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Conditions for XUV amplification considering recombination in clusters

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Abstract. We analyze the possibility of inversion generation and XUV amplification in a recombining scheme by various charged ions inside homogeneous clusters and discuss the conditions necessary for an efficient inversion generation in relation to the cluster density, the pulse duration and wavelength of the exciting laser pulse including the influence of the Coulomb explosion. We show that a high gain of $G \lessapprox 10^3\,\mathrm{cm}^{-1}$ for a wavelength of $\lambda_\mathrm{R} \approx 200$ Å in a time $\lessapprox 1$ ps can be expected. The measurable gain G_real depends on the cluster density N_cl. For $N_\mathrm{cl} \approx 10^{16}\,\mathrm{cm}^{-3}$ we expect $G_\mathrm{real} \gtrapprox 20\,\mathrm{cm}^{-1}$.

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Recently, it was shown that the interaction of rare-gas atoms with a short pulse (< 100 fs) high-intensity ($> 10^{15}\,\mathrm{W/cm^2}$) radiation caused the production of highly ionized ions leading to a prompt and, for special conditions, amplified XUV emission [1, 2]. The instantaneous optical field (tunnel) ionization produces electrons with sufficient energy to excite collisionally the upper laser level in multiply ionized rare gases (Ar, Kr, Xe).

A similar scheme for the optical field ionization and electron collisional excitation of atoms and a following prompt and amplified X-ray emission was discussed for the case where the rare gas atoms are bound in a van der Waals cluster [3]. Experiments [4, 5] have shown that the processes of ionization, excitation, and X-ray emission are more efficient inside small clusters. In addition, the possibility of excitations and especially an inversion generation by laser-driven motion of photoionized electrons and recombination inside the cluster was analyzed [6]. It was shown that a short-time high inversion density is possible via the recombination process where the excitation and the recombining electrons can be produced by electron collisions or by short-time optical field ionization. The inversion density increases in time [6] and reaches a maximum at a time strongly dependent on the cluster density, which is also a function of time because of the Coulomb explosion.

In this paper we want to discuss under which conditions clusters are advantageous for a XUV amplification in a recombining scheme. Therefore it is necessary to discuss the time behavior of the inversion and ion density in more detail. Taking into account the time dependence of the mean charge of the cluster we will show that the cluster lifetime after the excitation pulse increases with the initial ion density and also weakly depends on the laser pulse length. In this way we obtain an optimal inversion density as a function of the ion density and pulse length τ and we will discuss these conditions in relation to experiments.

1 Time dependence of the ion density

We consider the interaction of short-time high-intensity radiation ($\gtrapprox 10^{16}\,\mathrm{W/cm^2}$) with atoms bound in a cluster. We take into account an intensity $I_\mathrm{th}(Z)$ for (over-barrier) field ionization

$$I_\mathrm{th} = \frac{c}{128\pi}\frac{\varepsilon^4_{(Z+1)}}{(Z+1)^2\,e^6} \tag{1}$$

$\left(\varepsilon_{(Z+1)}\right.$ ionization energy for charge state $\left.Z+1\right)$

to produce ions with a charge $Z+1$ and quasi-free electrons, and assume an ionization rate of $> 10^{13}\,\mathrm{s}^{-1}$. Thus, after the end of the pulse, we expect a cluster of n ions with the same charge $Z+1$ and $n\,(Z+1)$ electrons with a mean kinetic energy (as a result of their ponderomotive motion) of

$$\varepsilon_\mathrm{e} = \frac{2\pi e^2}{mc\omega^2} I\,, \tag{2}$$

(ω circular frequency of the radiation field,
m electron mass, $I = I_\mathrm{th}$ intensity) .

For a cluster size of [6]

$$n \geqslant 8\,(8\pi)^{3/2}\left(\frac{e}{m\omega^2 r_0}\right)^3\left(\frac{I}{c}\right)^{3/2}\,, \tag{3}$$

(n number of atoms inside the cluster) where we have used the relation $R = r_0 n^{1/3}$ for the cluster radius R, the electrons oscillate inside the cluster. After a time $t \geqslant t_p$, a part of the electrons will recombine and produce ions with a charge Z and an excitation of the upper (and, of course, lower) XUV laser level. On the other hand, after a time t_i some (Z') electrons will have left the cluster and caused an overall positive charge Z' on the cluster, where $Z'(t)$ increases in time and determines the time t_c for the Coulomb explosion (cluster fission). As shown experimentally [7] a Na cluster is already unstable for relatively small charges Z', namely for:

$$n \leqslant 8Z'^2 . \tag{4}$$

To determine the time behavior of the density and its influence on the following (recombination) process, the evaluation of the Coulomb explosion dynamics is necessary, i.e., the calculation of the increase in the cluster radius and therefore decrease in the density as a function of time.

As an estimation we assume an initial homogeneous charge distribution of the spherical cluster. Thus, under the influence of the Coulomb repulsion the cluster radius expands homogeneously. The time evolution of the radius R can be determined by considering the force between a charge at the surface and the whole cluster charge located in the center. Under the (most interesting) condition that the increase of the cluster radius ΔR at time t will be small compared with R we obtain the simple formula

$$\frac{\Delta R}{R} \approx \frac{0.14}{A\, r_0^3} \int_{-\infty}^{t} \mathrm{d}t' \int_{-\infty}^{t'} \tilde{Z}^2\left(t''\right) \mathrm{d}t'' \tag{5}$$

where $\tilde{Z} = Z'/n$ (function of time) is the mean charge per ion, A is the atomic weight, r_0 is given in Å and t in femtoseconds. To take into account approximately the time behavior of $\tilde{Z}(t)$ we assume a Gaussian profile for the excitation laser pulse

$$I(t) = I_0 \mathrm{e}^{-t^2/\tau^2} \tag{6}$$

(FWHM $\approx 1.665\,\tau$) and simply assume that $\tilde{Z}(t) = \tilde{Z}_{\max}(t)$. This maximum mean charge $\tilde{Z}_{\max}$ is estimated assuming (as the most unfavorable case) the condition that in collision processes the ponderomotive energy of an electron can be immediately transferred to kinetic energy being sufficient to overcome the Coulomb attraction of the positively charged cluster, i.e.,

$$\frac{\tilde{Z}_{\max}(t)\, ne^2}{R} = 2\varepsilon_e \mathrm{e}^{-t^2/\tau^2} \quad (-\infty < t \leqslant 0) . \tag{7}$$

For $t > 0$, $\tilde{Z}_{\max}(t) = \tilde{Z}_{\max}(0)$ remains constant. Setting (2) and (7) into (5) yields

$$\begin{aligned}\frac{\Delta R}{R} &\approx \frac{1.18 \times 10^3 \lambda^4 I^2}{A\, r_0 n^{4/3}} \left(t^2 + t\tau\sqrt{\frac{\pi}{2}} + \tau^2\right) \\ &\leqslant \frac{1.18 \times 10^3 \lambda^4 I^2}{A\, r_0 n^{4/3}} \left(t + \frac{\tau}{\sqrt{2}}\right)^2 ,\end{aligned} \tag{8}$$

where the intensity I is given in $10^{16}\ \mathrm{W/cm^2}$ and the laser excitation wavelength λ in µm. Considering $(t \geqslant 0)$ (8) we can define the time $t = t_c = \frac{-\tau}{\sqrt{2}}$ as the effective starting point of the Coulomb explosion assuming constant $\tilde{Z} = \tilde{Z}_{\max}(0)$. This means that the increase of the cluster radius due to the action of a Gaussian pulse (6) is approximately the same as in the case of a Coulomb explosion with a time-independent charge starting at the time $-\tau/\sqrt{2}$ before the laser pulse maximum is reached.

Setting n according to the right side of (3) into (8) and assuming that the recombination process starts at the maximum of the excitation pulse we obtain

$$\frac{\Delta R}{R} \approx 1.53 \times 10^{-2} \lambda^4 \frac{r_0^3}{A} \left(\Delta t + \frac{\tau}{\sqrt{2}}\right)^2 , \tag{9}$$

where Δt is the recombination time, i.e. we define that the recombination starts (abruptly) at $t = 0$ where the maximum number of quasi-free electrons is reached.

$\Delta R/R$ is seen to be inversely proportional to the initial ion density $(\approx r_0^{-3})$ and, as the most important dependence in relation to the experiment, increases as λ^4 with the laser excitation wavelength. This means that an efficient interaction inside the dense cluster, i.e., before the Coulomb explosion leads to a drastic decrease of the density $(\Delta R/R \ll 1)$ is only possible by excitation with a short laser wavelength such as, for example, $\lambda = 0.248\ \mu\mathrm{m}$ (KrF excimer laser), as a simple numerical estimation shows.

We note, that for an optical field ionization as assumed, the cluster size n is, in principle, a free parameter. However, because the lifetime of the cluster also strongly depends on n, (see (8)) the value given by (3) is appropriate. For smaller n also smaller λ are necessary or, for the same λ, the pulse length τ must be shorter and optimized conditions can become impossible. In addition, for a given cluster size (3) an efficient electron collision excitation is in principle also possible. However, also for smaller n as given by (3) may be an interesting case to discuss.

2 Condition for optimized inversion density

Starting from n ions inside the cluster with a charge $Z+1$, which is determined by the maximum pulse intensity, the recombination process excites the upper and lower XUV laser level and produces an inversion state in the Z-fold charged ions, where the inversion density as a function of time Δt is given by [6]

$$\frac{N_e - N_g}{V} = \frac{N_{j'}^0}{V} \frac{1}{2} \frac{1-\xi}{1+\xi^r} \frac{\ln\left[1 + 2\left(1+\xi^r\right) W_j^0 \Delta t\right]}{\sqrt{1 + 2\left(1+\xi^r\right) W_j^0 \Delta t}} . \tag{10}$$

W_j^0 denotes the three-body collisional recombination rate for the upper laser level, $\xi = \frac{W_{j'}^0}{W_j^0}$, $\xi^r = \frac{W_{j'}^0}{W_j^0} + \frac{W_p^0}{W_j^0}$, j' lower laser level, p upper $(\neq j, j')$ levels, $N_{j'}^0 = n$ for full $(Z+1)$-fold ionization, provided that the decrease of the cluster density caused by Coulomb explosion can be neglected. Only in this case does an efficient recombination rate $\left(\approx r_0^{-6}!\right)$ occur. Therefore, the maximum recombination time Δt is on the one hand, limited by (9) where $\Delta R/R \ll 1$. On the other hand, the inversion density increases with Δt and reaches a maximum of

$$\left(\frac{N_e - N_g}{V}\right)_{\max} = \Delta N_{\max} = \frac{n}{V} \frac{1-\xi}{1+\xi^r} \times 0.37 \tag{11}$$

for

$$\Delta t^{\max} = \frac{3.2}{1+\xi^r}\frac{1}{W_j}\,. \tag{12}$$

For $\Delta t > \Delta t^{\max}$ the inversion density decreases mainly due to a further decrease of the charge state ($Z \to Z-1$) by recombination.

For an optimal inversion density Δt is given by (12) and so we can determine, setting $\lambda = 0.248\,\mu\text{m}$ and $\Delta t = \Delta t^{\max}$ using (9), the pulse length τ as a function of the (initial) ion density $\left(\approx r_0^{-3}\right)$.

$$\tau = 186\left(\frac{\Delta R}{R}\right)^{1/2}\frac{A^{1/2}}{r_0^{3/2}} \times\left\{1-3.1\times10^{-5}\frac{\varepsilon_j^2 I}{Z^2 j^2}\frac{r_0^{7.5}}{A^{1/2}\left(1+\xi^r\right)}\left(\frac{R}{\Delta R}\right)^{1/2}\right\}, \tag{13}$$

where the condition

$$1 < 3.2\times10^4\frac{Z^2 j^2}{\varepsilon_j^2 I}\frac{A^{1/2}\left(1+\xi^r\right)}{r_0^{7.5}}\left(\frac{\Delta R}{R}\right)^{1/2} \tag{14}$$

must hold. Otherwise the given $\Delta R/R$ value is reached for recombination times $\Delta t < \Delta t^{\max}$.

This means highly dense clusters favor a longer pulse length τ whereas for not so dense clusters short pulses are necessary or it is only possible to reach an inversion density smaller than the maximum value. To show this, we write $\Delta t = \Delta t^{\max} a$, where $a < 1$, and get an inversion density of

$$\Delta N = 1.36\frac{\ln(1+6.4a)}{\sqrt{1+6.4a}}\Delta N_{\max} \tag{15}$$

and the condition for τ in the form (13), where the second term in the brackets has to be multiplied by a.

To illustrate these conditions for some examples we took into account (low density) van der Waals- and (high density) metal clusters. The results are seen in Table 1. To fulfill relation (3) relatively large clusters are necessary. In principle, their experimental realization should be possible. As an example, sodium clusters with $n > 20\,000$ could be produced and detected [8]. The results in the last three rows of Table 1 are obtained setting $\Delta R/R = 0.2$. For rare-gas clusters even in the case of infinitely short excitation pulses the maximum inversion density cannot be reached, because for $\Delta t = \Delta t^{\max}$ the density inside the cluster is already remarkably reduced ($\Delta R/R > 0.2$). If pulses longer than the τ-value listed first are used, the recombination process starts when $\Delta R/R > 0.2$, i.e., the recombination is less efficient because of the cluster expansion. However, for Na, Cu, and Ag the Coulomb explosion occurs more slowly and consequently, the maximum inversion is reached for the listed τ-values. Therefore, higher density clusters seem to be favored under adequate conditions with respect to the atomic parameters (ε_j, j, ...).

However, the values of the gain are not only determined by the inversion density but also by the wavelength of the transition, the transition rate and, as the most important factor, by the linewidth of the transition. Because of their high density Ag and Cu clusters are characterized by a large Stark-broadened linewidth being in the order of the transition frequency itself. In these clusters efficient XUV generation may be possible during the progression of Coulomb explosion, however, the inversion density is reduced because $\Delta t \gg \Delta t^{\max}$. We will not further discuss Ag and Cu clusters in detail.

3 Gain in cluster experiments

The gain is defined by

$$G = \frac{A_{jj'}c^2}{8\pi\nu^2\Delta\nu}\frac{n}{V}\frac{N_e - N_g}{n} \equiv \tilde{G}\frac{N_e - N_g}{n}\,, \tag{16}$$

where $\tilde{G}$ is a function of the atomic parameters and, in particular, depends on the linewidth $\Delta\nu$, which in highly ionized dense clusters is given by the Holtsmark line broadening ($\leqslant 20\,\text{eV}$) arising from the Stark effect due to the neighboring ions. Taking into account the dependence on the quantum number this linewidth is proportional to the density $\frac{n}{V}$ and given by [9]

$$\Delta\nu_H = 8.8\times10^{-8}\frac{n}{V}\frac{f\left(j,j',l,l'\right)}{Z^2 I^{1/2}} \quad (\text{s}^{-1})\,, \tag{17}$$

where $f\left(j,j',l,l'\right) = \left\{ j^2\left(5j^2+1\right) - 3l_j\left(l_j+1\right) + j'^2\left(5j'^2+1\right) - 3l_{j'}\left(l_{j'}+1\right)\right\}$.

It thus follows that the advantage of the high cluster density $\frac{n}{V}$ causing a high inversion density (and consequently corresponding to (16) a high gain) will be partly compensated by a large linewidth. On the other hand, during the Coulomb explosion the ion density in the cluster drastically decreases. However, the gain remains constant for the case where the

Table 1. Comparison of rare gas and metal clusters. I is the peak intensity of a laser pulse with FWHM $= 1.66\,\tau$ necessary for $(Z+1)$-fold field ionization. Due to collisional recombination processes an inversion occurs between levels with the principal quantum numbers j and j' corresponding to a wavelength λ_R

Cluster type		rare gases			metal	
atom	Ar	Kr	Xe	Na	Cu	Ag
r_0/Å	3.8	4.0	4.4	2.1	1.4	1.6
Z	8	8	8	8	10	10
$j \to j'$	$3 \to 2$	$4 \to 3$	$5 \to 4$	$4 \to 3$	$5 \to 4$	$6 \to 5$
λ_R/Å	49.5	145	218	224	154	205
n	1.1×10^5	2.1×10^3	327	1.3×10^5	1.1×10^5	1.0×10^4
$I/10^{16}$ W/cm^2	119	9.6	3.3	40.3	17.0	4.3
$\Delta N/\Delta N_{\max}$	0...0.013	0...0.29	0...0.69	1	1	1
τ/fs	71...0	95...0	103...0	68	401	428

linewidth is determined by the Holtsmark broadening. That means for

$$\Delta\nu_{\mathrm{H}} > \Delta\nu_{\mathrm{D}}^{\mathrm{th}} + \Delta\nu_{\mathrm{D}}^{\mathrm{c}}\,, \tag{18}$$

where $\Delta\nu_{\mathrm{D}}^{\mathrm{th}}$ is the Doppler broadening due to the thermal motion of ions and $\Delta\nu_{\mathrm{D}}^{\mathrm{c}}$ the Doppler broadening due to Coulomb explosion [6]. The latter term increases with time.

For

$$\Delta\nu_{\mathrm{H}} < \Delta\nu_{\mathrm{D}}^{\mathrm{th}} + \Delta\nu_{\mathrm{D}}^{\mathrm{c}}\,, \tag{19}$$

the linewidth is independent of the density and given by the Doppler broadening and determines a critical density $\left(\frac{n}{V}\right)_{\mathrm{cr}}$, which, assuming $\Delta\nu_{\mathrm{D}}^{\mathrm{c}} \gg \Delta\nu_{\mathrm{D}}^{\mathrm{th}}$, is given by

$$\left(\frac{n}{V}\right)_{\mathrm{cr}} = 1.3\times 10^{23}\frac{Z^2\bar{Z}I}{A^{1/2}r_0^{3/2}\lambda_R}\frac{1}{f\,(j,\,j',\,l,\,l')}\ \ (\mathrm{cm}^{-3})\ . \tag{20}$$

For $\frac{n}{V} < \left(\frac{n}{V}\right)_{cr}$ the gain decreases proportional to the density. That means the relatively high gain inside the dense cluster will also be reached for a lower density. Its minimum value is given by (20).

However, the gain defined in (16) cannot be related to real experiments because the active volume is large compared with the volume of a single cluster. Thus the real gain G_{real}, inside a volume containing a large number of clusters can be expressed by the gain G (defined by (16)):

$$G_{\mathrm{real}} = \frac{N_{\mathrm{cl}}}{N_{\mathrm{ion}}}nG\,, \tag{21}$$

where N_{cl} is the number of clusters per unit volume and N_{ion} is the density of ions inside the cluster. Before Coulomb explosion has led to a significant cluster expansion we have

$$N_{\mathrm{ion}} = \left(\frac{4\pi}{3}r_0^3\right)^{-1}\ .$$

Setting the values of $Na^{(8+)}$ into (21) and assuming a cluster density of $10^{16}\ \mathrm{cm}^{-3}$ we obtain a gain of $G_{\mathrm{real}} \approx 0.049\,G$ before the Coulomb explosion. During the Coulomb explosion we get a maximum of $G_{\mathrm{real}} \approx 0.3\,G$. To estimate the value of the expected gain in cluster experiments we will calculate it in more detail for a typical example of a metal cluster consisting of various charged Na ions.

The energetic structure of the Na ions will be treated as hydrogen-like. This assumption is justified because for the generation of coherent radiation mainly transitions between Rydberg levels are chosen and due to their strong Holtsmark line broadening ($\leqslant 20\,\mathrm{eV}$) the energy levels can be considered to be degenerate with respect to the orbital quantum number l. Therefore, after three-body-collision recombination processes for each principal quantum number j we obtain j^2 equally populated sublevels (l, m-degeneracy). The linear gain G for a transition characterized by the upper (lower) principal quantum number j' (j'') can be found after summation of the gains over all possible sub-transitions $\left(j',\,l',\,m'\right) \to \left(j'',\,l'',\,m''\right)$ considering the selection rules and fixing $m' - m''$:

$$G_{j'\to j''} = \frac{8\pi^3\nu}{3hc\Delta\nu}\frac{\left(N_{j'} - N_{j''}\right)}{V}\sum_{\substack{l',m'\\ l'',m''}}\left|\mu_{j',l',m'\to j'',l'',m''}\right|^2\ . \tag{22}$$

Here $N_{j'} - N_{j''} = N_{j',l',m'} - N_{j'',l'',m''}$ is the inversion between a pair of sublevels, ν the transition frequency and $\Delta\nu$ the linewidth. The transition dipole moments $\left|\mu_{j',l',m'\to j'',l'',m''}\right|$ are calculated using the wave function for a hydrogen-like quantum system. As an example, in the case of a ($j' = j+1 \to j'' = j$) transition the sum (22) contains $2j\,(j-1)+1$ terms.

The results are shown in Table 2. It is seen that a gain of some 10^2 to $10^3\ \mathrm{cm}^{-1}$ can be expected for a time duration, corresponding to $(c\times G)^{-1}$ between 1 and 0.1 ps after the end of the recombination by an excitation with a laser intensity of some $10^{16}\ \mathrm{W/cm}^2$. The minimum density for this gain (after the Coulomb explosion) is given by $\left(\frac{n}{V}\right)_{\mathrm{cr}} \approx 10^{20}\ \mathrm{cm}^{-3}$ (more exactly see (20)) and then the gain decreases as the ion density decreases.

The principal difference between using gas targets or clusters is thus the time behavior of the stimulated XUV emission. For cluster targets we expect a high gain ($\approx 10^3\ \mathrm{cm}^{-1}$) for XUV amplification in the 1-ps region.

4 Conclusion

We have determined under which conditions, after the interaction of an intense laser field with atoms inside a cluster that caused a field ionization, an effective excitation of an upper level including an inversion generation by recombination inside the cluster an XUV emission occurs. We have shown, that the time behavior of the Coulomb explosion determines the optimum experimental condition (laser intensity, wavelength and pulse duration) in relation to the kind of clusters and the XUV radiation of interest. For wavelengths $\lambda_{\mathrm{R}} \approx (80\ldots 350)$ Å we expect a gain G of some $10^3\ \mathrm{cm}^{-1}$ for a time duration of $\lesssim 1$ ps. The measurable gain G_{real} depends on the cluster density N_{cl}. For $N_{\mathrm{cl}} \approx 10^{16}\ \mathrm{cm}^{-3}$ we expect values of $G_{\mathrm{real}} \gtrapprox 20\ \mathrm{cm}^{-1}$.

Table 2. Calculated gain for the $3\to 2$ and $4\to 3$ transition ($\Delta\varepsilon$ energy) in ionized sodium. ε is the ionization energy for the production of $(Z+1)$-time ionized Na, ε_j is the ionization energy for the level j.

Ionization		$Na^{(Z+1)}$	
Z	6	7	8
ε/eV	208	264	300
$I_{\mathrm{th}}/10^{16}\ \mathrm{W/cm}^2$	15.4	30.6	40.3
n	3.0×10^4	8.5×10^4	1.3×10^5
Transition to		$Na^{(Z)}$	
$\Delta\varepsilon\ (3\to 2)$ /eV		135	160
ε_3, ε_2/eV		100, 235	125, 285
$\lambda_R\ (3\to 2)$ /Å		95	84
$\Delta N/\Delta N_{\mathrm{max}}$		0.97	0.90
τ/fs		131 … 0	131 … 0
τ assumed/fs		70	70
G_{32}/cm^{-1}		6342	7200
$\Delta\varepsilon\ (4\to 3)$ /eV	35	45	55
ε_4, ε_3/eV	45, 80	55, 100	70, 125
$\lambda_R\ (4\to 3)$ /Å	351	273	224
$\Delta N/\Delta N_{\mathrm{max}}$	1	1	1
τ/fs	115	94	68
G_{43}/cm^{-1}	1460	2659	3316

Of course a lot of simplifying approximations have been made to describe this process, especially in relation to the exact rate of field ionization, the complex mechanism of the recombination process including collisions and ionization, and the wide range of the distribution of the cluster sizes including the exact time behavior of the Coulomb explosion and cluster fragmentation. Furthermore we neglected a coherent motion of ionized electrons leading to inelastic ionization of inner shell states. This process may be responsible for the experimentally observed X-ray emission in Xe clusters [10] where the cluster size was smaller than discussed in our model. It can be supposed that the influence of inelastic collisional excitations will further improve the conditions for an efficient XUV generation.

However, we believe that the basic idea, namely the excitation and inversion generation in a dense medium for a short-time $(\lesssim 1\,\mathrm{ps})$ XUV emission and amplification, is attractive, as we have shown in a first estimation.

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