Diffraction of X-ray pulse in crystals.

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Recently the investigation of the extremely short (subpicosecond) X-ray pulses interaction with crystals takes interest because of the development of linac-driven X-ray Free Electron Laser, operating in the SASE mode and X-ray Volume Free Electron Laser [1, 2].

According to the analysis [3] short X-ray pulse passing through a crystal is accompanied by the significant time delay of radiation. The $\delta-$pulse delay law for the Bragg diffraction is proportional to $\sim \left|\frac{J_1(at)}{t}\right|^2$, where J_1 - is the Bessel function, a - a coefficient will be defined below, t - time.

In the present paper the delay law dependence on the diffraction asymmetry parameters is analyzed. It is shown that the use of subpicosecond pulses allows to observe the phenomenon of the time delay of pulse in crystal and to investigate the delay law experimentally. It is also shown that the pulse delay law depends on the quanta polarization.

Let us consider the pulse of electromagnetic radiation passing through the medium with the refraction index $n(\omega)$. The wave packet group velocity is as follows:

$$v_{gr} = \left(\frac{\partial \omega n(\omega)}{c \partial \omega}\right)^{-1} = \frac{c}{n(\omega) + \omega \frac{\partial n(\omega)}{\partial \omega}}, \tag{1}$$

where c - is the speed of light, ω - is the quantum frequency.

In the X-ray range ($\sim$tens of keV) the index of refraction has the universal form $n(\omega) = 1 - \frac{\omega_L^2}{2\omega^2}$, ω_L is the Langmour frequency. Additionally, $n - 1 \simeq 10^{-6} \ll 1$. Substituting $n(\omega)$ to (1) one can obtain that $v_{gr} \simeq c\left(1 - \frac{\omega_L^2}{\omega^2}\right)$. It is clear that the group velocity is close to the speed of light. Therefore the time of wave packet delay in a medium is much shorter than the that needed for passing the length equal to the target width in a vacuum.

$$\Delta T = \frac{1}{v_{gr}} - \frac{1}{c} \simeq \frac{l}{c}\frac{\omega_L^2}{\omega^2} \ll \frac{l}{c}. \tag{2}$$

To consider the pulse diffraction in a crystal one should solve Maxwell equations that describe pulse passing through a crystal. Maxwell equations are linear, therefore it is convenient to use Fourier transform by time and to rewrite these equations as functions of frequency:

$$\left[-curl\ curl\ \vec{E}_{\vec{k}}(\vec{r},\omega) + \frac{\omega^2}{c^2}\vec{E}_{\vec{k}}(\vec{r},\omega)\right]_i + \chi_{ij}(\vec{r},\omega)\ E_{\vec{k},j}(\vec{r},\omega) = 0, \tag{3}$$

where $\chi_{ij}(\vec{r},\omega)$ - is the spatially periodic tensor of susceptibility, $i,j = 1,2,3$, repeated indices imply summation.

Making the Fourier transformation of these equations by coordinate variables one can derive a set of equations matching the incident and diffracted waves. When two strong waves are excited under diffraction (so-called two-beam diffraction case) the following set of equations for wave amplitudes determining can be obtained:

$$\begin{aligned} &\left(\tfrac{k^2}{\omega^2} - 1 - \chi_0\right)\vec{E}^s_{\vec{k}} - c_s\chi_{-\vec{\tau}}\vec{E}^s_{\vec{k}_\tau} = 0 \\ &\left(\tfrac{k_\tau^2}{\omega^2} - 1 - \chi_0\right)\vec{E}^s_{\vec{k}_\tau} - c_s\chi_{\vec{\tau}}\vec{E}^s_{\vec{k}} = 0 \end{aligned} \tag{4}$$

Here $\vec{k}$ is the wave vector of the incident wave, $\vec{k}_{\vec{\tau}} = \vec{k} + \vec{\tau}$, $\vec{\tau}$ is the reciprocal lattice vector; $\chi_0, \chi_{\vec{\tau}}$ are the Fourier components of the crystal susceptibility:

$$\chi(\vec{r}) = \sum_{\vec{\tau}} \chi_{\vec{\tau}} \exp(i\vec{\tau}\vec{r}) \tag{5}$$

$C_s = \vec{e}^{\ s}\vec{e}^{\ s}_{\vec{\tau}}$, $\vec{e}^{\ s}(\vec{e}^{\ s}_{\vec{\tau}})$ are the unit polarization vectors of the incident and diffracted waves, respectively.

The condition for the linear system (4) to be solvable leads to a dispersion equation that determines the possible wave vectors $\vec{k}$ in a crystal. It is convenient to present these wave vectors as:

$$\vec{k}_{\mu s} = \vec{k} + æ_{\mu s}\vec{N},\ \ æ_{\mu s} = \frac{\omega}{c\gamma_0}\ \varepsilon_{\mu s},$$

where $\mu = 1,2$; $\vec{N}$ is the unit vector of a normal to the entrance crystal surface directed into a crystal ,

$$\varepsilon_s^{(1,2)} = \frac{1}{4}[(1+\beta)\chi_0 - \beta\alpha_B] \pm \frac{1}{4}\left\{[(1+\beta)\chi_0 - \beta\alpha_B - 2\chi_0]^2 + 4\beta C_S^2\chi_{\vec{\tau}}\chi_{-\vec{\tau}}\right\}^{1/2}, \tag{6}$$

$\alpha_B = (2\vec{k}\vec{\tau} + \tau^2)k^{-2}$ is the off-Bragg parameter ($\alpha_B = 0$ when the Bragg condition of diffraction is exactly fulfilled),

$$\gamma_0 = \vec{n}_\gamma \cdot \vec{N}, \quad \vec{n}_\gamma = \frac{\vec{k}}{k}, \quad \beta = \frac{\gamma_0}{\gamma_1}, \quad \gamma_1 = \vec{n}_{\gamma\tau} \cdot \vec{N}, \quad \vec{n}_{\gamma\tau} = \frac{\vec{k} + \vec{\tau}}{|\vec{k} + \vec{\tau}|}$$

The general solution of (3,4) inside a crystal is:

$$\vec{E}^s_{\vec{k}}(\vec{r}) = \sum_{\mu=1}^{2} \left[\vec{e}^{\ s} A_\mu \exp(i\vec{k}_{\mu s}\vec{r}) + \vec{e}_\tau^{\ s} A_{\tau\mu} \exp(i\vec{k}_{\mu s\tau}\vec{r}) \right] \tag{7}$$

By matching these solutions with the solutions of Maxwell equation for the vacuum area we can find the explicit expression for $\vec{E}^s_{\vec{k}}(\vec{r})$ throughout the space. It is possible to discriminate several types of diffraction geometries, namely, the Laue and the Bragg schemes are the most well-known [4].

In the case of two-wave dynamical diffraction crystal can be described by two effective refraction indices

$$n_s^{(1,2)} = 1 + \varepsilon_s^{(1,2)},$$

$$\varepsilon_s^{(1,2)} = \frac{1}{4}\left\{ \chi_0(1+\beta) - \beta\alpha \pm \sqrt{(\chi_0(1-\beta) + \beta\alpha)^2 + 4\beta C_s \chi_\tau \chi_{-\tau}} \right\}. \tag{8}$$

The diffraction is significant in the narrow range near the Bragg frequency, therefore χ_0 and χ_τ can be considered as constants and the dependence on ω should be taken into account for $\alpha = \dfrac{2\pi\overrightarrow{\tau}(2\pi\overrightarrow{\tau} + 2\overrightarrow{k})}{k^2} = -\dfrac{(2\pi\tau)^2}{k_B^3 c}(\omega - \omega_B)$, where $k = \dfrac{\omega}{c}$; $2\pi\overrightarrow{\tau}$ - the reciprocal lattice vector that characterizes the set of planes where the diffraction occurs; Bragg frequency is determined by the condition $\alpha = 0$.

From (1,8) one can obtain

$$v_{gr}^{(1,2)s} = \frac{c}{n^{(1,2)}(\omega) \pm \beta \dfrac{(2\pi\tau)^2}{4k_B^2} \dfrac{(\chi_0(1-\beta) + \beta\alpha)}{\sqrt{(\chi_0(1-\beta) + \beta\alpha)^2 + 4\beta C_s \chi_\tau \chi_{-\tau}}}}. \tag{9}$$

In the general case $(\chi_0(1-\beta) + \beta\alpha) \simeq 2\sqrt{\beta}\chi_0$, therefore the term that is added to the $n_s^{(1,2)}(\omega)$ in the denominator (9) is of the order of 1. Moreover, v_{gr} significantly differs from c for the antisymmetric diffraction ($|\beta| \gg 1$). It should be noted that because of the complicated character of the wave field in a crystal one of the $v_{gr}^{(i)s}$ can appear to be much higher than c and negative. When β is negative the subradical expression in (9) can become equal to zero (Bragg reflection threshold) and $v_{gr} \to 0$. It should be noted that in the presence of the time-alternative external field a crystal can be described by the effective indices of refraction that depend on the external field frequency Ω . Therefore, in this case v_{gr} appears to be the function of Ω . This can be easily observed in the conditions of X-ray-acoustic resonance. The analysis done allows to conclude that center of the X-ray pulse can undergo the significant delay in a crystal $\Delta T \gg \frac{l}{c}$ that it is possible to investigate experimentally. Thus, when $\beta = 10^3$, $l = 0,1$ cm and $l/c \simeq 3 \cdot 10^{-12}$ the delay time can be estimated as $\Delta T \simeq 3 \cdot 10^{-9}$sec.

Let us study now the time dependence of delay law of radiation after passing through a crystal. Assuming that $B(\omega)$ is the reflection or transmission amplitude coefficients of a crystal one can obtain the following expression for the pulse form

$$E(t) = \frac{1}{2\pi} \int B(\omega) E_0(\omega) e^{-i\omega t} d\omega = \int B(t-t') E_0(t') dt'. \tag{10}$$

where $E_0(\omega)$ is the amplitude of the electromagnetic wave incident on a crystal

In accordance with the general theory for the Bragg geometry the amplitude of the diffractionally reflected wave for the crystal width that is much greater than the absorbtion length can be written [4]

$$B_s(\omega) = -\frac{1}{2\chi_\tau} \left\{ \chi_0(1+|\beta|) - |\beta|\,\alpha - \sqrt{(\chi_0(1-|\beta|) - |\beta|\,\alpha)^2 - 4\,|\beta|\,C_s \chi_\tau \chi_{-\tau}} \right\} \tag{11}$$

In the absence of resonance scattering the parameters χ_0 and $\chi_{\pm\tau}$ can be considered as constants and frequency dependence is defined by the term $\alpha = -\frac{(2\pi\tau)^2}{k_B^3 c}(\omega - \omega_B)$. So, $B_s(t)$ can be find from

$$B_s(t) = -\frac{1}{4\pi\chi_\tau} \int \left\{ \chi_0(1+|\beta|) - |\beta|\,\alpha - \sqrt{(\chi_0(1-|\beta|) - |\beta|\,\alpha)^2 - 4\,|\beta|\,C_s \chi_\tau \chi_{-\tau}} \right\} e^{-i\omega t} d\omega. \tag{12}$$

Fourier transform of the first term results in $\delta(t)$ and we can neglet it, because the delay is described by the second term. The second term can be calculated by the methods of theory of function of complex argument:

$$B_s(t) = -\frac{i}{4\chi_\tau} |\beta| \frac{(2\pi\tau)^2}{k_B^2 \omega_B} \frac{J_1(a_s t)}{t} e^{-i(\omega_B + \Delta\omega_B)t} \theta(t), \tag{13}$$

or

$$B_s(t) = -\frac{i\sqrt{|\beta|}}{2} \frac{J_1(a_s t)}{a_s t} e^{-i(\omega_B + \Delta\omega_B)t} \theta(t), \tag{14}$$

where

$$a_s = \frac{2\sqrt{C_s \chi_\tau \chi_{-\tau}}\omega_B}{\sqrt{|\beta|}\frac{(2\pi\tau)^2}{k_B^2}}, \Delta\omega_B = -\frac{\chi_0 (1 + |\beta|)\omega_B k_B^2}{|\beta| (2\pi\tau)^2}.$$

Since χ_0 and χ_τ are complex, both a_s and $\Delta\omega_B$ have real and imaginary parts. According to (12-14) in the case of Bragg reflection of short pulse (pulse frequency band width $\gg$ frequency width of the total reflection range) appear both the instantly reflected pulse and the pulse with amplitude undergoing damping beatings. Beatings period increases with $|\beta|$ grows and χ_τ decrease. Pulse intensity can be written as

$$I_s(t) \sim |B_s(t)|^2 = \frac{|\beta|}{2} \left| \frac{J_1(a_s t)}{at} \right|^2 e^{-2\,\mathrm{Im}\,\Delta\omega_B t} \theta(t). \tag{15}$$

It is evident that the reflected pulse intensity depends on the orientation of photon polarization vector $\overrightarrow{e_s}$ and undergoes the damping oscillations on time.

Let us evaluate the effect. Characteristic values are $\mathrm{Im}\,\Delta\omega_B \sim \mathrm{Im}\,\chi_0\omega_B$ and $\mathrm{Im}\,a \sim \frac{\mathrm{Im}\,\chi_\tau\omega_B}{\sqrt{\beta}}$. For 10 keV for the crystal of Si $\mathrm{Im}\,\chi_0 = 1,6 \cdot 10^{-7}$, for LiH $\mathrm{Im}\,\chi_0 = 7,6 \cdot 10^{-11}, \mathrm{Im}\,\chi_\tau = 7 \cdot 10^{-11}$, for LiF $\mathrm{Im}\,\chi_0 \sim 10^{-8}$. Consequently, the characteristic time τ for the exponent decay in (15) can be estimated as follows ($\omega_B = 10^{19}$):

for Si - $\tau \sim 10^{-12}$ sec, for LiF - $\tau \sim 10^{-10}$ sec, for LiH - $\tau \sim 10^{-9}$ sec!!

The reflected pulse also undergoes oscillations period of which increases with $|\beta|$ grows and decreasing of $\mathrm{Re}\,\chi_\tau$. This period can be estimated for $\beta = 10^2$ and $\mathrm{Re}\,\chi_\tau \sim 10^{-6}$ as $T\tilde{}10^{-12}$ sec (for Si, LiH, LiF).

When the resolving time of the detection equipment is greater than the oscillation period the expression (15) should be averaged over the period of oscillations. Then, for the time intervals when $\mathrm{Re}\, a_s t \gg 1$, $\mathrm{Im}\, \Delta\omega_B t \ll 1$ the delay law (15) has the power function form:

$$I_s(t) \sim t^{-3}.$$

References

[1] V.G.Baryshevsky, K.G.Batrakov, I.Ya.Dubovskaya J.Phys. D: Appl. Phys. 24(1991) 1250-1257.

[2] CERN COURIER 39, N4 (1999) 11-12

[3] V.G.Baryshevsky Izvestia AN BSSR ser.phys.-mat. N5 (1985) 109-112

[4] Z.G.Pinsker Dynamical scattering of X-rays in crystals (Springer, Berlin, 1988)