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SOURCES OF ULTRASHORT X-RAY PULSES IN THE INVESTIGATION OF THE STRUCTURE AND DYNAMICS OF NANOSYSTEMS

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Abstract

Free electron lasers are today one of the main sources of ultrashort X-ray pulses. The installations used in the world today are presented and the results of experiments and calculations with various nanosystems are presented.

INTRODUCTION

Free Electron Lasers (FELs) were invented by Madey [1] and then experimentally demonstrated by his group at Stanford University in the 1970s. These lasers use relativistic electrons propagating through a periodic system of magnets (undulator) to generate and amplify coherent electromagnetic radiation.

Initially, the operation of such lasers was demonstrated in infrared mode. After that, work continued on expanding the FEL in the direction of EUV and X-ray modes. To solve this problem, the radiation was additionally amplified using the spontaneous emission self-amplification (SASE) mode. In the SASE mode, the particles of the electron beam are grouped into microbunches when they pass through the undulator and interact in it with the radiation of the beam itself. It is these FELs that are currently actively used in SAR and have great prospects for further improvement.

FELs have the widest frequency setting range and can generate very high peak and average laser powers. The formation of attosecond X-ray pulses on XFELs is currently being reported [2]. The possibility of creating zeptosecond pulses has been reported [3]. The extremely high power, coupled with the excellent lateral coherence of these XFELs, provides a dramatic increase in peak brightness.

The advent of XFELs has ushered in a new era in X-ray and X-ray studies. A large number of such lasers have been built in the last 15 years.

FLACH is the first XFEL facility for photons with energy in the extreme ultraviolet (EUV) region and was built in 2005 at DESY, Hamburg. LCLS is the first hard X-ray FEL built in 2009 at the SLAC National Accelerator Laboratory, USA. The SACLA plant in Japan and the FERMI plant in Trieste represent the first generation of XFELs, which have demonstrated tremendous scientific potential and influence in broad fields of science. XFEL installations are currently expanding worldwide: PAL-XFEL in South Korea, SwissFEL in Switzerland, European XFEL (EuXFEL) in Germany, etc.

DIFFRACTION ANALYSIS USING X-RAY USP

One of the most common approaches that can be used to observe an object in four dimensions x, y, z, t is time-dependent femtosecond crystallography (TR-SFX).

Measuring the temporal dynamics of such processes includes 2 stages, see Fig.1. The first is the launch of the studied dynamic process, and the second is the collection of diffraction patterns with different time delays by irradiating the ultrashort pulses of the studied system. To study such processes, ultrashort pulses of high brightness are used, since a pulse of even shorter duration τ is required to study dynamic processes with characteristic times τ_T, i.e., the condition $\tau << \tau_T$ must be satisfied. A very bright USP source is necessary because in the short time τ of interaction of the pulse with the system under study, a sufficient amount of radiation has been scattered so that it can be detected. To implement this concept, difficulties arise due to the destruction of the sample under study due to the high brightness of the ultrashort pulses. Despite this, in 2000 Janos Hajdu and his colleagues showed how this difficulty can be overcome [4]. They calculated that a molecule exposed to an X-ray pulse would begin to explode on a time scale of about 10 femtoseconds. Thus, shorter light pulses can pass through the molecule, capturing information about the virtually unperturbed structure. And pulses bright enough will give rise to continuous diffraction patterns strong enough to be measured.

The first stage can be implemented in several ways. The most common method is the so-called *pump-probe*. This method, suitable for studying quantitatively reproducible dynamics, is based on the creation of a time-delayed sequence of two short pulses: a "pump" pulse (usually an optical laser or the first XFEL pulse) unbalances the substance, and a "probe" X-ray pulse is used to create or taking "snapshots" of the structure of a substance at a specific point in time during the dynamic response of the substance. The structural response of the system can then be traced as a function of time by repeatedly applying these two impulses to the substance with different relative delays.

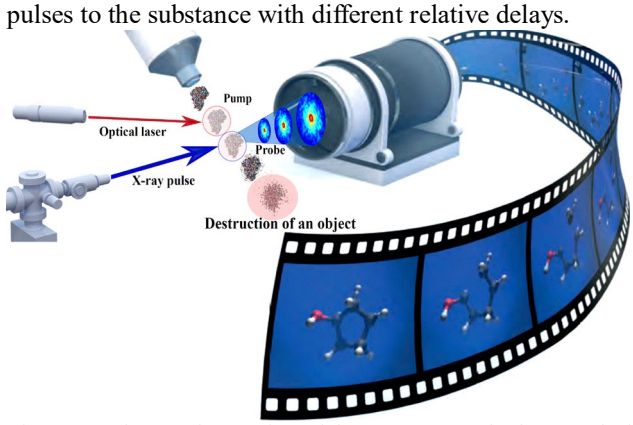

Figure 1: Scheme of operation of the TR-SFX method. Instead of an optical laser (Pump), there can be an X-ray pulse (the first pulse is Pump, and the second is Probe) [5].

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SCATTERING SPECTRA OF ULTRA-SHORT PULSES ON NANOSTRUCTURES

In [6-8], a theory of scattering of X-ray ultrashort pulses by nanostructures consisting of identical atoms was developed. In this theory, the basic expressions for calculations were obtained:

$$\frac{d^2W}{d\omega d\Omega_{\mathbf{k}}} = \frac{1}{(2\pi)^2}\frac{1}{c^3\omega}\left(N_a \mathrm{N}_e S(\omega,\mathbf{n},\mathbf{n}_0) + \delta_N(\mathbf{p})\mathrm{N}_e(\mathrm{N}_e-1)\mathrm{F}(\omega,\mathbf{n},\mathbf{n}_0)\right),$$

$$S(\omega,\mathbf{n},\mathbf{n}_0) = G(\omega,\mathbf{n},\mathbf{n}_0) - F(\omega,\mathbf{n},\mathbf{n}_0), \mathrm{G}(\omega,\mathbf{n},\mathbf{n}_0) = \frac{1}{N_e}\int \rho(\mathbf{r})\left|\mathbf{f}(\mathbf{r})\right|^2 d\mathbf{r},$$

$$F(\omega,\mathbf{n},\mathbf{n}_0) = \frac{1}{N_e^{\,2}}\left|\int \rho(\mathbf{r})\mathbf{f}(\mathbf{r})e^{-i\mathbf{kr}}d\mathbf{r}\right|^2, \mathbf{f}(\mathbf{r}_a) = \left[\tilde{\mathbf{E}}(\omega)\times\mathbf{n}\right],$$

$$\tilde{\mathbf{E}}(\omega) = \int_{-\infty}^{+\infty}\left(\mathbf{E}(\mathbf{r}_{a,e},t) - \frac{1}{2}\nabla_a\left(\frac{\mathbf{E}(\mathbf{r}_{a,e},t)}{c}\right)^2\right)e^{i\omega t}dt, \delta_N(\mathbf{p}) = \left|\sum_a e^{i\mathbf{p}\mathbf{R}_a}\right|^2. \quad (1)$$

Summation in Eq. (1) is carried out over all atoms of the considered system. We will use the electronic density $\rho(\mathbf{r})$ of the Dirac-Hartree-Fock-Slater model [9].

As an example, we present the results of calculations of the scattering spectra of USP on graphene. For this, we first need to determine the $\delta_N(\mathbf{p})$ factor, which is equal to [8]:

$$\delta_N(\mathbf{p}) = \frac{4\sin^2\left(\frac{\sqrt{3}}{2}L\mathbf{p}\mathbf{j}d\right)}{\sin^2\left(\frac{\mathbf{p}\mathbf{i}d}{2}\right)\sin^2\left(\frac{\sqrt{3}}{2}\mathbf{p}\mathbf{j}d\right)} \quad (2)$$

$$\left\{\cos\left(\frac{\mathbf{p}\mathbf{j}d}{\sqrt{3}}\right)\sin\left(\frac{\mathbf{p}\mathbf{i}dN}{2}\right) + \cos\left(\frac{\mathbf{p}\mathbf{j}d}{2\sqrt{3}}\right)\sin\left(\frac{\mathbf{p}\mathbf{i}d(N+1)}{2}\right)\right\}^2,$$

where L is the number of graphene ribbons, N is the number of the cells in graphene tape, d is the distance between atoms along the x axis, and $\boldsymbol{i}$ and $\boldsymbol{j}$ are unit vectors along the x and y axes, respectively, see Fig. 2.

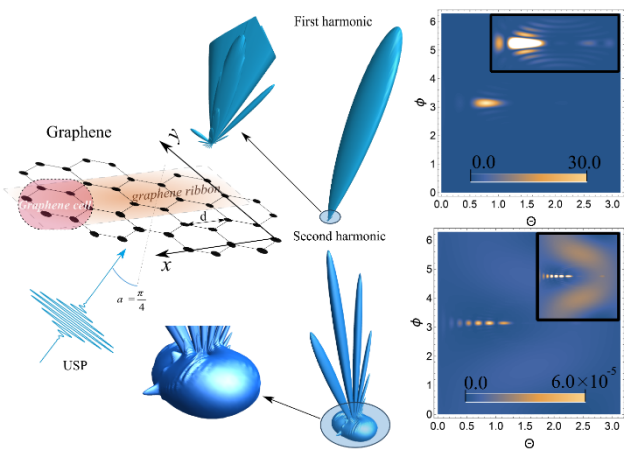

Figure 2: 3D radiation pattern of the scattering spectra USP (see Eq. (1)) and the contour plot for the spectra, where θ and ϕ are angles (in radians) with a spherical coordinate system, i.e. polar and azimuth angles, respectively. Inserts in contour plots show an enlarged (more contrasting) scatter spectrum. The first harmonic is in the figures above, and the second harmonic is below. A USP was selected with a frequency ω_0=100 au, a pulse duration γ corresponding to 43 as and amplitude E_0 = 1000 au. The numbers of the graphene ribbons and cells are $L=10$, $N=10$, respectively The angle of incidence α between $\mathbf{n}_0$ and the z axis was selected as $\alpha=\pi/4$ [8].

CONCLUSION

As a result, we obtain two diffraction patterns on the fundamental and second harmonics, both of which were derived from the same USP and on a given polyatomic system. As a result, diffraction analysis can be carried out by studying two diffraction patterns at once, which allows you to get more details about the research object.

ACKNOWLEDGEMENTS

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