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# HARD $X$-RAY EMISSION BY CLUSTERS IN AN INTENSE FEMTOSECOND LASER FIELD AT THE COLLECTIVE RECOMBINATION

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We consider the new mechanism of $X$-ray generation by clusters under irradiation by femtosecond laser pulses, the so called collective photo-recombination. We develop the theory of the photo-recombination of electrons that pass from atomic clusters at the outer ionization to the ground level of a homogeneously charged cluster. Such a cluster is considered as a quantum potential well. The dipole approximation is inapplicable for this process. We conclude that $X$-ray photons in the collective photo-recombination on a charged cluster as a whole have the energy that is much larger than that for the photo-recombination on separate atomic ions inside the cluster. For a typical cluster of $2.25 \cdot 10^6$ electrons, with the radius $R = 300$ Å and the number density of plasma electrons $n_e = 2 \cdot 10^{22}$ cm$^{-3}$, we obtain that at a 5 % outer ionization of this cluster, the energy of hard $X$-ray photons is 7.2 keV.

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The main experimental observables for energy transfer from laser pulse to a cluster and the major theoretical approaches are discussed in the recent review paper [1]. The interaction of an intense femtosecond laser pulse with a cluster is accompanied by efficient transformation of laser energy into hard $X$-rays [2, 3]. For example, these $X$-rays are produced at the radiation recombination by atomic $M$-shell emission from Xe clusters under their irradiation by a TW, 800 nm Ti:sapphire laser system delivering pulses with the duration 120 fs [4]. The peak laser intensity in this experiment was $7.4 \cdot 10^{17}$ W/cm$^2$ and the observed photon energy was $\sim 1$ keV. High-charged atomic ions inside the clusters are produced at the collisional ionization by hot electrons. In particular, the electron energy is higher than 100 keV at the irradiation of Ar clusters with a laser pulse of duration 28 fs and intensity $1 \cdot 10^{17}$ W/cm$^2$ [5]. The electrons in this experiment are accelerated via the betatron resonance mechanism. In the recent experiment [6], intense (up to a few $10^{17}$ W/cm$^2$) femtosecond (down to 40 fs) laser pulses are focused onto a partially clusterized argon gas jet. The interaction leads to $X$-ray emission that is spectrally resolved using a high-resolution time-integrated spectrometer in the $K$-shell range from 2.9 to 4.3 keV. The observed characteristic lines demonstrate highly charged atomic ions up to $Ar^{16+}$.

Various effects at the irradiation of large Van der Waals clusters by intense femtosecond laser pulses were reviewed in Ref. [7].

The emission of $X$-rays by clusters of moderate to large size ($10^4$–$10^6$ atoms) irradiated by an intense laser pulse ($10^{16}$–$10^{19}$ W/cm$^2$) is considered in Ref. [8]. Over-the-barrier ionization of the cluster atoms generates multicharged ions and electrons. For increasing the cluster size, more and more electrons remain trapped inside the cluster. $X$-rays are generated in the interaction of the trapped electrons with the cluster ions. Three such processes are considered in detail: direct photo-recombination of free electrons and ions, dielectronic recombination, and excitation of ions by electron impact followed by collisional recombination. It is shown that the main contribution to the $X$-ray emission under the above conditions is made by the electron impact excitation, while dielectronic recombination is less important, and photo-recombination hardly plays any role. The excitation is created before the cluster explodes, whereas emission of $X$-rays continues there-

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after.

Emission of $X$-rays by large clusters with $10^8$ to $10^{10}$ atoms irradiated by an intense laser pulse ($10^{16}$ to $10^{18}$ W/cm$^2$) is analyzed in Ref. [9]. A self-consistent model for the cluster evolution during and after irradiation is developed. The model takes the absorption of radiation, formation of multicharged ions, and cluster expansion into account. The model is applied to relate the parameters of the cluster beam and the laser pulse to those of the forming plasma. They find that the expansion of large clusters goes through different stages after the end of the laser pulse and is not accurately described by the Coulomb expansion model, which underestimates the cluster lifetime. For a more realistic description, the nonuniformity of the plasma must be considered. Two radiative processes are considered in detail: dielectronic recombination and excitation of ions by electron impact followed by photon emission. Under the above conditions, the contributions of both processes to $X$-ray emission are comparable. It is experimentally shown in [10] that there is a very significant enhancement in the yield of Ar $K$-shell $X$-rays in heterogeneous clusters that are made by coexpansion of Ar gas with water. The presence of water provides additional free electrons for collisional ionization, which leads to higher charge states and a larger fraction of Ar atoms with innershell vacancies, leading to enhancement in the $K$-shell $X$-ray yield. The change in the ionization intensity is argued to be due to the presence of easily ionizable water molecules doping the Ar clusters.

In our paper [11], various mechanisms of recombination of electrons with multiply charged atomic ions in atomic clusters irradiated by superintense femtosecond laser pulses are discussed: collisional recombination, dielectronic recombination, and direct photorecombination. All the recombination mechanisms are shown to take a time considerably longer than the laser pulse duration and, hence, they can develop only in a homogeneous, fairly rarefied cluster plasma after pulse termination.

This paper is devoted to the new mechanism of $X$-ray generation by clusters at their irradiation by femtosecond laser pulses, the so called collective photorecombination. We consider the photo-recombination of electrons that pass from atomic clusters at the outer ionization to the ground level of a charged cluster considering as a quantum potential well.

The potential energy of an electron in the field of a uniformly charged cluster with the radius $R$ and charge $Z$ is given by (here and hereafter, the atomic system of units is used, $e = \hbar = m_e = 1$)

$$U(r) = \begin{cases} -\dfrac{Z}{r}, & r > R, \\ \dfrac{Z}{R}\left(-\dfrac{3}{2} + \dfrac{r^2}{2R^2}\right), & r < R. \end{cases} \tag{1}$$

The cluster charge is a result of the outer ionization produced by the laser field.

The inequality $R \gg a_0/Z$ is always satisfied for atomic clusters. Here, $a_0 = \hbar^2/m_e e^2$ is the Bohr radius. Therefore, the ground and low-lying excited quantum states in potential (1) are described well by the potential of a spherical harmonic oscillator with the frequency

$$\omega_0 = \sqrt{\frac{Z}{R^3}} \ll U(r) \sim \frac{Z}{R}.$$

The energy of the ground state in this potential referenced to the bottom of the potential well is

$$\frac{3}{2}\omega_0 = \frac{3}{2}\sqrt{\frac{Z}{R^3}}.$$

This energy referenced to $U = 0$ is approximately

$$E = \frac{3}{2}\frac{Z}{R} \gg \omega_0$$

(see Fig. 1).

As an example, we consider a large atomic cluster with the radius $R = 300$ Å and with the typical number density of plasma electrons $n_e = 2\cdot 10^{22}$ cm$^{-3}$. This

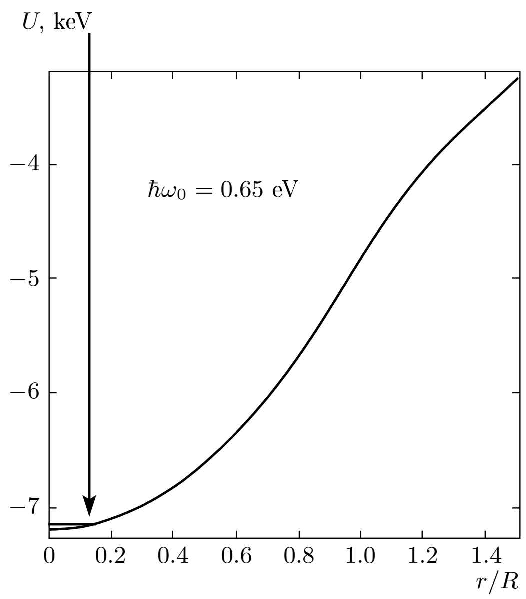


**Fig. 1.** The cluster potential $U(r)$ and the photorecombination schematic process, $N = 2.25 \cdot 10^6$, $Z = 10^5$, $R = 300$ Å

cluster contains $N = n_e(4\pi R^3/3) = 2.25\cdot 10^6$ electrons. In such a large cluster, approximately 5 % of electrons are typically ejected by laser field from the cluster. Hence, the charge of the cluster is $Z \approx 10^5$. The frequency of the harmonic oscillator is $\omega_0 = 0.64$ eV. The ionization potential of the ground level is $E = 7.2$ keV.

The normalized wave function of the ground state of the spherical harmonic oscillator is

$$\psi(r) = \left(\frac{Z}{\pi^2 R^3}\right)^{3/8} \exp\left(-\frac{\sqrt{Z/R^3}r^2}{2}\right). \qquad (2)$$

The typical radius of this state is $r_0 = (4R^3/Z)^{1/4}$. It is equal approximately to 5 Å in the above example. The wavelength of an $X$-ray photon emitted at the photo-recombination is $4\pi cR/3Z \approx 1.7$ Å. Thus, the photon wavelength is less than the radius of the ground state. Hence, the dipole approximation is inapplicable for describing photo-ionization and photo-recombination, although the considered problem is nonrelativistic.

To derive the photo-recombination cross section, we first calculate the cross section of the inverse process, i.e. of the photo-ionization. The differential cross section is

$$d\sigma_{ion} = \frac{p}{2\pi\omega c}\left|M_{fi}\right|^2 d\Omega, \qquad (3)$$

where $p$ is the electron momentum in the final continuum state, $\omega$ is the frequency of the absorbed photon, and $M_{fi}$ is the matrix element for the bound-free electron transition. According to the energy conservation law, we have $\omega = E + p^2/2$. We note that the electron energy in the final continuum state $p^2/2 \ll E$, because the photo-ionization cross section is exponentially small at $p^2/2 \sim E$ and $p^2/2 > E$.

The transition matrix element $M_{fi}$ is given by

$$M_{fi} = C\,(\mathbf{e}\cdot\mathbf{p}) \int \exp\left(-i\mathbf{p}\cdot\mathbf{r} + i\mathbf{k}\cdot\mathbf{r}\right)\psi(r)\,d\mathbf{r}, \qquad (4)$$

where $\mathbf{e}$ is the unit polarization vector of the absorbed photon and $\mathbf{k}$ is the photon wave vector, $|\mathbf{k}| = \omega/c$. The final electron state is described here by the plane wave $C\exp(i\mathbf{p}\cdot\mathbf{r})$. This is correct because the wave functions of the initial and final electron states overlap in the vicinity of small values of $r \sim r_0$; in this region, the potential of the charged cluster in Eq. (1) is practically constant. We expand the real continuum wave function of the final state with respect to plane waves in the semiclassical approximation, and hence the dimensionless normalized coefficient $C$ is

$$C = \sqrt{\frac{p}{(2E)^{1/2}}} \approx \sqrt{\frac{p}{(3Z/R)^{1/2}}} \ll 1.$$

The exponential $\exp(i\mathbf{k}\cdot\mathbf{r})$ cannot be expanded in the Taylor series because, as we have said, the dipole approximation is not valid, and significant values of $\mathbf{k}\cdot\mathbf{r}$ are of the order of unity. Significant values of $p$ in the integral in Eq. (4) are of the order of $k$. Hence, electrons are ejected mainly with the kinetic energy

$$\frac{k^2}{2} \sim \frac{\omega^2}{2c^2} \sim \frac{9}{8}\left(\frac{Z}{cR}\right)^2.$$

In the above example, this energy is approximately 50 eV.

To calculate the matrix element, we introduce the notation $\theta$ for the angle between the vectors $\mathbf{r}$ and $\mathbf{k}-\mathbf{p}$. Then

$$d\mathbf{r} = 2\pi r^2 dr\, dx,$$

where $x = \cos\theta$. Integrating Eq. (4) over $\theta$ yields

$$M_{fi} = \frac{4\pi C\mathbf{e}\cdot\mathbf{p}}{|\mathbf{k}-\mathbf{p}|}\int_0^\infty \psi(r)\sin\left(|\mathbf{k}-\mathbf{p}|r\right) r\,dr. \qquad (5)$$

We introduce the angle $\vartheta$ between vectors $\mathbf{k}$ and $\mathbf{p}$. Then

$$\mathbf{e}\cdot\mathbf{p} = p\sin\vartheta\cos\varphi$$

and Eq. (5) can be rewritten as

$$M_{fi} = \frac{4\pi pC\sin\vartheta\cos\varphi}{\sqrt{k^2+p^2-2kp\cos\vartheta}}\left(\frac{Z}{\pi^2R^3}\right)^{3/8} \times \\ \times \int_0^\infty \exp\left(-\frac{\sqrt{Z/R^3}\,r^2}{2}\right) \times \\ \times \sin\left(\sqrt{k^2+p^2-2kp\cos\vartheta}\,r\right) r\,dr. \qquad (6)$$

We here substituted the ground-state wave function from Eq. (2). Substituting Eq. (6) in Eq. (3), we derive the photoionization cross section by integrating over the angle $\varphi$:

$$\sigma_{ion} = \frac{16\sqrt{\pi}p^4}{3^{3/2}(ZR)^{3/4}c}\int_{-1}^{+1} ds \frac{1-s^2}{k^2+p^2-2kps} \times \\ \times \left|\int_0^\infty \exp\left(-\frac{\sqrt{Z/R^3}r^2}{2}\right) \times \right. \\ \left. \times \sin\left(\sqrt{k^2+p^2-2kps}\,r\right) r\,dr\right|^2. \qquad (7)$$

Here, $s = \cos\vartheta$.

The photo-recombination cross section can be found using the principle of detail equilibrium for direct and inverse processes:

$$\sigma_{rec} = \frac{2\omega^2}{c^2p^2}\sigma_{ion}. \qquad (8)$$

Substituting Eq. (7) in Eq. (8), we obtain

$$d\sigma_{rec} = \frac{24\sqrt{\pi}Z^{5/4}p^2}{R^{11/4}c^3 3^{1/2}}\int\limits_{-1}^{+1} ds \frac{1-s^2}{k^2+p^2-2kps} \times \times \left| \int\limits_0^\infty \exp\left(-\frac{\sqrt{Z/R^3}\,r^2}{2}\right) \times \times \sin\left(\sqrt{k^2+p^2-2kps}\,r\right) r\,dr \right|^2. \qquad (9)$$

Here, $k = 3Z/2cR$ because, as noted above, the electron energy $p^2/2$ is small compared to the ionization potential $3Z/2R$.

Changing the integration variable as $r = \left(R^3/Z\right)^{1/4} u$, we rewrite Eq. (9) in the form

$$\sigma_{rec} = \frac{8\sqrt{3\pi}(ZR)^{1/4}p^2}{c^3}\int\limits_{-1}^{+1} ds \frac{1-s^2}{k^2+p^2-2kps} \times \times \left| \int\limits_0^\infty \exp\left(-\frac{u^2}{2}\right) \times \times \sin\left(\sqrt{\sqrt{\frac{R^3}{Z}}\,(k^2+p^2-2kps)u}\right) u\,du \right|^2. \qquad (10)$$

We now introduce the dimensionless momentum

$$P = \left(R^3/Z\right)^{1/4} p$$

and dimensionless wave vector

$$K = \left(\frac{R^3}{Z}\right)^{1/4} k \approx \frac{3}{2c}\left(\frac{Z^3}{R}\right)^{1/4}.$$

In the above example, we have $K = 12.6 \gg 1$. Then Eq. (10) becomes

$$\sigma_{rec} = 8\sqrt{3\pi}\frac{(ZR)^{1/4}}{c^3}P^2\int\limits_{-1}^{+1} ds \frac{1-s^2}{K^2+P^2-2KPs} \times \times \left| \int\limits_0^\infty \exp\left(-\frac{u^2}{2}\right) \times \times \sin\left(\sqrt{K^2+P^2-2KPsu}\right) u\,du \right|^2. \qquad (11)$$

Integrating by parts in the integral over $u$, we rewrite this as

$$\sigma_{rec} = 2\sqrt{3\pi}\frac{(ZR)^{1/4}}{c^3}P^2\int\limits_{-1}^{+1} ds\left(1-s^2\right) \times \times \left| \int\limits_{-\infty}^\infty \exp\left(-\frac{u^2}{2}\right) \times \times \cos\left(\sqrt{K^2+P^2-2KPsu}\right) du \right|^2. \qquad (12)$$

We can further rewrite Eq. (12) in the form

$$\sigma_{rec} = 2\sqrt{3\pi}\frac{(ZR)^{1/4}}{c^3}P^2\int\limits_{-1}^{+1} ds\left(1-s^2\right) \times \times \left| \int\limits_{-\infty}^\infty \exp\left(-\frac{u^2}{2} - i\sqrt{K^2+P^2-2KPsu}\right) du \right|^2. \qquad (13)$$

The integral over $u$ is evaluated analytically:

$$\sigma_{rec} = 4\pi^{3/2}\sqrt{3}\frac{(ZR)^{1/4}}{c^3}P^2\exp(-K^2-P^2) \times \times \int\limits_{-1}^{+1} ds\left(1-s^2\right)\exp\left(2KPs\right). \qquad (14)$$

The integral over $s$ is also derived analytically:

$$\sigma_{rec}(P) = 16\left(\frac{\pi}{3}\right)^{3/2}\frac{R^{3/4}}{cZ^{5/4}}\exp(-K^2-P^2) \times \times \left(\mathrm{ch}(2KP) - \frac{\mathrm{sh}(2KP)}{2KP}\right). \qquad (15)$$

The dependence of the recombination cross section on $P$ is shown in Fig. 2 in the above case where $K = 12.6$. It can be seen that this dependence features a sharp maximum at $P = K$.

We further use the fact that $KP \gg 1$ as a rule. Then Eq. (18) simplifies to

$$\sigma_{rec}(P) = 8\left(\frac{\pi}{3}\right)^{3/2}\frac{R^{3/4}}{cZ^{5/4}}\exp\left[-\left(K-P\right)^2\right]. \qquad (16)$$

The maximum value of the recombination cross section is achieved at $P = K$:

$$\sigma_{rec}^{max} = 8\left(\frac{\pi}{3}\right)^{3/2}\frac{R^{3/4}}{cZ^{5/4}}. \qquad (17)$$

In the above example with $R = 30$ nm and $Z = 10^5$, this maximum cross section is equal to $4\cdot 10^{-5}$ a.u.$\sim 10^{-21}$ cm$^2$ at the electron energy of approximately 50 eV.

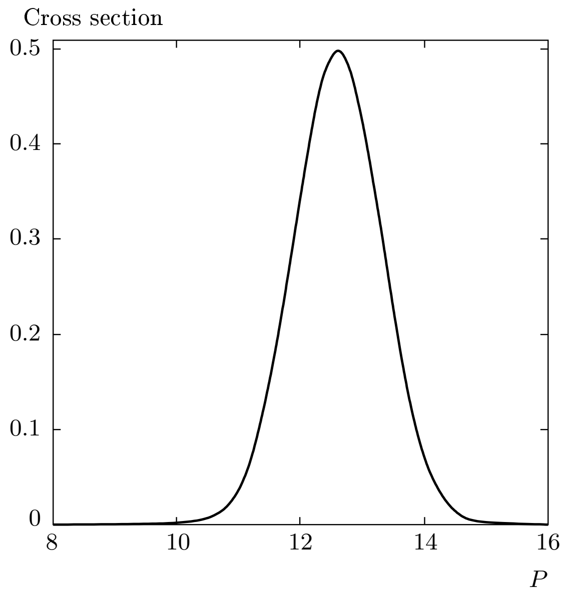


**Fig. 2.** The dependence of the dimensionless photo-recombination cross section $(3/\pi)^{3/2}Z^{5/4}c\sigma_{rec}(P)/16R^{3/4}$ on the dimensionless electron momentum $P = \left(R^3/Z\right)^{1/4}p$ for the case $K = 12.6$ (the atomic cluster with the radius $R = 300$ Å and the charge $Z = 10^5$)

The photo-recombination rate can be obtained from the cross section by multiplying it by the electron velocity $p \approx k$ and dividing by the cluster volume $4\pi R^3/3$. This gives the maximum photo-recombination rate

$$\nu_{rec} = \frac{(3\pi)^{1/2}}{c^2 R^{13/4} Z^{1/4}}. \qquad (18)$$

In the above example, this rate is $\nu_{rec} \approx 5 \cdot 10^3$ s$^{-1}$.

This quantity can be compared to the rate of the photo-recombination of an electron with the same energy per atomic ion inside the cluster. Unlike the collective photo-recombination, the dipole approximation is applicable in this case. The photo-ionization cross section with the production of an electron near the threshold is $\sigma_{ion} \sim 0.2/Z_i^2$ a.u. Here, $Z_i$ is the charge of an atomic ion. The cross section of the inverse process, i.e., photo-recombination, is found from Eq. (8) as

$$\sigma_{rec}(\text{ion}) = \frac{0.4\omega_i^2}{c^2 p^2 Z_i^2}, \qquad (19)$$

where $\omega_i$ is the frequency of the absorbed photon. In the hydrogen-like approximation, we obtain $\omega_i = Z_i^2/2$ and

$$\sigma_{rec}(\text{ion}) = \frac{0.05 Z_i^2}{c^2 E_e}, \qquad (20)$$

where $E_e$ is the electron kinetic energy. The photo-recombination rate is

$$\nu_{rec}(\text{ion}) = n_e p_e \sigma_{rec}(\text{ion}) = \frac{0.07 n_e Z_i^2}{c^2\sqrt{E_e}}. \qquad (21)$$

Substituting $E_e = 50$ eV and $Z_i = 3$, we find $\nu_{rec}(\text{ion}) \sim 3 \cdot 10^{11}$ s$^{-1}$.

We therefore conclude that the collective photo-recombination rate on a charged cluster as a whole is much less than the photo-recombination rate on separate atomic ions inside the cluster, but the photons in the collective process have the energy that is much larger than the energy for scattering on separate ions.

For the typical cluster with the radius $R = 300$ Å and with the number density of plasma electrons $n_e = 2 \cdot 10^{22}$ cm$^{-3}$, which contains $2.25 \cdot 10^6$ electrons, we obtain that at 5 % outer ionization of this cluster, the energy of hard $X$-ray photons is 7.2 keV.

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