



High-energy Coulomb explosions in ultra-dense deuterium: Time-of-flight-mass spectrometry with variable energy and flight length

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ARTICLE INFO

Article history:

Received 5 February 2009

Accepted 13 February 2009

Available online 24 February 2009

PACS:

79.20.Ds

61.46.-w

67.63.Gh

Keywords:

Deuterium

Ultra-dense deuterium

Coulomb explosion

Kinetic energy release

TOF-MS

ABSTRACT

High-density hydrogen is of great interest both as a fuel with the highest energy content of any combustion fuel, and as a target material for laser initiated inertial confinement fusion (ICF) [S. Badiel, L. Holmlid, J. Fusion Energ. 27 (2008) 296]. A much denser deuterium material named D(-1) can be observed by pulsed laser induced Coulomb explosions giving a well-defined, high kinetic energy release (KER). Neutral time-of-flight of the fragments from the material shows that the Coulomb explosions have a KER of 630 eV [S. Badiel, P.U. Andersson, L. Holmlid, Int. J. Hydrogen Energ. 34 (2009) 487]. By using ion time-of-flight-mass spectrometry (TOF-MS) with variable acceleration voltages and a few different values of laser pulse power, we now prove the mass and charge of the particles as well as the KER. In fact, the ions are so fast that they must be H⁺, D⁺ or T⁺. By using two different flight lengths, we prove with certainty that the spectra are due to D⁺ ions and not to photons or electromagnetic effects. The results also establish the fragmentation patterns of the ultra-dense D(-1) material in the electric field. The energy release of 630 ± 30 eV corresponds to an interatomic distance D-D of 2.3 ± 0.1 pm. This material is probably an inverted metal with the deuterons moving in the field from the stationary electrons, which gives a predicted interatomic distance of 2.5 pm, close to the measured value. Thus, we prove that an ultra-dense deuterium material exists.

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1. Introduction

The possible existence of dense hydrogen materials has been investigated for many decades. The focus of the studies has for a long time been on the formation of a metallic hydrogen (protium) material at extremely high pressures [1,2]. Experimental attempts have always started from ordinary covalently bonded hydrogen molecules, with a distance H-H of 74 pm. The hope has been to reach a hydrogen material with a density corresponding to maybe a 150 pm H-H distance. This may seem impossible: the covalent bond should change to a metallic bond, from a bond strength of 430 kJ mol⁻¹, probably over a considerable energy barrier to a metallic bond strength of approximately 330 kJ mol⁻¹ [3,4]. Metallic hydrogen was instead found by Nellis and co-workers [5,6] at high temperature and high pressure, where the dissociation of hydrogen molecules to H atoms due to the high temperature means a much more facile transition to the metallic bonded phase. Using another path via Rydberg matter (RM) of hydrogen to avoid the formation of covalently bonded H₂ molecules, it is also possible to produce a

hydrogen material with H-H bond distance of 150 pm corresponding to the metallic hydrogen density. This path employs a catalytic process at low pressure [7–9]. The catalyst used is a hydrogen transfer catalyst which readily dissociates H₂ to H atoms even at room temperature. At low pressure, hydrogen atoms desorb from the catalyst surface in the form of Rydberg atoms, forming RM clusters in desorption. These clusters are initially in higher electronic states (excitation levels), de-exciting by radiative processes to the material with H-H distance 150 pm. This material is named H(1) since the angular momentum of the metallic electrons is $l=1$ in this phase; the electronic state can also be considered to be formed from Bohr state H atoms with $l=n_B=1$ (the B is used to indicate the Bohr model), thus with an excitation level $n_B=1$. This material is the most energy-dense combustion fuel known [3]. It has also been proposed to be a target material for inertial confinement fusion (ICF) [10].

When this type of material H(1) is formed from ordinary hydrogen gas or its corresponding deuteron material D(1) is formed from pure deuterium gas, it is also possible to form a much denser material mainly consisting of D atoms. It is called D(-1) since it is probably a phase where the electrons and ions exchange their roles. In such a case, the distance scale is decreased due to the very different mass of the moving particles. A continuous material H(-1) is probably not formed, but short bonds H-D and H-H seem to exist. The background is described further in the theoretical section.

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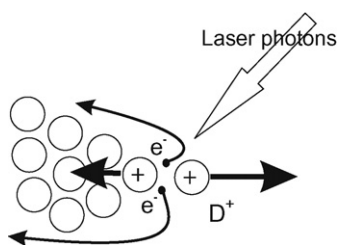


Fig. 1. A pictorial representation of the Coulomb explosion (CE) process in the material, in this case consisting of D atoms. One or two electrons are excited into higher localized non-shielding orbitals by the laser pulse. The CE process is finished in less than 1 fs.

2. Theory

The method we use to measure the bond distances in the dense hydrogen materials is laser induced Coulomb explosions (CE). A nanosecond laser pulse excites electrons in the material to give bare nuclei exposed to their full Coulomb repulsion, as shown pictorially in Fig. 1. The potential energy which pushes the two nuclei apart is

$$W = \frac{e^2}{4\pi\epsilon_0 d} \quad (1)$$

where d is the distance between the two ionic fragments. This kinetic energy release (KER) will be observed as kinetic energy in two different fragments, and the relative masses of the fragments will determine their fraction of the KER. Calculations of such energy distributions are utilized to interpret the results described below. The distance d for ordinary RM is given by

$$d = 2.9 n_B^2 a_0 \quad (2)$$

where n_B is the Bohr model principal quantum number for the electron in the atom, and a_0 is the Bohr radius equal to 52.9 pm. The factor 2.9 is found both from theory [11] and with high precision from rotational spectroscopy of RM clusters [12,13]. This means that at $n_B = 1$, the distance d is predicted to be 153 pm.

Coulomb explosions with energy of the order of 100 eV u^{-1} as observed here cannot take place in any type of known material. The limiting factor is the bond distance between the two atoms involved in the laser fragmentation process. For example, the bond distance between two Li atoms in a Li_2 molecule is 267 pm. This means that the maximum repulsion energy between two Li^+ from the dimer is 5.4 eV. He_2 has a bond distance of 108 pm in most of its low excited states. This gives a maximum KER of 13.3 eV if two ions can be formed at that distance, which requires a very intense laser. In Li metal, the interionic distance is approximately 300 pm. Even when ionized, the size of the ions in these two cases is considerable, since the inner electrons occupy space. The lowest Bohr orbit in Li with $Z=3$ has a diameter of 35 pm, corresponding to only 41 eV interionic repulsion if the ions could be that close, which is not possible by chemical bonding. Thus, KER of the order of 100 eV u^{-1} is only possible for the hydrogen nuclei in H, D and T, since there are no inner electrons in these cases. Thus their bond distance in a metallic phase can be very short, not determined by the repulsion due to overlap of any inner electrons.

Below, fragmentation processes corresponding to a common KER of 630 eV are reported. This corresponds to an interatomic distance of 2.3 pm, a factor of approximately 65 smaller than in the H(1) material studied previously [7–9]. We propose that this new material is dense atomic hydrogen (deuterium) of the type described by Ashcroft [14] and by Militzer and Graham [15]. In this dense atomic hydrogen the electrons can be considered to give the constant (negative) charged background, while the nuclei move within this charge density. (This state is either close in energy to the normal ground state D(1) or is in fact the ground state of condensed atomic

deuterium.) This description is the reverse of the ordinary description of a metal, where the electrons move in the dispersed positive potential due to the ions [16]. A switch from electronic motion to deuteron motion may be possible if the angular momenta and the energies of the system can be retained. The kinetic energy of an electron or a deuteron is given by $E_{kin} = 1/2(mv^2)$ and the angular momentum of its motion is in classical terms $l = mrv$, where r is the radius of the circular orbit. Assuming that both the kinetic energy and the angular momentum are conserved in a switch to the inverted state with the deuterons moving in the field from the stationary electrons, one finds directly:

$$\frac{r_d}{r_e} = \left(\frac{m_e}{m_d}\right)^{1/2} \quad (3)$$

This gives a scale change by the square root of m_e/m_d equal to $(2 \times 1836)^{-0.5} = 1/60.6$. Using the interatomic distance in D(1) and H(1) equal to 150 pm [7] we can apply this scale change to predict the bond distance in dense atomic deuterium to be 2.5 pm. As shown below, the experimental value for the type of dense atomic deuterium studied here is 2.3 ± 0.1 pm, which is quite close. If the material contains H atoms in the state H(–1), the mass ratio is different, equal to 1/42.8, predicting a bond distance of 3.5 pm.

Due to the high density of the D(–1) material, a factor of 2×10^5 higher than for H(1), the transport of energetic particles through the material is strongly impeded. In fact, the deuterons at 2.3 pm bond distance are close to the nuclear barrier, and a kinetic energy of 630 eV may be sufficient to give d–d fusion by tunneling. One important point to note is that neutral D atoms may slip away from the dense material with no strong Coulombic interaction, as proved by the very sharp neutral TOF peaks in Ref. [17] and below. However, in the experiments presented here we need to form D^+ ions to determine the ion kinetic energy by standard methods. In this case, ions need to leave this very dense phase D(–1). It can be expected that a Coulombic interaction with the ions in the material exists even after the Coulomb explosion, which may deflect and possibly capture the D^+ ions before they can leave the material. Since all ions have the same mass in D(–1), the KER but not the particle identity will be retained after single linear collisions. However, non-central collisions give lower energy. The ions leaving from the material may thus not be the original ones released but “boil off” from the surface of the material, but they will be slower if the collisions are not central. Further, in the case of collisions with several other ions, which is highly likely due to the high density, a large energy loss will take place. For example, a collision of one 630 eV D^+ with two static D^+ simultaneously may give around 280 eV in each of the two other ions (depending on geometry) and leave 70 eV in the first D^+ . Thus, collision events that slow down the D^+ ions will exist. This is the reason why relatively high voltages are used on the emitter, to clean the boundary layer outside the emitter surface.

The neutral TOF measurements [17] show that the interaction of one D^+ ion with several other ions during the acceleration phase in the Coulomb explosion is highly unlikely. The only such process observed is the one involving repulsion from two other charges named (2+), giving KER up to 1200 eV. Due to the well-defined TOF patterns observed, the possibility is very small that the 630 eV deuterons are due to several low-energy repulsions instead of one high-energy repulsion. This is of course also due to the short distances and short interaction times involved in the Coulomb explosions. This can be understood from Fig. 2, which shows that the repulsion between two charges is practically completed in less than a femtosecond. At such short time, the distance is already more than 100 pm, 50 times the original d–d distance in the material. Thus, this type of process is repeated many times during the 5 ns laser pulse. With the observed 10^7 Coulomb explosions per laser pulse in the $100 \mu\text{m}$ wide laser focus area, only one Coulomb repulsion event between two charges exists on average at any given time

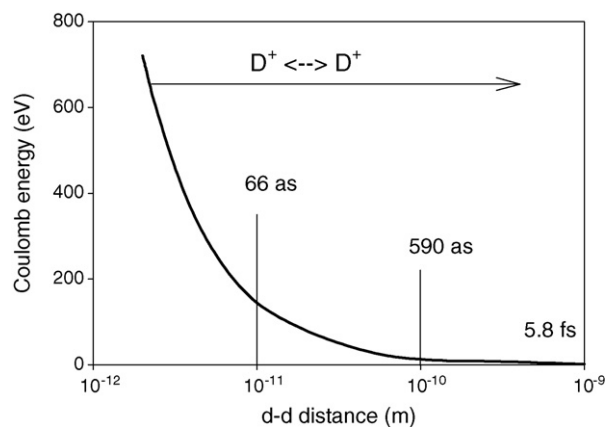


Fig. 2. Approximate potential energy of the $D^+–D^+$ repulsion from a distance of 2.3 pm. The time at a few distances after the start of the Coulomb explosion is shown.

in the focus volume. The distance where the interaction ceases is probably smaller than 100 pm due to shielding in the metallic material. This distance may anyway be compared to the size of the laser focus (beam waist), which is 10^6 times larger. Thus, the possibility of interaction between two Coulomb explosion processes also from the geometry is vanishingly small. It should also be noted that the Coulomb explosions are not due to ionization in the material, but only to excitation to higher electronic states in the condensed phase: the laser photon energy is only 2.2 eV relative to the theoretical electron energy of approximately -13.6 eV. The processes observed are single-photon processes, as seen from laser intensity variation. Thus, large scale ionization in the material does not take place.

3. Experimental

Our experimental setup has been described previously [7]. A Nd:YAG pumped dye laser with a power of <100 mJ per shot at 10 Hz and 564 nm wavelength is used, close to its maximum power. A lens with $f=400$ mm brings the laser beam to focus in the center of the UHV chamber. Close to the center of the apparatus, a K doped iron oxide catalyst (a hydrogen atom transfer catalyst) is used to produce H(RM) and D(RM) from normal hydrogen (1.5% deuterium) or pure deuterium gas at a pressure up to 2×10^{-5} mbar (uncorrected hot cathode gauge reading). The RM emitter is mounted in a Ta foil and can be heated by an ac current passing through the foil. The foil is cut away on the side facing the laser beam. In the present type of experiment, the emitter is moved close to the laser beam, which gives rise to the fast particles. Two similar detectors observe the fast particles released by the laser pulse, one at a distance of 101 mm and the other at 1120 mm from the laser focus. A cut through the short-path detector is shown in Fig. 3. In the detectors, the particles strike against a metal foil, forming energetic ions and photons at impact. A few cm above this point of impact, a Cu–Be Venetian blind dynode is kept at -6 to -10 kV and the energetic photons or ions expel electrons from its surface. These electrons are converted to photons in a plastic scintillator, and the resulting blue photons are observed by a photomultiplier (PMT). The pulses from the PMTs are amplified and measured by a fast time-of-flight MCS scaler (EG&G Ortec Turbo MCS). A photodiode at the laser, 2 m from the chamber, triggers the MCS scaler directly with a fast NIM pulse. The scattered laser pulse in the chamber is visible in the MCS time range due to the delay in the PMT. Calibration of the TOF signal with a digital oscilloscope is often made. Direct comparisons of the pulse at the laser and the scattered light from the chamber show that no other delay than that due to the velocity of light exists under these conditions. Since correct high-energy ion pulses can be observed at short times

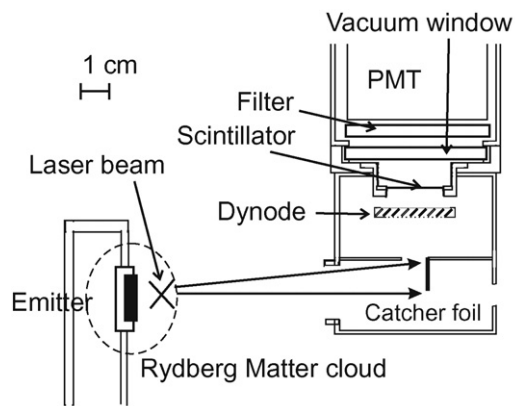


Fig. 3. Cut through the emitter in the center of the chamber and the short-path detector. The long-path detector has a similar construction. The laser beam touches the surface of the emitter in the present experiments.

after the laser pulse in both detectors, down to 50 ns in both cases, no further timing error for the ions exist. The delays in the PMTs of the order of 60 ns are known from the manufacturers; they are also measured independently and taken into account. The total timing error is within ± 10 ns [17]. This is completely negligible, especially for the experiments with long flight path. Usually, 1500 laser shots are accumulated in each experiment. The dwell time per channel is 5–200 ns in the spectra. The acceleration distance used in the calculations of ion TOF is determined to be 73 mm by the distance from the front of the emitter to front of the detector for the short flight path. We have checked that the signals observed are not due to photons that are delayed by some process in the emitter by removing the dark blue violet filter (BG37) normally mounted in front of the photomultipliers, in this way observing all photons. The thickness of the filters are up to 6 mm, which gives a transmission at the laser wavelength of only 1×10^{-5} .

4. Results and discussion

To determine the energetics of the Coulomb explosions accurately, we have now performed TOF-MS experiments of the ions released from the emitter surface by the laser pulse, with variable voltage applied to the emitter. This means that not only the TOF-MS spectra are studied but that the scale of the TOF-MS is varied by varying the acceleration voltage. To further prove our point concerning the KER and the ultra-high density of the material studied, we have used two different flight path lengths, 101 and 1120 mm. Thus, we can be absolutely certain that the signal we observe is due to ions with a mass, not to photons or electromagnetic processes of any kind.

As background information, a neutral TOF spectrum with short flight path is shown in Fig. 4. Such results have been published previously [17] and the results are easily interpreted from the KER of the CE process. From the calculated energy distributions between the fragments given in Table 1, the results of Fig. 4 can be interpreted. As can be seen, the neutral TOF spectrum displays mainly D atoms with a KER of 630 eV, thus from the material $D(-1)$. Repulsions from two charges (indicated 2+) are also apparent. However, one peak probably corresponding to $H(-1)$ with a KER of 390 eV is also observed.

In Figs. 5 and 6, TOF-MS experiments with short and long flight paths are shown respectively, with several values of the voltage applied to the emitter. A first observation from the results is that only H^+ , D^+ (and T^+) can arrive at these short flight times. H^+ and D^+ require a KER of the order of 100–200 eV to reach the detectors at the observed flight times, while He^+ requires more than 2 keV

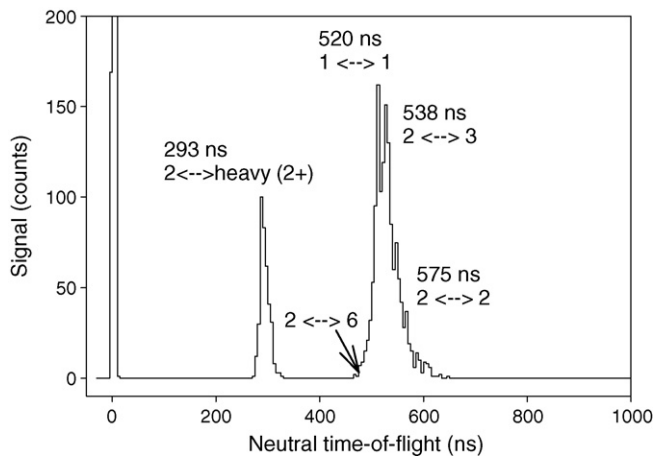


Fig. 4. Neutral TOF spectrum from laser probing of the D(-1) material close to the emitter surface. Short TOF path, H₂ gas admission. The off-scale peak to the left is the laser pulse rise. See further in Table 1.

Table 1

Neutral TOF calculated for the short TOF path in Figs. 4 and 5 at 630 eV kinetic energy release (KER) between mass m_1 with TOF t_1 and mass m_2 . The two last entries are for KER of 390 eV involving H atoms. The flight path is 101 mm. The number of charges is $1 \leftrightarrow 1$ if not indicated otherwise.

m_1 (u) \leftrightarrow m_2 (u)	No. of repelling charges	t_1 (ns)	E_{kin} (eV)
$2 \leftrightarrow \infty$	$1 \leftrightarrow 2$	293	1260
$2 \leftrightarrow \infty$		416	630
$2 \leftrightarrow 8$		460	503
$2 \leftrightarrow 5$		487	449
$2 \leftrightarrow 4$		504	419
$2 \leftrightarrow 3$		531	377
$2 \leftrightarrow 2$		582	315
$2 \leftrightarrow 1$		712	210
$2 \leftrightarrow 1$ (390 eV)		905	130
$1 \leftrightarrow 1$ (390 eV)		523	195

KER: such a high KER would mean a bond distance of 0.7 pm in the Coulomb explosion, much shorter than the diameter of He⁺ which is roughly 100 pm. This means that Coulomb explosions in any known material are excluded, and only hydrogen isotopic atoms (H, D, T) can approach closely enough since they have no inner electrons. This is described further in the theoretical section above. In Tables 2 and 3, the results are interpreted with calculated initial kinetic energies for 1 u (i.e., H⁺) and 2 u (i.e., D⁺) ions. The same

Table 2

Approximate assignments of the TOF peaks (start and maximum) in Fig. 5, including results for 400 V from a separate experiment [17]. No assignment of initial kinetic energy can be made in the case of H⁺ which should be in the range 195–390 eV. The odd masses are due to H atoms, for example mass 3 u is mainly HD.

Emitter voltage	TOF (ns)	eV/u	eV/2 u	Assignment fragmentation	E_{kin} (eV) calc.
Start of peak					
400 V	332	260	730	D ⁺ (-1)(2+) 2 \leftrightarrow 3	754
300 V	362	230	630	D ⁺ (-1)(2+) 2 \leftrightarrow 2	630
200 V	395	230	570	D ⁺ (-1) 2 \leftrightarrow 20	572
100 V	457	190	450	D ⁺ (-1) 2 \leftrightarrow 5	449
50 V	524	165	360	D ⁺ (-1) 2 \leftrightarrow 3	377
25 V	576	145	305	D ⁺ (-1) 2 \leftrightarrow 2	315
0 V	667	120	240	D(-1) 2 \leftrightarrow 1	210
-25 V	806	98	180	D ⁺ (-1) 2 \leftrightarrow 1	210
Peak maximum					
400 V	356	200	610	D ⁺ (-1) 2 \leftrightarrow 50	605
300 V	376	210	585	D ⁺ (-1) 2 \leftrightarrow 20	572
200 V	419	185	495	D ⁺ (-1) 2 \leftrightarrow 7	489
100 V	500	155	370	D ⁺ (-1) 2 \leftrightarrow 3	377
50 V	571	135	300	D ⁺ (-1) 2 \leftrightarrow 2	315
25 V	640	115	245	D ⁺ (-1) 2 \leftrightarrow 1	210
0 V	762	90	180	D(-1) 2 \leftrightarrow 1	210
-25 V	958	75	130	D ⁺ (-1) 2 \leftrightarrow 1 (3.7 pm)	130

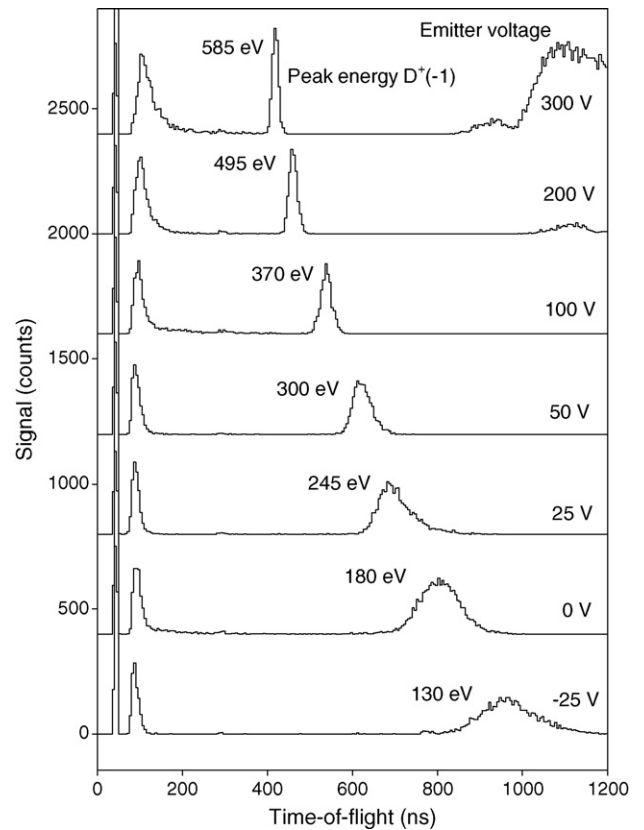


Fig. 5. TOF-MS spectra with variable acceleration voltage on the emitter with short TOF path. Admission of D₂ gas, initially also some H₂. Time sequence is from the top down, but with spectra at 25 and 50 V taken after the others. The off-scale peak to the left is the laser pulse rise. See analysis in Table 2.

procedures were used to evaluate the results as in Refs. [17,18], with constant electric field strength between the plane emitter and the front of the detector at 73 mm distance to accelerate the ions which have an initial kinetic energy (to be determined) from the Coulomb explosion process. In the case of the long-path results, the acceleration region is small relative to the long TOF path and any non-linearity of the acceleration gives insignificant effects. In the case of the short-path data, a non-constant field strength may have some influence but previous results [17,18] and calibrations show

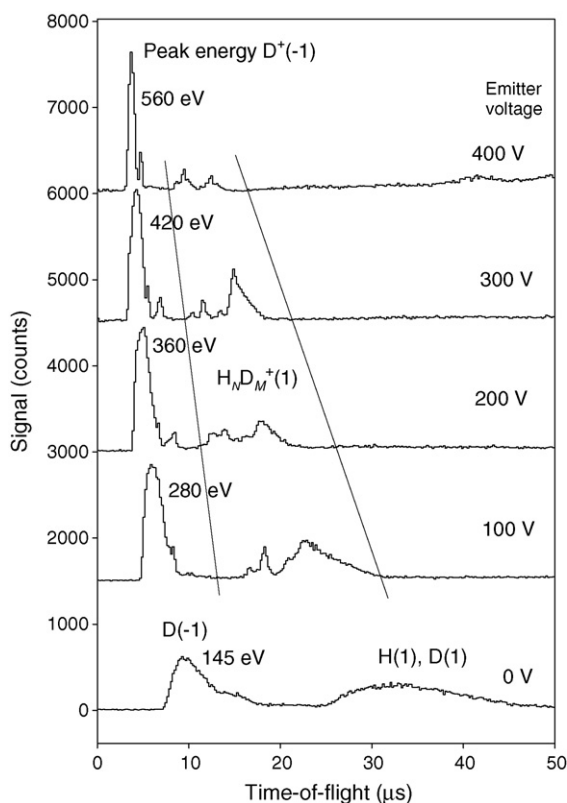


Fig. 6. TOF-MS spectra with variable acceleration voltage on the emitter with long TOF path. No gas admission during this experiment, D absorbed in the emitter. Time sequence is from the top down. See analysis in Table 3. Cluster ions at 8–30 μs are from the close-packed H(1), D(1) layer on the emitter [18].

that the procedure used is accurate. (See also the evidence from the simultaneous two-detector experiments below.) One important requirement is that the calculated initial energies of the ions should be similar in the two experiments: the experiments are performed with several months distance in time and the condition of the emitter is not identical in these two cases; however, the energetics of the processes should not change strongly. (The fragmentation patterns, see also below, vary with the density of the material and very sensitively with the depth of the laser focus in the material and will certainly be different for the two experiments.) A comparison of the results in the tables shows then that the main part of the very fast TOF peaks is due to $\text{D}^+(-1)$ with KER of 630 eV. Some $\text{H}^+(-1)$

Table 3

Approximate assignments of the TOF peaks (start and maximum) with long flight distance in Fig. 6. This experiment was done several months after that in Table 2 under different conditions. No assignment of the initial kinetic energy can be made in the case of H^+ which should be in the range 195–390 eV. The odd masses are due to H atoms, for example mass 1 u is H. (2+) means repulsion of one charge (the observed ion) from two charges.

Emitter voltage	TOF (μs)	eV/u	eV/2 u	Assignments	E_{kin} (eV) calc.
Start of peak					
400 V	3.1	300	1000	$\text{D}^+(-1)(2+)2 \leftrightarrow 8$	1006
300 V	3.3	320	900	$\text{D}^+(-1)(2+)2 \leftrightarrow 8$	898
200 V	3.9	240	670	$\text{D}^+(-1)(2+)2 \leftrightarrow 2$	630
100 V	4.7	200	500	$\text{D}^+(-1)2 \leftrightarrow 10$	503
0 V	7.3	123	246	$\text{D}(-1)2 \leftrightarrow 2$	315
Peak maximum					
400 V	3.7	100	560	$\text{D}^+(-1)2 \leftrightarrow 10$	559
300 V	4.3	75	420	$\text{D}^+(-1)2 \leftrightarrow 6$	419
200 V	4.9	80	360	$\text{D}^+(-1)2 \leftrightarrow 4$	377
100 V	5.9	90	280	$\text{D}^+(-1)2 \leftrightarrow 2$	315
0 V	9.5	72	145	$\text{D}(-1)2 \leftrightarrow 1$ (3.7 pm)	130

with KER of 390 eV is also possible, but only in the first part of the TOF peaks at high emitter voltages. This agrees with the more preliminary results in [17]. The main peaks cannot be described by $\text{H}^+(-1)$ as seen from the different kinetic energies derived for the long and short flight paths, and from the unsystematic variation with emitter voltage for the long-path detector. In fact, the initial kinetic energy derived from the TOF cannot be explained if H^+ would be the ion observed, since the energies vary much more than the factor of two (e.g., 195–390 eV) possible for a reasonable fragmentation scheme: the many low-energy data for H^+ cannot be explained. The conclusion that the best fit is found for D^+ is independent of if a variation of the KER fragmentation is assumed in the calculation as done here, or if no such variation with the repelling partner is assumed to exist as done in Ref. [17] for simplicity.

The reason why the distribution of the KER changes with the emitter voltage must be discussed. With a high voltage, the RM cloud surrounding the emitter is reduced in density and size due to weak field ionization of the RM. This has been observed in numerous experiments. In such a case, the laser pulse mainly interacts with the denser material closer to the emitter surface, which gives repulsions from heavy masses as seen in Tables 2 and 3. With low voltage and thus low field strength, the RM cloud is more extended, and also the more diffuse or filamentary parts of the RM material exist and are exposed to the laser. This means CE processes further out in the RM cloud and thus smaller repelling masses and lower kinetic energy to the D^+ fragments observed. Note that the experiments in Figs. 5 and 6 have both been done from high voltage to low, to avoid too large drifts and changes in the RM cloud during the experiments: large such changes are easily observed if the experiments start at low voltage. The alternative explanation that the bond distances instead of the fragmentation pattern change with the applied voltage can be rejected directly for at least three reasons: (1) the weak external field cannot influence the bond distances in the $\text{D}(-1)$ ultra-dense condensed material, which probably is metallic, (2) the fragmentation changes in the same type of pattern with laser pulse power (laser intensity) (see further below), and maybe most important (3) the fragmentation patterns are of the same type as (and derived from) the patterns observed in numerous experiments by neutral TOF as in Ref. [17] and in Fig. 4. Such neutral TOF experiments use no external field but the same type of fragmentation is observed, with different fragmentation patterns depending on the material density at the point sampled by the laser focus.

In Tables 2 and 3, only the best-fit agreement of the fragmentation is included. Since the TOF-MS peaks are much broader than the neutral TOF peaks in Fig. 4, it is apparent that the ejected ions undergo more collisions than the neutrals both in the material and the cloud outside the emitter. This is expected due to the extremely dense material $\text{D}(-1)$, as described above. Thus, both delays and broadening are expected. The best fit in the tables in a few entries includes the fragmentation of $\text{D}-\text{H}$ at a distance of 3.7 pm, which is the distance observed for the adsorbed $\text{H}(-1)$ on the $\text{D}(-1)$ material. Another possibility not included in the assignments is the release of T atoms from bond distance 3.7 pm. We conclude that the broad TOF-MS peaks may easily contain peaks due to other types of processes including collisions, but the overall agreement with the $\text{D}(-1)$ fragmentation patterns is good evidence for the high-energy KER and the short bond distances in $\text{D}(-1)$.

From the TOF-MS results, it is further clear that heavy ions like K^+ with very high kinetic energies or light particles with low energy do not agree with the experimental results. For heavy ions, the variation of the TOF with emitter voltage would be very small. This is concluded since they could not be formed directly in CE processes, but would be formed by collisions with even more energetic particles. (Note that the initial interionic distance is large for heavy ions, giving only low KER.) Thus, their kinetic energy would not vary

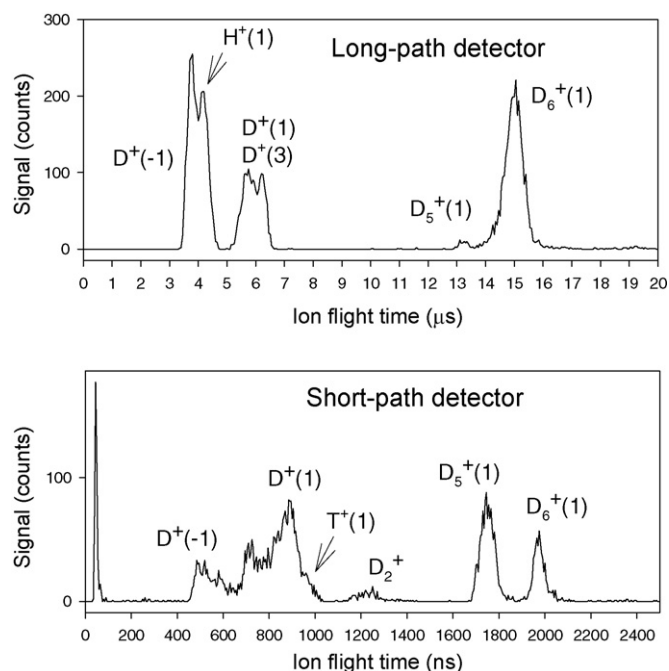


Fig. 7. Nearly simultaneous TOF-MS spectra for the long and short TOF paths, with low laser power (around 10 mJ per shot). Emitter voltage was 400 V. For interpretation, see Table 4.

directly with the emitter voltage. For a light particle like a muon (μ^+ with $m = 0.113$ u and lifetime $2.2 \mu\text{s}$), the variation with voltage would be much stronger. Also in this case, a direct variation of the kinetic energy with the applied field cannot be understood easily. In fact, such a small mass could not arrive as late as the observed peaks, and would not be observable at emitter voltage -25 V in Fig. 5. Thus, it is clear that the peaks observed are due to ions D^+ from $D(-1)$ with small additional amounts of H^+ .

The fragmentation pattern in the Coulomb explosions also varies, as expected, with the laser power. At high laser power of 50–100 mJ per pulse, the high-energy processes observed by the fast particles in Fig. 5 for example, become stronger. Here, the focus is on the well-defined Coulomb explosion processes. Thus, in Fig. 7 an experiment with laser power of only 10 mJ per pulse is shown with the emitter at 400 V, with analysis given in Table 4. One interesting point here is that the measurements with the two detectors are made close in time, with only one hour between the two sets of

Table 4

Calculated TOF for comparison with TOF-MS spectra in Fig. 7, measured with the two detectors in the same experiment at 10 mJ laser pulse power. Acceleration potential on emitter is 400 V. The entries under heading excitation level (-1) are for ion–heavy repulsion and ion–ion repulsion respectively, with KER 630–315 eV for D and KER 390–195 eV for H and T. The KER is 9.4 eV for $n_B = 1$ and 1.04 eV for $n_B = 3$.

Long-path	TOF (μs)		
	Level	(1)	(3)
Ion			
D^+	3.60–4.34	5.93	6.05
H^+	2.91–3.38	4.20	4.28
T^+	5.05–5.85	7.26	7.41
Short-path	TOF (ns)		
	Level	(1)	(3)
Ion			
D^+	351–444	782	850
H^+	293–358	556	600
T^+	507–620	955	1044

experiments. Thus, they are almost simultaneous and show a close similarity, even if material is continuously removed from the emitter surface layer by the laser initiated Coulomb explosions and thus the sampled spot is not entirely constant in composition. In Table 4, the TOF expected for the values of KER from fragmentation of the material composed of H, D or T are shown. In the material type indicated (-1) like in $D(-1)$, the two TOFs given are those for D^+ –heavy $^+$ repulsion of 630 eV, and for D^+ – D^+ repulsion giving 315 eV to each D^+ . It is directly seen that the distributions of ion flux in Fig. 7 agree with an origin from $D(-1)$ and not from $H(-1)$ or $T(-1)$. (These two materials are assumed to have the same bond distances since their nuclei are fermions.) Only one contribution observed with the long-path detector seems to be from H, and that is in the form of $H(1)$ with 9.4 eV initial KER [7–9]. With the short-path detector, the peak at 955 ns (partly overlapping) may be due to T^+ from $T(1)$ on the emitter. In fact, no other particle can give this signal but it must of course be confirmed in other experiments (submitted). Under these low power conditions, strong contributions to the signal can be observed both from $D(-1)$ and $D(1)$, and it is proved once more that the fast particles are due to energetic Coulomb explosions in the material $D(-1)$.

Neutral TOF results give more precise distances in the $D(-1)$ material [17], and the uncertainty in the distance $2.3 \mu\text{m}$ is $\pm 0.1 \mu\text{m}$ from the KER of 630 ± 30 eV, found from the uncertainty ± 10 ns in the TOF. The corresponding results for the $H(-1)$ distance is $3.7 \mu\text{m}$ from the KER of 390 eV.

In Fig. 6, some further peaks are observed in the spectra at slightly longer times. Most of these peaks are due to ions containing both H and D atoms, formed at $n_B = 1$ in the RM cloud. Such ions from a similar phase $H(1)$ (containing only H atoms) in $n_B = 1$ has been described recently [18]. The ions described there have a close-packed form, in the case of H atoms having the form of the regular bodies H_4 , H_6 and H_{12} besides the dimer H_2 . The same principle of cluster formation is apparently seen here, but complicated by the existence of both D and H in the cluster ions. In the neutral spectrum, the peak centered at $32 \mu\text{s}$ is due to $H(1)$. The KER in this case is 9.4 eV, which is correct for $n_B = 1$ [7]. A few small peaks $H^+(1)$ and $D^+(1)$ also seem to be identifiable in the TOF-MS spectra in Fig. 6. In Fig. 7, slower cluster ion peaks are also observed. The slower peaks shown are due to clusters D_5^+ and D_6^+ both emitted from $D(1)$ with 9.4 eV energy as in the case in Ref. [18]. The formation of a five-atom ion cluster is not observed in studies of ion emission from $D(1)$ or $H(1)$ at lower emitter voltages [18]. It is likely due to further collisional or field induced fragmentation from D_6^+ . Since no odd masses are observed, the mixing with H in this case is negligible. The agreement of the results taken with long and short TOF paths in Fig. 7 shows that any variation of the acceleration field strength gives insignificant deviations from the calculations which use a constant electric field. Such an effect would be most visible for low KER, thus for $D^+(1)$ and for the cluster ions D_5^+ and D_6^+ , but no such effect is observed. In Fig. 5, much faster particles are observed at TOF of only 50–100 ns. Such high-energy particles with kinetic energy of $>10 \text{ keV u}^{-1}$ can also be observed with the long flight path, and will be further described elsewhere.

5. Conclusions

The high deuteron KER of 630 eV is unmistakable and the structured and well resolved TOF and TOF-MS spectra are interpreted in detail. The use of TOF-MS with several emitter voltages for two different flight paths, one over 1 m long, and with different laser pulse power gives conclusive evidence for both mass, charge and kinetic energy of the particles. It is shown that the high kinetic energy release observed can only be found for hydrogen atoms (H, D, T). The fragmentation in the Coulomb explosions is modeled in detail. The KER of $630 \text{ eV} \pm 30 \text{ eV}$ corresponds to a distance of $2.3 \pm 0.1 \mu\text{m}$

between two deuterons. Thus a novel phase of ultra-dense condensed atomic deuterium is observed. The value of 2.3 pm is slightly smaller than the theoretically predicted value of 2.5 pm. The KER of 390 eV observed in relation to H atoms indicates an interionic distance of 3.7 pm. In this case, theory predicts 3.5 pm; thus the experimental distance is slightly larger than theory.

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