# ON DYNAMICS OF LARGE FINITE SYSTEMS. FROM CLUSTERS 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, energetics, level structure, electronic-vibrational spectroscopy, response and nuclear-electronic dynamics of large finite systems [1-17]. Recently, these dynamic studies were extended for the adiabatic nuclear dynamics of multicharged clusters, which manifests unique fragmentation patterns, such as cluster fission and Coulomb explosion [18-23]. These features are unique for finite systems and do not have an analogue in the dynamics of the corresponding bulk matter. Concurrently, a fascinating analogy was established between the Coulomb explosion of multicharged atomic clusters and the nuclear dynamics of finite, ultracold gases, i.e., optical molasses in the temperature domain of $T = 10 – 100\ \mu K$.

## II. FROM FISSION TO COULOMB EXPLOSION

The fragmentation of multiply charged finite systems driven by long-range Coulomb forces, i.e., nuclei [26], clusters [19,28,29], droplets [30], and optical molasses [24] raises some interesting questions regarding the energetics and dynamics of dissociation:

(1) How does a finite system respond to a large excess charge or 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 liquid drop model (LDM) [26,30], where a classical 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 $X < 1$, thermally activated fission over the barrier prevails. At the Rayleigh instability limit of $X = 1$ the barrier height is zero [26,30]. Many features of nuclear and metal cluster fission require the account for quantum shell effects. Nevertheless, the simple LDM expression $X = Z^2e^2 / 16\pi\gamma R_n^3 = (Z^2n) / (Z^2/n)_{cr}$ with $(Z^2/n)_{cr} = 16\pi\gamma r_0^3 / e^2$ (where $\gamma$ is the surface tension, Z the total charge, R the system's radius and $r_0$ the constituent radius) provided the conceptual framework for the fission of charged finite systems. All the ubiquitous phenomena of fission were realized for the fissibility parameter below the Rayleigh instability limit of X = 1, i.e., nuclear fission [31], the fission of metal clusters [32], and of hydrogen-bonded clusters [33]. Beyond the fissibility limit (X > 1) barrierless fission and other dissociative channels can open up. Recently we have transcendented 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 [34]. On the basis of molecular dynamics simulations we explored 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. We established that 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).

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 [32]) of charged droplets (i.e., X = 0.7 – 1.0 for hydrogen bonded systems [33]) and of multiply charged metal clusters (X = 0.85 ± 0.07 for $\mathrm{Na}_n^{+z}$ clusters [32]) pertains to the fission limit (X < 1). 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. The increase of E(Coulomb) can be attained by cluster multielectron ionization in ultraintense (intensity I = $10^{16}$ – $10^{20}$ 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.

## 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 = $10^{15}$ – $10^{19}$ Wcm$^{-2}$) laser pulses (Fig. 2). The multielectron ionization mechanism of clusters is distinct from that of a single constituent, involving two sequential processes of inner ionization (due to semiclassical barrier suppression mechanism for each constituent with a small contribution of impact ionization, which yields a nonequilibrium plasma within the cluster) and of outer ionization (induced by barrier suppression for the entire cluster and by quasiresonance effects) [19-21]. Femtosecond [19–21] 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. ‘Pure’ electron dynamics constitute new dynamic processes in chemistry and physics. For the intensity domain of I = $10^{16}$–$10^{17}$ Wcm$^{-2}$ the cluster molecules loose all their valence electrons, while for the highest intensity range of $10^{18}$–$10^{19}$ Wcm$^{-2}$ both valence and inner shell electrons can be stripped off. The Coulomb instability of a highly charged finite cluster triggers simultaneous and concurrent ultrafast Coulomb explosion on the time scales of 10–100 fs (Fig. 2). 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 and were confirmed by molecular dynamics simulations with attosecond time steps describing fs dynamics. Ultrahigh ion energies in the energy range 1keV – 1MeV are released by cluster Coulomb explosion (Fig. 2).

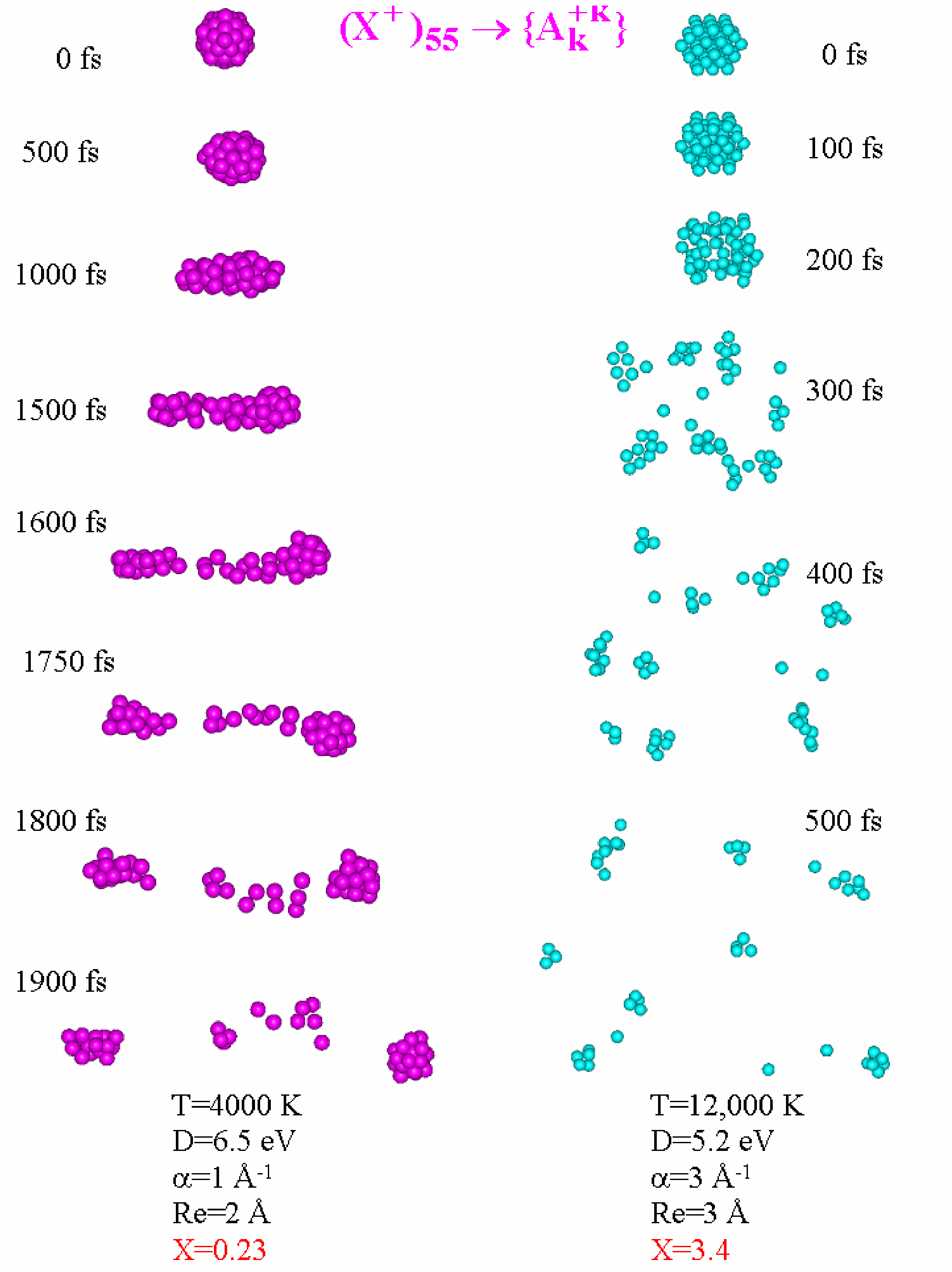

Fig. 1. Time resolved nuclear dynamics of the fragmentation of highly charged $(A^+)_{55}$ Morse clusters (mass of A is 100 amu). The potential parameters, the X values and the times, are marked on the two panels, and the time t = 0 corresponds to the T jump to the final temperature. Note the dramatic difference between the spatially isotropic Coulomb explosion (X = 4.2) and the spatially unisotropic fission (X = 0.23).

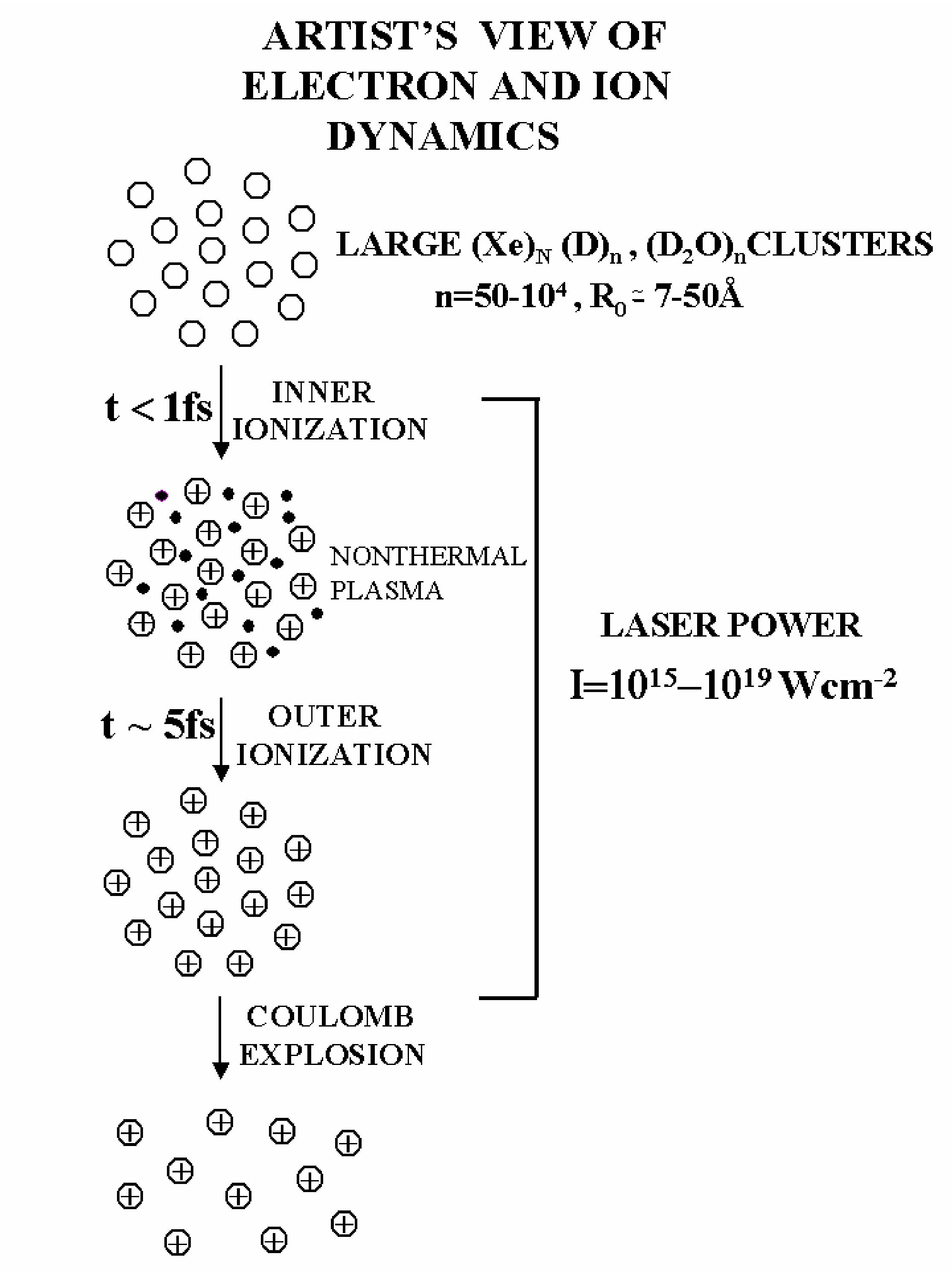

Fig. 2. A schematic description of ultrafast electron and ion dynamics for molecular clusters in ultraintense laser pulses.

## IV. NUCLEAR FUSION DRIVEN BY CLUSTER COULOMB EXPLOSION

During the last two years compelling experimental [35,36] and theoretical [20-23] evidence was advanced for nuclear fusion driven by Coulomb explosion (NFDCE) of an assembly of deuterium containing clusters. In an assembly of deuterium or tritium containing exploding charged clusters the collisions of the high-energy d or t nuclei in the 1–10keV domain originating from Coulomb explosion of different clusters may induce dd or dt nuclear fusion reaction, i.e., $d+d \rightarrow {}^3He + n + 3.27$ MeV or $d + t \rightarrow {}^4He + n + 17.6$ MeV, in the chemical physics laboratory. The dd fusion reaction driven by Coulomb explosion of totally ionized $(D_2)_n$ clusters (cluster radius $R_0 =$ 10–50Å and number of constituents $n = 400 - 2x10^4$) in an intense laser field ($I \simeq 5x10^{17}Wcm^{-2}$) was experimentally observed in the Lawrence Livermore laboratory [35,36]. The Tel Aviv work [20,23] proposed and demonstrated by molecular dynamics simulations that an effective way to produce energetic d or t nuclei for fusion involves multielectron ionization and Coulomb explosion of molecular heteroclusters of deuterium/tritium bound to heavy atoms. Adequate clusters for high-energy Coulomb explosion involve multielectron ionized heavy water clusters $(D^+D^+O^{6+})_n$, $(D^+T^+O^{6+})_n$ or heavy methane clusters $(D_4^+C^{q+})_n$, $(T_2^+D_2^+C^{q+})_n$ (with $n = 100–10^5$, $R_0 =$ 10–80Å and $q = 4$ or 6), where the heavy multielectron ions ($O^{6+}$, $C^{4+}$ or $C^{6+}$) act as energetic triggers for driving the light $D^+$ or $T^+$ ions to considerably higher kinetic energies than for totally ionized homonuclear $(D_2)_n$ or $(DT)_n$ clusters of the same size. For example, the maximal energy, $E_M$, of deuterons produced by Coulomb explosion of a homonuclear $(D^+)_{3376}$ cluster ($R_0$ = 25.4Å) is $E_M$ = 1.9keV, while the heteronuclear $(D^+D^+O_6^+)_{2172}$ cluster with the same radius ($R_0$ = 25Å) provides deuterons with $E_M$ = 10.8keV. An extreme way to obtain highly effective energetic triggers for driving of deuterons to very high energies, can be accomplished by multielectron ionization of mixed $(Xe)_m(D_2)_n$ clusters or of $(DI)_n$ heteroclusters, where heavily charged $Xe^{+26}$ or $I^{+26}$ ions, with huge charges , can be produced in very intense ($I = 10^{19}Wcm^{-2}$) laser fields. In addition, kinematic effects, which manifest a sharp energy maximum of the light ion kinetic energy spectra emerging from deuterium containing molecular heteroclusters, provide a supplementary contribution to the efficiency of the NFDCE [20-23]. The Tel Aviv work determined the cluster size dependence of the fusion reaction yields (Y) per laser pulse (Fig. 3), e.g., for the NFDCE of $(D_2O)_n$ and $(DTO)_n$ clusters with $R_0$ = 30Å, $Y = 10^6$ for the dd reaction and $Y = 10^8$ for the dt reaction. Thus a theoretical and computational framework for the feasibility of the NFDCE of deuterium containing clusters was established.

## 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 the 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 in the Chemical Physics laboratory for the production of short (100ps–10ns) neutron pulses [22,35], 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 CNO cycle in hot stars. The CNO (carbon-nitrogen-oxygen) cycle of nuclear fusion, which supplies energy to the hot stars, is catalyzed by $^{12}C^{6+}$, which is regenerated [22,23]. The $^{12}C^{6+}+H^{+}$ NFDCE can be induced by multielectron ionization of sufficiently large methane clusters ($R_0$ = 120Å), providing information on the cross sections and dynamics of elemental nuclear processes in astrophysics.

Some novel 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 T = 100nk–100 μK, involving gases in magneto-optical traps, optical molasses and Bose-Einstein condensates. A striking analogy was established between the nuclear dynamics of ultracold optical molasses (T = 10–100 μK) and Coulomb explosion of multicharged atomic clusters [24]. These studies, together with exploration of superfluidity of Helium–4 clusters, bridge between the dynamics of clusters and ultracold large finite systems.

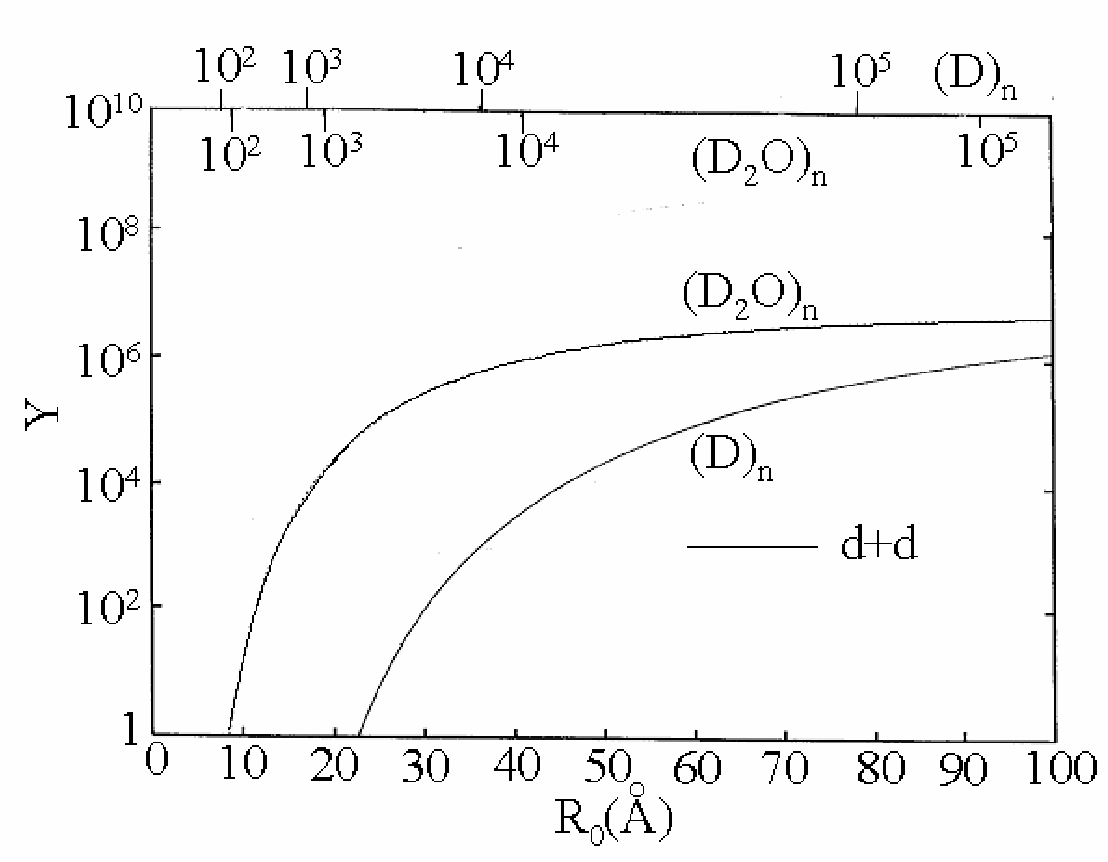

Fig. 3. The cluster size dependence of the neutron yields (Y) for dd NFDCE per laser pulse (under the experimental conditions of reference 35) calculated from molecular dynamics simulations for multielectron ionization and Coulomb explosion of homonuclear $(D)_n$ and heteronuclear $(D_2O)_n$ clusters ($n = 10^2 - 10^5$) in very intense ($I = 10^{17}$ Wcm$^{-2}$) laser fields (reference 21). The number of atoms corresponding to a cluster radius $R_0$ is shown on the upper scale for the homonuclear and heteronuclear clusters. The NFDCE for the heteronuclear clusters manifests a considerably larger neutron yield than for the homonuclear clusters of the same size, exhibiting energetic and kinematic effects, discussed in the text.

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