Hard X-ray emission by atomic clusters in an intense femtosecond laser field at the collective recombination

V P Krainov and A V Sofronov
Moscow Institute of Physics and Technology, 141700 Dolgoprudny, Moscow Region, Russian Federation
E-mail: vpkrainov@mail.ru

Abstract. The new mechanism of x-ray generation by large atomic clusters at their irradiation by femtosecond laser pulses has been considered, so called collective photo-recombination. We develop the theory of the photo-recombination of electrons which leave atomic clusters at the outer ionization and then get into the ground level of a homogeneously charged cluster. The latter is considered as a quantum potential well. The dipole approximation is inapplicable for this process. The conclusion has been made that x-ray photons in the collective photo-recombination on the charged cluster as on a whole have the energy which is much larger than that for the photo-recombination on separate atomic ions inside the cluster. 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 one obtains that at 5% outer ionization of this cluster the energy of hard x-ray photons is 7.2 keV.

1. Introduction

The multiply inner ionization of atoms occurs at the irradiation of large atomic clusters consisting of several thousands atoms of noble gases (Ar, Kr, Xe) by the front of femtosecond laser pulses having the intensity of the order of $10^{16}$ W/cm$^2$. Electrons are heated quickly due to Brunel mechanism at which an electron is ejected by laser field from cluster and returns back to cluster approximately in a half of laser period, but with the energy of the order of the ponderomotive energy. An electron can be heated also due to induced inverse bremsstrahlung inside the cluster. This process dominates at the rear side of the laser pulse when due to cluster expansion the laser frequency is equal to the Mie frequency of surface plasma oscillations. Then the internal electric field is much larger than the external electric laser field. Hot electrons can collide with atomic ions inside the cluster and produce their subsequent multiply ionization. Electrons of outer atomic shells diminish their energy at the transition to the inner shells. As a result, x-ray photons of high energies are emitted [1].

In this paper we suggest the new mechanism of generation of hard x-ray radiation. Hot electrons leave cluster, and then they can be captured by another positively charged cluster. The value of cluster charge depends on the cluster size. For example, small deuterium clusters with the diameter of 2 nm in the laser field with the intensity of $10^{16}$ W/cm$^2$ quickly are transformed into positive spherical ball consisting only of deuterons [2-3]. The cluster diameter increases up to 4 nm only in 30 fs. An electron can be capture into the ground quantum state of the Coulomb potential well of the charged cluster with...
the ejection of high energy x-ray photon. The depth of the potential well is equal to several keV depending on the cluster parameters and parameters of laser pulse. Our task is derivation of rate of this recombination process.

2. Photo-recombination cross section and rate on the Coulomb field of an atomic cluster

At the outer ionization there are two possibilities of spatial distribution of electrons inside the charged cluster. The first possibility corresponds to electrons which are distributed uniformly over the cluster volume. In the second case electrons are attracted to the cluster centre so that the central part of the cluster becomes neutral region while on the cluster surface only positively charged atomic ions occur.

We consider the first possibility having in mind that hot electrons can quickly move over the whole volume of the ionized cluster. The potential energy of an electron in the field of the uniformly charged cluster with the charge $Z$ and radius $R$ is of the form

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

(1)

We use the atomic system of units $e = \hbar = m_e = 1$. In the realistic case $R > > a_b / Z$ ($a_b = \hbar^2 / me^2$ is the Bohr radius for a hydrogen atom) we obtain the potential of the spherical harmonic oscillator with the frequency $\omega_b = \sqrt{Z / R^3}$ so that the energy of the ground state accounted from the bottom of the potential well is

$$\frac{3\omega_b}{2} = 1.5 \sqrt{Z / R^3}.$$  

(2)

This energy being accounted from the zero of the potential well is approximately equal to

$$E = -\frac{3Z}{2R} > > \sqrt{Z / R^3}.$$  

(3)

Let us consider the typical example of large atomic cluster with the radius of 30 nm and the number density of free electrons of $n = 2 \times 10^{22} \text{ cm}^{-3}$. This cluster contains $2.25 \times 10^6$ electrons. In the typical situation approximately 5% of these electrons are ejected out of the cluster. Hence, the charge of the cluster is equal to $Z = 10^5$. The oscillator frequency is equal to $\omega_b = \sqrt{Z / R^3} = 0.64 \text{ eV}$. The energy of the ground state accounted from the zero the potential is $|E| = 3Z / 2R = 7.2 \text{ keV}$.

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

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

(4)

The radius of this state is

$$r_0 = \left( 4R^3 / Z \right)^{1/4}.$$  

In the above example it is equal to 0.50 nm. The wavelength of x-ray photon emitted at the photo-recombination is equal to $4\pi c R / 3Z = 0.17 \text{ nm}$. Thus, it is less than the radius of the ground state, and it is much less in comparison to the cluster radius. Hence, the dipole approximation for ionization and recombination processes is inapplicable.

First we derive the cross section for the inverse process – photo-ionization. Then the photo-recombination cross section can be derived using the principle of detailed equilibrium. The differential cross section for photo-ionization is of the form

$$d\sigma_{\text{ion}} = \frac{v}{2\pi\alpha} |M_f|^2 d\Omega,$$  

(5)

where $v$ is the velocity of the electron, $\alpha$ is the fine structure constant, and $|M_f|^2$ is the magnetic transition probability.
where \( \nu \) is the velocity of non-relativistic electron in the continuum, and the quantity

\[
\omega = -E + \frac{\nu^2}{2}
\]

is the photon frequency; it should be noted that the electron kinetic energy \( \frac{\nu^2}{2} \) is much less in comparison to the ionization potential \( E \) though electrons can be ejected from the cluster with the energies of the order of several keV. However, the rate of these photo-recombination processes is exponentially small. The mechanism of collective photo-recombination is shown in Figure 1.

![Figure 1. Mechanism of collective photo-recombination for a typical cluster](image)

The quantity

\[
M_{fi} = (e\nu) \int \exp(-i\nu r + ikr) \psi_i \psi_f r dr
\]

is the transition matrix element in which the final electron continuum state is described by the plane wave, i.e. by the wave function \( C \exp(ikr) \). Such assumption is valid since in the vicinity of the wave function of the ground state where the overlap with the continuous wave function of the final state takes place, the potential of the cluster is practically the horizontal line. The dipole approximation is inapplicable in this case due to large value of the charge \( Z \). Therefore it is incorrect to expand the exponent \( \exp(ikr) \) in Taylor series. The quantity \( e \) is the photon polarization vector, the quantity \( k \) is the photon wave vector, which is perpendicular to the vector \( e \), and \( k = \omega / c \). Electrons for which the photon momentum \( k \) and the electron momentum \( p \) are of the order of magnitude are of the main role in the considered processes of bound-free transitions. Indeed, the integral (6) is exponentially small at larger values of the electron energy because of strongly oscillating integrand.

The constant \( C \) in the continuum wave function can be found using WKB approximation taking into account that the overlap of the initial and final wave functions occurs near the cluster centre

\[
C = \sqrt{\frac{p}{2E}}
\]
Let us introduce the notation $\theta$ for the angle between the vectors $r$ and $(k - v)$; $\cos \theta = x$. Changing $dr = 2\pi r^2 drdx$, one obtains after integration over the angle $\theta$:

$$M_{fi} = \frac{4\pi C(ev)^2}{|k - v|} \int_{0}^{\pi} \sin(|k - v| \cdot r) \cdot \psi(r) \cdot rdr.$$  \hspace{1cm} (8)

Introducing the angle $\vartheta$ between the vectors $k$ and $v$, we rewrite this expression in the form

$$M_{fi} = \frac{4\pi C v \sin \vartheta \cos \varphi}{\sqrt{k^2 + v^2 - 2kv \cos \vartheta}} \int_{0}^{\infty} \left[ \sin \left( \sqrt{k^2 + v^2 - 2kv \cos \vartheta} \cdot r \right) \right] \cdot \psi(r) \cdot rdr.$$  \hspace{1cm} (9)

Substituting (4) into (9), we derive this radial integral

$$M_{fi} = \frac{4\pi^{1/4} C v \sin \vartheta \cos \varphi}{\sqrt{k^2 + v^2 - 2kv \cos \vartheta}} \int_{0}^{\infty} \left[ \sin \left( \sqrt{k^2 + v^2 - 2kv \cos \vartheta} \cdot r \right) \right] \cdot \exp \left( \frac{Z}{R^3} r^2 / 2 \right) \cdot rdr.$$  \hspace{1cm} (10)

Further we rewrite this integral using the parity of the integrand

$$M_{fi} = \frac{2\pi^{1/4} C v \sin \vartheta \cos \varphi}{\sqrt{k^2 + v^2 - 2kv \cos \vartheta}} \int_{-\infty}^{\infty} \left[ \sin \left( \sqrt{k^2 + v^2 - 2kv \cos \vartheta} \cdot r \right) \right] \cdot \exp \left( \frac{Z}{R^3} r^2 / 2 \right) \cdot rdr.$$  \hspace{1cm} (11)

Let us introduce the notation $\sqrt{k^2 + v^2 - 2kv \cos \vartheta} = \beta$; then the integral (11) can be rewritten in the form

$$M_{fi} = \frac{\pi^{1/4} C v \sin \vartheta \cos \varphi}{i\beta} \int_{-\infty}^{\infty} \exp \left( i\beta r - \frac{Z}{R^3} r^2 / 2 \right) \cdot rdr + c.c.$$  \hspace{1cm} (12)

Now we simplify this expression

$$I = \int_{-\infty}^{\infty} \exp \left( i\beta r - \frac{Z}{R^3} r^2 / 2 \right) \cdot rdr =$$

$$= \int_{-\infty}^{\infty} \exp \left[ -\left( \frac{Z}{4R^3} \right)^{1/4} r - i\beta \left( \frac{R^3}{4Z} \right)^{1/4} \right] \cdot rdr \times$$

$$\times \exp \left[ -\beta^2 \left( \frac{R^3}{4Z} \right)^{1/2} \right].$$

Changing the integral variable

$$\left( \frac{Z}{4R^3} \right)^{1/4} r - i\beta \left( \frac{R^3}{4Z} \right)^{1/4} = w$$

we obtain the simple expression for the integral $I$:

$$I = i\beta \sqrt{2\pi} \left( \frac{R^3}{Z} \right)^{3/4} \exp \left[ -\beta^2 \left( \frac{R^3}{4Z} \right)^{1/2} \right].$$

Thus, the transition matrix element is of the form

$$M_{fi} = 2^{3/4} \pi^{3/4} C v \sin \vartheta \cos \varphi \left( \frac{R^3}{Z} \right)^{3/8} \exp \left[ -\beta^2 \left( \frac{R^3}{4Z} \right)^{1/2} \right].$$  \hspace{1cm} (12)

Then we derive the differential cross section, Eq. (5). Integrating this cross section over angles of photo-electron ejection, one obtain the total cross section $\sigma_{t,o}$. The integral over angle $\varphi$ is derived elementary ($s = \cos \vartheta$):
The remaining integral in Eq. (13) is derived taking into account that \( k = \omega / c = 3Z / 2Rc \), since the contribution of the electron energy can be neglected in comparison with the ionization potential:

\[
J = \int_{-1}^{1} \left( 1 - s^2 \right) \exp \left[ 3(ZR)^{1/2} vs / c \right] ds =
\]

\[
= \int_{-1}^{1} \exp \left[ 3(ZR)^{1/2} vs / c \right] \frac{2scds}{3(ZR)^{1/2} v} =
\]

\[
= \int_{-1}^{1} d \exp \left[ 3(ZR)^{1/2} vs / c \right] \frac{2c^2}{9ZRv^2} =
\]

\[
= \frac{4c^2}{9ZRv^2} \cosh \left( 3(ZR)^{1/2} v / c \right) - \frac{2c^2}{9ZRv^2} \int_{-1}^{1} \exp \left[ 3(ZR)^{1/2} vs / c \right] ds =
\]

\[
= \frac{4c^2}{9ZRv^2} \cosh \left( 3(ZR)^{1/2} v / c \right) - \frac{4c^3}{27(ZR)^{3/2} v^3} \sinh \left( 3(ZR)^{1/2} v / c \right).
\]

Thus,

\[
\sigma_{ion} = \frac{32\pi^{3/2} R^{9/4} v^2 c}{27Z^{11/4} E} \exp \left[ -2 \left( k^2 + v^2 \right) \sqrt{R^3 / 4Z} \right] \times
\]

\[
\times \left[ \cosh \left( 3\sqrt{ZR} v / c \right) - \frac{c}{3(ZR)^{1/2} v} \sinh \left( 3\sqrt{ZR} v / c \right) \right].
\]

The photo-recombination cross section can be found using the well known principle of detailed equilibrium:

\[
\sigma_{rec} = \frac{2\omega^2}{c^2 v^2} \sigma_{ion},
\]

or

\[
\sigma_{rec} = 16 \left( \frac{\pi}{3} \right)^{3/2} \left( \frac{R}{cZ^{5/4}} \right)^{3/4} \exp \left[ -9 \frac{Z^3}{4c^4} \sqrt{\frac{Z}{R}} \right] \exp \left[ -v^2 \sqrt{R^3 / Z} \right] \times
\]

\[
\times \left[ \cosh \left( 3\sqrt{ZR} v / c \right) - \frac{c}{3\sqrt{ZR} v} \sinh \left( 3\sqrt{ZR} v / c \right) \right].
\]

It vanishes both for zero energy of an electron in continuum \( (v = 0) \) and for the infinite electron energy.

It follows from Eq. (16) that the cross section has the sharp maximum when the photon momentum is equal to the initial electron momentum. The maximum cross section is equal to

\[
\sigma_{rec} = 8 \left( \frac{\pi}{3} \right)^{3/2} \left( \frac{R}{cZ^{5/4}} \right)^{3/4}.
\]

In the above example when the initial electron energy is 50 eV, the maximum cross section is equal to \( 10^{-21} \text{ cm}^2 \).
3. Conclusion

The photo-recombination rate can be obtained from the cross section by multiplying by the electron velocity and dividing by the volume of the atomic cluster. In the above example this rate is equal to $5 \times 10^3 \text{ s}^{-1}$. Meanwhile the photo-recombination rate of these electrons on atomic ions inside the cluster for above cited parameters of cluster and laser pulse is much larger and it is equal to $3 \times 10^{11} \text{ s}^{-1}$. It should be noted that the dipole approximation is valid with high accuracy for photo-recombination on atomic ions. Though the photo-recombination rate on the charged cluster as a whole is very small, the effect can be measured since the photon energies differ strongly. Indeed, in the mechanism of collective photo-recombination x-ray photons have much larger energies. In the above example the energy of x-ray photon is equal to 7.2 keV.

In the recent experiment [4] Ar clusters were irradiated by laser pulses with the intensity of $10^{17} \text{ W/cm}^2$ and pulse duration of 40 fs. Spectrometer discovered the hard x-ray radiation with the photon energies from 2.9 to 4.3 keV. The observed characteristic lines demonstrated transitions in multicharged atomic ions of argon with the charge multiplicities up to 16+. We can hope that photons with larger energy which are explained by the mechanism of collective photo-recombination can be also observed though the number of these photons is small. In our opinion, it is possible also to observe this process of collective photo-recombination in metal clusters irradiated by a laser field [5].

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