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Cluster ions D_N^+ ejected from dense and ultra-dense deuterium by Coulomb explosions: Fragment rotation and D^+ backscattering from ultra-dense clusters in the surface phase

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ABSTRACT

The two forms of condensed atomic deuterium, dense deuterium D(1) and ultra-dense deuterium D(−1), can be studied by laser-induced Coulomb explosion time-of-flight mass spectrometry and neutral time-of-flight. In the present study pulsed laser intensity below 10^{14} W cm^{-2} is used. Cluster ions D_N^+ from D(1) are observed with $N = 3, 4, 12$ and 17, thus not in close-packed forms. Clusters $D_N(1)$ are mainly in the form of chains of D_2 and D_3 groups, a shape derived from the D(−1) material which D(1) is spontaneously converted to. Only atomic ions D^+ with initial kinetic energy of hundreds of eV are observed from D(−1). Half of these ions are ejected from the emitter surface, half of them penetrate into the ultra-dense D(−1) layer on the emitter surface. This second half of the ions is reflected completely from the surface layer formed by ultra-dense D(−1) strongly bonded clusters $D_3(-1)$ and $D_4(-1)$.

1. Introduction

Dense deuterium D(1) [1–5] and ultra-dense deuterium D(−1) [1,2,5–10] have been studied in our laboratory with laser-induced mass spectrometry (TOF-MS) of ions formed by characteristic Coulomb explosions (CE). Similar time-of-flight methods for neutral fragments have also been used. D(1) and D(−1) are two forms of the same material, which rapidly interconvert, with D(1) being of the general Rydberg matter (RM) [11–14] type. They are the lowest energy state of RM with a small barrier towards inter-conversion [7,10]. Due to the extremely high density of D(−1), of the order of 10^{29} cm^{-3} (140 kg cm^{-3}) it is believed to be very useful as target material for inertial confinement fusion (ICF) using intense pulsed lasers [1,2,6,7,9,15]. Recent results show fusion without ignition in this material [9]. The ultra-dense material is only formed by deuterons, making the fusion process D + D attainable more easily. The main beneficial properties as fusion targets are the extremely high density and also the high-energy deuterons released in the material by laser pulses [8]. The amount formed regularly in the laboratory is less than 1 μg [7] since this amount is enough to give ignition in a fusion reaction [16].

The catalytic process of forming ultra-dense deuterium D(−1) starts from D(RM) at high excitation levels D(3) and D(4), falling down in energy to dense deuterium D(1) which is spontaneously converted into ultra-dense deuterium D(−1) [1,2,7]. The present study is concerned with the ion clusters which can be observed by laser initiated CE, and with the clusters which exist in the ultra-dense surface layer on the catalytic emitter. The excitation level is generally indicated as D(l), where l is the angular momentum quantum number. This is the main quantum number describing the material. The basis for the description of D(−1) is that this material is similar to an inverted form of D(1), where the deuterons and the electrons have exchanged their roles. This is based on the general ideas of dense hydrogen materials by Ashcroft and other authors [17,18]. The reason for this inversion is probably the fact that the deuterons are bosons and thus do not resist the inversion to the ultra-dense material. In the case of protium, no separate ultra-dense form has been observed so far. The quantum mechanical basis for D(−1) was recently discussed by Winterberg [16,19]. The D(−1) material is expected to be both superfluid and

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superconductive at room temperature and is probably formed by exchange forces between the deuterons [16,19]. Only hydrogen isotope atoms are expected to give an ultra-dense material form, since the inner electrons prevent this inversion for all other atoms.

2. Theory

The main emphasis in the present study is on the material forms D(1) and D(−1) which constitute the lowest energy state of D(RM). They thus lack any excited state properties of general RM. They are stable for days at low pressure in the laboratory [7]. The conduction band electrons in D(1) are excited and delocalized as in general RM. Thus, the properties of D(1) are those of a condensed metallic phase, not of separate Rydberg species. The difference between RM and an ordinary metal is that the conduction electrons in an ordinary metal have orbital angular momentum $l = 0$, while RM is characterized by $l \geq 1$. In RM, the potential for the conduction electrons is neither central due to their comparable distances to several ions, nor of the form $1/r$ [14]. This means that the only good quantum number is the orbital quantum number l (spin quantum numbers also exist). Since the principal quantum number for a free atom n does not exist, a condition $l < n$ does not exist either. To distinguish between the quantum numbers for free atoms and the condensed phase, the excitation level n_B (B for Bohr model) is introduced, numerically equal to the orbital electron angular momentum l. RM at excitation levels $n_B = 3$–8 is normally observed in the form of planar six-fold symmetric clusters with magic number $N = 7$, 19, 37, 61 and 91 [14,20,21]. Many experimental methods have been used to characterize and study the processes that form Rydberg species and RM in desorption from the RM emitter materials, as well as the RM cluster phase directly. Rotational spectroscopy gives accurate information about planar RM cluster forms and bond distances at excitation levels $n_B = 4$–8, in good agreement with other experiments and theory [20,22].

Theoretical classical calculations with electron correlation taken into account [14] show that, even though the RM electrons are delocalized in the RM clusters, it is still possible to describe them as moving in stable circular orbits that are scaled by n_B^2. Further, it was concluded that bonding may only exist when all the electrons have the same excitation level in the RM cluster: dephasing will otherwise take place. Thus each classically stable orbit determines the interionic bond length $d = 2.9 n_B^2 a_0$, where a_0 is the Bohr radius. The approximate scale factor 2.9 was determined in quasi-classical stability calculations of RM clusters [14]. It was recently determined with high precision by rotational spectroscopy of RM clusters [22] and shown to vary slightly with cluster size N and excitation level n_B [20].

When a laser pulse passes through the dense materials D(1) and D(−1), the photons may excite (displace) one bonding electron between two adjacent atoms so that two ions become exposed to each other. A simple picture of the process proposed to take place in D(1) is shown in Fig. 1. In the case of D(−1), a similar process takes place but with the electrons initially only shielding the deuterons in each D_2 pair in the chain cluster. Coulomb repulsion makes the ions move apart rapidly, in <1 fs for D(−1). When the CE takes place, the ions fly apart with almost all their repulsion energy as kinetic energy release (KER) in the ionic fragments. Thus, it is possible to determine the initial repulsion energy between the ions by measuring the kinetic energy of the fragments at a distance from the actual explosion event. Then, the distance between the ions before the break-up i.e. the bond length is found directly from the Coulomb formula as

$$r = \frac{1}{4\pi\varepsilon_0}\frac{e^2}{E_{kin}} \quad (1)$$

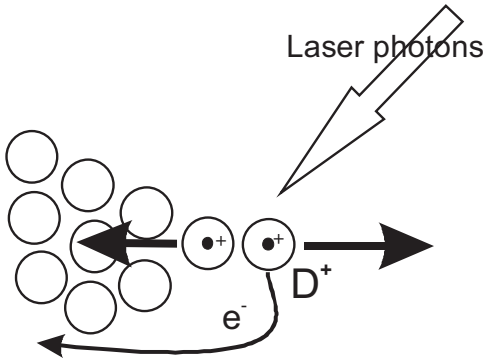

Fig. 1. Schematic drawing of a Coulomb explosion (CE) process in dense deuterium D(1). After removal of an electron by a laser pulse, two deuterons are exposed to each other.

where ε_0 is the vacuum permittivity, e the unit charge and E_{kin} the sum kinetic energy for the two fragments (KER) from the CE. The fraction of the KER that is observed on each fragment depends on the mass ratio of the fragments. The kinetic energy is determined most easily by measuring the time-of-flight (TOF) of the particles and converting this quantity to kinetic energy. This requires that the mass of the particle is known or can be inferred, which of course is simplified when working with just deuterium.

Dense deuterium D(1) is in most respects similar to H(1), which has been studied in several publications [4,23–25]. This means that the interatomic bonding distance d_1 derived from the observed KER of 9.4 eV is close to 2.9×52.9 pm ≈ 153 pm [24,25]. The geometry of the orbitals at $l = 1$ is not strictly planar, giving various cluster forms. The H(1) ion and neutral clusters can be both planar, as for higher RM levels, and also close-packed 3D like tetrahedrons and octahedrons [4].

The internal form of the ultra-dense material D(−1) is not known very well. For example, the observed pairing of the electrons in the D(−1) clusters is not well understood, while the pairing of the deuterons probably is due to exchange forces [16,19]. It is likely that the deuterons rotate around the center of mass of the pair. This explains the transformation between D(−1) and D(1) as due to angular momentum conservation with a switch from orbital electron to orbital deuteron motion [5] as expected for an inverted material. The energy level of D(−1) is close to that of D(1), since the inter-conversion between these two forms is facile in the experiments. The observed general KER of 630 eV (varying with the detailed CE fragmentation process) gives a bond distance of 2.3 ± 0.1 pm [1,2,5]. This is close to the expected distance in an inverted material of $d_{-1} = (m_e/m_D)^{1/2}\, d_1$ equal to 2.5 pm [2,5].

3. Experimental

The apparatus and the methods used here have been described in several publications: see examples in refs. [24,26]. A few different constructions of the central emitter part giving the dense and ultra-dense hydrogen materials have been used, while the detector part in the UHV chamber is the same. The emitter is a sample of an industrial iron oxide catalyst doped with K (initially at 8 wt%) [27,28]. It is of the (now obsolete) styrene catalyst type Shell S-105, but several other similar catalysts work. The emitter is either mounted in a Ta foil holder with a flat surface area of 3×10 mm^2 exposed to vacuum, or held in the opening of a heated tube for the gas feed. The Ta foil holder shown in Fig. 2 can be moved perpendicularly to the laser beam, and a voltage up to 500 V may be applied to the emitter in this construction. In the other construction, the emitter can be moved around the center of the chamber in all directions. The emitter is heated by an ac current through the Ta foil or through the gas feed tube to a temperature <500 K. Deuterium gas (>99.8%