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DIFFRACTION AND SCATTERING OF IONIZING RADIATIONS

Computer Simulation of Images of Photonic Crystals for Hard X Rays in a Transmission Scheme. Near Field[1]

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Abstract—We have developed a method of calculation of transmission of hard x-ray radiation through a perfect and well oriented photonic crystal consisting of closely packed spheres of matter. The method is based on using an approximate solution of paraxial equation for small distances. The recurrent formula is obtained for a transmission of radiation on one period of crystal. A computer program is elaborated for a simulation of images of photonic crystals in a near field, particularly, just behind the crystal. The calculation is performed for silica spheres of 500 nm diameter. It is shown that the standard phase contrast technique is not valid for these objects, because a strong change of intensity takes place inside the volume of crystal due to a scattering of radiation by separate spheres.

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INTRODUCTION

Photonic crystal is a name for natural or artificial materials with a periodic change of electron density under the condition that the value of period is inside the interval from a tenth of a micron to one micron. Natural crystals of such a type are opals. Precious opals consist of silica globules $SiO_2 \cdot nH_2O$, which are ordered in a three-dimensional lattice. Artificial photonic crystals are usually created from silica SiO_2 spheres of necessary size, which are closely packed in three-dimensional lattice. For this reason they are called synthetic opals.

There exist photonic crystals of other types, including both two-dimensional and one dimensional ones. Several different techniques are used for a synthesis of artificial photonic crystals. The most simple and widely used technique is a self-assembly approach for colloid particles on the vertical surface [1]. In this process one tries to realize a situation when all colloid particles become spheres of the same radius and are ordered in closely packed structure.

In closely packing the identical spheres on the plane there arises a triangular (hexagonal) structure in which the centres of three neighbouring spheres create a equilateral triangle with a side length equal to a sphere diameter D. Each sphere creates six such triangles with its all neighbors. In packing a second layer the spheres are placed in a triangle centres but fill up only three from six possible positions. The difference arises in packing a third layer. In it the spheres can occupy the same positions as in the first layer. Such a structure is called *ABABAB*. It is a hexagonal close-packed lattice. In a horizontal plane the spheres fill rows with a period D along a chosen row, and with a period $p = D\cos30° = 0.866D$ normally to the row. The spheres in neighbouring rows are shifted relatively to one another on $D/2$. The period along a vertical is equal to $h = D(8/3)^{1/2}$.

In another variant of packing the spheres of third layer fill the triangle centres which were empty in the second layer. In such a way the third layer is not equivalent to the first layer. This structure is called *ABCABC*. It is a face-centered cubic lattice, in which the horizontal plane is correspondent to a direction 111.

Both structures have practically the same density, and both ones are realized in the synthetic opals, moreover they coexist with one another frequently. Unfortunately, the self-assembly approach for colloid particles create practically always the crystals with a large amount of defects of random types. Therefore the problem of developing the methods of diagnostic of photonic crystals structure is very important. One can find a review of various techniques in [1].

The methods using the x-ray radiation of high intensity from the synchrotron radiation sources of third generation are the most interesting. Particularly, the method of small angle x-ray diffraction is widely used, in which the structure of diffraction spots from a small region of a crystal is experimentally detected on a large distance from it ([2–5] and references therein). The structure of diffraction spots allows one to reveal a symmetry of lattice and possible defects, but it does not give any information about the process itself of radiation scattering in such an object.

[1] The article was translated by the authors.

The alternative method is a technique of direct imaging of photonic crystals with a resolution which allows one to observe images of separate periods of structure locally in each point [6, 7]. It becomes possible after a development of **HRXRM** (high resolution x-ray microscopy) technique [7, 8], based on using the compound refractive lens (**CRL**) [9, 10]. With CRL it is possible not only to resolve an increased distribution of radiation intensity just behind the crystal, but also to obtain a diffraction pattern on a short distance, namely, at the focal length of the lens.

The development of experimental HRXRM technique makes actual the task of theoretical calculation of radiation intensity distribution behind the crystal, i.e. in a near field. There is no reports in literature about a solution of this task. Standard methods of x-ray diffraction in usual crystals are not valid because the crystal has a very large period compared to a wavelength of radiation, and the Bragg condition is met for a large number of reciprocal lattice vectors, but scattering by one sphere is not weak.

A situation is close to the electron transmission microscopy or to the effect of channeling the fast particles in crystal [11]. On the other hand, the task can be solved by the method which is closed to a calculation of the x-ray phase contrast [12]. However, this method has to be modified with the aim to take into account a strong radiation scattering inside the volume of photonic crystal. The present work is devoted to a development of the method of accurate calculation of transmission of hard x-ray radiation through a thick photonic crystal.

FORMULATION OF THE PROBLEM AND A METHOD OF COMPUTING

The synchrotron radiation beams for the sources of third generation have a very small angular divergence, which can be neglected in the task of beam transmission on a small distance about the crystal thickness. When the parallel beam (a plane wave) propagates through the photonic crystal, a good contrast is obtained only for a definite orientation of the crystal. This fact is easy to understand from an analogy with the channeling effect. It is clear that it has to be the orientation with a small period along the beam direction.

Let a coherent monochromatic wave of x-ray radiation is incident on the entrance surface of the photonic crystal which is normal to the beam direction and coincides with the z-axis of Cartesian coordinate system. The amplitude of the electric field of this wave is equal to

$$E(x, y, z) = A(x, y, z)\exp(ikz), \quad k = 2\pi/\lambda. \qquad (1)$$

Here λ is a wave length of radiation. If the wave is plane, then the wave amplitude on the entrance surface ($z = 0$) is constant, let $A(x, y, 0) = 1$. Our task is to calculate a wave amplitude distribution on the exit surface of the crystal, i.e. after a transmission of the radiation beam though the crystal.

For hard x rays a paraxial approximation is met with a high accuracy, therefore instead of solution of Maxwell's equation for the electric field amplitude, it is sufficient to solve the paraxial equation for the wave function $A(x, y, z)$ [13], which can be written in the form

$$\frac{dA}{dz} = -ik\eta\rho(x, y, z)A + \frac{i}{2k}\left(\frac{d^2A}{dx^2} + \frac{d^2A}{dy^2}\right). \qquad (2)$$

Here, for the sake of simplicity it is assumed that the photonic crystal consists of closely packed spheres of chemically homogeneous material, for example, SiO_2. Then a complex parameter $\eta = \delta - i\beta = 1 - n$, where n is a matter complex index of refraction, i.e. with taking into account an absorption. The function $\rho(x, y, z)$ is equal to 1 in the points of matter, i.e. inside the spheres, and it is equal to 0 in the points where matter is absent.

For the arbitrary function $\rho(x, y, z)$ the task remains a very complicated. But for a short period of the crystal along the beam, i.e. in the most interesting for us case, an addition approximation can be made. We will assume that on the distance equal to the period h, the wave function is changed very slowly, and this change can be neglected. Then (2) can be averaged on the period, and the function $k\eta\rho(x, y, z)$ can be replaced by

$$a(x, y) = k\eta s(x, y), \quad s(x, y) = \frac{1}{h}\int_0^h dz'\,\rho(x, y, z'). \qquad (3)$$

In such an approximation the coefficient in the first term of the right-hand part of (2) does not depend of z, and we can eliminate it by a substitution $A = B\exp(-iaz)$. Then we obtain instead of (2)

$$\frac{dB}{dz} = \frac{i}{2k}\left(\frac{d^2B}{dx^2} + \frac{d^2B}{dy^2}\right) + O(z). \qquad (4)$$

The second term in the right-hand part of equation does not written in an explicit form. It contains all terms which are obtained from a differentiation of $\exp(-iaz)$ on the coordinates x and y. Therefore it contains both terms proportional to z and terms proportional to z^2. It is important for us that for a small value of z just linear over z terms are essential in the first turn.

It is evident that in a solution of equation we can arbitrarily choose the coordinate origin of z axis. We will choose the origin each time in such a point z_0, where we know the function $B(x, y, z_0)$, i.e. in (4) the coordinate z should be understanded as distance from the point where a solution is known by us. The next approximation consists in that we can neglect the second term in (4) in integration on a small distance along the z-axis, because it leads to a change of second order of smallness.

In such an approximation we obtain a parabolic equation for the empty space which has a well known solution [13]. It can be written as a convolution over the coordinate x and y for the known solution $B(x, y, 0)$ and the Kirchhoff propagator $P_2(x, y, z) = P(x, z)P(y, z)$, where

$$P(x,z) = \frac{1}{(i\lambda z)^{1/2}} \exp\left(i\pi \frac{x^2}{\lambda z}\right). \quad (5)$$

For the initial wave function $A(x, y, z)$ the equation takes the form

$$A(x,y,z) = \exp(-ia(x,y)z) \times \int dx'dy' P_2(x-x', y-y', z) A(x', y', 0). \quad (6)$$

The physical sense of the obtained solution can be formulated as follows. Let us know the solution (2) in the plane (x, y) inside the object at some point z_0 on the optical axis. By choosing the new coordinate origin in this point we can obtain a solution on the some distance from this point, i.e. in the point $z_0 + z$, by using the equation (6). Therefore (6) has a sense of recurrent relation which can be used many times, and we need to choose the propagation interval z each time as not large one.

We note that (6) describes effectively two processes. First, the wave function is transmitted on the distance z through the empty space. Then it is multiplied by the phase factor which takes into account a phase shift due to a propagation of rays through the matter. This phase shift is the same as it is obtained in the geometrical optics approximation. But (6) is more than the geometric optics because it contains in addition a convolution which is correspondent to a diffraction of radiation in a propagation on the distance z in the empty space.

In a standard phase contrast technique the equation (6) is applied to the total thickness of the object. If in front and behind the object there exist large distances of empty space, then the factor describing a convolution does not play an essential role. It simply adds to the distance in front the object the crystal thickness, but the object itself is assumed to have a zero thickness but with a correct phase shift according to the geometrical optics.

It can be shown that more correct is the approximation in which a half crystal thickness is added to the distance in front the object and the same to the distance behind the object. The phase factor in the phase contrast theory is called a transmission function, and just it describes the object. But if we consider the equation (6) as recurrent, then the object thickness should be divided on the equal parts (layers), and in each layer a multiplication by a phase factor for the layer can be made in the middle of layer.

Then first a propagation for the empty space on half layer thickness is calculated, then many times a multiplication by the phase factor and a propagation on total layer thickness, but at the end of cycle, once again, a propagation on half layer thickness is calculated. In the case of photonic crystal a minimum thickness of layer is correspondent to the crystal period h along the z axis. But one can consider thicker layers. To make the analytical estimation for the accuracy of considered approximation is rather complicated. One can consider some object variants for which an analytical (accurate) solution of equation (2) exists. Such a case was considered by us, but due to its complexity we don't consider it in this paper.

It is important to note that the neglected in (4) terms is proportional not only the distance of propagation z, but also the derivatives on the coordinates x,y from the function $a(x, y)$. And in the case of photonic crystal these derivatives are equal to infinity at the sphere boundaries. Though the derivatives are divergent only on the lines and are large in a narrow region, but just these regions give a main contribution to the image. This is why the problem of accuracy estimation of approximation is a complex task in case of photon crystals. We note that for a validation of geometrical optics approximation the smallness of phase shift derivations over transverse coordinates is also necessary.

SPECIFIC EXAMPLE

For a practical realization of the calculation method described above we consider the photonic crystal consisting of SiO_2 spheres of diameter D and having a structure of type *ABABAB*. The beam direction (z axis) coincides with the axis of hexagonal symmetry. The period of crystal along the z axis is equal to $h = D(8/3)^{1/2}$. Figure 1 shows the function $s(x,y)$ as a black-white linear contrast in limits of calculating region. The black and white colors are used for $s = 0$, and $s = 1$ correspondingly.

For a calculation of the convolution of two functions the rule is used that Fourier image of convolution is equal to a product of Fourier images of the functions. For a calculation of Fourier images we use the Fast Fourier Transformation (**FFT**) procedure with the set of points 2048×1024. So first the Fourier image of

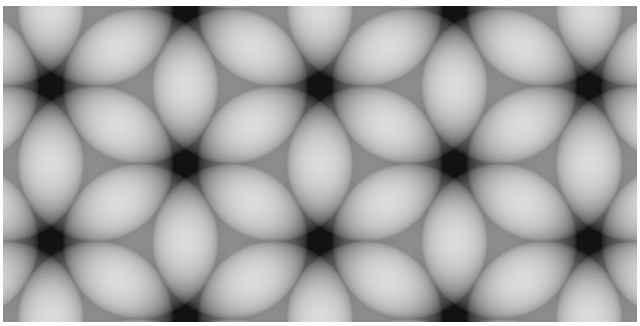

Fig. 1. The function $s(x, y)$ within the calculating area.

complex wave function is calculated on a calculating set of points. Then it is multiplied by the Fourier image of the Kirchhoff propagator which has an analytical form. Then the reverse Fourier transformation is made. The result was obtained on the same calculating area. This area had a size of $2D$ vertically and $4D$ horizontally. The period of structure is equal to D vertically and $1.732D$ horizontally.

A peculiarity of this structure is an existence of three empty region near each sphere, which is not filled in neighbouring layers. As a result the crystal has empty channels on a total thickness, i.e. on arbitrary number of layers. The boundaries of these channels have a hexagonal symmetry with a very sharp change of electron density. Therefore this case is most complicated for a calculation and is most interesting from the physical point of view.

Since the structure is periodic, and a scattering (refraction) of rays occurs on very small angles, it is not necessary to consider a large area containing many periods because the structure can be always periodically multiplied. Moreover, this multiplication is indeed necessary to make on each iteration step. The reason is that in the FFT procedure the result is calculated in such a manner that the wave function is equal to zero outside the calculation area, i.e. similar to using a slit in front of a crystal.

The result shows a diffraction by the slit which is absent in a periodic crystal. Fortunately the FFT procedure on the small distances of propagation describes well a slit diffraction only near the boundaries, and does not disturb almost the wave function in the centre of calculating area. We just use this property. After each iteration (a propagation on the one period along the z-axis) the central period of transverse distribution (the unit cell) is selected from the whole distribution of the complex wave function, and then the rest area is recalculated from a periodic condition.

Such a procedure allows one to avoid an accumulation of wave function distortion due to a slit diffraction. It is of interest that the same procedure can be used in calculation of wave function propagation behind the crystal in empty space, if a wave function is periodical with a small period. If one considers formally infinite transverse sizes of crystal then a periodicity of the wave function is conserved on any distances. We note that in this way it is impossible to obtain the intensity distribution on very large distances behind the crystal because very many iterations are necessary.

On the other hand, in propagating on a small distance we cannot increase the step of set of points. This step has to be several times (ideally tens times) smaller than the first Fresnel zone diameter which is equal to $2(\lambda z)^{1/2}$. Moreover, for spheres of small diameter the transmission function $\exp[-ia(x, y)z]$, and then the wave function $A(x, y)$ change sharply near the boundaries, and a small step is necessary for a correct description of this change.

In this work we are interested only in a near field, i.e. intensity distribution just behind the crystal for various crystal thicknesses which are defined by a number of periods. For very long distances a different calculation scheme has to be used for obtaining the diffraction pattern. This question will be considered in a separate work.

RESULTS OF CALCULATION

The computer program was written with taking into account a possibility to calculate a propagation through both a photonic crystal and empty space. Since the plane wave stays the same in propagation the initial state of the wave function was determined as the transmission function $T(x, y) = \exp[-ia(x, y)z]$, where $z = h$, i.e. for the layer as one crystal period. Then a definite number of iterations was made cyclically. Each iteration consists of two operations: a transmission through empty space on one period and a multiplication by $T(x, y)$.

The result as the complex wave function was written to the file in each iteration. The function can be used for a continuation of calculation and creation of graphics. On the last iteration the multiplication by $T(x, y)$ was not done but a distance can be changed arbitrarily. In the mode of calculation continuation before the cycle the wave function was read from the file and was multiplied by $T(x, y)$. On each iteration the program showed two-dimensional maps of intensity distribution together with the figures of intensity distribution along the central sections (vertical and horizontal).

The most informative were the black-white maps of two-dimensional radiation intensity distribution. To conserve an information these maps were created with a linear gray scale so that the black and white colors were chosen for the minimum and maximum values correspondingly. The minimum I_{min} and maximum I_{max} values are changed monotonously with a number of iterations (number of periods).

Figure 2 shows the calculation results for $\lambda = 1$ Å and $D = 500$ nm as such maps which show the central fragment of size 1024×512 points for 5, 10, 20 and 40 iterations. The number of iterations are shown on the pictures. Initially $I_{min} = I_{max} = 1$. With increasing the number of iterations the intensity becomes stronger in some places and weaker in other places. One can see that increasing takes place on the spheres boundaries, but in the parts with a large amount of material the intensity is decreased. However, a reason of such decreasing is not an absorption but a scattering, i.e. a refraction of rays to the boundary area. The rays can not leave this area due to neighbouring spheres.

At small number of iterations one can see clear interference fringes which are characteristic for an

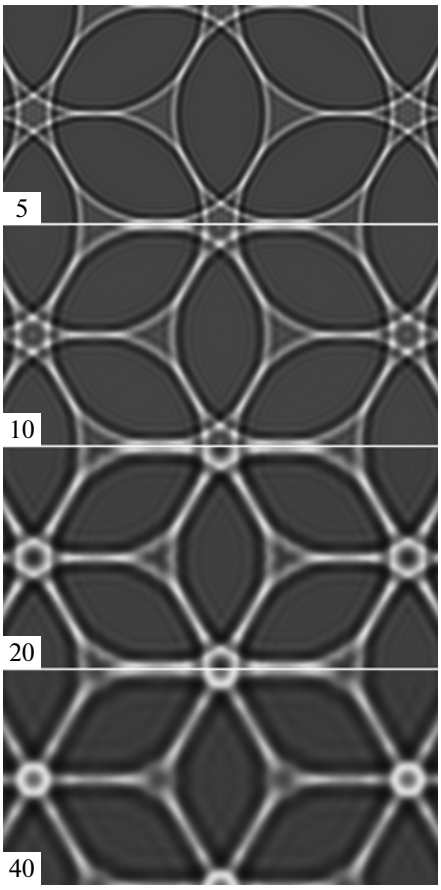

Fig. 2. The maps of radiation intensity distribution for various crystal thicknesses. The thickness is measured by a number of periods which is shown on the panels.

interference of rays refracting by the object and rays going outside the object, but the contrast is weak. With increasing the number of iterations such fringes become smoothed due to multiple scattering, but other fringes arise with a larger period.

Figure 3 shows the curves of I_{min}, I_{mid}, I_{max} (minimum, middle and maximum intensity values) dependence on the number of iterations. The middle value is changed very weakly and this change is completely determined by absorption. On the curves for minimum and maximum values one can see a fracture of almost linear dependences at approximately 25-th iteration. The maximum value increases faster that the minimum decreases. The reason is that the areas of maximum intensity cover a smaller area and are located in the area of empty channels.

The result of calculation shows that the standard phase contrast technique which does not take into account the intensity change on the total object thickness can not be used to the photonic crystal because an essential intensity change takes place for the photonic crystal with a thickness of several periods. It is of interest what happens for larger thicknesses of crystal. This calculation can be made easily, but at present time the photonic crystals with a number of period more than

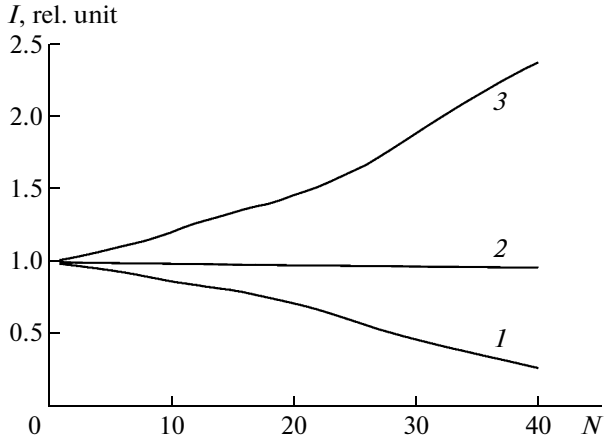

Fig. 3. Dependence of the intensity values I_{min} (*1*), I_{mid} (*2*) and I_{max} (*3*) on the number of iterations *N*.

100 don't exist. On the other hand, the more accurate analysis of error accumulation is necessary. This question is out of scope of our task and it will be considered in the next work.

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