Track membranes and their replicas as high-frequency phase-contrast objects in X-ray optics

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Abstract. Possibility of using polymer track through membranes and membrane replicas as elements of X-ray optics for the visualization of microobjects with high spatial resolution is discussed. It is shown that samples prepared on the basis of track membranes and their replicas can be used in a wide X-ray range, including soft spectral regions (≥ 1 nm) as phase screens or model phase test objects for X-ray microscopy. Highly porous membranes, being diffuse weakly absorbing samples in the form of a single layer or stack of several films, influence coherent properties of the primary X-ray beam. Optical constants of the material of available porous membranes, their thickness, density and size allow one to vary optical characteristics of the phase screens in a wide frequency range, including visible region of spectrum and the X-ray band where many sources of synchrotron radiation work. The issues of wave field concentration by phase structures with narrow through channels, the effect of pore diameter on phase velocity in the channels and spreading (in transverse plane) of the phase pattern for membranes with extremely narrow pores, limiting the resolution of the phase screen used as X-ray test object, is studied in this work in detail. Numerical experiments have been performed by solving parabolic wave equation with tabular values of optical constants for membrane material.

Keywords: X-ray optics, phase-contrast X-ray microscopy, test nanoobjects, track membranes, parabolic equation method in X-ray optics

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1. INTRODUCTION
As it is known, the phase contrast method in optics was proposed by Dutch physicist F. Zernike in 1934 as a replacement of the "shadow method" being used for quality control of astronomical mirrors [2]. As soon as in 1935, he examined the application of the method of microscopic phase contrast imaging of refractive objects [3]. In 1953, F. Zernike received Nobel prize for the development of the method and creation of the first phase contrast microscope. The history of this discovery is presented in his Nobel lecture published, with minor changes, in 1955 in Science magazine [2]. In experimental optics the method of phase contrast relatively quickly became in demand and already before 1953 first reviews and theoretical papers appeared on Zernike's method [4, 5]. In the monograph by M. Franson [6], optical schemes and the principle of the phase contrast microscope designed for studying various phase objects in the visible spectral band (transparent micro-objects, reflecting layers with weakly pronounced surface relief, samples of biological tissues, etc.) are discussed. Methods and optical circuits for the phase-to-amplitude contrast conversion are being considered.

Recently phase contrast method got further development in X-ray microscopy [7]. Study of low-contrast biological objects at the cellular level, problem of testing microelectronic and nanotechnology products has become an impetus for development and improvement of optical schemes working on the principle of X-ray phase contrast microscope. Successes in this branch of experimental physics were achieved in recent years thanks, to a large extent, to the emergence of powerful X-ray sources and progress in focusing X-ray optics. Details are considered in the review [7]. Note that the estimated spatial resolution of X-ray microscopy lies between optical and electron microscopy. However, by the radiation dose imposed on the sample during the session, phase contrast microscopy is the most delicate and nondestructive technique compared with any research mode of the electronic microscopes.

The choice of the contrast type depends on the properties and size of the object itself, its preparation, technical characteristics of the equipment, radiation source, etc. Speaking on studying the internal structure of micro- and nano-objects, the test result at a wavelength is determined by the optical properties of the object – first of all, total refraction index of the sample, in which the X-ray beam propagates [8, 9]:

\[ n(\lambda) = 1 - \delta + i\beta. \]  

Here \( \delta, \beta \) are the optical constants of the material, wavelength dependent. Usually they are small additions to unity in the expression for the refraction index (1). To the present time, dispersion of the refractive index of various substances and materials in a wide X-ray spectral region is well studied, and the quantities \( \delta(\lambda) \) and \( \beta(\lambda) \) can be found with the required accuracy from literary sources or well-known network resources [10]. In a classic X-ray band, at moderate photon energies \( E \leq 10 \text{ keV} \), radiation absorption in the sample is determined mainly by the photoelectric effect by the electrons transition from an atomic shell in the Coulomb field of the nucleus [11] (taking into account the field screening by the internal shells). Total absorption due to the photoelectric effect is described by the value \( \beta \), and transmission coefficient of the sample at the wavelength \( \lambda \), for a parallel beam, is given by the following expression:

\[ T = \exp(-\mu L) = \exp(-4\pi\beta L/\lambda), \]  

where \( \mu \) is the linear absorption coefficient, \( L \) is the thickness of the sample. The real correction \( \delta(\lambda) \) in (1) is positive, it arises as a result of the collective electron response of
each atom of the sample in the X-ray radiation wave field, like in the case of electrons in ionized plasma. Similar to the case of plasma in a high-frequency field, the real part of the refraction index (1) can be written in the form:

\[ 1 - \delta = 1 - \frac{1}{2} \left( \frac{\omega_c}{\omega} \right)^2, \quad (3) \]

where \( \omega = 2\pi c/\lambda \) is the circular frequency, \( c \) is light velocity, \( \omega_c \) – so-called critical frequency, of plasma, coinciding with the circular frequency of the electron gas free vibrations:

\[ \omega_c = \frac{4\pi N_e^2 e^2}{m c^2} \quad (4) \]

(here, \( e \) and \( m \) are the charge and mass of the electron, \( N_e \) is electron density). Note that in contrast to the photoelectric effect all the electrons of the atom in a wide wavelength range (outside the absorption jumps) make approximately the same contribution to the real part of dielectric permittivity of the medium and therefore in the value of \( \delta(\lambda) \), regardless of which shell contains the electrons. Using Eq. (4), the expression for the real part of deviation \( \delta(\lambda) \) in Eq. (1) can be written in another form:

\[ \delta(\lambda) = \left( \frac{e^2}{2\pi mc^2} \right) \lambda^2 \quad (5) \]

or, since \( N_e = ZN_{at} \), where \( Z \) is the atomic nucleus charge, \( N_{at} \) – atomic density, in the more familiar form [8-10] it reads:

\[ \delta(\lambda) = \frac{e^2}{2\pi mc^2} \lambda^2 Z \approx \frac{e^2}{\pi mc^2} \lambda^2 N_{at} f, \quad (6) \]

where \( f \) is the so-called atomic scattering function of the material (its real part), almost equal to the effective atomic number (outside the X-ray absorption jumps); here \( e^2/\pi mc^2 = (2.81E^{-13}) \text{sm} \) – classical radius of the electron [8-10].

2. WHEN AN OBJECT IN X-RAY OPTICS CAN BE CONSIDERED AS PHASE SCREEN?

An ideal X-ray phase contrast object, \( L \) being maximum structural thickness of its elements, when a parallel X-ray beam with a wavelength \( \lambda \) passes through, provides, firstly, a noticeable (of order of unity) phase shift \( 2\pi \delta L/\lambda \) compared with the phase of the wave covering the same distance \( L \) in free space and, secondly, not very large absorption, so the beam attenuation in the sample does not prevent observing the differential phase pattern of the object in the transmission beam, i.e.:

\[ 2\pi \delta L/\lambda \approx 1, \quad 4\pi \mu L/\lambda << 1. \quad (7) \]

In addition, for a given wavelength and experiment geometry, the measured phase shifts should not be too large that is an ideal for observation object must not be too thick-the first of the relations (7). Otherwise, if the reverse condition \( 2\pi \delta L/\lambda >> 1 \) is satisfied, the measurement result will be ambiguous or utterly dependent on the sample tilt and jitter, beam angular characteristics, experiment layout, radiation monochromaticity degree etc. To a first approximation, the object can be considered rather phase than amplitude one when the inequality \( \delta/\beta \geq 1 \) begins to fulfil. If these two optical constants are of the same order the material phase shift over the characteristic damping length \( \lambda/4\pi\beta \) equals 0.5 radian. This limiting length can be considered as a conditional boundary range when the object becomes phase one with decreasing X-ray wavelength.

As an example, in Fig. 1a is depicted the ratio \( \beta/\delta \) spectral dependence for polyethylene terephthalate (PETP) – polymer material of porous track membranes considered in this article (Fig. 2). The intersection of this curve with the dashed line marks the wavelength when, in terms of characteristic amplitude, the attenuation distance of a homogeneous

![Fig. 1](image-url)
polymer layer becomes equal to a quarter-wavelength phase plate.

Consider the table values of the optical constants in X-ray spectral region [10], limiting ourselves to the elements with a small atomic number $Z$. These are Li, Be, B, C, Mg, Al, Si, Sc with atomic numbers from $Z = 3$ to $Z = 21$, frequently used as refractive materials for manufacturing focusing optics, thin films, filters and coatings in soft and hard bands of X-ray spectrum. In a wide wavelength range (0.1-10 nm) thin layers of these elements can be considered as phase objects, except the areas near the edge of absorption. Also, out of photoabsorption jumps, the optical constants of these materials have a power wavelength dependence law, the exponent being constant over the entire aforementioned range. It turns out that the intersection points of the curves $\delta(\lambda)$ and $\beta(\lambda)$ on the wavelength axis (in the vicinity of $K$ absorption jumps) set the boundary values of waves $\lambda_c$, locating on a single curve $\lambda_c(Z)$ for the elements with different $Z$ (Fig. 1b). The shorter is X-ray wavelength compared with $\lambda_c(Z)$, the easier it is to examine the object with this atomic number by the methods of phase contrast. For a given sample composition, it holds approximately: $\beta/\delta \sim (\lambda/\lambda_c)^2 << 1$, that is, for small wavelengths compared with critical $\lambda_c$, the object obviously has phase properties. Note that the equality of real and imaginary additions to unity in the Eq. (1) holds for any $Z$ in some wavelength region near the edge of the absorption band $\lambda_{as}$ [10] (see Fig. 1b), therefore $\lambda/\lambda_{as}(Z) << 1$ ratio is a sufficient condition to consider given object as purely phase, and not amplitude screen. For substances containing different elements in their spectral formulas we should consider effective (averaged over ensemble) atomic number $Z$.

3. TEST OBJECTS IN X-RAY MICROSCOPY

As in the hard X-ray phase contrast microscopy of greatest interest for the researcher are transparent three-dimensional objects with characteristic linear dimensions of the order of 10-0.1 microns [7], high requirements are made to the performance of modern X-ray microscopes. This applies above all to the spatial resolution of the device, which should be at several tens of nanometers level over the entire field of view, with the absence of noticeable chromatic aberration. As “phantoms” at the initial setup and research stages in the imaging X-ray microscopy usually simplest test objects are used, whose geometry and size of elements are known from independent sources or gauge measurements. It can be a thin thread of boron or polypropylene fiber, spherical latex or polystyrene microparticles, perforated substrate – sample holder with micro-holes used in transmission electronic microscopy [14], various kinds of microgrids, fragments of a transmission diffraction grating or Fresnel phase zone plate, chips of various microstructures, etc. These micro-objects are used mainly for demonstration purposes, for qualitative microscope performance assessment. Quantitative measurements, such as determination of the image contrast frequency characteristics, are conducted using more
complex tests. Special metrological structures having elements with different controlled sizes are used, such as so-called Siemens star [15] – high contrast radial microstructure, designed to work in a given X-ray spectral band.

In this paper we propose to use as phase test objects for imaging X-ray microscopy porous track membranes (Fig. 2) [1] or their inorganic thin-film replicas, more persistent objects when working with intense sources of X-ray radiation. Track membranes, as structurally heterogeneous samples, satisfy the above requirements presented to the test objects when working in hard spectral region in phase contrast mode. Here we'll consider samples with round cylindrical channels and axes perpendicular to the plane membrane surface, although there are other types of track membranes (with different axis orientation and more complex pore shape [17]). Pore diameter $D$, varying from 10-20 nm to several microns, depending on the manufacturing conditions, sample can be considered almost constant across the membrane area ($\Delta D/D << 1$), pore walls, to a first approximation, are assumed perfectly smooth. Track membranes with such pore geometry can be considered as reference perforated samples with calibrated through holes randomly distributed over the film area. Pore density $N$ depends on the dose being loaded during the exposing the polymer film to heavy ions and can vary with the top limit of the order of $10^9 - 10^{10}$ cm$^{-2}$ [1]. It is possible to make films with low etched hole density, even with a single micro- or nanohole in a sample with the area of the order of 1 cm$^2$ [18].

It should be noted that, thanks its unique properties, simple and controlled micron pore structure and submicron track membranes attracted the attention of researchers in various related fields of science and technology from the very beginning of this technology development (see reviews [19, 20]). Besides, the opportunity to localize radiation field or particle flux in areas with very small transversal dimensions – of the order of the track membrane pore cross section, has been used in experimental research (coordinate detector resolution, recording element development in contact lithography [21] and photography [22], spatial resolution control methods, astigmatism compensation in raster translucent microscopes [23], etc.). In X-ray optics, several works appeared in which track membranes were used as strong supporting structures for thin X-ray filters or as selective spectral filters having high transparency in ultra-soft X-ray spectrum but effectively blocking longer wavelength ultraviolet, visible and IR background radiation from the observed object (Sun, laboratory plasma source, etc.) [24-27].

Earlier, an attempt was undertaken to carry out X-ray microscopic studies of track membranes with through pores with moderate (low compared with diffraction limit) resolution in the amplitude contrast mode [28]. And finally, in a recently published work V.I. Balykin with colleagues [29] considered the possibility of subwavelength light localization by passing an excited atom through a nanohole in the track membrane. The issue of fixing the phase of X-ray radiation using track membranes, interesting for applications in phase contrast microscopy, to our knowledge, so far was not studied.

When using the simplest test objects such as fine threads, fine meshes or perforated screens with micro-holes, microscope resolution usually is evaluated “by eye” by contrast images of structural elements of the test object or, more precisely, by blurring its edges. In a track membrane with identical cylindrical through pores that are used to test a microscope, as metrological dimensions of the elements can be considered pore diameter $D$, average distance between pores $\bar{D} = 0.5 N^{-1/2}$, determined by the...
pore density $N$, and a variable size of jumpers that appear in the object in case of closely spaced pores or at their intersection. Note that calibrated pore density in a track membrane gives an easy way to estimate magnification of the microscope.

In this paper, we restrict ourselves with consideration of the phase-amplitude evolution of an X-ray plane wave, in submicron spectrum band, passing through a flat layer of a homogeneous material (polyethylene terephthalate) with a single cylindrical through pore. In our simulation, the pore axis was perpendicular to the sample surface, which was considered to be parallel to the incident wave front. The wavelength selected for the calculations corresponds to the characteristic X-ray emission line of copper $\text{CuK}_\alpha$ equal to 0.154 nm (8.047 keV). Since the spectral properties of all materials in this classic X-ray spectral regions are well known [10], the results obtained using this data are easy to recalculate for other wavelengths and materials.

4. WAVE FIELD CALCULATION IN THE FILM WITH A SINGLE PORE

For numerical simulation of X-ray propagation through a single hole in the membrane we use the method of parabolic equation ("parabolic wave equation, PWE" in English literature). Physical meaning of the parabolic equation approximation has been set out in the pioneering work of Leontovich, Fock and Malyuzhinets [30-33]. This powerful computational tool is widely used in diffraction theory for solving problems of underwater acoustics, radio wave propagation, remote sensing, in fiber optics, etc. [34,35]. Basics of the PWE method and examples of its use are included in textbooks and monographs on electrodynamics and physical optics [36-39]. Taking into account geometry of the object and properties of X-ray radiation, our problem can be considered in the scalar approximation [40-42], so we write down the wave equation for the electric field strength in the following form:

$$\frac{\partial^2 E}{\partial z^2} + \Delta_\perp E + k^2 n^2(\vec{r}) E = 0, \quad \Delta_\perp = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}. \quad (8)$$

Without repeating classical derivation, recall the basic assessments. In the case of a homogeneous space: $n(\vec{r}) = \text{const}$, equation (8) has a solution in the form of a plane wave:

$$E(x, z) = Ae^{ikn(x\sin\theta + z\cos\theta)}, \quad \text{propagating at an angle } \theta \text{ to the z axis, } x \text{ being lateral coordinate.}$$

At small angles $\theta < < 1$ (paraxial approximation) the solution can be written as

$$E(x, z) = e^{-ikz}u(x, z), \quad \text{where the wave amplitude } u(x, z) \approx Ae^{-ikz}$$

has a characteristic oscillation period $\Lambda_\perp \approx \frac{2\pi}{kn\theta}$ in the transverse direction $x$ and $\Lambda_\perp \approx \frac{4\pi}{kn\theta}$ along the longitudinal axis $z$. Besides, $\Lambda_\perp \gg \Lambda_\parallel \gg \lambda$, i.e. $n(x, z)$ is a function slowly varying in the propagation direction along the $z$ axis, namely: $\partial u / \partial z \ll \partial u / \partial x$.

By using this ratio and turning to the case of three spatial variables $(x, y, z)$, we obtain from (8) for the wave amplitude an equation of evolutionary type (Leontovich "parabolic" equation [30-32], or "transversal diffusion equation" in Malyuzhinets’ terminology [33]):

$$2ikn \frac{\partial u}{\partial z} + \Delta_\perp E + k^2 n^2 u = 0. \quad (9)$$

In the case of axial symmetry that we are interested in, it is convenient to rewrite it in cylindrical coordinates:

$$2ikn \frac{\partial u}{\partial z} + \frac{\partial^2 u}{\partial r^2} + \frac{1}{r} \frac{\partial u}{\partial r} + k^2 n^2 (r, z) u = 0. \quad (9a)$$

Parabolic equations (9-9a) describe propagation of the package electromagnetic waves with wave vectors directed mainly along the axis $z$. Applicability of the parabolic equation method in a particular field of physics depends on specific conditions: material (optical constant) and the nonuniformity scale of the environment, object geometry and boundary conditions. In X-ray optics, application of
the parabolic equation method for practical problems has been developed in [40-44].

In short-wavelength diffraction and propagation problems, phase of the solution to the wave equation (8): \( \arg E(r,z) = k z + \arg u(r,z) \) is a fast oscillation function of longitudinal variable \( z \), while slowly changing argument of the complex wave amplitude \( \arg u(r,z) \) characterizes its deviation from the phase of a plane wave \( e^{iknz} \).

Computational advantages of the parabolic equation method, noted in classical works of Malyuzhinets and Tappert [33, 34], are due to transition to slowly varying wave amplitude \( u(r,z) \). Instead of a full boundary value problem for the elliptic equation (8), we will solve a Cauchy problem with initial conditions at the entrance to the computation domain. Eliminating the main oscillating factor \( e^{iknz} \) sharply reduces computational costs in the short-wave propagation problems. A useful feature of the method is the ability of equivalent truncation of the computational domain by setting exact transparency conditions at its boundaries, resulting from accurate matching the numerical solution with the exact analytical solution in free space [44]. Finite difference approximation of parabolic equation (9) can be performed using absolutely stable implicit six-point Crank-Nicolson scheme [40,45,46]. This approach provides high accuracy and avoids computational error accumulation with distance.

An elementary estimate of the approximation error, following from the comparison of particular solutions of the exact wave equation \( E(x,z) = Ae^{ik(x \sin \theta)} \) with their “parabolic” approximation \( u(x,z) \approx Ae^{-\frac{a^2}{2}} \), bounds the distance \( z \leq d^2/\lambda^3 \) where the latter gives accurate amplitude and phase values of the approximate solution. Here \( \lambda = 2\pi/k \) is the wavelength of X-ray radiation, \( a \) — characteristic transverse object size or non-uniformity scale of the environment; in our case \( a = D \). This condition, as well as the estimates of the finite-difference scheme steps \( b \ll a, \tau \ll a^2/\lambda \) are not restrictive in the problems of X-ray optics; besides, there are efficient analytical techniques for increasing computational range and angular sector of the parabolic equation [40-42]. Another inaccuracy, due to backscattering neglection and overcome with the method of coupled waves, is not critical in X-ray optics problems due to small variations of relative refraction index.

5. PHASE VARIATIONS NEAR NANOPORE IN A TRACK MEMBRANE

Consider phase distortion of a plane wave passing through an extremely narrow pore in the membrane. Such blurring limits spatial resolution of a screen with nanoholes used as an X-ray phase-contrast test object. Our calculations were performed by numerical solution of the parabolic wave equation [40,41] using tabular values of the optical constants for the membrane material at the copper \( \text{CuK}_\alpha \) wavelength (0.154 nm). For polyethylene terephthalate at this wavelength \( \delta(\lambda) = 4.5 \times 10^{-6} \) and \( \beta(\lambda) = 1.0 \times 10^{-8} \). Corresponding values of absorption length \( L_e \) and quarter-wave thickness phase plate \( L_\phi \) for homogeneous PETP and the selected wavelength are equal to \( L_e = \lambda/4\pi\beta = 1.22 \text{ mm} \) and \( L_\phi = \lambda/4\delta = 8.5 \mu\text{m} \).

Here, we present calculated phase and amplitude of the wave field \( u(r,z) \) of the X-ray radiation passing through a single pore of diameter \( D = 30 \text{ nm} \) in a film with thickness of \( L = 22.5 \mu\text{m} \). For convenience, numerical results are presented in color scale (Fig. 3ab) and contour mode (Fig. 4).

Two-dimensional phase and amplitude wave field spatial distribution produced by the object under study allow qualitative assessment of a membrane with nanoholes as a test object for
X-ray microscope. More detailed quantitative information on the test object and functional possibilities of nano-perforated films is reached by studying linear plots of phase and amplitude distributions for membranes with different pore diameters and variable thickness. In Fig. 5-6, for a selected sample thickness 22.5 μm and a pore diameter of 30 nm, phase distribution and field amplitude along the pore axis and in the transverse direction on the back surface of the sample are presented.

The following Figs. 7-9 demonstrate how the phase deviations from the plane wave front in the vicinity of a single pore vary when reducing its diameter from the maximum (200 nm), to the average (50 nm) and minimum (10 nm). The selected smallest diameter value approximately corresponds to the smallest pore size that can still be etched in a thin track membrane by irradiation of the sample with heavy ions. In these figures, spatial phase distribution near the pore presented in different visualization modes: false colors, radial plot on the back side of the sample, grayscale image and contour map of phase distribution – highlight main features of the wave front distortions from the incident wave one.

Numerical calculations of the wave amplitude and phase shift by passing the membrane with a single hole have been made for samples of three thicknesses $L = 22.5, 10$ and 5 μm and a set of pore diameters $D$ in the range from 5 nm to a few micrometers. As expected, for large pores the wave phase portrait at the output face of the membrane reproduces with good accuracy geometry and size of the pore opening, while for small diameters, starting from some threshold, transversal phase blurring of the wave packet was observed near and beyond the channel.

![Fig. 3. Spatial distribution of wave field phase (a) and amplitude (b) in the vicinity of a single 30 nm pore in 22.5 μm thick PETP film. The scale is compressed along the pore axis by a factor of 103. Boundaries of the sample and the pore channel are shown by thin light line.](image1)

![Fig. 4. Contour representation of the phase shift in a single pore with diameter of 30 nm (compare Fig. 3a).](image2)

![Fig. 5. Axial (a) and radial (b) phase distribution on the perforated membrane back surface. $D = 30$ nm, $L = 22.5$ μm. The input end of the sample is shifted from the left boundary (Fig. 3a) by 1000 nm.](image3)

![Fig. 6. Axial (a) and radial (b) amplitude distribution on the back surface. $D = 30$ nm, $L = 22.5$ μm. The input end of the sample is shifted from the left boundary (Fig. 3a) by 1000 nm.](image4)
Fig. 7. Spatial phase distribution of X-ray beam passing a pore of 200 nm diameter in a PETF sample of 10 μm thickness.

For extremely small pores, the diameter of the phase perturbation zone was more than by an order of magnitude higher than D. To find the functional dependency of the phase blur values on the pore parameters pores of the test sample (i.e., L and D values), turn attention to the plots, where numerical data of the phase blurring on the rear side of membranes with different thicknesses (Figs. 10-11). For the sake of easier analysis of the calculation results in different variable ranges, the data are presented in different scales: in relative (normalized to outlet. For extremely small pores, the diameter of the phase perturbation zone was more than by an order of magnitude higher than D. To find the functional dependency of the phase blur values on the pore parameters pores of the test sample (i.e., L and D values), turn attention to the plots, where numerical data of the phase blurring on the rear side of membranes with different thicknesses (Figs. 10-11). For the sake of easier analysis of the calculation results in different variable ranges, the data are presented in different scales: in relative (normalized to

Fig. 8. Phase distribution near the pore of 50 nm diameter for a sample 10 μm thick.

pore diameter, Figs. 10 a, b) and absolute form (Fig. 11 a).

For each of the presented plots, three regions can be distinguished where the phase blurring near the exit of the channel behaves differently with changing pore diameter. When D value is large – about 0.05 μm and more, the phase pattern of the hole in the transverse direction is equal to this diameter, the transmitted wave front is flat, except small edge effects), and the average phase coincides with the free-space phase of the primary plane wave. It is clearly seen in the color Fig. 7a - the exit aperture looks monochrome and its “color” matches the “color” of the incident wave phase. In this case, radiation impinging on the sample, diffracts only at

Fig. 9. Phase distribution near the pore of 10 nm diameter for a sample 5 μm thick.

Fig. 10. Diameter of the phase jump region at the end of the sample, measured at half maximum of the axial phase plot (Figs. 7-9, b) in linear (a) and logarithmic scales (b), for three different samples of a thickness of 22.5, 10 and 5 μm.
the pore inlet edges and almost all radiation "falls" into the channel, weakly interacting with the pore walls. Another behavior of phase curves, due to propagation in the sample material is observed for small pore diameters ranging from almost zero values to several tens of nanometers. In this case, the size of the phase blur region at the output plane of a given sample thickness is almost constant, independent of the hole diameter \( D \). With changing thickness of the membrane \( L \), the transverse phase blur diameter increases approximately as a square root of \( L \). A transition region between these two modes is observed, when so-called diffraction beam length \( D^2/\lambda \) is approximately equal to the channel length \( L \), i.e., the dimensionless wave parameter \( \lambda L/ D^2 \), the inverse Fresnel number, of the order of unity. In our problem, in normalized variables \( X = \lambda L/ D^2 \) and \( Y = 2\delta L \sqrt{L_0/ L} \), all numerical values of the phase blur on the back side of samples with different thicknesses are described by a single curve (Fig. 11b). In this figure, lines 1 and 2 are drawn — linear interpolation results for large (1) and small (2) pores, outside the transition area. Curve 1 corresponds to the law \( 2\Delta r_i = D \), valid for pores with diameters about 0.1 \( \mu m \) and more, while the horizontal line 2 describes the fact that for small pore diameters phase localization (or blurring) at the back of the sample near the nanohole is independent of \( D \) and grows as \( L \) with the increasing film thickness. Note that for all our samples, the angular size of the region of localization of the phase perturbation is equal by the order of magnitude to \( 2\Delta r_i / L \approx \theta_{cr} = \sqrt{2\delta} = 0.003 \), where \( \theta_{cr} = \sqrt{2\delta} \) is the critical angle of the external total reflection from the nanohole walls for a given wavelength.

Analyzing the results of calculations presented in Fig. 11b and returning to the original problem statement, it can be stated that a track membrane with nanoholes, as a test object for phase-contrast X-ray microscope, satisfies, with a margin, modern spatial requirements for these devices [7], even in near-field mode, when working with relatively large test samples pores, i.e. in the case of large Fresnel numbers. The plots of phase blurring in Figs. 10-11 allow one to choose for the experiment an appropriate track membrane for testing X-ray microscope with necessary spatial resolution.

It should be noted as well that in this problem the main variable determining the solution is Fresnel number of the pore, of course, taking into account frequency dispersion of the sample material. The role of tilting the membrane with a nano-hole from the beam direction is not so important as we consider relatively thin test objects. It is similar to the interference colors of fine soapy or polymer films when the color sequence is weakly dependent on the orientation of the film relative to the light source and observer.

From Fig. 11b and requirement not to be too low-contrast object we can infer that an optimal by spatial resolution phase screen must have through pores whose dimensions satisfy the conditions \( \lambda / D \approx \theta_{cr} = \sqrt{2\delta} \) or \( D / L \approx \theta_{cr} = \sqrt{2\delta} \), then the same order of magnitude, as noted above, there will have
angular wavefront blur equal to \(2\delta r/L\) at the pore exit.

6. PHASE VELOCITY OF WAVES PROPAGATING ALONG THE AXIS OF CYLINDRICAL NANOHOLES

In large pores of a track membrane X-ray radiation propagates with light velocity, like in free space. In homogeneous membrane material, phase velocity is greater: 
\[v_\phi = c/1-\delta \approx (1+\delta)c.\]

With increasing pore diameter \(D\) from several nanometers to the values of about 0.1 \(\mu\)m, phase velocity \(v_\phi(D)\) increases from \(v_{\phi0}(D)\) to the light velocity in vacuum \(c\). Using parabolic wave equations, we can easily determine this relationship: \(v_\phi = v_\phi(D)\) for a selected wavelength. From color plots of the phase deviation near the nanopore, by varying diameter \(D\), we find the length \(l/\pi/2\) where the phase shift versus plane wave in free space equals \(\pi/2\) (Fig. 12a). In other words, we define a quarter-wave thickness on the axis of the perforated plate and determine its dependence on the pore diameter. Based on these results, one can calculate relative wave deceleration along the channel axis with increasing its diameter \(D\):
\[
\Delta V_\phi(D)/\Delta V_{\phi0} = V_\phi(D) - C/(V_{\phi0} - C)
\]
(see Fig. 12b).

From this model problem of X-ray transmission through a material layer with a narrow cylindrical channel, we can conclude that porous track membranes with parallel pores is capable to act as a transparent phase screen providing spatial phase modulation. Fine phase modulation in the sample plane allows one to use track membranes as inhomogeneous phase test objects with deep phase modulation with spatial frequencies up to \(2\cdot10^{-2}-10^{-3}\) \(\text{nm}^{-1}\), which corresponds to the spatial resolution requirements of modern X-ray phase contrast microscopy [7].

7. TRACK MEMBRANE AS X-RAY DIFFUSER – FILTER FOR SPECKLE SUPPRESSION

Slightly absorbing X-ray frosted screen is an optical an analogue of an ordinary thin matte plate widely used in the visible spectrum band (“frosted glass”, “ground glass”) for visualization of the optical image, changing the light field characteristics in lighting systems, in various kinds of light diffusers or as a way to control coherent properties of the incident laser beam. In the X-ray band, there is also demand for the use of matte reflective surfaces, frosted screens and transparencies, not only as test objects but also to reduce the grazing angle specular beam reflection, to change spatial coherence of synchrotron radiation, for suppressing speckles in imaging optics, etc. [47-49]. The aforementioned numerical results on the light wave transmission through a single nanopore film allow one to draw some conclusions on the phase of the wave passing through an ensemble of randomly distributed pores, i.e. for a realistic track membrane, not a single pore model sample.

A track membrane with relatively large pores, when the Fresnel number of the pore cavities is greater than unity \((D^2/\lambda L \geq 1)\), for hard X-rays presents a weakly absorbing transparency modulating the wave phase with a shift \(\Delta\phi = 2\pi\delta L/\lambda \approx 1\) in the pore openings compared to the uniform layer of membrane. The fraction of the sample area where the transmitted wave experiences phase shift...
commensurate with the membrane "nominal porosity" \( P_N = \pi D^2 N / 4 \), where \( N \) is the pore density. For highly porous membranes, taking into account the mutual pore intersection, effective membrane porosity is equal to \( P_{\text{eff}}(P_N) = 1 - e^{-P_N} \) [50]. Therefore, a stack of identical track membranes consisting of \( n(P_N) \geq 1/P_N \) layers laid “back to back” serves as an X-ray diffuser, destructing transversal coherence of the incident beam. In order to not to have interfering radiation transmitted through different pores for large transverse correlation length of the incident beam, one can use highly porous track membranes with tilted pores and wide tilt angle distribution that are usually made for filtering gases or liquids. Such membranes usually have the tilt angles of the pore axes (two- or one-dimensional) within ±30 degrees to the membrane surface. Tilted pores provide reduced role of mutual pore crossings and improved membrane performance in standard filtering problems [1]. With a reasonable number of layers about 5 to 10, a stack of porous membranes with a thickness of 5-10 μm still weakly absorbs the radiation and can be considered as a perfect matte screen for hard X-rays -n the through mode. This type of structural heterogeneous filter is now in demand in experimental X-ray microscopy with powerful 3rd or 4th generation X-ray sources.

According to estimates, track membranes with extremely narrow pores and small Fresnel numbers can play the role of ideal matte phase screens transparent for high spatial frequencies even in single- or double layer options if the pore density and diameter satisfy the requirements \( N \geq 10^{10} \text{ sm}^{-2}, D \leq 50 \text{ nm} \). Note that high-density small-pore samples with wide angular distribution of the axes relative the sample surface are effective and most preferred for the use as a device called X-ray speckle suppressor [51].

8. CONVERSION OF PHASE CONTRAST TO AMPLITUDE

Another interesting feature observed in our model problem of penetrating of a flat X-ray wave through a film with a narrow single pore is the conversion of phase contrast in the amplitude variations in the sample itself and its surroundings. Figs. 3b and 6a,b illustrates this effect. An essential amplification of the field amplitude inside and behind the nanohole near its axis takes place for well-known diffraction effects. First, due to the difference in refractive indices refraction occurs, and a wide channel can capture in the pore a significant part of the input radiation flux. Secondly, due to interference of the wave propagating inside the pore, with a plane wave passing through the material sample, not only phase disturbance occurs but also spatial redistribution of the wave field amplitude. Because of the presence of the nanohole, the plane wave impinging onto the film becomes inhomogeneous in the sample and behind it – phase and amplitude spatial modulation occurs. Quantitatively, the amplitude modulation effect by a nanohole can be described by calculating the transmission coefficient of radiation through the pore, defined as the integral of the squared field amplitude over the pore cross-section, with appropriate normalization by the incident power stream. Note that the wave field has the form \( E(r,z) = e^{ikz} u(r,z) \), therefore this definition is correct for transmitting holes although, generally speaking, a high value of the field amplitude does not always mean an intense radiation flux, as it is, for example, in the case of evanescent waves at the edge of a hole or wave field inside a volume (closed) resonator.

Let us turn to Fig. 13 illustrating numerical results that show the appearance of amplitude
contrast on the sample and the increase of the transmission coefficient in a lavsan film with a diameter in the nanometer range. The points of Curve 1 show how the pore transmittance, determined in a standard way (power ratio past stream to that falling on the input aperture) depends on the pore diameter. For very small (several nanometers in diameter) and large (micron-sized) pores, transmittance does not differ much from one, as could be expected. But for the samples with intermediate values of diameter, the transmittance curve $I/I_0$ has a pronounced maximum at $D \approx 20-30$ nm. In this case, the radiation flux density through the pore, averaged over its section, at the output end of the membrane by an order of magnitude exceeds the flow density in the incident cross section. For large pores with diameters of the order of 1 μm or more, phase conversion in the amplitude contrast on track membrane models is not observed.

The points of Curve 2 in Fig. 13 are plotted by summation radiation flux at the membrane output around the nano-hole over a circle of diameter $4D$ with the same normalization procedure. Because the membrane material itself with such thickness $L$ absorbs almost nothing, the large part of Curve 2 corresponds to a constant value $I/I_0 = 16$. But for small values $D$, an increase of transmission curve with decreasing diameter is observed that shows what area of the entrance surface around the nanohole begins to capture the incident wave due to refraction and diffraction at the entrance edge of the pore. Line 3 in the figure, depicted by squares, shows the maximum local values of the field amplitude squared modulus on the pore axis, depending on its diameter and normalized to the input value.

The appearance of amplitude contrast with uniform illumination of the sample in this task, due to phase modulation of the wave front near the nanohole, accompanied by the radiation flux density spatial redistribution near the pores does not contradict with the law of energy conservation.

This article does not cover wave field evolution (phase and amplitude) when propagating further behind the sample. But on the basis of the above results it can be stated that the appearance of amplitude contrast assures visualization of the phase object with nanopores as a test transparency for X-ray microscope calibration.

It can also be noted that a high X-ray flux density provided by nanopores with allows, in principle, to achieve a simple way of contact X-ray lithography (without preparing special templates) and make replicas of track membranes with deep and extremely narrow pores (tens of nanometers in diameter).

9. INORGANIC TRACK MEMBRANES AND THEIR REPLICAS AS PHASE TEST OBJECTS

One of the weak points of phase objects made of polymer film is their relatively low thermal and radiation durability. Organic polymeric
materials, even those having benzene rings (cyclic polymers) and crosslinked macromolecules with bulk bonds with the formation of a net structure have too low heat and radiation resistance to serve as material for power optics and function in intensive beams of X-ray (especially, synchrotron) radiation. However, with moderate fluxes of ionizing radiation PETP films have long been used in X-ray experiments, mainly as filter material, substrates or windows of radiation detectors. Simple estimates show which limiting intensity of the incident beam and what absorbed radiation dose allow working with a test object made of a PETP polymer film. With monochromatic illumination at a wavelength of 0.154 nm, the admissible X-ray flux density by uniform illumination of a 22.5 μm thick test sample lies within 10 to 20 W/sm². This assessment takes into account that a film of this thickness only absorbs about 2% of the incident radiation, and heat loss of the sample occurs with large efficiency in the near infrared spectral band from both sides of the film [52]. Due to radiolysis of PETP under an ionizing beam with such a limiting for this polymer flux density [53], the "radiation" of the considered test object is about 1 minute (excluding temperature effects). Really, in the image registration schemes of X-ray microscopes with CCD coordinate receivers, the flux density is less than this limit values by orders of magnitude. Therefore, prolonged operation of the proposed in this article polymer test object is possible without significant deterioration of its performance. Anyway, such a test object from PETP film, due to more efficient heat transfer, is not so vulnerable under X-ray beam as most biological microstructures under study [54].

In conclusion, we make the following brief comment. Thin film porous objects with topology typical for track membranes are manufactured by track technology (including nano-porous objects) not only from organic polymers, but also basing on inorganic substrates (glasses, mica, oxide films, etc. [55]), more suitable for experiments with intense X-ray beams, than polymer objects. It can be also deep replicas, including thermo- and radiation resistant metal or ceramic structures obtained by using polymer track membranes as source matrices (templates) with narrow cylindrical or conical pores [56,57]. Strictly speaking, the term "replica" of the track membrane is now applied in a broader sense than was stated in technical dictionaries: "An exact copy of the sample surface on which the structure of the material is clearly expressed".

Track replica manufacturing method appeared almost simultaneously with the birth of track technology [54]. Metallic or carbon thin-film replicas of the surface of the samples with the tracks of fast heavy ions in a solid massive sample or polymer film previously were intended mainly for density measuring density of the tracks and their lateral dimensions with the use of electron microscope. From some time, the researchers learned to make volumetric replicas of the tracks etched not only on the surface, but throughout the depth of the sample irradiated with ions [55,56]. At present, various secondary structures based on deep replicas of track membranes ("template method") received wide spread in various technical and applications as substantive functional elements in nanotechnology [1,57].

Replicas of track membranes, as phase objects of X-ray optics, can be of three types. The first type is a thin-film surface replica (no bulk structures), providing phase variations much less than unity (low contrast phase test objects capable to model some properties of biological micro-objects by X-ray exposure).
Of the second kind are deep (volumetric) replicas reproducing to some degree the structure of the original track membrane template (positive replica) [58,59]. And finally, the third kind reversed (negative) copies in which hollow pores of the track membrane are replaced by a substance for example, metal [55,56] or dielectric (cyacrine glue allowing to realize one of the simplest ways to make bulk replicas from track membranes [60, 61]). Strictly speaking, negative replicas are geometrically more similar to biological microstructures than track hollow membranes themselves with micro- and nanopores. For replicas of this type, X-ray propagation through a nano-column or nano-tip essentially differs from the radiation transport through a hollow pore of the same size, due to the differences in the refractive indices of the material structures. This fact is illustrated in Figs. 14 and 15 versus the results presented in Figs. 3-6. We can compare the amplitude and phase of the diffracted light at a wavelength of 0.154 nm for two objects of the same shape. In the former case (Figs. 3-6) it was a single hollow cylindrical pore with diameter of 30 nm in a thick PETP film. The latter case presents a similar but reverse (negative) structure: PETP nano-rod with the diameter of 30 nm, length of 22.5 μm and the same orientation relative to the incident wave (Figs. 14-15). The hollow pore serves as an X-ray concentrator amplifying by several times wave field amplitude at the output rod end and providing a noticeable phase shift, of the order of 0.4 radian. In the case of polymer nano-rod (Figs. 3-6), radiation is not being concentrated, but noticeably scattered into free space out of the rod, and at the exit end of the element we see a considerable (almost five times) decrease in amplitude. In the spatial phase pattern, only a weak and localized in the nano-rod cross section phase modulation is seen (by fractions of radian). For the same reason, a biological or other micro-object wrapped in a thin film looks more contrast in phase-contrast X-ray microscopy, provided the real part of the film refractive index is greater than the index of micro-object under study.

Therefore, volumetric positive replicas of track membranes, as test objects for X-ray microscopy, have significant advantages over with similar negative samples. Besides, secondary replication of a negative replica allows one to produce another positive copy – analogue of the primary track membrane, but with a matrix made of a better material suitable for operations with intensive X-ray beams.
contrast test sample for an X-ray microscope working in a hard region of the spectrum. By numerical solution of the parabolic wave equation, phase shift and amplitude distribution of X-ray radiation penetrating polymer material in the sample and free space near a nano-pore. It is shown that transverse phase blurring at the output end of the membrane can be approximately described by a universal function of the dimensionless Fresnel number defined for a single pore. The issue of phase-to-amplitude conversion in the membrane material and pore cross sections at is studied for various hole diameters. The obtained results on the phase shift magnitudes and their spatial localization near the pore channels allow us to propose track membranes not only as test objects but also as X-ray diffuser or speckle suppressor in imaging optical systems with coherent or partially coherent light sources illuminating micro-objects under study.

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