International Journal of Mass Spectrometry 282 (2009) 70–76

Contents lists available at ScienceDirect

International Journal of Mass Spectrometry

journal homepage: www.elsevier.com/locate/ijms

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. Badiei, 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. Badiei, 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.

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 H_2 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 H_2 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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doi:10.1016/j.ijms.2009.02.014

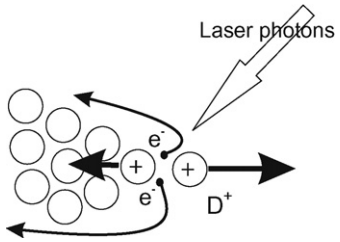

Fig. 1. A pictorial representation of the Coulomb explosion (CE) process in the material, in this case consisting of D atoms. One or two electrons are excited into higher localized non-shielding orbitals by the laser pulse. The CE process is finished in less than 1 fs.

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

$$W = \frac{e^2}{4\pi\varepsilon_0 d} \tag{1}$$

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

$$d = 2.9\, n_B^2 a_0 \tag{2}$$

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:

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

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 μm wide laser focus area, only one Coulomb repulsion event between two charges exists on average at any given time