EXCURSIONS IN CLUSTER SCIENCE; FROM DYNAMICS OF LARGE FINITE SYSTEMS TO ULTRACOLD CLOUDS

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I. PROLOGUE

During the last two decades, the chemical physics group of Tel-Aviv University explored the structure, energetics, spectroscopy and dynamics of clusters, focusing on the energy landscapes, spatial structures and shapes, phase changes, superfluidity, energetics, level structure, electronic-vibrational spectroscopy, size effects, response and nuclear-electronic dynamics of large finite systems [1-22]. Recently, our dynamic studies were extended for the adiabatic nuclear dynamics of multicharged atomic and molecular clusters, which manifest unique fragmentation patterns, such as cluster fission and Coulomb explosion [23-34]. Concurrently, a fascinating analogy was established between Coulomb explosion of multicharged clusters and nuclear dynamics of finite, ultracold gases, i.e., optical molasses, in the temperature domain of $T = 10\mu K-100\mu K$ [35, 36]. Cluster science constitutes the art of building bridges, i.e., bridging between the structure, energetics, thermodynamics, response and dynamics of molecular and condensed phase systems in terms of size scaling laws [11-14], bridging between the electron-nuclear dynamics and response of clusters and of nanostructures [37], and bridging between nuclear dynamics of clusters and of ultracold, large, finite, quantum systems [35,36].

II. FROM FISSION TO COULOMB EXPLOSION

The fragmentation of multiply charged finite systems driven by long-range Coulomb (or pseudo-Coulomb [35,36]) forces, i.e., nuclei [38], clusters [23-34,39-42], droplets [43,44], and optical molasses [35,36], raises the following interesting questions regarding the energetics and dynamics of dissociation:

- (1) How does a finite system respond to a large excess charge or to an effective charge?
- (2) What are the topography and topology of the multidimensional energy landscape that guide the system's shape evolution and fragmentation?
- (3) What are the fragmentation channels and under what conditions are they realized?
- (4) What is the interplay between fission, i.e., instability towards dissociation of the finite system into two (or a small number of) fragments and Coulomb explosion into a large number (~n, where n is the number of constituents) of ionic species?

The ubiquity of fission phenomena of droplets, nuclei, and clusters was traditionally described by the classically liquid drop model (LDM) [38, 44], where a classically charged drop deforms through elongated shapes to form separate droplets. The fissibility parameter X = E(Coulomb)/2E(surface) characterizes the relative contribution of repulsive (Coulomb) and cohesive (surface) energies to the fission barrier, separating between the bound initial states and the fission products. For $X \le 1$, thermally activated fission over the barrier prevails. At the Rayleigh instability limit of X = 1 the barrier height is zero [38, 44]. Many features of nuclear and metal cluster fission require to account for quantum shell effects. Nevertheless, the simple LDM expression $X=Z^2e^2/16\pi R_0^3=(Z^2/n)/(Z^2/n)_{cr}$, with the proportionality factor $(Z^2/n)_{cr} = 16\pi R_0^3/e^2$ (where γ is the surface tension, Z the total charge, R_0 the system's radius and r_0 the constituent radius), provided the conceptual framework for the fission of charged finite systems. All the diverse phenomena of fission were realized for fissibility parameters below the Rayleigh instability limit of X = 1, i.e., nuclear fission [45], the fission of metal clusters [42], and of hydrogen-bonded clusters [43]. Beyond the fissibility limit (X > 1) barrierless fission and other dissociative channels open up. We have transcended the Rayleigh instability limit (X = 1) for Coulomb instability of large finite systems, demonstrating the prevalence of a qualitatively different fragmentation pattern of Coulomb explosion beyond the Rayleigh instability limit [28]. We studied the fragmentation patterns and dynamics of highly charged Morse clusters by varying the range of the pair potential and of the fissibility parameters. The instability of multicharged Morse clusters directly reflects on covalently or dispersion-bound chemical and biophysical finite systems. The Rayleigh instability limit separates between nearly binary or tertiary spatially unisotropic fission for X<1 and spatially isotropic Coulomb explosion into a large number of ionic fragments for X>1 (Fig. 1).



Figure 1. Time resolved nuclear dynamics of the fragmentation of highly charged (A⁺)₅₅ Morse clusters (mass of A is 100 amu). The two panels show superimposed temporal patterns of the fragmentation, where each color corresponds to a different time for a one-color snapshot, as marked on the two panels. The Morse potential parameters and the fissibility parameter X are marked on the panels. The time t = 0 corresponds to the T jump to the final temperature (see text). Note the dramatic difference between the spatially isotropic Coulomb explosion (for X = 4.9) on the upper panel, and cluster fusion (for X = 0.24) on the lower panel. (See colored figure on back cover).

We explored Coulomb the of multicharged [23-34,39-44], or effectively charged [35,36], finite systems

instability (Fig. 2). The majority of the currently available experimental information on the Coulomb instability of nuclei (i.e., X = 0.7 for ^{235}U and X = 0.9 for the recently discovered Z = 114 element [45]), of charged droplets (i.e., X = 0.7 - 1.0 for hydrogen bonded systems [43]), and of multiply charged metal clusters (X = 0.85 ± 0.07 for Na^{+z} clusters [42]) pertains to the fission limit, i.e., X < 1 (Fig. 2). How can the Rayleigh limit for the Coulomb instability of a finite system be overcome?

The X >> 1 domain can be accomplished either by a marked enhancement of the repulsive Coulomb energy, or by a dramatic reduction of the cohesive surface energy (Fig. 2). The increase of E(Coulomb) can be attained by cluster multielectron ionization in ultraintense (peak intensity $I = 10^{15} - 10^{20}$ Wcm⁻²) laser fields (section III), while the dramatic decrease of E(surface) can be accomplished in three-dimensional, ultracold optical molasses (section V), where pseudo-Coulomb forces result in isotropic cloud expansion, in analogy with Coulomb explosion [35,36].

The traditional view of Coulomb explosion involves uniform ion expansion. Such is the case for the explosion of multicharged homonuclear clusters (e.g., $(D_2)_{n/2}$ or $(Xe)_n$) with the expansion of (e.g., D^+ or Xe^{q+}) ions retaining a uniform spatial distribution (as is the case for X > 1 in Fig. 1), with an energy distribution being proportional to the square root of the energy, up



Figure 2. A classification of fragmentation patterns of multicharged and effectively charged large, finite systems.

light-heavy heteroclusters consisting of light and heavy ions, e.g., vertically ionized heteroclusters of hydrogen iodide, $(H^+I^{q+})_n$ or $(D^+I^{q+})_n$ (q = 7–35) [34]. In this case, kinematic overrun effects of the light ions (e.g., H^+ or D^+) will result in thin, two-dimensional shells of these light ions, with the monolayer expansion occurring on the femtosecond time scale (Fig. 3). Such an expanding nanoshell of light ions, corresponding to transient soft matter, is analogous to a 'soap bubble' characterized by negative surface tension and is being driven by Coulomb pressure. This transient halo of an expanding, regular monoionic spherical nanointerface manifests transient self-organization on the molecular level in complex systems [34]. Future experimental interrogations of these novel phenomena will emerge from the exploration of the energetics of the light ions in the Coulomb explosion of multicharged light-heavy heteroclusters, involving a narrow energy distribution with a low-energy cut-off [34]. An exciting experimental approach pertains to the application of ultrafast electron diffraction methods [46] for the exploration of the transient structure of the exploding clusters [34].



Figure 3. A 2-dimensional picture of the spatial structure of Coulomb expanding $(H^+I^{25+})_{2171}$ light-heavy heteroclusters at t = 0, 7.4 fs and 14 fs, obtained from molecular dynamics simulations. Black squares (**■**) represent I^{25+} ions, while circles (O) represent H^+ ions. This pictorial representation reveals the formation of narrow expanding shells of the light ions.

III. ULTRAINTENSE LASER – CLUSTER INTERACTIONS

Table top lasers delivering an energy of 1 Joule per pulse on the time scale of ~ 100 fs, can deliver a power of ~ 10^{20} Wcm⁻², constituting the highest light intensity on earth. Highly charged molecular clusters can be prepared by the irradiation of a cluster beam by ultrashort (tens of fs) and ultraintense (intensity I = 10^{15} – 10^{20} Wcm⁻²) laser pulses (Fig. 4).



The extreme cluster multielectron involves ionization process the removal of valence electrons or complete stripping of all the electrons in light first-row atoms or molecules [25-27.29], or the formation of highly charged ions, e.g., up to Xe³⁶⁺. from heavy atoms [29-31,41]. The compound multielectron ionization mechanism of clusters is distinct from that of a single constituent. It involves three sequential processes of inner ionization (due to the semiclassical barrier suppression mechanism for each constituent with a contribution of impact ionization), the formation and response of a nonequilibrium, high energy (100eV-3keV) nanoplasma within the cluster, and outer ionization (induced by barrier suppression for the entire

cluster and by quasiresonance effects) [24,30,31]. Femtosecond [24,30,31] electron dynamics of inner ionization on the time scale of ~ 1–5 fs and of outer ionization on the time scale of ~ 5–20 fs results in multielectron ionization. For the intensity domain of I = $10^{16}-10^{17}$ Wcm⁻² the cluster molecules loose all their valence electrons, with the nanoplasma being persistent, while for the highest intensity range of $10^{18}-10^{19}$ Wcm⁻² both valence and inner shell electrons can be stripped off, with the nanoplasma being completely depleted [31]. The Coulomb instability of a highly charged finite cluster triggers simultaneous and concurrent ultrafast Coulomb explosion [24-29,32,33,41] on the time scales of 10–200 fs (Fig. 4). Analytical expressions for the fs time scales of Coulomb explosion and of (divergent) scaling laws for

the energetics of the highly charged ions were derived [25-27,32,33] were and confirmed bv molecular dynamics simulations with attosecond time steps describing fs dynamics. Ultrahigh ion energies in the range of 1keV-1MeV are released by cluster Coulomb explosion, as portrayed in Fig. 5 for deuterium containing homonuclear and heteronuclear clusters, where deuteron energies in the range of 1-100keV can be obtained.



Figure 5. Energy distributions of D⁺ ions from Coulomb explosion of $(D^+)_n$ homonuclear clusters (n = 2.44·10⁴) and deuterium containing heteronuclear clusters (C⁶+D₄⁺)_n (n = 2171) and (C⁺⁴+H₃⁺I^{q+})_n (n = 2171, q = 25 at I = 10¹⁹ Wcm⁻², and q = 35 at I = 10²⁰ Wcm⁻²).

A significant implication of these high ion energies pertains to nuclear fusion reactions of highly energetic D^+ (as well as T^+ or H^+) ions produced by Coulomb explosion of multicharged clusters in extreme multielectron ionization in ultraintense laser fields, which will be addressed in section IV. Cluster dynamics is moving from ultrafast femtosecond to picosecond nuclear dynamics, towards ultrafast attosecond to femtosecond electron dynamics, and towards electron-nuclear dynamics in ultraintense laser fields. 'Pure' electron dynamics constitutes new dynamic processes in chemistry and physics. Ultrafast cluster dynamics is not limited to the dynamics of ions on the time scale of nuclear motion, but is extended to the realm of electron dynamics, which bypasses the constraints imposed by the Franck-Condon principle [47].

IV. NUCLEAR FUSION DRIVEN BY CLUSTER COULOMB EXPLOSION

Eighty years of search for table-top nuclear fusion, driven by bulk or surface chemical reactions, which involved catalytic dissociation or electrochemical productions of deuterium, reflects on a multitude of experimental and conceptual failures [29]. In 1926 the German physicist Fritz Paneth reported on the apparent observation of helium from hydrogen absorbed on powdered palladium, which might have originated from nuclear fusion. A year later this claim was retracted. In 1935 Adalbert Farkas and Ladislaus Farkas, the founders of physical chemistry in Israel, worked on ortho- and parahydrogen and on deuterium



Figure 6. dd nuclear fusion driven by Coulomb explosion of deuterium clusters. Top: Multielectron ionization in ultraintense laser fields (I > 10^{17} Wcm⁻²) strips the $(D_2)_{n/2}$ clusters of all their valence electrons via consecutive inner and outer ionization. Parallel and concurrent with outer ionization, cluster Coulomb explosion of $(D^+)_n$ clusters occurs. In the size domain of n = $459-7.6\cdot10^4$ the D⁺ average energy increases from 0.3keV to 9.0keV. Energetic deuterons (D⁺ ions) emerging from different clusters in the cluster beam undergo dd nuclear fusion. Bottom: Energy dependence of the cross sections $\sigma(dd)$ for dd fusion adopted from the data of reference [48].

chemistry in the Department of Colloid Science at Cambridge University, England, where they found shelter as refugees from Germany. When passing deuterium gas through a palladium tube, they seemed to observe traces of helium, which might have originated from dd (D^++D^+) nuclear fusion. However, a search for neutron emission in this system, conducted by Lord Rutherford at the request of the Farkas brothers, was negative and eliminated any possibility of nuclear fusion. In this category of negative results for nuclear fusion belongs the widely publicized 1989 "cold fusion" controversy, which did not provide any acceptable scientific information.

These spectacular failures

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are not surprising as, to the best of our knowledge, no theoretical evidence is available to support any valid mechanism of nuclear fusion driven by chemical reactions in infinite bulk or surface systems [29]. The fragmentation dynamics of large finite systems involves an alternative avenue for the induction of nuclear fusion by chemical reactions, e.g., the dd (D^++D^+) nuclear fusion reaction $D^++D^+ \rightarrow {}^{3}He^{2+}+n(2.45MeV)+3.27MeV$, with the production of neutrons (n). Coulomb explosion of extremely multicharged finite molecular systems strives towards the exploration of new areas that are alien to the majority of the chemical physics community. These areas involve nuclear fusion driven by Coulomb explosion of deuterium containing homonuclear and heteronuclear clusters [25-27,29,31-33,49-51]. High-energy Coulomb explosion of an assembly of multicharged, deuterium containing, molecular clusters produces high-energy (1–100keV) deuterons (Fig. 5) in the energy domain of nuclear physics. The high energy deuterons originating from different clusters undergo dd nuclear fusion. During the last four years compelling experimental [49,50] and theoretical [25,26,32] evidence was advanced for nuclear fusion driven by Coulomb explosion (NFDCE) in an assembly of deuterium clusters (Fig. 6). Completely ionized $(D^+)_n$ clusters are produced by multielectron ionization of homonuclear $(D_2)_{n/2}$ $(n = 500-4 \cdot 10^4, R_0 = 10-75 \text{Å})$ clusters in ultraintense laser fields (I > 10¹⁷ Wcm⁻²), stripping the clusters from all their electrons (section III). For Coulomb explosion of very large homonuclear deuterium $(D^+)_n$ clusters (n = 3.8 · 10⁴ and cluster radius $R_0 = 72$ Å), the average deuteron (D^+) energy is $E_{av} = 9 \text{keV}$ and the maximal energy is $E_M = 13 \text{keV} [25,26,32]$. For these deuteron energies the cross section for dd (D⁺+D⁺) nuclear fusion is $\sigma(dd) \approx 10^{-28} \text{cm}^2$ (Fig. 6) [48], being sufficiently high to induce the dd fusion reaction [25,26,32]. Collisions between energetic deuterons, which originate from Coulomb explosion of different deuterium clusters (Fig. 6), result in NFDCE [25,26,32], which was experimentally observed in the Lawrence-Livermore laboratory [49,50]. Our theoretical and computational work [25-27,29-34] proposed and demonstrated that an effective way to produce highly energetic d nuclei



Figure 7. Cluster size dependence of neutron yields per laser pulse for dd NFDCE in an assembly of $(CD_4)_n$ (n = 55–4213) heteroclusters (solid curves), and of $(D_2)_{n/2}$ (n = 55–33573) homonuclear clusters (dashed curves) in the laser intensity range I = 10^{16} – 10^{19} Wcm⁻². Data adopted from reference 33. The NFDE for heteronuclear clusters manifests a considerably larger neutron yield than for homonuclear clusters, as discussed in the text.

 $(D^+ \text{ ions})$ for nuclear fusion involves multielectron ionization and Coulomb explosion of molecular heteroclusters of deuterium bound to heavy atoms. Highly ionized heteroclusters high-energy for Coulomb explosion involve heavy water clusters $(D^+D^+O^{q+})_n$ (q = 6-8), heavy methane clusters $(C^{q+}(D^{+})_{a})_{n}$ (q = 4-6), or deuterated $(D^{+}I^{q+})_{n}$ hydroiodic clusters (q = 7-35) in the size domain of $n = 55-4 \cdot 10^3$ ($R_0 = 10-40$ Å). A dramatic energy enhancement of deuteron energy from these heteroclusters. as compared to from homonuclear deuterons deuterium clusters, is manifested (Fig. 5). For Coulomb explosion of heteroclusterstheheavymulticharged ions (e.g., C⁴⁺, C⁶⁺, O⁶⁺, O⁸⁺) act as energetic triggers driving the light D⁺ clusters to considerably higher kinetic energies than for totally ionized deuterium clusters of the same size. In addition, kinematic effects, which manifest a sharp energy maximum in the vicinity of E_M , in the energy spectra of the D⁺ ions from heteronuclear clusters, provide a supplementary contribution to the efficiency of the NFDCE [25,27,33,34]. The effects of energetic and kinematic triggering on the energetics of the D⁺ ions in Coulomb explosion of multicharged deuterium containing homonuclear and heteronuclear clusters are manifested by the neutron yields for NFDCE (Fig. 7) calculated [26,32,33] under the conditions of the Lawrence-Livermore experiment [49,50]. The neutron yields per laser pulse (Fig. 7) for laser intensities of I > 10¹⁷ Wcm⁻² are higher by 2–3 orders of magnitude for Coulomb explosion of (CD₄)_n clusters than for (D₂)_{n/2} clusters of the Saclay group [51], which demonstrated a marked enhancement of neutron yields from dd fusion in an assembly of Coulomb exploding (CD₄)_n clusters, as compared to (D₂)_{n/2} clusters.

An extreme way to attain highly effective energetic and kinematic triggering for driving deuterons to very high energies can be achieved for Coulomb explosion of deuterated methyl iodide $(C^{6+}D_3^+I^{q+})_n$ and hydroiodic acid $(D^+I^{q+})_n$, which produces heavily charged I^{q+} ions in very intense laser fields, i.e., q = 25 at I = 10¹⁹Wcm⁻² and q = 35 at I = 10²⁰Wcm⁻² [34]. For such iodine containing heteroclusters (with $n \approx 4213$, q = 25 and $R_0 \approx 40$ Å) the deuteron energies are $E_{av} = 40$ keV, being considerably higher than those for homonuclear $(D_2)_{15000}$ ($E_{av} \approx 2.8$ keV) and heteronuclear $(CD_4)_{4213}$ clusters ($E_{av} = 10$ keV) of the same size. When the D⁺ energy increases by a numerical factor of 3, the cross section for dd fusion and the neutron yield increase by 2-3 orders of magnitude (Fig. 6) [48]. We infer from the data of Fig. 7 and from the foregoing discussion that the neutron yields for Coulomb explosion of deuterium containing homonuclear and heteronuclear clusters with $R_0 = 40$ Å are predicted to be $Y = 10^3$ neutrons/laser pulse for $(D_2)_{15000}$, $Y = 10^5$ neutrons/laser pulse for $(CD_4)_{4213}$, and $Y = 10^8$ neutrons/laser pulse for $(DI)_{4213}$. A semi-quantitative confirmation of these predictions was provided from experiments for Coulomb exploding $(D_2)_n$ and $(CD_4)_n$ clusters [51]. The dream of table-top nuclear fusion in the chemical physics laboratory came true.

IV. PERSPECTIVES

The NFDCE of molecular clusters induced by multielectron ionization and Coulomb explosion involves a "cold-hot" fusion mechanism, where the cluster beam constitutes a cold (or even ultracold) target, while Coulomb explosion of the assembly of clusters provides the high energy required to induce nuclear fusion. Of considerable interest are the perspectives of the NFDCE of deuterium (or tritium) containing homonuclear and heteronuclear clusters for the production of short (100ps–1ns) neutron pulses [26,50], which may be instrumental in the exploration of time–resolved structural studies of biomolecules or large molecules. The utilization of NFDCE of deuterium containing heteronuclear clusters will greatly enhance the intensity of the neutron pulse. In addition, some nuclear fusion reactions involving protons and heavy nuclei, e.g., the ${}^{12}C^{6+}+H^+ \rightarrow {}^{13}N^{7+}+\gamma$ reaction, are of astrophysical interest for the carbonnitrogen-oxygen (CNO) cycle in hot stars. The CNO cycle of nuclear fusion, which supplies energy to the hot stars, is catalyzed by ${}^{12}C^{6+}$, which is regenerated. The ${}^{12}C^{6+}+H^+$ NFDCE [27] can be induced by multielectron ionization of sufficiently large methane clusters ($R_0 = 120$ Å), providing information on the cross sections and dynamics of elemental nuclear processes in astrophysics.

Some novel and fascinating phenomena relating to cluster size effects and dynamics pertain to the nuclear dynamics and phase changes in finite, ultracold, gases in the temperature domain of



Figure 8. Nuclear dynamics of the spatial extension of optical molasses of Rb, as reported in reference 35. (a) A photograph of the irradiated cloud at t = 0. (b) Distribution of excited atoms in an irradiated cloud at t = 0. (c) Time dependence of the radius of the irradiated cloud. Characteristic expansion time $\tau = 1.4$ ms. (d) Time dependence of the volume of the irradiated cloud. (e) Time dependence of the density of the irradiated expanding cloud. Characteristic expansion time $\tau = 1.8$ ms.

 $T = 100nK - 100\mu K$, involving gases in magnetooptical traps, optical molasses and Bose-Einstein condensates [36]. Astriking analogy was established between the nuclear dynamics of ultracold optical molasses (T = $10-100 \mu$ K) and Coulomb explosion of multicharged atomic clusters [35,36]. The optical molasses involve a cloud of trapped, laser irradiated, neutral atoms, e.g., Rb, in a magnetic trap (Fig. 8) which is characterized by a density of $\rho = 10^{11} - 10^{13}$ atoms/cm³ and by an interatomic distance of $r_0 \simeq 10^4$ Å. When the magnetic trap is suppressed, the cloud expands via the radiative trapping force, which prevails between radiation-emitting and reabsorbing atoms [35]. An isomorphism was established [35,36] between the radiative trapping force and the electrostatic Coulomb force, with the effective charge characterizing the radiative trapping force being $\sim 4.10^{-5}$ e, with e being the electron charge. The theory of the dynamics of cluster Coulomb explosion of multicharged molecular clusters [23,25,32,33] was applied for the expansion of optical molasses [35,36]. While the Coulomb explosion time of $(Xe^+)_n$ clusters is 10⁻¹³s [23], the expansion time of optical molasses of Cs atoms was predicted to be 10 orders of magnitude longer, i.e., $\sim 10^{-3}$ s [35]. This estimate is in accord with experiments (Fig. 8) [35,36]. These studies, together with the exploration of superfluidity of helium-4 clusters [22], bridge between the dynamics of clusters and ultracold, large finite quantum systems.

During the last decade, cluster science explored new fascinating scientific territories, bridging between cluster electron-nuclear dynamics and nuclear dd fusion, and bridging between cluster dynamics and ultracold quantum clouds.

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