# Angular dispersion boost of high order laser harmonics interacting with dense plasma clusters

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Periodic surface gratings or photonic crystals are excellent tools for light diffraction and direction. However, this method is less effective in the case of extreme ultraviolet (XUV) light due to the high absorption of any material in this frequency range. In the paper we research the possibility of angluar boost of a radiation in the XUV range by scattering on suitable spherical clusters. Within the work the analytical model was developed with help of the Drude dielectric function of the plasma and the Mie scattering theory. The model was constructed in the quasi-static approximation since the ionization time is shorter than the pulse duration, which is much shorter than the plasma expansion time. Within the model we use the limiting forms of the Bessel functions since we are only interested in particle sizes smaller than the incident wavelength. The resonance parameters of the target was estimated using the tenth harmonic of titan:sapphire laser and the scattered field enhancement in the resonance case in comparison with the first harmonic was found. Using the same resonance conditions for a single cluster, we simulate diffraction by an array of such clusters using code CELES. Obtained results show a significant boost of the scattered field in the resonance case for large angles, which corresponds to the Bragg-Wolfe diffraction theory, - the ability to control high harmonics of laser radiation in XUV range using an ionized cluster gas.

## Introduction

Finite size targets interacting with high-intensity coherent radiation are a well-studied phenomenon of linearly excited surface plasmon oscillations. Absorption and scattering of incident light in this case can be described with good accuracy using the Mie theory, which predicts the existence of a resonance corresponding to multipole oscillations of a part of the free electrons of the target relative to positively charged ions. In the resonance regime, effective excitation of surface plasmons can lead to a significant enhancement of the internal and external fields at the natural frequency of the cluster. This can lead to amplification of the field scattered at large angles relative to the initial direction of the incident wave.

Within micrometer wavelengths, photonic crystals and gratings can be used to guide or diffract electromagnetic waves [6], while similar X-ray manipulations can use crystals with atoms regularly spaced a few nanometers apart, as scattering centers [1]. At the same time, a large gap between these wavelength ranges, called XUV (extreme-ultraviolet) or hard ultraviolet, turns out to be difficult to manipulate.

It is well-known fact that high-order laser harmonics can be generated using a short intense laser pulse when interacting with dense solid surfaces [7]. In the intensity range under consideration (order of $10^{18}$ W/cm$^2$), the conversion coefficient is at best about $10^{-4}$, which gives the intensity of high-order harmonics no higher than $10^{14}$ W/cm$^2$, which is not enough to ionize the target and generate plasma with a purely imaginary refractive index. To solve this problem, it is proposed to use a prepulse.

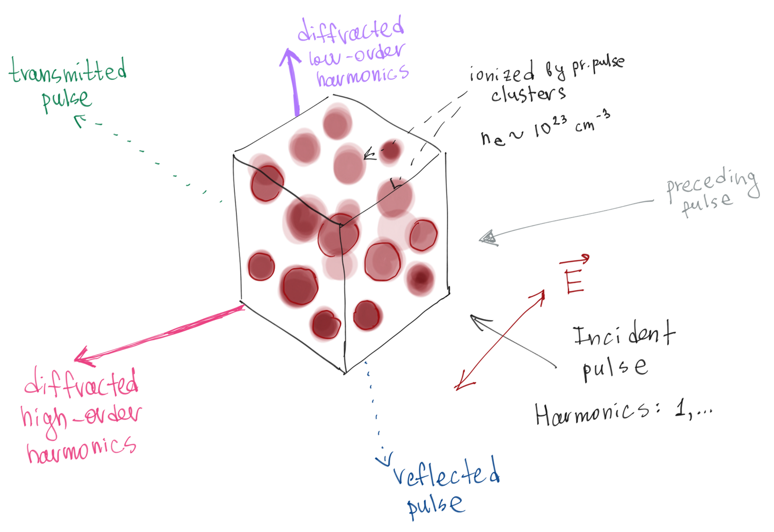



**Figure 1:** An interaction scheme. The plane of polarization is parallel to one of the faces of the cubic region. The sizes of spherical clusters are on the order of a few nanometers, and the distance between them is at least hundreds of nanometers. The distribution of clusters inside the cubic region is generally arbitrary, the clusters do not intersect the faces of the region.

A generalized interaction scheme is shown in Fig. 1. The harmonics that the main pulse contains have different intensities at different angles, which leads to an angular dependence of the output radiation shape.

## Analytical model

Let us consider a single spherical cluster of radius $a$ irradiated with a short femtosecond pulse $\tau$ in duration and $I_h \approx 10^{14}$ W/cm$^2$, obtained as a result of the transformation laser harmonics with a conversion factor of $10^{-4}$. The Drude model gives a representation of the plasma dielectric function:

$$\varepsilon(\omega) = 1 - \left(\frac{\omega_{pe}}{\omega}\right)^2 \frac{1}{1+i\beta_e}, \qquad \omega_{pe} = \sqrt{\frac{4\pi e^2 n_e}{m_e}}, \tag{1}$$

where $\omega$ is the considered harmonic frequency, $\omega_{pe}$ is the electron plasma frequency, $e$ and $m_e$ are the electron charge and mass, $n_e = Zn_i$ — electron density, where $Z$ is the average degree of ionization, $n_i$ is the ion density. $\beta_e = v_e/\omega$ and $v_e$ is the electron-ion collision coefficient in the Spitzer approximation. Under solid-state plasma conditions, the cluster ion density is of the order of $n_i = 6 \cdot 10^{22}$ cm$^{-3}$, while the electron density of the cluster must be higher than the critical one for a given frequency $n_c = \omega^2 m_e / 4\pi e^2$. For the 10th harmonic of laser radiation $\lambda_{10} = 83$ nm, we obtain the condition $n_e > n_c = 1.3 \cdot 10^{23}$ cm$^{-3}$, which agrees with condition on the ionic density at an average degree of ionization $Z > 2$. Mie theory can be used to describe the scattered field and the field inside a scattering object. For a spherical cluster, we can write the solution of the corresponding equation using the spherical Bessel and Hankel functions of the $n$th order, including the associated Legendre polynomials [2].

Let us consider a plane wave propagating along the $z$ axis of the Cartesian coordinate system, polarized along the $x$ axis. Further, the flat one can be expanded into a series using the generalized Fourier expansion. In the case of an isotropic medium, we have the following form of the scattered field coefficients [2]:

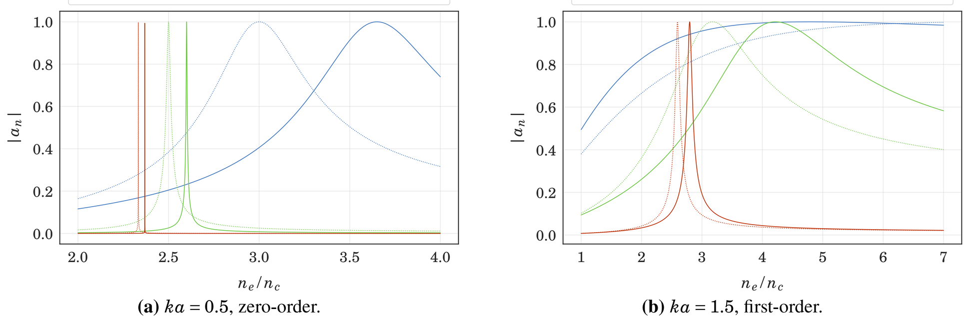
$$\mathbf{E}_s = \sum_{n=1}^{\infty} E_n \left[ i a_n(ka,m) \mathbf{N}^{(3)}_{e1n} - b_n(ka,m) \mathbf{M}^{(3)}_{o1n} \right], \qquad E_n = i^n E_0 \frac{2n+1}{n(n+1)} \tag{2}$$

$$a_n(x,m) = \frac{m\psi_n'(x)\psi_n(mx) - \psi_n'(mx)\psi_n(x)}{m\xi_n'(x)\psi_n(mx) - \psi_n'(mx)\xi_n(x)}, \tag{3}$$

$$b_n(x,m) = \frac{\psi_n'(x)\psi_n(mx) - m\psi_n'(mx)\psi_n(x)}{\xi_n'(x)\psi_n(mx) - m\psi_n'(mx)\xi_n(x)}, \tag{4}$$

where $\psi_n(\rho) = zj_n(\rho)$, $\xi_n(\rho) = zh_n(\rho)$ — Riccati-Bessel functions, $h_n = j_n + i\gamma_n$ — spherical Hankel functions of the first kind, $x = ka$ — dimensionless cluster radius, $m = \sqrt{\varepsilon}$ — complex refractive index (1).

In the case of spherical symmetry, the amplitude of the scattered field is maximum for $m^2 = -(n+1)/n$ and $ka \ll 1$, which gives the corresponding set of resonant densities in the collisionless case $n_e = n_c(2n+1)/n$. This can be obtained using the zero asymptotic approximation of the Bessel functions [2], as a result of which the coefficients (3, 4) are greatly simplified. Such an approximation can be used instead of (3, 4) for objects of sufficiently small radius, but already at $ka \sim 1$ it ceases to be reasonable, especially for large $n$. Instead, in this case, higher-order approximations obtained in a similar way [2] are better suited.




**(a)** $ka = 0.5$, zero-order. **(b)** $ka = 1.5$, first-order.

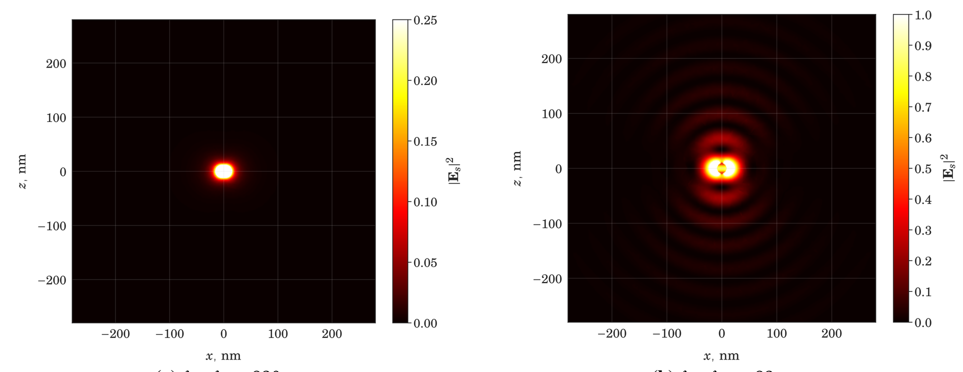
**Figure 2:** Coefficients of spherical harmonics in zero and first order approximation, $\beta_e = 0$. The "exact" curves are built using full expansions.

Such approximations make it possible to estimate the resonance parameters of the cluster, in particular, the electron density and radius. Wavelength $\lambda_{10} = \lambda_L/10 = 83$ nm corresponds to the resonant electron density $n_{el} \approx 5.7 \cdot 10^{23}$ cm$^{-3}$ for $ka = 0.7$ and $n_{el} \approx 3.9 \cdot 10^{23}$ cm$^{-3}$ for $ka = 0.3$, with medium ionization degrees $Z|_{ka=0.7} \approx 9$ and $Z|_{ka=0.3} \approx 6$.

## Single cluster

To check the resulting analytical model, we calculated the values of the complex refractive index $m$, which correspond to the previously obtained conditions for the resonance electron density $n_{el}$ at $\lambda_{10} = 83$ nm, $ka = 0.7$: $m = 1.851i$ (**??**). The complex refractive index is purely imaginary, since the collision coefficient $v_e$ in the case under consideration is somewhat lower than the harmonic frequency, so the interaction can be considered collisionless [5].

For this case, the scattered electric field (2) was calculated for $\lambda = \lambda_L$ and $\lambda = \lambda_{10}$ in order to compare the resonant and nonresonant cases. It can be seen that in the resonance case (**??**) the scattered field is a diverging spherical wave, the field amplitude in the vicinity of the cluster is much higher than in the absence of resonance (**??**), where Scattered waves as such are practically not observed, which indicates that the incident wave in the nonresonant case practically does not interact with the cluster.




**(a)** $\lambda = \lambda_L = 830$ nm. **(b)** $\lambda = \lambda_{10} = 83$ nm.

**Figure 3:** $ka = 0.7$ ($a \approx 8.9$ nm); $|\mathbf{E}_s|^2$ in the plane of polarization of the incident wave.

## Multiple clusters

Within the framework of the stationary Mie scattering theory, many clusters were considered in the form of an extended cylindrical gas jet (hereinafter referred to as a target) with a regular and quasi-regular spatial configuration in order to study the possibility of scattering by such structures at large angles of hard ultraviolet radiation, in particular, corresponding to high-order harmonics.

A primitive cubic lattice with a distance between neighboring nodes $d$ was chosen as a regular distribution. The quasi-regular distribution was constructed by introducing random shifts of the coordinates of nodes with an arbitrary shift norm in the range $0 \le |\Delta d| \le \eta d$, where $0 \le \eta < 0.5$ is the degree of irregularity. Then, for a multiple of $d = b\lambda$, $b \in \mathbb{N}$, the distance between adjacent nodes after making shifts:

$$b(1-\eta)\lambda \le d_{\text{irreg}} \le b(1+\eta)\lambda \tag{5}$$

In the quasi-regular case, the simulation was carried out several times in order to average and obtain a generalized picture of the scattered field. The program code CELES [3] was used for calculations.

## Diffraction theory

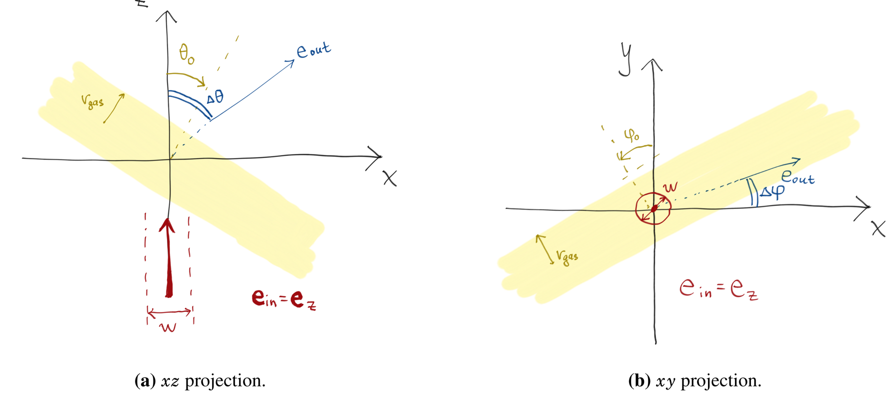
The diffraction condition in the case of a three-dimensional regular grating with elastic scattering takes the form [4]:

$$\begin{cases} (\mathbf{D}_x, \mathbf{e}_{\text{out}} - \mathbf{e}_{\text{in}}) = h\lambda \\ (\mathbf{D}_y, \mathbf{e}_{\text{out}} - \mathbf{e}_{\text{in}}) = k\lambda \\ (\mathbf{D}_z, \mathbf{e}_{\text{out}} - \mathbf{e}_{\text{in}}) = l\lambda \end{cases} \tag{6}$$

where $h, k, l$ are Miller indices represented by integers, $\mathbf{D}_i$ is a vector connecting adjacent lattice nodes along the $i$ direction, $\mathbf{e}_{\text{in}}$ is the unit vector of the direction of the incident radiation, $\mathbf{e}_{\text{out}}$ is the unit vector of the direction of the transmitted radiation. Passing to spherical coordinates related to $\mathbf{e}_{\text{in}}$ so that in Cartesian representation $\mathbf{e}_{\text{in}} = \mathbf{e}_z$ , 6 can be converted as follows, given that $|\mathbf{D}_x| = |\mathbf{D}_y| = |\mathbf{D}_z| = d$ for the considered cubic lattice:

$$\begin{cases} \cos\theta_0 \sin\Delta\theta \cos(\Delta\varphi - \varphi_0) - \sin\theta_0 (\cos\Delta\theta - 1) = \dfrac{h'\lambda}{d} \\ \sin\Delta\theta \sin(\Delta\varphi - \varphi_0) = \dfrac{k'\lambda}{d} \\ \sin\theta_0 \sin\Delta\theta \cos(\Delta\varphi - \varphi_0) + \cos\theta_0 (\cos\Delta\theta - 1) = \dfrac{l'\lambda}{d} \end{cases} \tag{7}$$

where $\Delta\theta$, $\Delta\varphi$ are the angles characterizing the deviation of the direction of the diffracted radiation relative to the incident one, $\theta_0$, $\varphi_0$ are the angles characterizing the rotation of the target (grid) in space , $h'$, $k'$, $l'$ — new Miller indices (4). Using 7, we can obtain the angular distribution of diffracted radiation for given initial parameters $d$, $\lambda$, $\theta_0$, $\varphi_0$.




**(a)** $xz$ projection. **(b)** $xy$ projection.

**Figure 4:** General scheme of interaction of incident radiation with a grating. $\theta_0$, $\varphi_0$ — characterize the target angles in space, $\Delta\theta$, $\Delta\varphi$ — angles of deflection of the direction of the diffracted radiation relative to the incident, $r_{\text{gas}}$ is the radius of the gas jet representing the target, $w$ is the diameter of the Gaussian beam of incident radiation. $\Delta\theta$ is counted counterclockwise around $y$, $\Delta\varphi$ — counterclockwise around $z$.

## Monochromatic radiation scattering

## Wavepacket scattering

## Conclusions

We found that a periodic structure of dense plasma clusters turned out to be a suitable element for efficient directional scattering of radiation in the XUV range. Since many spherical scatterers require calculations in three-dimensional space, a stationary model was proposed and the range of cluster radii was determined, within which the electron density is quasi-stationary during interaction with an external field. When the ionization is such that the electron concentration is close to resonance for given initial parameters, the scattering efficiency increases significantly and reaches several percent in the case of a single cluster. For many clusters, the efficiency of angular dispersion increases with the number of rows of clusters and can reach several percent in the case of certain directions.

The obtained angular distributions of diffraction maxima for scattering by a set of regularly spaced clusters are well described using the Laue theory, while introducing a small irregularity into the distribution of clusters weakens the most intense diffraction directions, which differ from the direction of the transmitted radiation, by no more than 25%. In the case of non-monochromatic radiation, the efficiency of amplification of the angular dispersion decreases in accordance with the spectral distribution of the field amplitude, the attenuation can reach about three times.

## References

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