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Ionic recoil energies in the Coulomb explosion of metal clusters

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Abstract. The photoionization of metal clusters in intense femtosecond laser fields has been studied. In contrast to an experiment on atoms, the interaction in this case leads to a very efficient and high charging of the particle where tens of electrons per atom are ejected from the cluster. The recoil energy distribution of the atomic fragment ions was measured which in the case of lead clusters exceeds 180 keV. Enhanced charging efficiency which we observed earlier for specific pulse conditions is not reflected in the recoil energy spectra. Both the average and the maximum energies decrease with increasing laser pulse width. This is in good agreement with molecular dynamics calculations.

PACS. 36.40.Vz Optical properties of clusters – 36.40.Gk Plasma and collective effects in clusters – 52.40.-w Plasma interactions (nonlaser)

1 Introduction

Recently, the interaction of intense femtosecond laser light pulses with metal clusters has opened up a novel direction in cluster physics. Due to the strong nonlinear behavior of nano–sized matter new and interesting phenomena have been observed. The high atomic density within a cluster provides the possibility to benefit from solid state many particle effects in a molecular beam. It was shown, that the absorption of laser energy by the cluster beam can reach up to 100% [1]. Remarkably nuclear fusion was observed when irradiating deuterium clusters by ultrashort light pulses [2]. Thus it seems possible that new light and neutron sources and eventually an X–ray laser can be realized, built up from this concept.

When a cluster absorbs an essential fraction of the laser light, which typically is applied with pulse intensities in the 10^{14}–10^{18} W/cm^2 range, the energy deposited into the particles may lead to huge ionization states (*e.g.* $z = 20$ per atom for platinum [3] and xenon [4] clusters), and X–ray emission [5]. The Coulomb repulsion between the charges results in high kinetic energies of ions [6] and electrons [7]. Several theoretical models were introduced to explain the results on rare gas clusters. McPherson *et al.* [8] described the produced inner–shell vacancies leading to prompt X–ray emission by quasi–particle effects. Whereas Rose-Petruck *et al.* [9] introduced an ionization ignition model, the hydrodynamic approach by Ditmire *et al.* [10] treats the cluster as a spherical nanoplasma. Last and Jortner [11] explained the enhanced charging by a quasiresonance ionization mechanism. Furthermore they found that the recoil energy of the resulting ionic fragments increases superlinearly with the cluster size and is proportional to the square of the charge, *e.g.*, a 40 fold charged atomic ion from Xe_{55} has a final kinetic energy of 70 keV. For comparison, Ditmire *et al.* [12] measured recoil energies up to 250 keV from the explosion of xenon clusters having approximately 400 atoms.

When considering the interaction of light with metal clusters the delocalization of the valence electrons should allow collective electron motion. Excitations in the energy range close to the corresponding dipole resonance have been found to exhibit huge absorption cross sections of about 1 Å$^2/s$–electron [13]. Recently several attempts were made to calculate the optical response of metal clusters to the irradiation by intense laser fields [14–16]. Some calculations agree that excitations close to and on the plasmon resonance lead to enhanced ionization. This holds true for systems which initially are off–resonance, but, upon expansion due to Coulomb forces, match the photon energy after a given time.

In this contribution we report on the measured kinetic energy spectra of ions ejected from the Coulomb explosion of metal clusters irradiated by intense femtosecond laser pulses with variable laser pulse duration. These results are compared with molecular dynamics calculations.

2 Experimental setup

A Ti:sapphire chirped–pulse amplification laser system delivers sub–100 fs pulses at a wavelength of 800 nm (corresponding to a photon energy of 1.54 eV) with a pulse energy of up to 25 mJ. The light pulses are focused into the time–of–flight (TOF) chamber of a cluster

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beam machine [17]. With a $f/50$ lens peak intensities up to 1.5×10^{16} W/cm^2 can be achieved. A variation of the distance between the compressor gratings in the amplifier leads to a change in the pulse width from 80 fs up to 10 ps.

A Pulsed Arc Cluster Ion Source (PACIS) [18,19] is used to produce the metal clusters. The coexpansion of a metal arc plasma and helium carrier gas into the vacuum results in a supersonic cluster beam. The neutral part is irradiated by the laser pulses, which enters the interaction region perpendicularly to the molecular beam axis. With this setup no distinct mass selection can be achieved. However, by tuning the source conditions, the size distribution as measured by the ionic part of the beam, can be adjusted. For the experiment described below, we will use three size ranges: (a) $N = 1...50$ (small), (b) $N = 5...150$ (medium–sized), and (c) $N = 50...1000$ (large). Note that this bracketing is extremely rough. Still we will see a significant dependence of the recoil energies on this size modification. The kinetic energy of the resulting ions is determined by the flight time through a field free drift tube of 40 cm length. In order to reduce the noise caused by secondary electrons and moreover to restrict the detection of ions to only those originating from the Rayleigh region of the laser focus, the entrance of the flight tube is confined by an adjustable narrow slit.

3 Molecular dynamics calculations

In order to describe the motion of the cluster ions after exposure to the strong laser field a molecular dynamics code was developed. With the intention of calculating the Coulomb explosion of charged particles with up to several hundreds of atoms, the modeling of the physical processes has to be strongly simplified. The cluster consists of N particles representing the heavy atomic cores. Any contribution of the electrons, *i.e.* the ion–electron and the electron–electron interactions, is neglected. The particle motion is calculated based on Newton's laws. In contrast to our earlier approach in Ref. [20], the two body Morse potential function is used in the code which should give a better representation of the binding forces. Furthermore, the geometrical ground state configuration is explicitly calculated where the parameters of the potential are fitted to referenced data in order to reflect binding length, energy and oscillation frequency of the neutral dimer. The system is driven by potential gradients of the molecular and Coulomb fields. In the numerical algorithm a kinetic energy correction is implemented to force energy conservation. The initial geometrical configuration must be generated by a dynamic structure simulation. First the atoms are randomly distributed in a sphere and subsequently allowed to relax. A damping term in the dynamical equations cools the system and it will find a structural energy minimum within a few picoseconds. Since the number of isomers increases rapidly with the cluster size an ensemble of starting configurations is used in the calculations.

The Coulomb explosion of the cluster is ignited by starting the charging process by, *e.g.*, tunneling or field ionization. Assuming a metallic plasma, and thus homogeneous charge distribution, all atoms in the cluster are equally charged. The time dependence of the ionization is given by a charging function having two adjustable parameters, see section 4. The accessible data of the model are the explicit positions and velocities for all particles at any time of the calculation. Hence the simulation yields information about the kinetic/potential particle energy spectrum, the cluster temperature, and the ion density function.

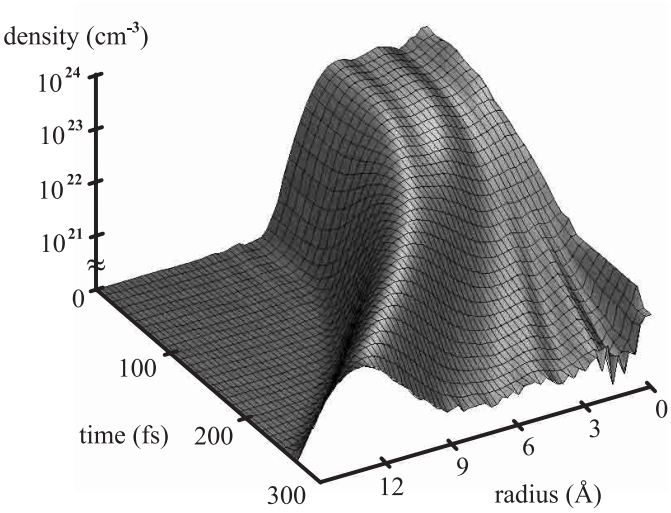

Fig. 1. Calculated time-dependent ion density of a Pt_{55} which is charged up to $z = 110$ within 300 fs. A significant expansion starts only after 80 fs.

As an example, the resulting ion density of a highly charged Pt_{55} cluster is illustrated in Fig. 1. Note that the position of the charged particles remains constant in the first 80 fs and then the cluster undergoes a rapid expansion. This time scale is in good agreement with those found by Reinhard *et al.* [21] using the non–linearized time–dependent local density approximation.

4 Results and discussion

In earlier mass and charge resolved experiments [22] we have shown that metal clusters under conditions chosen here disassemble completely into atomic ions and electrons. Therefore, a time–of–flight spectrum of such a process without using any acceleration field directly maps the energy distribution of the ions. In principle the electron energy can also be resolved, however, this is not a subject of this contribution. Fig. 2a shows a typical time–of–flight spectrum from large lead clusters irradiated with laser pulses of 3×10^{15} W/cm^2. The data exhibit a fast feature for short flight times which is mainly caused by the arrival of energetic electrons, and possibly the generation of high energy photons. The broad feature beyond 1 μs corresponds to the arrival of ions, which after conversion to the energy scale leads to the kinetic energy spectrum,

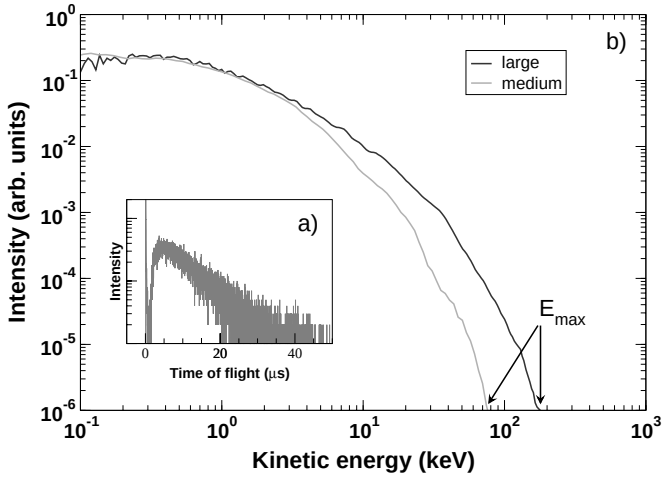

Fig. 2. (a) Time–of–flight spectrum of the fragment ions expelled from the interaction of 140 fs laser pulses with large neutral lead clusters. The corresponding energy spectrum is given in (b), showing a maximum kinetic ion energy E_{max} of 180 keV. For medium–sized lead clusters the distribution shifts to lower energies reducing E_{max} by more than a factor of two.

Table 1. Average ($\langle E \rangle$) and maximum (E_{max}) kinetic energies of platinum and lead ions generated from the Coulomb explosion of clusters exposed to strong laser fields (130 fs, $6{\times}10^{15}$ W/cm^2). The cluster size ranges are defined in the experimental section.

	Pb_N			Pt_N
cluster size	small	medium	large	small
$\langle E \rangle$ (keV)	2	4.7	5.8	1.5
E_{max} (keV)	30	75	180	20

see Fig. 2b. Note, that a significant number of ions have energies up to $E_{max} = 180$ keV. The average ion energy $\langle E \rangle$ [23] is found to be 5.8 ± 0.5 keV.

By tuning the source parameters, *i.e.* mainly the opening time of the gas valve and the discharge voltage, the cluster size distribution can be varied from large to smaller N. We found that under comparable laser pulse conditions $\langle E \rangle$ and E_{max} drop with decreasing cluster size. As an example Fig. 2b also shows the energy spectrum of medium–sized sized particles. For these clusters E_{max} exhibits a value which is more than a factor of two lower when compared to the larger species, see also table 1. Note that the reduction in cluster size slightly tilts the overall shape of the energy distribution.

In recent contributions [3,20] we have shown that plasmon excitations in metal clusters can lead to an enhanced charging of the particle. Briefly, the cluster having N atoms is treated as a metallic sphere, where the ion density is given by the Wigner–Seitz radius r_s. Ionization is supposed to yield an uniform charge distribution leading to an expansion of the sphere. From the position of the ions the atomic density can be calculated giving a time dependent $r_s(t)$. This serves as an input for the calculation of the optical response within the random phase approximation [24] giving for every time step the plasmon energy of the cluster. The results show that during the first 100 fs the charging process initially induces a shift of the plasmon energy to higher values [20]. During the subsequent expansion the decreasing ion and thus electron density shifts the collective resonance to lower energies until it matches the laser photon energy. We found that for a platinum cluster of 58 atoms the time scale for this process is about 600 fs. On resonance the charging efficiency is strongly enhanced due to multiplasmon excitations. The results of the model calculations give a reasonable explanation for our measured charge state enhancement at longer pulse widths in the experiments on platinum, gold and lead clusters.

The question arises whether the enhanced charging upon pulse length increase is also reflected in the ionic recoil energies. To explore this aspect we have extracted kinetic energy spectra from the molecular dynamics calculations, see section 3. Starting with the nuclear ground state configuration an ionization function is used to simulate the charging process, *i.e.* $z(t) = z^*[1-\exp(-4(t/\tau)^2)]$ where z^* is the final atomic charge state and τ is comparable to the pulse width in the experiment. We take the maximum charge state observed in the charge resolved experiments on platinum clusters [3], see Fig. 4b, for a rough estimate of z^*. In order to account for the spread in the neutral particle abundance an ensemble of clusters is used in the calculations. The resulting spectra yield the kinetic energy distribution for a given ionization function. The results are displayed in Figs. 3a and 3b for small platinum clusters. In the case of $\tau = 50$ fs, E_{max} exceeds

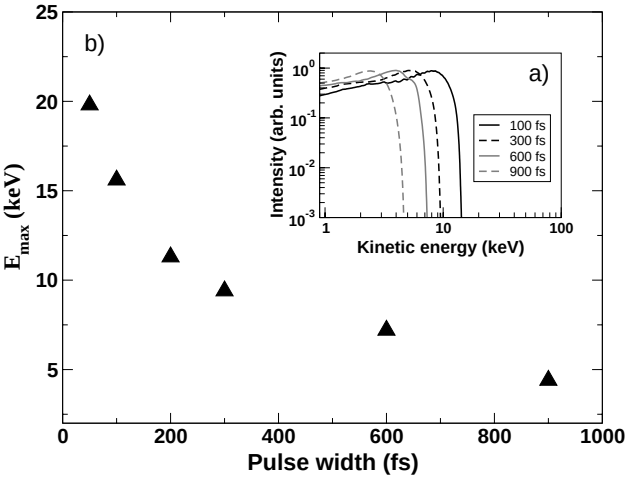

Fig. 3. (a) Calculated recoil energy spectra for the Coulomb explosion of small platinum clusters as a function of the pulse width parameter τ. The corresponding maximum charge state z^* is taken from Fig. 4b, see also Ref. [3]. (b) Resulting maximum kinetic energies as function of the pulse width. Note that the enhanced ionization at $\tau = 600$ fs is not clearly reflected in the kinetic energies of the fragment ions.

19.8 keV. By increasing τ the spectrum shifts to lower energies reaching a value of $E_{max} = 4.4$ keV for $\tau = 900$ fs.

For comparison, the experimental results for small platinum clusters are given in Fig. 4a. Clearly the measured E_{max} lie slightly above the calculated values, but the pulse width dependence agrees well. Note, that in going from 140 to 600 fs the maximum charge state per atom increases from $z^* = 12$ to 17, see Fig. 4b and falls off for even longer pulse widths. The enhancement of z^* at 600 fs obviously has no counterpart in the distribution of E_{max}. Statistical errors of the measurements are primarily caused by the fluctuations of the cluster source producing different particle sizes. The deviations of the measured from the calculated maximum recoil energies are mainly due to the assumption of a homogeneous charge distribution. Furthermore, to obtain a sufficient charge resolution it is necessary to discriminate ions with high recoil energy. Therefore the observed maximum charge states are possibly underestimated leading to lower recoil energies in our calculations. In addition, at this point it cannot be ruled out that contributions from a hydrodynamic type of expansion might play a role.

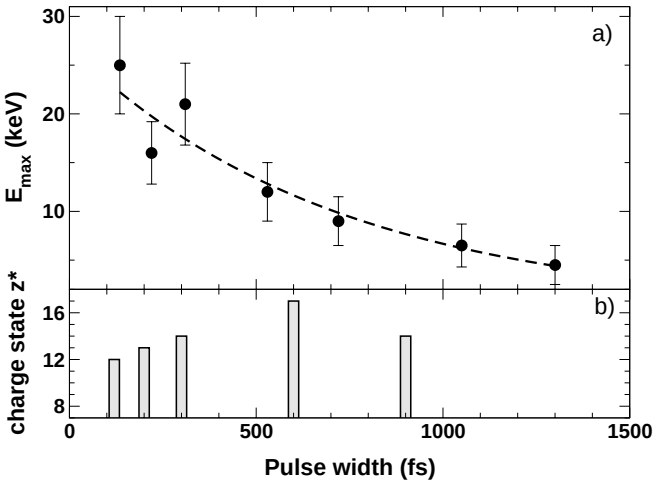

Fig. 4. (a) Measured maximum kinetic ion energy of small platinum clusters irradiated by laser pulses with constant energy of 15 mJ as a function of the pulse width (*e.g.* 3×10^{15} W/cm^2 @ 130 fs). The dashed line shows an exponential fit to the data. In contrast to the increase in z^* which finds a maximum at 600 fs, see (b), $E_{\max}$ shifts to lower values with increasing pulse width.

The calculated, as well as the experimental results show that the charge enhancement for longer pulse widths does not lead to a significant increase of the energy of the atomic ions. Instead, the energy distribution shifts to lower values with increasing pulse width. Thus the additional charging due to plasmon enhanced ionization is not sufficient to compensate for the decrease in potential energy upon expansion. Due to the small interatomic distances in a metallic particle the recoil energy spectra should show larger values of E_{max} when compared to rare gas clusters. Still, Ditmire *et al.* [12] found slightly higher ion energies of up to 250 keV for xenon clusters having a size of about 400 atoms. However, their laser intensity was one order of magnitude higher than in our experiment on large lead clusters, indicating that indeed the metal clusters might carry increased ionic recoil energies. This finding is confirmed by calculations by Last *et al.* [25] which predict an ion energy of 4.5 keV for a 550–fold charged Xe_{55}. For Pt_{55}, on the other hand, our calculation gives a value of 7.2 keV.

5 Conclusion

In conclusion, the exposure of small metal clusters to laser pulses with intensities in the 10^{14}-10^{16} W/cm^2 range leads to a Coulomb explosion where the atomic fragment ions reach energies of up to 180 keV in the case of lead. The results show that the enhanced charging upon increasing the pulse width observed in earlier experiments is not reflected in the kinetic energy of the ions. It was found that E_{max} decreases with cluster size as well as with increasing width of the laser pulse. The calculations, based on a classical molecular dynamics approach and a reasonable assumption on the ionization process, clearly show that a high ground state density of the atoms is mainly responsible for huge kinetic energies.

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