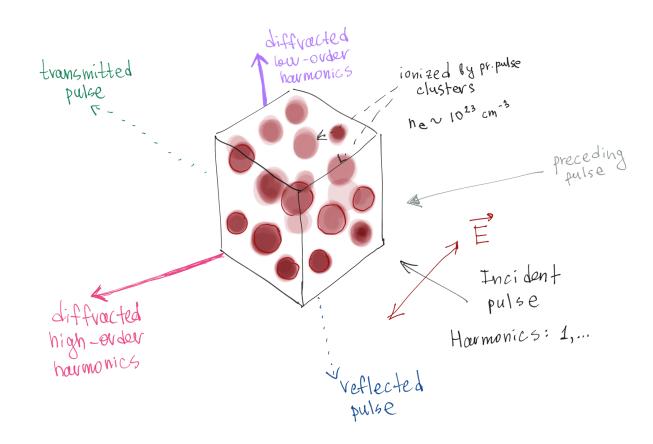
# L.A. Litvinov<sup>1</sup>, A.A. Andreev<sup>1, 2</sup>

<sup>1</sup>Saint Petersburg State University, Saint Petersburg, Russia <sup>2</sup>Ioffe Physico-Technical Institute, Saint Petersburg, Russia

Periodic surface gratings or photonic crystals are excellent tools for light manipulation. 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 quasistatic 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

Within micrometer wavelengths, photonic crystals and gratings can be used to guide or diffract electromagnetic waves, while similar X-ray manipulations can use crystals with atoms regularly spaced a few nanometers apart, as scattering centers. 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. To solve this problem, we propose the use of arrays of spherical nanoclusters for directed scattering of hard ultraviolet radiation (Fig. 1).



**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.

#### **Analytical model**

Consider a single cluster of radius a irradiated with a short femtosecond pulse with  $I_h \approx 10^{14}$  W/cm<sup>2</sup> intensity. 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,  $n_e = Z n_i$  — electron density, 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.

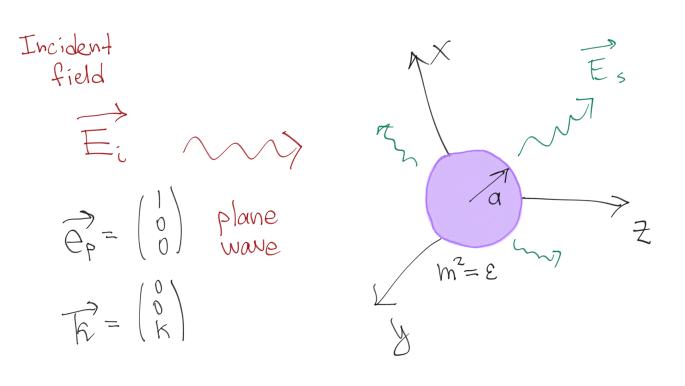


Figure 2: Base model scheme.

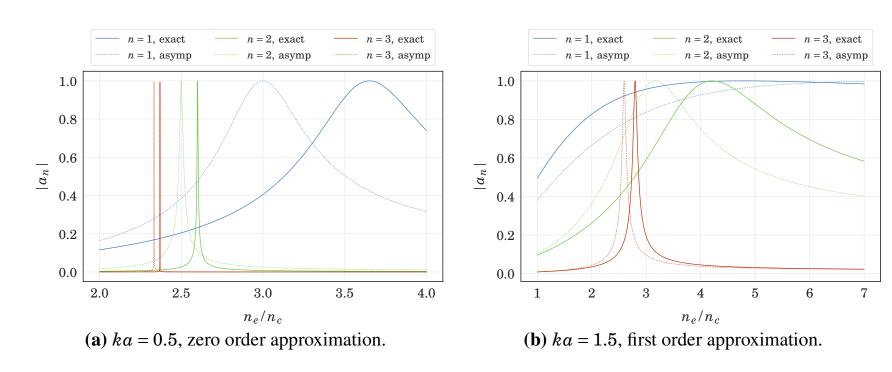
Mie theory can be used to describe the scattered field and the field inside a scattering object. Consider a spherical cluster and x-polarized plane wave propagating along the z axis (Fig. 2). Using generalized Fourier expansion in the case of an isotropic medium, we have the following form of the scattered field coefficients [1]:

$$\mathbf{E}_{s} = \sum_{n=1}^{\infty} E_{n} \left[ i a_{n} (ka, m) \mathbf{N}_{e1n}^{(3)} - b_{n} (ka, m) \mathbf{M}_{o1n}^{(3)} \right], \quad E_{n} = i^{n} E_{0} \frac{2n+1}{n(n+1)}, \quad (2)$$

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

$$b_n(x, m) = \frac{\psi'_n(x)\psi_n(mx) - m\psi'_n(mx)\psi_n(x)}{\xi'_n(x)\psi_n(mx) - m\psi'_n(mx)\xi_n(x)},$$
(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,  $x = k\alpha$  — dimensionless cluster radius,  $m = \sqrt{\varepsilon}$  — complex refractive index.

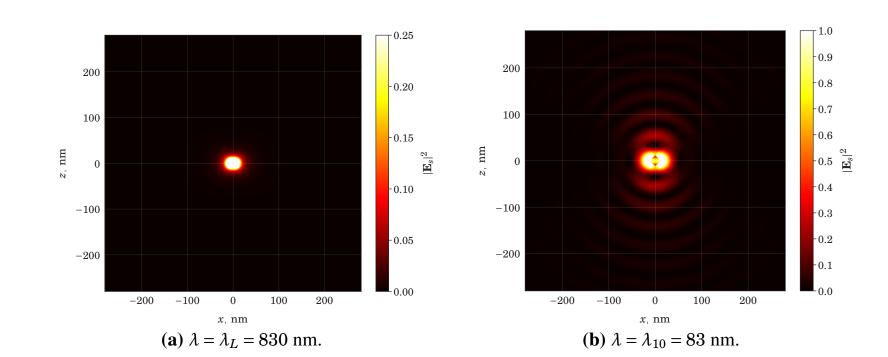


**Figure 3:** Coefficients of spherical harmonics in zero and first order approximation,  $\beta_e = 0$ . The "exact" curves are built using full expansions.

Using approximations of the Bessel functions [1] we can significantly simplify the coefficients (3, 4) and obtain resonance conditions for the electon density and cluster radius.

#### Single cluster

To check the analytical model, we calculated the values of the complex refractive index m, using resonance conditions from first order approximation at  $\lambda_{10} = 83$  nm,  $k\alpha = 0.7$ : 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 [2].

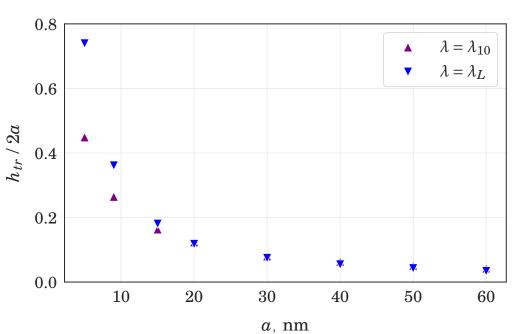


**Figure 4:**  $k\alpha = 0.7$  ( $\alpha \approx 8.9$  nm);  $|\mathbf{E}_s|^2$  in the plane of polarization of the incident wave.

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. In the resonance case the scattered field amplitude near the cluster is much higher than in the absence of resonance, where scattered waves as such are practically not observed.

#### Stationary model justification

In the general case, the calculation of the interaction of a high-intensity laser pulse with a group of dense spherical clusters located in three-dimensional space requires long and complex non-stationary calculations due the changes of the electron density of clusters in time. To check the scale of such change, we simulated the evolution of the electron density distribution for single one-dimensional cluster using LPIC++ [3].



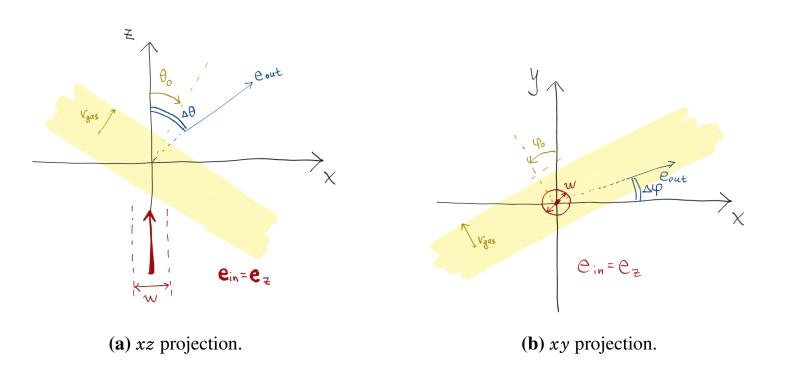
**Figure 5:** Asymptotic behavior of the average total thickness of the transition layer at  $0 \le t \le 10T$  with respect to the target radius.  $n_c$  used in the construction corresponds to the critical density for the wavelength  $\lambda = \lambda_{10}$ .

## **Diffraction theory**

The diffraction condition in the case of a three-dimensional regular grating with elastic scattering [4] can be converted as follows:

$$\begin{cases}
\cos\theta_{0}\sin\Delta\theta\cos\left(\Delta\varphi-\varphi_{0}\right)-\sin\theta_{0}(\cos\Delta\theta-1)=\frac{h'\lambda}{d} \\
\sin\Delta\theta\sin\left(\Delta\varphi-\varphi_{0}\right)=\frac{k'\lambda}{d} \\
\sin\theta_{0}\sin\Delta\theta\cos\left(\Delta\varphi-\varphi_{0}\right)+\cos\theta_{0}(\cos\Delta\theta-1)=\frac{l'\lambda}{d}
\end{cases}$$
(5)

where  $\Delta\theta$ ,  $\Delta\varphi$  are the angles characterizing the deviation 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 (6),  $\mathbf{e}_{\text{in}} = \mathbf{e}_z$  is the fixed direction of the incident radiation, d — distance between clusters. Using (5), we can obtain the angular distribution of diffracted radiation for given initial parameters d,  $\lambda$ ,  $\theta_0$ ,  $\varphi_0$ .



**Figure 6:** 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,  $r_{\rm gas}$  is the radius of the gas jet representing the target, w is the diameter of the Gaussian beam of incident radiation.

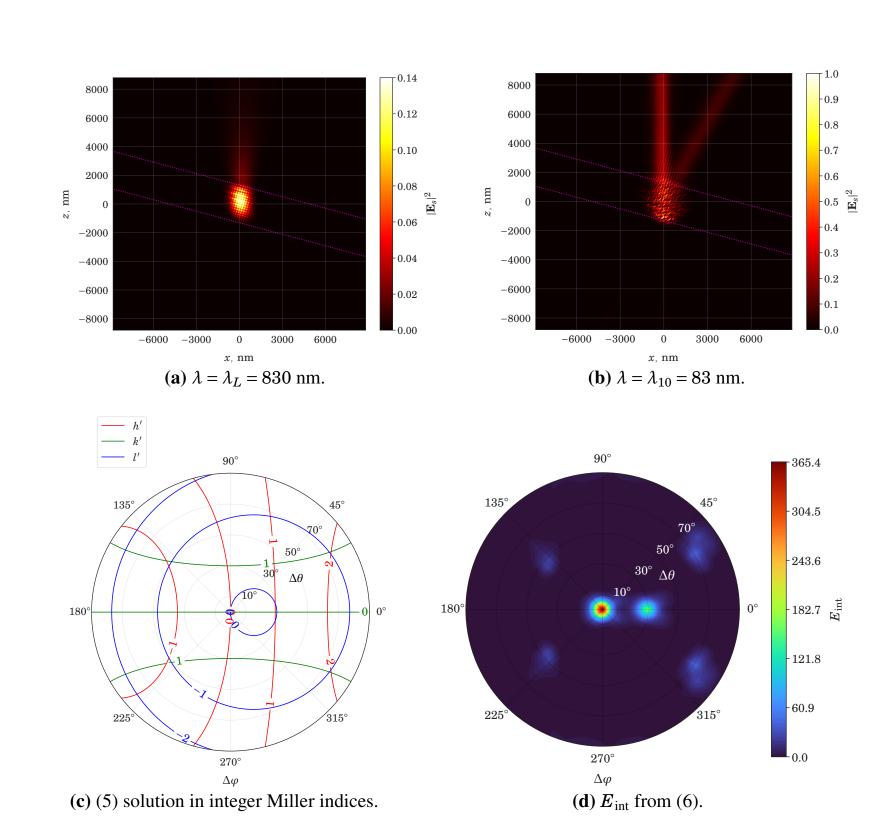
## **Monochromatic radiation scattering**

Within the framework of the stationary Mie scattering theory, many clusters were considered in the form of an extended cylindrical gas jet with regular and quasi-regular spatial configuration. 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. The program code CELES [5] was used for calculations.

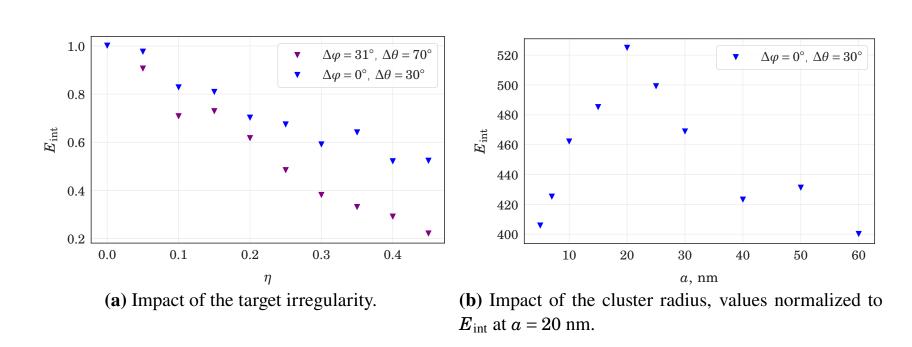
$$E_{\text{int}}(\eta, \lambda, V(\Delta\theta, \Delta\varphi), E_0, \varphi_0, \theta_0) = \int_{V(\Delta\theta, \Delta\varphi)} |\mathbf{E}_s(\eta, \lambda, E_0, \varphi_0, \theta_0)|^2 dV, \quad (6)$$

$$\mathbf{c} = \mathbf{c} \left( \mathbf{x}, \Delta \theta, \Delta \varphi \right) = M_y(\Delta \theta) M_z(\Delta \varphi) \mathbf{x}, \quad \mathbf{x} = \left( x \ y \ z \right)^T, \tag{7}$$

$$V(\Delta\theta, \Delta\varphi) = \{\mathbf{x} : c_x^2 + c_y^2 \le \rho^2, \ b_1^2 \le |\mathbf{x}|^2 \le b_2^2\}.$$
 (8)

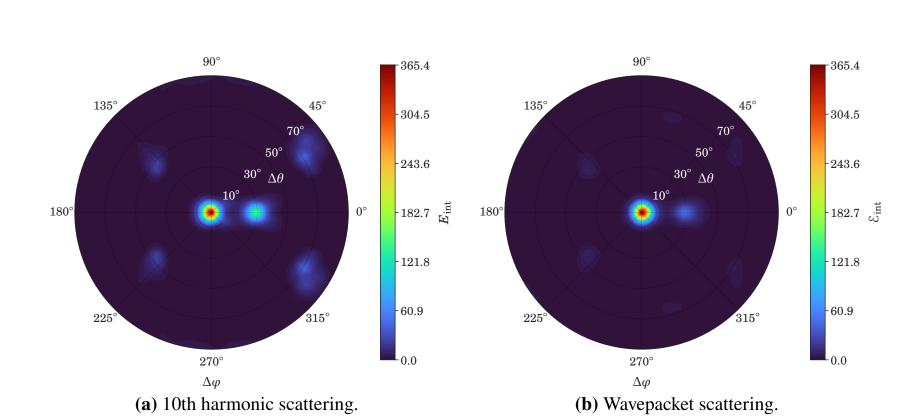


**Figure 7:** 10th harmonic scattering for a=20 nm and  $d=2\lambda_{10}$ ,  $\varphi_0=0^\circ$ ,  $\theta_0=15^\circ$ ,  $\lambda=\lambda_{10}=83$  nm,  $\Delta\theta\in[0,\pi/2]$ .

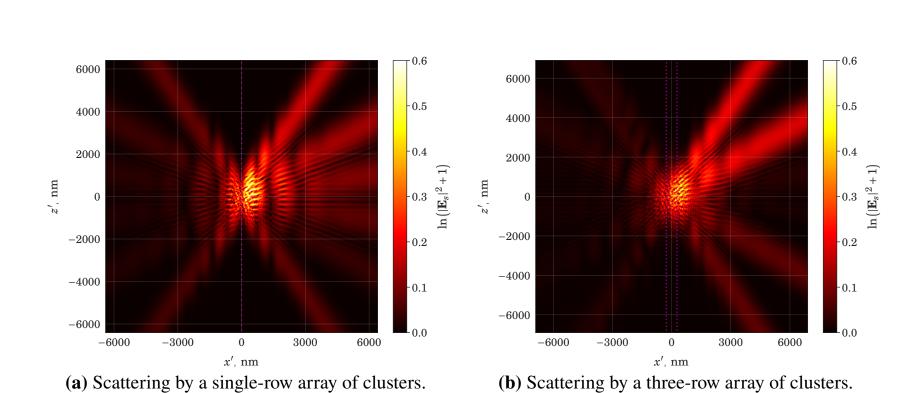


**Figure 8:** Scattering attenuation depending on the target irregularity and the cluster radius.

#### Wavepacket scattering



**Figure 9:** Angular scattering diagram of a Gaussian wave packet and the 10th harmonic.  $\theta_0 = 15^\circ$ ,  $\varphi_0 = 0^\circ$ ,  $d = 2\lambda_{10}$ , cluster radius a = 20 nm.



**Figure 10:** Scattering of a harmonic with  $\lambda \approx 89$  nm by an array of clusters,  $\varphi_0 = 0^\circ$ ,  $\theta_0 = 30^\circ$ ,  $\alpha = 30$  nm, the distance between clusters is  $d = 3\lambda$ , the incident field is directed from the lower left corner to the upper right at an angle  $\theta_0$  with respect to the normal to the target.

## 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. When the ionization is such that the electron density is near the 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 causes the attenuation of the directed energy up to four times for the most intense diffraction orders. In the case of non-monochromatic radiation, the angular dispersion boost decreases in accordance with the spectral distribution of the field amplitude, the directed energy weakens up to three times.

## References

- [1] C. F. Bohren and D. R. Huffman, *Absorption and scattering of light by small particles*. Wiley Science Paperback Series, 1998.
- [2] Z. Lécz and A. A. Andreev, "Angular dispersion boost of high order laser harmonics with carbon nano-rods," *Optics Express*, vol. 28, no. 4, pp. 5355–5366, 2020.
- [3] R. E. W. Pfund, R. Lichters, and J. Meyer-ter-Vehn, "LPIC++ a parallel one-dimensional relativistic electromagnetic particle-in-cell code for simulating laser-plasma-interaction," in *AIP Conference Proceedings*, AIP, 1998.
- [4] C. Kittel, *Introduction to Solid State Physics*. Wiley, New York, 1986.
- [5] A. Egel, L. Pattelli, G. Mazzamuto, D. Wiersma, and U. Lemmer, "CELES: CUDA-accelerated simulation of electromagnetic scattering by large ensembles of spheres," *J Quant Spectrosc Radiat*, vol. 199C, pp. 103–110, 2017.