2360\pi r^2 \tau[I_M(0) - I_M(h)], \tag{14}$$

where r and h are given in cm. Using Eqs. (13) and (14) and considering the case of partial light absorption, i.e., $h<Z_A$, one obtains $W_{abs}=\pi r^2 h\rho^{(p)}(1.83E^{(a)})=\pi r^2 h \rho^{(p)}E^{(p)}$, which results in

$$W_{abs} = N^{(p)}E^{(p)}, \tag{15}$$

where $N^{(p)}=\pi r^2 h\rho^{(p)}$ is the number of ions (deuterons) in the plasma filament. This last expression, which also follows from energy conservation arguments, demonstrates the independence of the absorbed energy W_{abs} on the laser intensity $I_M(0)$ for weak light absorption and for the CVI conditions (when $E^{(p)}$ is independent of I_M). In the foregoing analysis of the CVI domain, the decrease of $I_M(z)$ with increasing z leads to the decrease of the border radius $R_0^{(I)}$, Eq. (10). Thus, at some value of z, $R_0^{(I)}$ may become smaller than R_0, violating the CVI conditions. Accordingly, Eq. (13) is valid for $R_0^{(I)}(I_m(z))>R_0$. Using Eqs. (10) and (13) the validity condition for Eq. (13) is

$$Z_A - z > \frac{7.3 \times 10^{17}}{\alpha\bar{B}\rho^{(p)}\rho\tau^{0.24}}, \tag{13b}$$

where $\rho^{(p)}$ is in cm^{-3}, ρ in Å^{-3}, τ in fs and $I_M(0)$ in W cm^{-2}. We note that this limit, Eq. (13b), is independent of $I_M(0)$. For typical values of $\tau=25$ fs, $\rho^{(p)}=2\times10^{19}$ cm^{-3} we obtain $Z_A-z=0.025$ cm.

In the saturation domain of the ion kinetic energy $E^{(a)}$, $R_0^{(I)} \ll R_0$, Eqs. (8) and (10) provide

$$E^{(p)} = 2.0 \times 10^{-15}\alpha\beta^{-2}\bar{B}\rho\tau^{1.24}I_M. \tag{16}$$

Substituting this expression into Eq. (7) one obtains the exponential distance (z) dependence

$$I_M(z) = I_M(0)\exp[- az], \tag{17}$$

where

$$a = 8.6 \times 10^{-19}\rho^{(p)}\alpha\bar{B}\rho\beta^{-2}\tau^{0.24}. \tag{17a}$$

The units in Eq. (17a) are as follows: $\rho^{(p)}$ in cubic centimeters, ρ in Å^{-3}, τ in fs, and a in cm^{-1}. The absorbed energy by the plasma filament, Eq. (12), for the saturation domain is

$$W_{abs} = 2360\pi r^2\tau(1 - \exp[- ah])I_M(0). \tag{18}$$

Equation (18) implies that in the kinetic energy saturation domain the absorbed energy is proportional to the incident laser peak intensity $I_M(0)$ and the flux $S(0)$. This saturation regime is expected to be realized at lower intensities. Such a linear dependence of W_{abs} versus $I_M(0)$ was found by Santra and Greene [32], who analyzed experimental data for Xe_n clusters at moderately low intensities of $I_M \leq 10^{14}$ W cm^{-2}. At these intensities mostly singly charged Xe^+ ions were produced [32]. This bears similarity to the case of D^+ ions from $(D)_n$ clusters, where the kinetic energy saturation condition is expected to be satisfied.

The parameters for the plasma filament under the experimental conditions of the Lawrence-Livermore experiments [10,37] with $\tau=35$ fs correspond to deuteron density $\rho^{(p)}=2\times10^{19}$ cm^{-3} [37], filament length $h=0.2$ cm, and filament radius $r=0.1$ cm [10]. For a rough estimate at $I_M(0)=10^{18}$ W cm^{-2} [where Eq. (10) gives $(R_0)_I=195$ Å] we choose $R_0=60$ Å ($R<R_0^{(I)}$), which corresponds to the CVI domain. Under these conditions $E^{(a)}=5.90$ keV and Eq. (13a) results in $Z_A=0.43$ cm, which is almost twice as large as h and indicates the lack of absorption. Of course, when $Z_A<h$, the absorption becomes strong, i.e., $I_M(z=h) \ll I_M(0)$, and the linear approximation, Eq. (13), has to be extended. In the general case, the local intensity $I_M(z)$ and the deuteron energy $E^{(a)}(z)$ were found by a numerical solu-

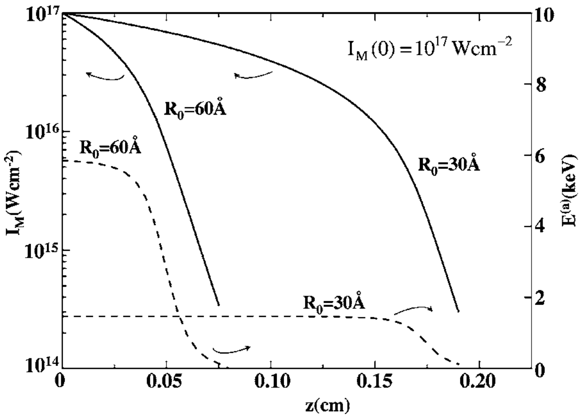

FIG. 2. The local laser peak intensity $I_M(z)$ obtained from the solution of Eq. (7), and the average deuteron kinetic energy $E^{(a)}$ obtained from molecular dynamics simulations for an assembly of $(D_2)_{n/2}$ clusters with $R_0=30$ Å ($n=5650$) and $R_0=60$ Å ($n=45\,200$) in a plasma filament ($\ell=0.2$ cm and $\rho^{(p)}=2\times10^{19}$ cm^{-3} [10,37]). The incident peak intensity was $I_M(0)=10^{17}$ W cm^{-2}.

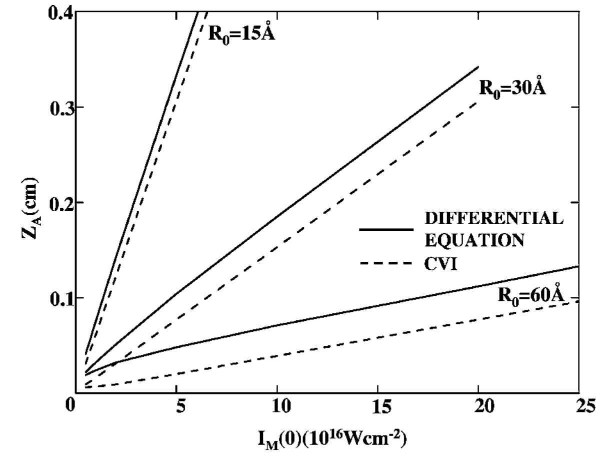

FIG. 3. The total absorption distance Z_A for an assembly of $(D_2)_{n/2}$ clusters with $R_0=15$, 30, and 60 Å, respectively, obtained from the solution of Eq. (7) (solid lines) and from the CVI expression, Eq. (13a) (dashed lines).

tion of Eq. (7). The results are presented in Fig. 2 for $\tau=25$ fs and $\rho^{(p)}=2\times10^{19}$ cm^{-3} of the Lawrence-Livermore experiment [37], and for the initial intensity of $I_M(0)=10^{17}$ W cm^{-2}. Clusters with a smaller radius of $R_0=30$ Å absorb about 50% of the energy flow S in the middle of the plasma filament, at $z\approx0.1$ cm (Fig. 2). However, this intensity decrease is not accompanied by a noticeable decrease in the $E^{(a)}$ energy. The $E^{(a)}$ energy decrease starts only at $z\approx0.16$ cm, when I_M drops below the level of 10^{16} W cm^{-2}, and where a persistent nanoplasma is produced within the clusters [26]. Clusters with the larger radius of $R_0=60$ Å strongly absorb the light so that the total absorption is reached in the middle of the plasma filament, at $z=0.1$ cm (Fig. 2). In this case the drop in the energy flow (or the intensity variable I_M) is accompanied by the steep decrease of the deuteron energy $E^{(a)}$, Eq. (9).

The total absorption distances $Z_A(I_M(Z_A)=0)$ (as a zero level we accept the atom ionization intensity $I_M=5\times10^{14}$ W cm^{-2} [35]), which were determined by the numerical solution of Eq. (7) and by the CVI condition, Eq. (13a), are presented in Fig. 3 for the parameters presented above. The CVI approach, Eq. (13a), overestimates the light attenuation, providing smaller Z_A values. The "exact" solution, Eq. (7), shows that the light is totally absorbed inside the plasma filament $h=0.2$ cm at the laser intensities $I_M(0)<3\times10^{16}$, 10^{17}, and 4×10^{17} W cm^{-2}, for a cluster size of $R_0=15$, 30, and 60 Å, respectively. The experimentally determined average cluster radius for almost total absorption is $R_0\approx24$ Å (Fig. 2 of Ref. [10]) which, according to our results, corresponds to the laser intensity of $I_M(0)\approx6\times10^{16}$ W cm^{-2}. This laser intensity lies between the average peak intensity estimated as $I_M\approx10^{16}$ W cm^{-2} and the maximal peak intensity of $I_M=5\times10^{17}$ W cm^{-2} in the Lawrence-Livermore experiment [10]. The energy absorbed in the plasma filament for the condition of 90% absorption was found to be $E^{(p)}\approx5$ keV per atom [10]. Our theoretical treatment, Eqs. (9) and (12), indicates that in the absence of absorption such energy can be provided by Coulomb explosion of clusters with $R_0\approx40$ Å, a radius which is considerably larger than the radius of $R_0=24$ Å reported by Zweiback *et al.* [10]. It should be emphasized that our theoretical results, at least for this case, are based on simple electrostatic arguments of the CVI approach [27,30], and on our findings that the ion kinetic energy $E^{(a)}$ is larger than the electron energy $E^{(e)}$, as confirmed by the experimental results of Zweiback *et al.* [10].

V. ENERGY DISTRIBUTION IN THE COULOMB EXPLOSION OF AN ASSEMBLY OF CLUSTERS

Light absorption effects significantly affect the kinetic energy distribution $P(E)(\equiv P(E^{(a)}))$ of the product ions by increasing the number of low-energy ions, whereas the maximal energy of the product ions is expected to be close to the maximal energy $(E_M(I_M(0))$ in the absence of absorption. For the kinetic energy distribution $P^{(s)}(E)$ of a single cluster we take a two component fitting equation [30]

$$P(E)=C_1P_1(E)+(1-C_1)P_2(E), \tag{19}$$

where $P_1(E)$ is the low-energy component

$$P_1(E)=\frac{4\sqrt{2}}{\pi E^{(a)}}\Big/[1+(2E/E^{(a)})^4] \tag{20}$$

with the average energy $E^{(a)}$ determined by Eq. (9). $P_2(E)$ is the CVI energy distribution

$$P_2(E)=\frac{3}{2E_M}(E/E_M)^{1/2},\quad E\leqslant E_M$$

$$P_2(E)=0,\quad E>E_M \tag{21}$$

with the maximal energy E_M [30] being

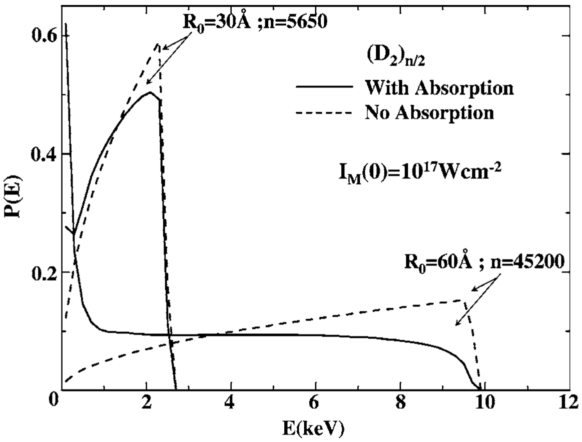

FIG. 4. The effect of light absorption on the distribution of the kinetic energies of D^+ ions in the Coulomb explosion of an assembly of $(D_2)_{n/2}$ clusters. Calculations were conducted for $R_0=30$ Å ($n=5650$) and for $R_0=60$ Å ($n=45200$). Solid lines: Molecular dynamics simulations for the kinetic energy distribution of the D^+ ions, accounting for the attenuation of the laser intensity by the solution of Eq. (7). Laser incident peak intensity $I_M(0)=10^{17}$ W cm^{-2} and laser pulse duration $\tau=25$ fs. Parameters of plasma filament as in Fig. 2. Dashed lines: Molecular dynamics simulations for the kinetic energy distribution of the D^+ ions at a fixed laser local peak intensity $I_M(z)=10^{17}$ W cm^{-2}. These data without absorption effects correspond to the CVI conditions.

$$E_M = \alpha \frac{4\pi}{3}\bar{B}\rho R_0^2 \Bigg/ \left[1+\left(\frac{\tilde{\beta}R_0}{R_0^{(I)}}\right)^4\right]^{1/4} \qquad (22)$$

with $\tilde{\beta}=0.60$ for $\tau=25$ fs. The coefficient C_1 in Eq. (19), which determines the contribution of the first, low-energy term $P_1(E)$, Eq. (20), is given by

$$C_1 = \left(\frac{3}{5}E_M - E^{(a)}\right) \Bigg/ \left(\frac{3}{5}E_M - \frac{1}{2\sqrt{2}}E^{(a)}\right). \qquad (23)$$

The effect of light absorption on the energy distribution of the filament D^+ nuclei is demonstrated in Fig. 4. For two different cluster sizes ($R_0=30$ Å and $R_0=60$ Å) subjected to $I=10^{17}$ W cm^{-2}, the smaller cluster, $R_0=30$ Å, represents the case of weak absorption (Fig. 2), when the filament energy distribution differs only slightly from the energy distribution of a single cluster in the absence of absorption (Fig. 4). At $R_0=60$ Å, the absorption is strong and the laser light is totally absorbed at $z=0.1$ cm (Fig. 2). In this case the filament energy distribution exhibits marked deviations from the distribution of a single cluster subjected to $I_M(0)$, with a sharp rise in the low-energy range (Fig. 4), which leads to a low-energy maximum. A low-energy divergence of $P(E)$ is excluded on the basis of the preservation of the normalization of the energy distribution. The location of the low-energy rise in the energy distribution $P(E)$ provides a new criterion in the assessment of the contribution of the absorption, with the high-energy maximum at $E\sim E_M$ representing weak absorption, while a low-energy rise at $E \ll E_M$ manifests strong absorption. Such a behavior of $P(E)$ for weak absorption is demonstrated in the experiments of Sakabe *et al.* [38]. In the experiments of Madison *et al.* [14] the average energy of the hot ions is $E_{av}=2.1$ keV, whereas the maximum in $P(E)$ lies at a much lower energy of ~ 0.2 keV. The shift of the distribution maximum to low energies, which is comparable to E_{av}, probably indicates strong light absorption. However, the deformation of the shape of the energy distribution can also be affected by the cluster size distribution, which has to be taken into account in order to allow for a more precise analysis of the energy distribution reported by Madison *et al.* [14]. The energetics of the Coulomb explosion in an ensemble of size-distributed clusters will be considered in a forthcoming paper [39].

VI. LIGHT ABSORPTION BY A SINGLE CLUSTER

Our treatment of laser light absorption in a plasma filament rests on the assumption that the light absorption by a single cluster is not sufficiently strong to attenuate the intensity within a single cluster. In what follows, we shall establish the conditions for the validity of this assumption. The attenuation of the laser light within a single $(D)_n$ cluster is expected to be weak provided that

$$E_{tot} \ll E_{f_\ell}, \qquad (24)$$

where E_{tot} is the total energy absorbed by the cluster, while $E_{f\ell}$ is the laser energy flow through the cluster. Following the analysis of Sec. II we set $E_{tot}=nE^{(p)}$, where $n=(4\pi\rho/3)R_0^2$ is the number of atoms and $\rho=0.05$ Å^{-3}, while $E^{(p)}$, the energy absorbed per atom, is given by Eq. (4). The laser energy flow through a cluster of radius R is $E_{f\ell}=\pi R^2\int_{-\infty}^{\infty}I(t)dt$. Ignoring the cluster expansion on the time scale of strong energy absorption (i.e., the time scale of outer ionization), we express $E_{f\ell}$ by Eq. (14) in the form $E_{f\ell}=2360\pi R_0^2\tau I_M$ Equation (24) results in the condition for weak light attenuation within the cluster

$$\rho R_0 E^{(p)} \ll 1.77\times 10^{-13}\tau I_M, \qquad (25)$$

where $E^{(p)}$ is expressed in eV, R_0 in Å, τ in fs, and I_M in W cm^{-2}. In the CVI domain, $R_0 \leq R_0^{(I)}$, the condition for weak light attenuation is ensured, provided that the inequality, Eq. (25), is satisfied for $R_0=R_0^{(I)}$. According to Eqs. (9) and (12), the absorbed energy at $R_0=R_0^{(I)}$ is $E^{(p)}=3.74\bar{B}\rho(R_0^{(I)})^2$ Substituting this expression for $E^{(p)}$ into Eq. (2) and presenting $R_0^{(I)}$ by I_M and τ, Eq. (10), we obtain

$$I_M \ll 5\times 10^{21}\tau^{-1.72}, \qquad (26)$$

where I_M is in units of W cm^{-2} and τ in fs. Equation (26) provides the upper limit of the intensity which ensures weak single-cluster absorption in the CVI domain. For the pulse width of $\tau=25$ fs the laser peak intensity has to be smaller than 2×10^{19} W cm^{-2}, which corresponds to a cluster border radius of $R_0^{(I)}=870$ Å. This result indicates that in the CVI domain the attenuation of the laser field inside a single cluster will be manifested only for very high intensities (and for large clusters with $n\sim 10^8$). Accordingly, the effects of light attenuation in the CVI domain are expected to be small under any experimental and computational conditions. Next we consider the non-CVI "cold" nanoplasma domain where $R_0 \gg R_0^{(I)}$. In this case, $E^{(p)} \propto I_M$, according to Eq. (16). Equation (25) provides a condition for weak attenuation, which is valid for any laser intensity I_M, and which sets a lower limit for the cluster radius in the form

$$R_0 \ll 88.5/(\alpha\beta^{-2}\bar{B}\rho^2\tau^{0.24}) \quad (27)$$

with α and β being expressed by Eqs. (11a) and (11b), respectively. For $\tau=25$ fs, Eq. (27) results in the condition $R_0 \ll 330$ Å in the non-CVI "cold" nanoplasma domain. The cluster size corresponds to $n \sim 5\times10^6$. From the practical point of view we assert that for the cluster sizes considered in the present work the attenuation of light within a single cluster is expected to be small, also in the non-CVI regime. From the point of view of methodology, a search for laser light absorption by a single deuterium cluster in an electromagnetic trap should be conducted for the non-CVI domain. The estimates of $R_0 \simeq 330$ Å ($n\sim5\times10^6$) for the prevalence of single cluster absorption can be realized for large, but physically accessible, deuterium clusters.

VII. CONCLUDING REMARKS

The absorption of ultraintense laser radiation by an assembly of clusters of the same size is different in different regions of laser intensities and cluster sizes, which govern distinct domains of the cluster size dependence of the energetics of Coulomb explosion. These domains are determined by the CVI border radius $R_0^{(I)}$, whose dependence on the laser intensity is $R_0^{(I)}(I_M) \propto I_M^{1/2}$, according to Eq. (10). While in our previous work [30,35] we considered the energetics of Coulomb explosion with increasing the cluster radius at a fixed laser intensity I_M, it will now be useful to consider a fixed cluster size R_0 and define from Eq. (10) the border intensity $I_M^{(R)}$ for which the CVI breaks down at a fixed value of R_0. This border intensity is given by

$$I_M^{(R)} = 1.42\times10^{15}\tau^{-1.24}R_0^2. \quad (28)$$

When the CVI validity conditions, i.e., $R_0 < R_0^{(I)}(I_M)$ or $I_M > I_M^{(R)}(R_0)$, are obeyed along the entire plasma filament up to $z=h$, the absorbed energy, Eq. (15), does not depend on the initial light intensity $I_M(0)$ and absorption does not significantly affect the ion energy. In the opposite domain, when $R_0 \gg R_0^{(I)}(I_M(0))$ or $I_M(0) \ll I_M^{(R)}(R_0)$ the intensity $I_M(z)$ decreases exponentially with z, according to Eq. (17). This decrease of $I_M(z)$ is accompanied by the decrease of the ion energy. Total absorption is realized for $ah \gg 1$. When the absorption is weak, i.e., $ah \ll 1$, the absorbed energy is proportional to the initial intensity $I_M(0)$, Eq. (18). In the intermediate domain, when $R_0 \gtrsim R_0^{(I)}(I_M(0))$ the local intensity $I_M(z)$

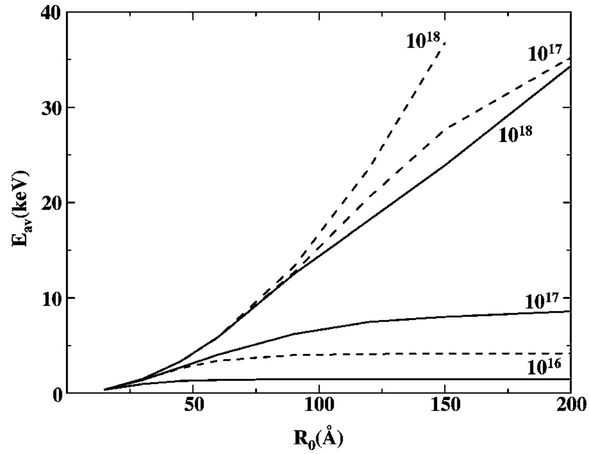

FIG. 5. The effect of light absorption on the cluster size dependence of the average energies of the D^+ ions in the Coulomb explosion of $(D_2)_{n/2}$ clusters. Laser incident peak intensities $I_M(0) = 10^x$ W cm^{-2} ($x=16-19$) are marked on the curves. Solid lines: Molecular dynamics simulations accounting for the attenuation of the laser intensity by the solution of Eq. (7). Laser pulse duration $\tau=25$ fs. Parameters of plasma filament as in Fig. 2. Dashed lines: Molecular dynamics simulations in the absence of absorption.

becomes smaller than the border intensity $I_M^{(R)}(R_0)$ at some distance $z<h$, and the absorption features cannot be described in a simple way.

Light absorption may affect the dependence of the ion energy on the cluster radius R_0. In the case of weak absorption, the average ion energy $E^{(a)}$ increases as R_0^2 in the CVI domain, while in the $R_0 \gg R_0^{(I)}$ domain this dependence reaches saturation [Eq. (9)]. Such behavior is also manifested when absorption effects are significant. However, the saturation onset is exhibited at a lower value of R_0 than in the absence of absorption, and the energies in this domain are smaller (Fig. 5). The early transition from the $E^{(a)} \propto R_0^2$ dependence to the saturation behavior in an assembly of clusters may be responsible for the presence of a maximum in the neutron yield dependence on the cluster radius R_0 observed in the NFDCE experiments of Zweiback *et al.* [10]. Direct calculations of the absorption effects of the ultraintense laser radiation on the neutron yield, which will be presented in a forthcoming paper [39], will provide information on the cluster size dependence of the neutron yield.

ACKNOWLEDGMENTS

This research was supported by the Deutsche Forschungsgemeinschaft (DFG) SFB 450 on "Analysis and Control of Ultrafast Photoinduced Reactions" and by the Binational German-Israeli James Franck Program on Laser-Matter Interaction.

[1] T. Ditmire, T. Donnelly, A. M. Rubenchik, R. W. Falcone, and M. D. Perry, Phys. Rev. A **53**, 3379 (1996).
[2] T. Ditmire, J. W. G. Tisch, E. Springate, M. B. Mason, N. Hay, R. A. Smith, J. Marangos, and M. H. R. Hutchinson, Nature (London) **386**, 54 (1997).
[3] T. Ditmire, E. Springate, J. W. G. Tisch, Y. L. Shao, M. B. Mason, N. Hay, J. P. Marangos, and M. H. R. Hutchinson, Phys. Rev. A **57**, 369 (1998).
[4] M. Lezius, V. Blanchet, D. M. Rayner, D. M. Villeneuve, A. Stolow, and M. Yu. Ivanov, Phys. Rev. Lett. **86**, 51 (2001).
[5] E. Springate, N. Hay, J. W. G. Tisch, M. B. Mason, T. Ditmire, M. H. R. Hutchinson, and J. P. Marangos, Phys. Rev. A **61**, 063201 (2000).
[6] J. Schulz, H. Habnitz, T. Laarmann, S. Gürtler, W. Laasch, A. Swiderski, Th. Möller, and A. A. B. de Castro, Nucl. Instrum. Methods Phys. Res. A **507**, 572 (2003).
[7] M. Krishnamurthy, D. Mathur, and V. Kumarappan, Phys. Rev. A **69**, 033202 (2004).
[8] T. Laarman, A. R. B. de Castro, P. Gürtler, W. Laasch, J. Schulz, H. Wabnitz, and T. Möller, Phys. Rev. Lett. **92**, 143401 (2004).
[9] J. V. Ford, O. Zhong, L. Poth, and A. W. Castleman, Jr., J. Chem. Phys. **110**, 6257 (1999).
[10] J. Zweiback, R. A. Smith, T. E. Cowan, G. Hays, K. B. Wharton, V. P. Yanovsky, and T. Ditmire, Phys. Rev. Lett. **84**, 2634 (2000).
[11] D. A. Card, E. S. Wisniewski, D. E. Folmer, and A. W. Castleman, Jr., J. Chem. Phys. **116**, 3554 (2002).
[12] G. Grillon, Ph. Balcou, J.-P. Chambaret, D. Hulin, J. Martino, S. Moustaizis, L. Notebaert, M. Pittman, Th. Pussieux, A. Rousse, J.-Ph. Rousseau, S. Sebban, O. Sublemontier, and M. Schmidt, Phys. Rev. Lett. **89**, 065005 (2002).
[13] S. Teuber, T. Döppner, T. Fennel, J. Tiggesbäumker, and K. H. Meiwes-Broer, Eur. Phys. J. D **16**, 59 (2001).
[14] K. W. Madison, P. K. Patel, M. Allen, D. Price, and T. Ditmire, J. Opt. Soc. Am. B **20**, 113 (2003).
[15] K. W. Madison, P. K. Patel, D. Price, A. Edens, M. Allen, T. E. Cowan, J. Zweiback, and T. Ditmire, Phys. Plasmas **11**, 270 (2004).
[16] K. W. Madison, P. K. Patel, M. Allen, D. Price, R. Fitzpatrick, and T. Ditmire, Phys. Rev. A **70**, 053201 (2004).
[17] I. Last and J. Jortner, Phys. Rev. A **62**, 013201 (2000).
[18] K. Ishikawa and T. Blenski, Phys. Rev. A **62**, 063204 (2000).
[19] J. Liu, R. Li, P. Zhu, Zh. Xu, and J. Liu, Phys. Rev. A **64**, 033426 (2001).
[20] P. B. Parks, T. E. Cowan, R. B. Stephens, and E. M. Campbell, Phys. Rev. A **63**, 063203 (2001).
[21] V. P. Krainov and A. S. Roshchupkin, Phys. Rev. A **64**, 063204 (2001).
[22] I. Last and J. Jortner, Phys. Rev. A **64**, 063201 (2001).
[23] V. P. Krainov and M. B. Smirnov, Phys. Rep. **370**, 237 (2002).
[24] Ch. Siedschlag and J. M. Rost, Phys. Rev. A **67**, 013404 (2003).
[25] Z. Jurek, G. Faigel, and M. Tegze, Eur. Phys. J. D **29**, 217 (2004).
[26] I. Last and J. Jortner, J. Chem. Phys. **120**, 1348 (2004).
[27] I. Last and J. Jortner, J. Chem. Phys. **121**, 3030 (2004).
[28] I. Last and J. Jortner, J. Chem. Phys. **121**, 8329 (2004).
[29] Ch. Jungreuthmayer, M. Geissler, J. Zanghellini, and T. Brabec, Phys. Rev. Lett. **92**, 133401 (2004).
[30] I. Last and J. Jortner, Phys. Rev. A **73**, 013202 (2006).
[31] D. Bauer and A. Macchi, Phys. Rev. A **68**, 033201 (2003).
[32] R. Santra and Ch. V. Greene, Phys. Rev. Lett. **91**, 233401 (2003).
[33] T. Taguchi, T. M. Antonsen, and H. M. Milchberg, Phys. Rev. Lett. **92**, 205003 (2004).
[34] Ch. Siedschlag and J. M. Rost, Phys. Rev. Lett. **93**, 043402 (2004).
[35] I. Last and J. Jortner, J. Chem. Phys. **120**, 1336 (2004).
[36] M. Eloy, R. Azambuja, T. M. Mendonca, and R. Bingham, Phys. Plasmas **8**, 1084 (2001).
[37] J. Zweiback, T. E. Cowan, R. A. Smith, J. H. Hartley, R. Howell, C. A. Steinke, G. Hays, K. B. Wharton, J. K. Krane, and T. Ditmire, Phys. Rev. Lett. **85**, 3640 (2000).
[38] S. Sakabe *et al.*, Phys. Rev. A **69**, 023203 (2004).
[39] I. Last and J. Jortner (unpublished).