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About new applications of parametric X-radiation for crystallography

I. D. Feranchuk[a]* and A. Ulyanenkov[b]†

[a]*Byelorussian State University, F. Skariny Av. 4, 220080 Minsk, Republic of Belarus, and* [b]*X-ray Research Laboratory, Rigaku Corp., 3-9-12 Matsubara-cho, Akishima-shi, Tokyo 196-8666, Japan. E-mail: ilya@fer.minsk.by*

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Abstract

A theoretical analysis of the parametric X-radiation (PXR) produced by the nonrelativistic electrons in an X-ray tube with a monocrystalline anode is presented. It is shown that PXR can be considered as a source of quasi-monochromatic X-radiation for laboratory diffraction experiments. The intensity of the PXR is comparable with the intensity of the characteristic radiation, but the frequency of the former can be varied in a wide range. Possible applications of the phenomenon for crystallography are discussed.

1. Introduction

Parametric X-rays produced by a charged particle moving uniformly in a crystal were theoretically predicted more than 20 years ago (Ter-Mikaelian, 1972; Baryshevskii & Feranchuk, 1971, 1973, 1983; Garibyan & Yang, 1972) and were observed experimentally with ultrarelativistic electrons (Adishchev *et al.*, 1985, 1986).

Various applications of PXR to structure investigations have been suggested (Feranchuk, 1979; Baryshevskii & Feranchuk, 1980; Feranchuk & Ivashin, 1989) but they were mostly regarded as exotic methods. The main reason for this is the necessity of using a linear accelerator to obtain the maximal spectral intensity output of PXR (Baryshevskii & Feranchuk, 1985). Therefore, it is considered that PXR cannot compete with the main sources for structure investigations, *i.e.* synchrotron radiation (SR) from a recycling electron beam in an accelerator, and the characteristic radiation (CXR) from X-ray tubes under laboratory conditions.

The coherent radiation produced by the movement of charged electrons through a single crystal has been intensively studied in recent decades. There are many works devoted to the investigation of different types of coherent radiation produced during the interaction of low-energy electrons with a crystal, such as coherent *Bremsstrahlung* (CB) (Korobochko *et al.*, 1965; Reese *et al.*, 1984; Sáenz & Überall, 1985) and channelling radiation (CR) (Andersen *et al.*, 1981; Vorobiev & Vorobiev, 1975; Sáenz & Überall, 1985). The main purpose of this paper is to demonstrate theoretically that PXR may be applied under laboratory conditions, *i.e.* that PXR from low-energy electrons, like other sources of radiation, may be considered as an intense source of X-rays for crystal structure investigations.

We show that PXR can be produced by nonrelativistic electrons in an X-ray tube with a monocrystalline anode or a monocrystal attached to the anode plate, and that the spectral intensity of such radiation is no less than that of CXR. The spectral intensity of radiation arising *via* any radiation mechanism is known to decrease proportionally to E^2 with decreasing particle energy E (Baryshevskii & Feranchuk, 1985). PXR follows the same behaviour; this fact has been confirmed experimentally by Freudenberger *et al.* (1995). However, PXR peaks still remain sharp with respect to the background, even for nonrelativistic electrons, as was first discussed by Baryshevskii *et al.* (1975). The energy of electrons in conventional X-ray tubes is about three orders of magnitude less than the optimal electron energy for PXR (Baryshevskii & Feranchuk, 1985) and consequently the corresponding spectral intensity is six orders less. However, the electron current in tubes is about 10^8–10^9 times higher than in ordinary linear accelerators (Freudenberger *et al.*, 1995). Thus, the total intensity of the monochromatic X-radiation produced in an X-ray tube by the PXR process proves to be of the same order as the CXR intensity.

This argument can be clarified if the CXR is considered to result from the resonant scattering of the pseudophotons corresponding to the electromagnetic field of the electron beam on the atoms of the anode material (Ter-Mikaelian, 1972; Akhiezer & Berestetzkii, 1969); then PXR can be treated as the result of diffraction of the same pseudophotons on the crystallographic planes of the monocrystalline anode. The frequencies of the diffracted pseudophotons are determined by the crystal-lattice periods and are not related to the characteristic frequencies of the separate atoms. This presents the possibility of tuning the PXR spectrum by varying the angle between the crystallographic planes and the electron velocity.

Having considered the nature of PXR, we propose several configurations of PXR diffraction experiments.

† On leave from Institute of Nuclear Problems, Bobruiskaya Str. 11, 220080 Minsk, Republic of Belarus.

In the first scheme, the investigated sample is attached to the anode of the tube; the lattice periods and structure amplitudes of the probed material are defined from the PXR spectrum. In the second set-up, PXR from an X-ray tube with a monocrystalline anode can be considered as a source of quasi-monochromatic radiation with tunable frequency, which may be important for some diffraction experiments. Finally, we shall show that PXR can be used for increasing the intensity of the ordinary CXR lines of an anode material. As will be shown below, the orientation of the anode plate relative to the direction of the electron velocity can be chosen in such a way that the characteristic frequency of the atoms of the anode material coincides with one of the PXR spectral lines. In this case, an increase of the radiation intensity is expected.

2. Qualitative analysis

The electromagnetic field of a charged particle can be represented as a beam of virtual quanta ('pseudophotons') with nearly uniform frequency distribution; such a beam is a potential source of real photons with a 'white' spectrum (Ter-Mikaelian, 1972; Akhiezer & Berestetzkii, 1969). Furthermore, any radiation mechanism can be considered to convert the pseudophotons to the real photons in some spectral interval depending on the interaction between the particle and the external field. This approach allows one to introduce the universal formula for the estimation of the spectral angular distribution of photons produced by the electron beam interacting with some external field or medium (Baryshevskii & Feranchuk, 1983, 1985):

$$\frac{\partial^2 N_s}{\partial\omega\partial\mathbf{n}} \simeq \frac{\alpha}{2\pi}(\mathbf{e}_s\mathbf{v})^2 R(\omega, \mathbf{n}, E) L^2_{\rm coh}(\omega, \mathbf{n}, E)\omega I. \qquad (1)$$

Here α is the fine structure constant, $\mathbf{e}_s$ is the polarization vector of the photon with frequency ω, the unitary vector $\mathbf{n}$ defines the direction of photon emission, I is the number of electrons with energy E and velocity $\mathbf{v}$ passing through the interaction region in unit time, and the unit system $\hbar = c = 1$ is chosen. Despite the fact that this estimation does not take into account electron-beam collective effects, such as in the case of a free-electron laser, it is in good agreement with the results for coherent *Bremsstrahlung* reported by Reese *et al.* (1984) and Sáenz & Überall (1985).

The dimensionless coefficient $R(\omega, \mathbf{n}, E)$ defines the probability of transformation of a pseudophoton to a real photon with wave vector $\mathbf{k} = \omega\mathbf{n}$. The magnitude of the probability depends on the specific features of the radiation mechanism and satisfies the inequality

$$R(\omega, \mathbf{n}, E) \leq 1. \qquad (2)$$

The coherent length $L_{\rm coh}$, introduced by Galitzky & Gurevich (1964) and Bolotovskii & Voskresenskii (1966) for the qualitative analysis of the radiation, is the universal characteristic of any radiation process, which is defined by the kinematics of the interaction between the electron and radiation fields. In the general case, the magnitude of $L_{\rm coh}$ can be estimated as the lesser value of the following (Galitzky & Gurevich, 1964; Bolotovskii & Voskresenskii, 1966)

$$L_{\rm coh} = \min\{L, L_{\rm abs} = 2/\omega\varepsilon'', \\ L_{\rm el} = (p_z - p_{fz} - k_z)^{-1} \equiv q_z^{-1}\}, \qquad (3)$$

where $q_z = p_z - p_{fz} - k_z$ is the projection of the transmitted momentum on the direction of the electron velocity; $\mathbf{p}$ and $\mathbf{p}_f(E_f)$ are the momenta of the electron at the initial and final states, $E_f = E - \omega$, and L is the sample length along the electron trajectory. As the condition $\omega \ll E$ is satisfied, the parameter $L_{\rm el}$ is expressed as

$$L_{\rm el} \simeq 1/\omega[1 - (\mathbf{vn})\varepsilon'].$$

Here, ε' and ε'' are the real and imaginary parts of the medium refraction index, respectively. From the physical point of view, the value $L_{\rm coh}$ defines the length of the electron path in the medium where the emitted photons are coherent.

The simple formula (1) shows that, keeping the energy of the electron constant, the ratio of intensities for various radiation mechanisms is defined by the transformation coefficient (2). This coefficient achieves its maximum value for Čerenkov radiation at $q_z = 0$ and within the spectral interval where the inequality $\varepsilon' > 1$ is satisfied. For PXR, $R(\omega, \mathbf{n}, E)$ coincides with the coefficient of the Bragg reflection from the crystallographic planes and tends to unity for photons with vector $\mathbf{k}$ near the Ewald sphere (Baryshevskii & Feranchuk, 1983). The maximum value of the PXR intensity corresponds to the case (Baryshevskii & Feranchuk, 1971, 1973, 1983) when the Bragg diffraction condition for some reciprocal vector $\mathbf{g}$,

$$(\mathbf{k} + \mathbf{g})^2 = k^2, \qquad (4)$$

is fulfilled simultaneously with the Čerenkov condition

$$p_z - p_{fz} - k_z = 0. \qquad (5)$$

The first condition (4) dictates a value of R close to unity, $R \simeq 1$, whereas equation (5) makes the value $L_{\rm coh}$ maximal. This situation is possible only for relativistic particles with $E \gg m$; for electrons, the optimal value, $E_{\rm opt}$, is about 50 MeV (Baryshevskii & Feranchuk, 1985).

Evidently, for the case $E \gg m$ and $\omega \ll E$, when condition (5) is not fulfilled, $L_{\rm el}$ is proportional to $(E/m)^2$ and decreases proportionally to E^2 with diminishing energy. At the same time, the factor $(\mathbf{e}_s\mathbf{v})^2$, being defined by the square of the characteristic angle of the radiation, increases proportionally to E^{-2} (see Baryshevskii & Feranchuk, 1983). This means that the spectral intensity of radiation depends on the charged-particle energy as follows [confirmed experimentally for PXR by Freudenberger *et al.* (1995)]:

$$\frac{\partial^2 N}{\partial \mathbf{n} \partial \omega} \sim E^2. \qquad (6)$$

The estimation (6) indicates the essential feature of the spectral intensity function, namely the behaviour of the latter is defined solely by the kinematics of the radiation process not by the mechanism of radiation. As a consequence, the ratio of the peaks in the spectral angular distribution of the radiation intensity (1), being defined by the coefficient $R(\omega, \mathbf{n}, E)$, remains almost constant in the wide range of the electron energy.

This simple result is the basis of our proposal to use PXR spectral lines from nonrelativistic electrons with energy $E \simeq 20$–50 keV, which are usual for laboratory X-ray tubes. The proposed model predicts the losses of radiation intensity produced by one electron, by the factor 10^{-6}–10^{-7} compared with its maximum value. However, this decrease can be compensated by increasing the electron current from 10^{-8}–10^{-9} A, as in linear accelerators (Freudenberger *et al.*, 1995), to $\sim$1 A, which is an ordinary value for X-ray tubes.

3. Theory

The detailed quantitative analysis of the PXR intensity should take into account the quantum effects according to Baryshevskii & Feranchuk (1983). In previous publications, the PXR intensity was calculated either for the case of ultrarelativistic electrons ($E \gg m$) or neglecting the quantum recoil in the radiation process ($E \gg \omega$) (Freudenberger *et al.*, 1995; Nitta, 1991; Stepanov *et al.*, 1996). Both conditions are not fulfilled for the case of nonrelativistic particles discussed here.

Fig. 1 shows the possible geometry of the experiment. The anode of the X-ray tube is assumed to be the monocrystal or the monocrystal plate attached to the anode ($\mathbf{g}$ is the arbitrary reciprocal-lattice vector). The photons are registered in the direction $\mathbf{n}$ with beam angular resolution $\Delta\Omega$ defined by the exit window of the tube or the collimating slit; $J = eI$ is the electron current, $\mathbf{v}$ is the electron velocity depending on the tube voltage U.

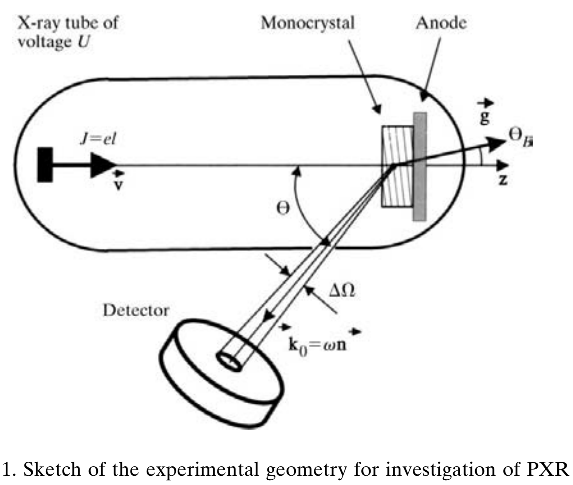

Fig. 1. Sketch of the experimental geometry for investigation of PXR from an X-ray tube.

The spectral angular distribution of the number of photons emitted within the crystal by the electron beam in unit time is defined by the following formula (Baryshevskii & Feranchuk, 1983):

$$\frac{\partial^2 N_s}{\partial \omega \partial \mathbf{n}} = I\omega^2 \sum \int |T_{fi}|^2 \delta(E - E_f - \omega) \frac{\mathrm{d}\mathbf{p}_f}{(2\pi)^6}, \qquad (7)$$

where

$$T_{fi} = \textstyle\int \mathrm{d}\mathbf{r} \exp[i(\mathbf{p} - \mathbf{p}_f)\mathbf{r}]\, u_{p_f} [\gamma \mathbf{A}^{(-)}_{k_0 s}(\mathbf{r})]^* u_{pi}.$$

Here, E_f and $\mathbf{p}_f$ are the energy and momentum, respectively, of the electron in the final state, and γ_i are the Dirac matrices. The matrix element in (7) is calculated with the wavefield $\mathbf{A}^{(-)}_{k_0 s} = [\mathbf{A}^{(+)}_{-k_0 s}]^*$ corresponding to the diffraction of the plane wave of unitary amplitude which exits the crystal along the direction $(-\mathbf{k}_0)$. This result has been discussed previously by Baryshevskii & Feranchuk (1983) and Stepanov *et al.* (1996) and actually represents the analogue of the reciprocity theorem in optics. As usual, we suppose the summation on the final and averaging over the initial electron states defined by the spinors u_i and u_f. For clarity, we consider the Laue case (transmitted geometry) for the reflected wave in the monocrystalline anode and use the well known expression for the functions $\mathbf{A}^{(+)}_{\mathbf{k}s}(\mathbf{r})$,

$$\mathbf{A}^{(+)}_{\mathbf{k}s}(\mathbf{r}) = \sum_{\mathbf{g}} \mathbf{A}_{gs} \exp[i(\mathbf{k} + \mathbf{g})\mathbf{r}]. \qquad (8)$$

To calculate the PXR intensity from the nonrelativistic electrons, the amplitudes $\mathbf{A}_{gs}$ can be found within the framework of the kinematical diffraction theory (Feranchuk & Ivashin, 1985, 1989; Nitta, 1991). In this case, the Čerenkov condition (5) cannot be fulfilled simultaneously with the Bragg condition (4) and the amplitudes of the wavefields are expressed as (Nitta, 1991)

$$\mathbf{A}_g = (2\pi/\omega)^{1/2} [\chi_g/(k_g^2 - \omega^2)][\mathbf{k}_g(\mathbf{g}\mathbf{e}_s) - \omega^2 \mathbf{e}_s], \qquad (9)$$

where

$$\mathbf{k}_g = \mathbf{k}_0 + \mathbf{g}.$$

The simple matrix element for this particular case was calculated by Freudenberger *et al.* (1995) and by Nitta (1991) (see also Ter-Mikaelian, 1972; Feranchuk & Ivashin, 1989). We use these results to derive the expression for the spectral angular distribution of the photons determined by one of the diffraction amplitudes $\mathbf{A}_g$ in the vicinity of the Bragg peak of the reciprocal-lattice point:

$$\begin{aligned}\frac{\partial^2 N}{\partial\omega\partial\mathbf{n}} &= (\alpha/2\pi)[|\chi_g|^2/(k_g^2-\omega^2)^2]\{\omega^4[v^2-(\mathbf{nv})^2]\\ &\quad -2\omega^2(\mathbf{vk}_g)[\mathbf{vg}-(\mathbf{vn})(\mathbf{gn})]\\ &\quad +(\mathbf{k}_g\mathbf{v})^2[\mathbf{g}^2-(\mathbf{gn})^2]\}\\ &\quad \times L_{\rm abs}[1-\exp(-L/L_{\rm abs})]\delta\{(2mE)^{1/2}\\ &\quad -[2m(E-\omega)-k_{g\perp}^2]^{1/2}-k_{0z}-g_z\}\omega I. \qquad (10)\end{aligned}$$

Equation (10) differs from the analogous result of Freudenberger *et al.* (1995) in the argument of the δ function. In our case, this argument corresponds to the nonrelativistic expression for the electron energy $E=p^2/2m=eU$. Certainly, this formula is consistent with the qualitative estimation (1) because of the tendency $\delta(q_z)\to L_{\rm coh}$ at the maximum of the radiation peak, and the considerable decrease of the transformation coefficient,

$$R(\omega,\mathbf{n},E)\ll 1,$$

is determined by the essential remoteness of the wave vector $\mathbf{k}_0$ from the Ewald sphere.

Let us consider the most important features of the radiation. The radiation frequency can be found from the dispersion equation defined by the argument of the δ function,

$$\omega^2+2m\omega(1-v\cos\Theta)+g^2-2mvg\cos\Theta_B+2\omega(\mathbf{gn})=0. \qquad (11)$$

Here we use the notation in Fig. 1. Taking into account the condition $m\gg g$, the expression for radiation frequency is written as

$$\omega_g\simeq\frac{g}{1-v\cos\Theta}\left[v\cos\Theta_B-\frac{g}{2m}-\frac{gv^2\cos^2\Theta_B}{2m(1-v\cos\Theta)^2}-\frac{(\mathbf{gn})}{m(1-v\cos\Theta)}\right], \qquad (12)$$

where

$$v=(2eU/m)^{1/2}.$$

Because g is a discrete value, the radiation is quasi-monochromatic and the width of each line in the radiation spectrum is defined by the angular aperture of the detector and the dispersion of the electron energies within the beam:

$$\Delta\omega/\omega_g\simeq[(\Delta\Theta)^2+(\Delta v/v)^2]^{1/2}. \qquad (13)$$

To discuss the most important question about the intensity of the radiation, we use the brightness of the X-ray source, a characteristic which is conventionally used in the field of synchrotron radiation (Hart, 1996). Therefore, formula (10) should be integrated over the photon angles in the region of $\Delta\Omega=10^{-6}$ sr and over the frequency in the interval $\Delta\omega/\omega=10^{-3}$ near the value ω_g, calculated from (12). For nonrelativistic electrons, the condition $\omega<g$ is valid and, neglecting the high-order terms in (10), the expression for the number of PXR quanta contributing to one of the diffraction peaks can be found as

$$\frac{\partial N_g}{\partial\Omega}\simeq\frac{\alpha}{2\pi}|\chi_g|^2\omega_g Lv^3\cos^2\Theta_B\sin^2(\Theta-\Theta_B)I, \qquad (14)$$

where I is the number of electrons passing through the crystal per second and we suppose $L\le L_{\rm abs}$.

4. Applications

Formulae (12), (13) and (14) define the principal characteristics of the PXR from X-ray tubes: the frequencies and widths of spectral lines and their intensities. Specific features of PXR allow one to consider several applications of this radiation in laboratory diffraction experiments.

Firstly, PXR from an X-ray tube can be used as a quite intensive source of quasi-monochromatic radiation with tunable frequency. Below, a numerical example is given of a quantitative estimation of this possibility. A germanium crystal of thickness $L=100$ µm is assumed to be attached to the anode of an X-ray tube with voltage $U=50$ kV. For the crystallographic plane (111) in the geometry of Fig. 1 with $\Theta_B=0$ and $\Theta=\pi/2$, the following parameters should be used:

$$|\chi_g|^2=2.177\times10^{-8};\quad v=0,44;\quad g=6.53\,\text{keV};$$
$$\omega_g=2.079\,\text{keV}.$$

Then the PXR intensity can be estimated by

$$\frac{\partial^2 N_{[\rm PXR]}}{\partial\Omega}\simeq 1.45\times10^7 J\,(\text{A})$$
$$[\text{photons s}^{-1}\,\text{mrad}^{-2}\,(0.1\%\ \text{bandwidth})^{-1}]. \qquad (15)$$

The estimation (15) can be compared with the analogous characteristics for synchrotron radiation (Hart, 1996),

$$\frac{\partial^2 N_{[\rm SR]}}{\partial\Omega}\simeq 10^{13}E^2\,(\text{GeV})\times J\,(\text{A})$$
$$[\text{photons s}^{-1}\,\text{mrad}^{-2}\,(0.1\%\ \text{bandwidth})^{-1}]$$

and for the Cu $K\alpha$ characteristic radiation from the conventional X-ray tube (Mirkin, 1961),

$$\frac{\partial^2 N_{[\rm CXR]}}{\partial\Omega}\simeq 10^7-10^8 J\,(\text{A})$$
$$[\text{photons s}^{-1}\,\text{mrad}^{-2}\,(0.1\%\ \text{bandwidth})^{-1}].$$

The *Bremsstrahlung* (BS) under the same conditions causes the background (noise) for the PXR spectra and has the intensity

$$\frac{\partial^2 N_{[\rm BS]}}{\partial\Omega}\simeq 10^5 W\,(\text{kW})$$
$$[\text{photons s}^{-1}\,\text{mrad}^{-2}\,(0.1\%\ \text{bandwidth})^{-1}].$$

Thus, the parameters of the X-ray beam from the standard tube are comparable with the characteristics of the radiation originating from PXR, but an essential advantage of the latter is the possibility of varying smoothly the radiation frequency by means of rotating the anode of the X-ray tube.

Secondly, as is evident from (12) and (14), the PXR resonant frequencies are determined by the lattice parameters and the intensities of spectral lines are directly related to the structure amplitudes. This presents the possibility of solving the inverse problem, *i.e.* analysing the crystal structure on the basis of the PXR spectrum if the investigated sample is attached to the anode.

The proposed method can actually be considered as a means to merge two points, namely where the X-rays are generated by electrons and the region of interaction between the X-rays and the electron density of the probed crystal. This technique allows one to avoid the intermediate path for the transportation of the X-ray beam and decrease the loss of beam intensity. Besides, in many cases, the analysis of the PXR spectrum permits one to decrease the total measurement time, as has been discussed by Feranchuk (1979) and Feranchuk & Ivashin (1989).

Thirdly, an attractive possibility presents itself when the PXR resonant frequency ω_g in (12) coincides with the characteristic frequency ω_0 of the atoms in the anode material. Of course, this requirement sets the values g and Θ_B with quite high precision. But, if the condition is fulfilled, the value $|\chi_g(\omega_0)|^2$ becomes $\eta \simeq 10^3$ times as large. The parameter η is defined by the ratio of the characteristic radiation intensity to the *Bremsstrahlung* background next to the centre of the characteristic line (Mirkin, 1961).

As follows from (14), the PXR spectral intensity should also increase by η times near the frequency ω_0, giving rise to the possibility of creating quite an intense source of quasi-monochromatic X-rays for laboratory experiments.

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