Ionization of cluster atoms in a strong laser field

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Inner and outer multiple ionization of clusters by a superintense ultrashort laser pulse is studied. The barrier-suppression mechanism governs inner field ionization in this case, while impact ionization can be neglected. Outer ionization produces a static Coulomb field inside the ionized cluster. This field increases the charge multiplicity of the atomic ions produced inside the cluster approximately by a factor of 1.5. Various models are suggested for the charge distribution inside the cluster.

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I. INTRODUCTION

A specific hot plasma is produced in the irradiation of cluster beams by superintense femtosecond laser pulses. Under typical experimental conditions, a large cluster contains 10^3–10^7 atoms. Laser pulses with the intensity in the range 10^{16}–10^{19} W/cm^2 and durations of 30–100 fs are considered here (these laser parameters are realized in many experiments [1–6]). The local solid number density of atoms inside the cluster is in the range 10^{22}–10^{23} cm^{-3}, while the average gaseous number density of these atoms inside the cluster beam is in the range 10^{18}–10^{19} cm^{-3}. This large difference between the local and the average number density leads to different behavior of cluster targets and solid targets. In contrast to solid targets, the laser pulse is not reflected by the cluster beam, and it penetrates freely through each cluster [7–9]. On the other hand, the local solid number density of the clusters provides a high initial number density of the forming plasma.

These peculiarities of cluster beams are of importance for applications of superintense laser pulses. Cluster targets provide effective x-ray generation [3,4,6,10], unlike gaseous targets. The conversion efficiency of laser energy into x-ray radiation [11] reaches several percent [5,12]. The cluster plasma contains multicharged atomic ions, and the energy of the released ions may be as high as 1 MeV [13–15]. The energy spectrum of the atomic ions has a nonthermal form; it is characterized by a sharp cutoff at high energies.

The theoretical analysis of the interaction between an intense laser pulse and a cluster beam allows us to understand various aspects of the behavior of a cluster plasma. This interaction is accompanied by strong inner and outer ionization of the clusters. The barrier-suppression mechanism of ionization is realized (see Refs. [16–18]). Some fraction of the released electrons leaves the cluster, which acquires a positive charge Q. The subsequent redistribution of the charge inside the cluster leads to the formation of a self-consistent static electric field. This field causes additional ionization of the atomic ions; this ionization is most efficient at the cluster surface in the direction of the laser polarization. As a result, multicharged atomic ions with charge $Z=12$–36 are produced [19,20]. On the other hand, additional ionization of small clusters (size <10 Å) is determined by collisions between neighboring atomic ions and electrons [21].

The ionized clusters are unstable. The cluster size increases due to Coulomb and hydrodynamic expansion; the clusters are transformed into a uniform plasma within 1–50 ps. The cluster expansion occurs after the end of the laser pulse. The role of impact ionization is small since the electron number density quickly drops during the cluster explosion. The skin depth is of the order of several hundred angstroms, so that it exceeds the cluster size (even a cluster consisting of 10^6 Xe atoms has a radius ~ 250 Å [16]).

A model of cluster ionization that explains the high charge of the atomic ions produced is developed. This charge is determined both by barrier-suppression ionization of the atoms and by the self-consistent static electric field. Within the framework of this model we find the key parameter that is responsible for the enhancement of ionization.

II. MECHANISMS OF INNER IONIZATION

Two mechanisms for the laser-cluster interaction can be of importance. The first one is impact ionization. We use the Lotz formula [22] to estimate the contribution to electron production from this mechanism (atomic units are used as a rule in this section, $e=m_e=\hbar=1$),

$$\sigma = 2.17 f_i \frac{\ln(E_e/J_Z)}{E_e J_Z} \text{a.u.}, \quad E_e > J_Z. \tag{1}$$

Here E_e is the kinetic energy of the incident electron, J_Z is the ionization potential of an atomic ion with the charge multiplicity Z, and f_i is the number of electrons in the valence shell. The rate of impact ionization is

$$w = \langle n_e \sigma \sqrt{2E_e} \rangle, \tag{2}$$

where n_e is the electron number density in the cluster and the brackets $\langle \cdots \rangle$ denote an average over the electron velocity distribution. We assume that the electron number density is constant inside the cluster and zero outside the cluster.

Electrons with energies $E_e = v_e^2/2 > Q/R$ (R is the cluster radius) leave the cluster; therefore the velocity distribution of

TABLE I. Ionization potentials J_Z of Xe ions [26] and their approximated values $\overline{J_Z}$ [Eq. (12)].

Z	J_Z (eV)	$\overline{J_Z}$	Z	J_Z (eV)	$\overline{J_Z}$
1	12.1	1.6	2	21.21	6.2
3	32.1	14.0	4	46.7	24.8
5	59.7	38.8	6	71.8	55.8
7	92.1	76	8	105.9	100
9	171	125	10	202	155
11	233	188	12	263	223
13	294	262	14	325	303
15	358	349	16	390	397
17	421	448	18	452	502
19	549	560	20	583	620
21	618	684	22	651	750
23	701	820	24	737	893
25	819	969	26	897	1050
27	1385	1130	28	1491	1215
29	1587	1303	30	1684	1395
31	1781	1489	32	1877	1590
33	1987	1690	34	2085	1790
35	2211	1900	36	2302	2010
37	2554	2120	38	2639	2240
39	2728	2360	40	2812	2480

the electrons inside the cluster can be written in the form

$$f(v_e) = \begin{cases} \dfrac{3}{4\pi}\left(\dfrac{R}{2Q}\right)^{3/2}, & v_e^2 < \dfrac{2Q}{R} \\ 0, & v_e^2 > \dfrac{2Q}{R}. \end{cases} \tag{3}$$

Here Q is the total charge of the ionized cluster. This function is normalized by the condition

$$\int f(v_e) d\mathbf{v}_e = 1. \tag{4}$$

Substituting Eq. (3) into Eq. (2), one obtains

$$w = 4.60\sqrt{\frac{R}{Q}}\frac{f_i n_e}{J_Z}\ln\left(\frac{Q}{RJ_Z}\right) \quad \text{a.u.} \tag{5}$$

As an example, we consider an ionized Xe cluster consisting of the atomic ions Xe^{12+}; the ionization potential of the cluster is $Q/R=20$ keV (see Table I). The electron number density is $n_e \approx Zn_i = 2\times 10^{23}$ cm^{-3}. It follows from Eq. (5) that $w=0.04$ fs^{-1}, i.e., total ionization is reached during the laser pulse.

The second ionization mechanism is produced by the laser field and the self-consistent electric field. This mechanism is described within the frame of barrier-suppression ionization [23]: a valence electron is liberated when the effective potential barrier disappears and this electron escapes to infinity. Barrier-suppression ionization dominates for laser intensities higher than 10^{16} W/cm^2 for ground states of atoms [24]. The threshold electric field strength F for this process is given by the Bethe formula [23]

$$F_{\rm th} = \frac{J_Z^2}{4Z}. \tag{6}$$

Additional electric fields appear inside the ionized cluster. There are three kinds of such fields. First, adjacent atomic ions and electrons interact with a test atomic ion. The second type results from collective electron oscillations inside the cluster. Finally, the self-consistent electric field produced by the cluster charge should be taken into account.

The electric-field strength generated by neighboring charged particles can be estimated as $F_{\rm n} \sim \delta Z/r_{\rm W}^2 = Qr_W/R^3$, where δZ is the average number of electrons per atomic ion that leave the cluster, and $r_W = N/R^{1/3}$ is the Wigner-Seitz radius, N is the number of atoms in the cluster. A typical value of the self-consistent electric-field strength is $F_{\rm S} \sim Q/R^2$. The ratio $F_{\rm n}/F_{\rm S}$ is of the order of $\sim N^{-1/3} \ll 1$. Hence, one can neglect the field $F_{\rm n}$.

A dynamic electric field results from oscillations of the free electrons inside the cluster driven by the laser field. The vector potential $\mathbf{A}$ produced by the electron oscillations is determined by the formula

$$\mathbf{A}(\mathbf{r}) = \frac{1}{c}\int \frac{\mathbf{v}(\mathbf{r}')\rho(r')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}'.$$

The dynamic electric field strength is

$$|\mathbf{F}_{\rm d}| = \left|\frac{1}{c^2}\frac{\partial}{\partial t}\int \frac{\mathbf{v}(\mathbf{r}')\rho(r')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}'\right| \sim \frac{\dot{v}_a Q_{\rm e}}{c^2 R}. \tag{7}$$

Here $Q_{\rm e} = NZ - Q$ is the number of electrons inside the cluster. The local electron velocity $\mathbf{v}(\mathbf{r})$ has been replaced by the average electron velocity $v_{\rm a}$. The average electron acceleration in the cluster can be estimated as

$$\dot{v}_{\rm a} \sim a\omega^2, \tag{8}$$

where a is the displacement of the electron number density under the action of the laser field. This value is of the order of $a \sim F/\omega_p^2$ (ω_p is the plasma frequency) [25]. Substituting Eq. (8) into Eq. (7), one obtains

$$\frac{F_{\rm d}}{F} \sim \left(\frac{R}{\lambda}\right)^2 \ll 1.$$

Here λ is the laser wavelength. Thus, the dynamic field and the field from adjacent atomic ions and electrons can be neglected. However, the Coulomb-barrier lowering due to neighboring ions can reduce the number of photons required to remove further electrons. Estimations show that this effect is also small. The additional field inside the cluster is produced mainly by the self-consistent electric field of the ionized cluster.

The field inside the cluster is diminished in comparison to the external laser field by ω_p/ω times, since the laser frequency ω is less than the plasma frequency ω_p. But this fact does not change above conclusions.

III. CHARGE DISTRIBUTIONS OF ATOMIC IONS IN THE CLUSTER

We have found that the charge multiplicity of atomic ions inside the cluster is determined by the superposition of the laser field $\mathbf{F}(t)$ and the Coulomb electric field $\mathbf{F}_S(r)$ inside the ionized cluster. The Poisson equation for the total electric field $\mathbf{F}_S(r)+\mathbf{F}(t)$ inside the cluster can be written in the form

$$\operatorname{div}[\mathbf{F}_S(r) + \mathbf{F}(t)] = \operatorname{div}\mathbf{F}_S(r) = 4\pi[n_i Z(r) - n_e(r)]. \quad (9)$$

Here n_i is the initial number density of the atomic ions, $Z(r)$ is the charge of the atomic ions produced at a distance r from the cluster center, and $n_e(r)$ is the number density of the free electrons in the cluster. Since the laser wavelength λ is much larger than the cluster size R, the laser field strength $\mathbf{F}(t)$ does not change inside the cluster. Therefore one can ignore the spatial dependence for the laser field $\mathbf{F}(t)$ in Eq. (9). According to the Bethe formula [23] for barrier-suppression ionization, the connection between the charge multiplicity $Z(r)$ of the atomic ion and the electric-field strength $F_S(r)$ is given by

$$F \cos\theta \cos\omega t + F_S(r) = \frac{J_Z^2}{4Z}. \quad (10)$$

Here J_Z is the ionization potential of an atomic ion with charge multiplicity Z, ω is the laser frequency, and θ is the angle between the laser field strength and the radius–vector $\mathbf{r}$. Liberation of electrons proceeds when the laser field reaches its maximum and occurs faster than the laser field period. Therefore we can replace the current laser field strength $F(t)\cos\omega t$ by its amplitude $F(t)$ in Eq. (10). First we assume that the superposition of these two fields is independent of the angle between their directions. Below we demonstrate that this assumption does not change results. It follows from Eq. (10),

$$F(t) + F_S(r) = \frac{J_Z^2}{4Z}. \quad (11)$$

In the case of heavy atoms, the ionization potential J_Z of the atomic ion can be approximated by the formula

$$J_Z = 0.057 Z^2 \text{ a.u.}, \quad Z > 5. \quad (12)$$

A comparison of this formula with experimental values of the ionization potentials has been carried out in Table I. Substituting Eqs. (12) and (11) into Eq. (9), we get finally

$$\frac{1}{r^2}\frac{d}{dr}\left[r^2\left(\frac{Z^3(r)}{1230} - F\right)\right] = 4\pi[n_i Z(r) - n_e(r)]. \quad (13)$$

The boundary condition is of the form $F_S(0)=0$: the self-consistent field strength is zero at the cluster center. Equation (13) includes the dependence of the electron number density n_e on the distance r from the cluster center.

We now consider several possible distributions $n_e(r)$ and solve Eq. (13) for these cases. The simplest version takes the radial distribution of the electron number density $n_e(r)$ in the form $n_e(r)=n_{0e}Z(r)$, where $Z(r)$ and n_{0e} are an ion charge and a constant. The charge Q of the ionized cluster and the constant n_{0e} are related by

$$Q = (n_i - n_{0e})\int_0^R Z(r) 4\pi r^2 dr, \quad (14)$$

where R is the cluster radius. Taking into account Eq. (14), we rewrite Eq. (13) for $Z(r)$ in the form

$$\frac{1}{r^2}\frac{d}{dr}\left[r^2\left(\frac{Z^3(r)}{1230} - F\right)\right] = \frac{Z(r)Q}{\int_0^R Z(r')r_2'dr'}. \quad (15)$$

Let us introduce the dimensionless variables

$$y = \frac{Z(r)}{Z(0)} \quad \text{and} \quad x = \frac{r}{R}, \quad (16)$$

where $Z(0)=10.71F^{1/3}$ is the charge of the atomic ion in the absence of the self-consistent electric field $\mathbf{F}_S$. Then Eq. (15) is reduced to

$$\frac{1}{x^2}\frac{d}{dx}\{x^2[y^3(x) - 1]\} = \frac{ky(x)}{\int_0^1 y(u)u^2 du},$$

$$y(0) = 1, \quad k \equiv \frac{Q}{FR^2}.$$

The solution $y(x,k)$ of Eq. (17) depends only on the one-dimensionless parameter k. This parameter is the ratio of the external laser field strength $\mathbf{F}$ over the self-consistent field strength $\mathbf{F}_S$. The solution may be approximated also analytically, if $y(x)$ is expanded in a Taylor series for $0<x<1$:

$$y(x) = 1 + ax - bx^2,$$

$$a = \tfrac{2}{3}(\sqrt{1+k} - 1),$$

$$b = \tfrac{1}{9}(2 + k - 2\sqrt{1+k}).$$

It is seen that this expansion is valid under the condition $b \ll a$. For example, in the case of the Bethe approximation for outer ionization we have $k=4$ [16]. Then it follows from Eq. (18),

$$y(x) = 1 + 0.824x - 0.170x^2. \quad (19)$$

In Fig. 1 we present the numerical solution of Eq. (17) for $k=4$ (thick solid line) and its approximation by formula (19) (dotted line). It is seen that both solutions practically coincide. In particular, according to Eq. (19), $y(1)=1.654$ for $k=4$. Thus, according to this model, the charge of atomic ions at the cluster periphery is about 1.65 times larger than that at the cluster center.

We now consider another version of the charge distribution inside the cluster assuming the charge density to be constant, i.e.,

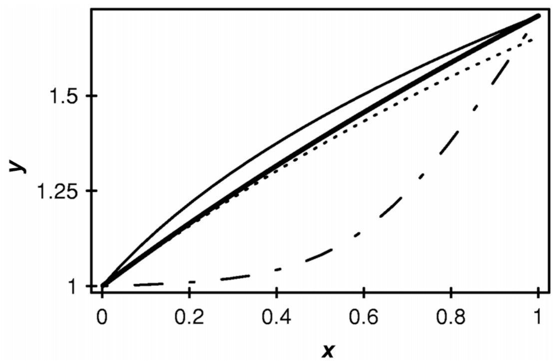

FIG. 1. The dependence of the reduced charge y [see Eq. (16)] on the reduced distance from the cluster center x for $k=4$: the thick solid line is the numerical solution of Eq. (17), the dotted line is described by formula (19), the thin solid line corresponds to formula (23), and the dash-dotted line relates to the numerical solution of Eq. (25) with $\beta=32.3$.

$$n_i Z(r) - n_e(r) = \frac{3Q}{4\pi R^3}. \qquad (20)$$

Substituting Eq. (20) into Eq. (13), one can obtain, instead of Eq. (17),

$$\frac{1}{r^2}\frac{d}{dr}\left[r^2\left(\frac{Z^3(r)}{1230} - F\right)\right] = \frac{3Q}{R^3}. \qquad (21)$$

Using the dimensionless variables (16), this equation can be rewritten in analogy with Eq. (17),

$$\frac{1}{x^2}\frac{d}{dx}\{x^2[y^3(x) - 1]\} = 3k, \quad y(0) = 1. \qquad (22)$$

Its solution is of the form

$$y(x,k) = (1 + kx)^{1/3}. \qquad (23)$$

In particular, $y(1)=1.71$ for $k=4$. The function $y(x,k)$ Eq. (23) for $k=4$ is given in Fig. 1 by a thin solid line. It follows that both models give closely related charge distributions.

We now consider yet another version of the charge distribution assuming the electron number density to be constant (n_e= const) inside the cluster. Then we have for the total charge of the ionized cluster Q, instead of Eq. (14),

$$Q = n_i\int_0^R Z(r)4\pi r^2 dr - n_e\frac{4}{3}\pi R^3.$$

Equation (13) is transformed into

$$\frac{1}{r^2}\frac{d}{dr}\left[r^2\left(\frac{Z^3(r)}{1230} - F\right)\right] = 4\pi n_i Z(r) - \frac{3n_i}{R^3}\int_0^R Z(r)4\pi r^2 dr + \frac{3Q}{R^3}. \qquad (24)$$

It can be rewritten, using the dimensionless variables defined above, as

$$\frac{1}{x^2}\frac{d}{dx}\{x^2[y^3(x) - 1]\} = 3k + \beta\left(y(x) - 3\int_0^1 y(u)u^2 du\right),$$

$$y(0) = 1.$$

Here the following notation is used:

$$\beta = 134.6\frac{n_i R}{F^{2/3}}. \qquad (26)$$

Equation (25) differs from Eq. (22) by the additional second term on the right-hand side of Eq. (25). This term increases the effective value of k.

In contrast to the previous two cases, the solution of Eq. (25) depends on two parameters, k and β. For example, in the case of a Xe cluster ($n_i=0.0024$ a.u.) with $N=10^6$ atoms we have $R=464$ a.u. Taking for the laser field strength $F=10$ a.u., we find $\beta=32.3$. The solution of Eq. (25) for $k=4$ is shown in Fig. 1 by a dash-dotted line. In contrast to the last model, the first two models lead to almost identical results. Indeed, in the last model, an excess cluster charge is concentrated mostly near the cluster surface; it creates a weak electric field inside the cluster.

IV. DISCUSSION

Above, we used a one-dimensional model of barrier-suppression ionization of atomic ions. However, the total electric-field strength depends on the angle θ between the self-consistent cluster field and the laser field

$$F_{\mathrm{tot}} = \sqrt{[F + F_{\mathrm{S}}(r)\cos\,\theta]^2 + F_{\mathrm{S}}^2(r)\sin^2\,\theta} = \sqrt{F^2 + F_{\mathrm{S}}^2(r) + 2FF_{\mathrm{S}}(r)\cos\,\theta}. \qquad (27)$$

It is seen that the total electric-field strength varies from $\sqrt{F^2+F_{\mathrm{S}}^2(r)}$ up to $F+F_{\mathrm{S}}(r)$. We estimate the accuracy of the above model of a constant electron number density n_e inside the cluster. Taking the total field strength as $\sqrt{F^2+F_{\mathrm{S}}^2(r)}$, we obtain the Bethe formula in the form [instead of Eq. (11)]

$$\sqrt{F^2 + F_{\mathrm{S}}^2(r)} = \frac{Z^3(r)}{1230}. \qquad (28)$$

Then, Eq. (22) is transformed into the following equation for $y(x)$:

$$\frac{1}{x^2}\frac{d}{dx}[x^2\sqrt{y^6(x) - 1}] = 3k, \quad y(0) = 1. \qquad (29)$$

The solution of this equation has the form

$$y(x) = [1 + (kx)^2]^{1/6}. \qquad (30)$$

For $k=4$ we find $y(1)=17^{1/6}\sim 1.60$.

This solution is shown in Fig. 2. It is seen that now the effect of the Coulomb electric field F_{S} is smaller than in the case when both fields are parallel (solid line); however, the difference between the two cases [compare Eq. (23)] is only a few percent. In addition, we show in Fig. 3 the ratio of the ion charges for the cases when the laser field and the Cou-

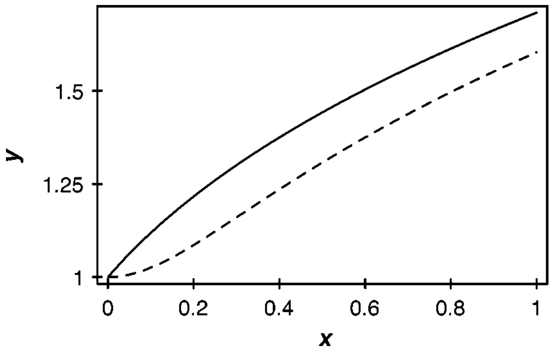

FIG. 2. Reduced ion charge y as a function of the reduced distance $x=r/R$ from the cluster center: the solid line corresponds to the case when the cluster and laser fields are parallel, Eq. (23), to each other; the dashed line relates to the case, if cluster and laser fields are perpendicular, Eq. (30), to each other.

lomb electric field are parallel and perpendicular to each other. This ratio is a function of the distance $x=r/R$ from the cluster center.

The hierarchy of times for the various processes during the laser pulse determines the establishment of an equilibrium ionization at each stage of the laser pulse. In particular, the equilibrium of electrons inside the cluster is maintained by ionization processes. A typical time of electron release from an atomic ion by barrier-suppression ionization is the time of electron motion through the barrier [16,17]; it does not exceed atomic times. This time is estimated as 0.1 fs; thus the ionization equilibrium with respect to the liberation of atomic electrons is established promptly, as soon as the potential barrier disappears.

The outer ionization of the cluster requires a longer time because an electron has to pass a longer way through the barrier. A typical time for the establishment of this ionization equilibrium is less than a half laser period (i.e., $\sim$1 fs) for clusters consisting of 10^6 atoms. Redistribution of electrons inside the ionized cluster proceeds approximately within the same period of time, i.e., the self-consistent cluster field establishes itself during the first period of the laser field. Thus, equilibrium ionization in the cluster is established on the leading edge of the laser pulse.

This hierarchy of ionization times is important for the formation of the cluster plasma. This plasma consists of multicharged ions and electrons which are locked in the self-consistent electric field. We consider large clusters when the number of cluster atoms exceeds $N=1000$. Then, even for very high intensity of the laser pulse, only a small part of the cluster electrons is released, while most of the liberated electrons remain locked inside the cluster.

When the laser field is switched off, ionization processes largely stop. Since cluster expansion proceeds after the end

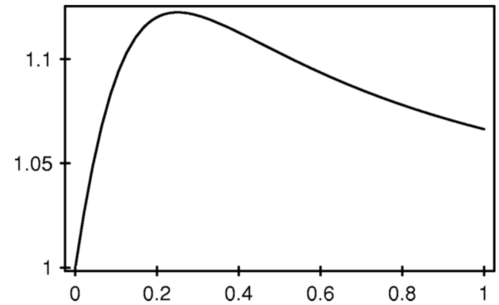

FIG. 3. Ratio of ion charges (ordinate) for the cases when the laser and cluster fields are parallel, Eq. (23), and perpendicular, Eq. (30), to each other, as a function of the reduced distance (abscissa) $x=r/R$ for $k=4$.

of the laser pulse, the ionization processes during this stage are weak. Moreover, the rate of impact processes drops sharply with cluster expansion, because the expansion process is accompanied by a sharp decrease of the electron number density.

Above, we ignored cluster expansion during the laser pulse. For moderately sized clusters and strong fields, this assumption can be violated partially. Nevertheless, the ionization processes as described above hardly change. Indeed, according to the Bethe formula (6) for the outer ionization of the cluster, the cluster charge is $Q\sim R^2$; therefore the parameter $k=Q/(FR^2)$, which is responsible for the increase of ionization due to the self-consistent cluster field, is independent of the cluster radius R. Hence, cluster expansion does not affect the ionization processes inside the cluster.

V. CONCLUSION

The above analysis demonstrates the barrier-suppression type of inner and outer ionization of clusters by a superintense femtosecond laser pulse. This process proceeds at the leading edge of the laser pulse. It is accompanied by redistribution of the cluster electrons and creates a self-consistent electric field. The latter produces additional inner ionization. As a result, the charge multiplicity of the atomic ions is higher at the cluster surface than at its center, approximately by a factor of 1.6–1.7. This result follows from various models of the charge distribution inside the cluster.

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