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ADIABATIC SOLUTION IN COHERENT X-RAY DIFFRACTION AND OPTICS

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Abstract

By the example of experimental data on X-ray diffuse scattering from rough multilayer grating we show the presence of irreversible decoherence of quantum states of X-ray photons in the process of dynamical diffraction. This effect proves out as violation of the reciprocity principle in experimental data and is caused by the inevitable presence of large-scale structural imperfections of the sample. On the one hand, these imperfections lead to domination of the diffuse-scattering channel. On the other hand, the total integrated scattering keeps being invariable as compared with the case of perfect sample structure according to the Rayleigh adiabatic solution.

Recent development of high-brilliant synchrotron radiation (SR) sources of X-rays allows one to apply new experimental techniques for the purposes of studies of matter. In particular, since emittance of SR sources is small, it is possible to obtain spatially coherent beams and to perform coherent diffraction experiments [1]. Significant scientific progress in X-ray intensity fluctuation spectroscopy [2, 3] as well as in X-ray coherent diffraction and holography [4] was made in the last decade. Further development in these applications is restricted to some degree by the lack of suitable X-ray optics. Presently used optical devices remarkably worsen spatial coherence of incident X-ray beam [5, 6]. We show by the example of X-ray diffraction from a multilayer grating (MG) that this effect is caused by the presence of large-scale (tens of microns) smooth imperfections of structure. Although these imperfections do not result in decrease of integrated diffraction intensity, their presence causes irreversible decoherence of quantum states of X-ray photons. In diffraction experiments, this effect can be observed as a violation of the reciprocity principle [7, 8].

Already Rayleigh examining reflection of sound at a corrugated surface has attracted attention to the following adiabatic feature of the wave equation [9]: if surface curvature is sufficiently low, in spite of the value of surface deviation from ideal plane, the integrated reflection coefficient remains unchanged and corresponds well to reflection from ideal plane surface. This effect is true not only in the case of X-ray reflection from rough surface [9], but in the case of X-ray diffraction from a wide class of objects, provided that the adiabatic condition for deviations from an ideal structure is met.

Let us consider this effect by the example of X-ray diffraction from an MG [10–13]. The principal scheme of an ideal MG is shown in Fig. 1(a). The real structure of the MG is schematically depicted in Fig. 1(b). It is evident that the MG, as depicted in this Figure, can not provide regular X-ray diffraction.

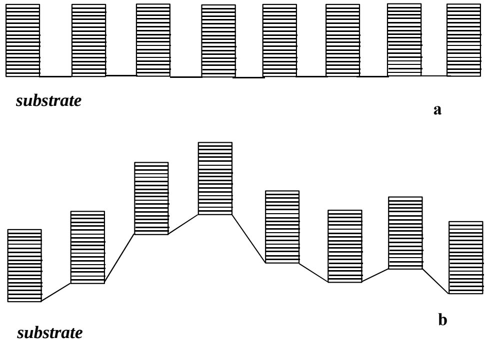

Fig. 1. Schematic representation of MG: (a) ideal MG; (b) real MG.

However, taking into account the manifold compression of the genuine horizontal scale in this Figure, one sees clearly that such an MG can reflect X-rays without loss of integrated scattering as compared with the perfect MG. Indeed, let the scattering potential of the perfect MG can be described by some function $V_0(\boldsymbol{r})$. Let the solution $E_0(\boldsymbol{r})$ of the wave equation

$$(\Delta + k^2)E_0(\boldsymbol{r}) = V_0(\boldsymbol{r})E_0(\boldsymbol{r}), \qquad (1)$$

be known, where Δ is the Laplacian and $k = 2\pi/\lambda$ is the wavevector. Let the large-scale smooth imperfections can be described by some function $\Delta z(x, y)$. The z axis is normal to the multilayer lateral planes; the axes x and y are directed parallel to these planes (Fig. 1). Then, the real scattering potential has the following form:

$$V(x, y, z) = V_0(x, y, z + \Delta z(x, y)).$$

Further, replacing the variables $x \to \tilde{x}$, $y \to \tilde{y}$, $z + \Delta z(x, y) \to \tilde{z}$ and using the smoothness condition, i.e. neglecting the derivatives $\partial V_0/\partial x$ and $\partial V_0/\partial y$, it is easy to obtain that wave equation (1) remains valid in these new variables. Thus, appearance of adiabatic phase shifts in the diffracted wave that is all that is caused by the large-scale smooth imperfections. At the same time, the total integrated scattering keeps being invariable. Moreover, the X-ray diffraction pattern preserves its general form, slightly spreading in the diffraction space.

Note that the modern technology does not prevent the large-scale imperfections discussed. Indeed, since the X-ray wavelength is very short, the problem of coherent X-ray optics manufacture is essentially more difficult as compared with the visible spectrum, for example. On the other hand, existence of large-scale imperfections can be revealed only if spatial coherence of incident beam is quite high [7]. Nevertheless, if the last condition is met, there are no reasons to expect that influence of large-scale imperfections can be considered as perturbation [9].

The perturbation theory is frequently used for calculation of X-ray diffraction from rough objects, and the "statistical" part of scattering potential is considered as the disturbance [14]. It may be noted that even if structural imperfections can be considered as perturbation it is not evident that Maxwell's equations have a stable solution [15]. Especially, it is not expected in the case discussed, when the statistical part of scattering potential dominates and structural imperfections can not be considered as perturbation. Moreover, it would be reasonable to assert that such solution simply does not exist in this case. The conception of scattering potential arises as a result of reduction of the many-particle task of scattering to the one-particle task. The "statistical nature" of potential means that this procedure can not be performed correctly. Firstly, the system "X-ray photon + scattering atoms" can not be considered as an isolated one. Secondly, this system gets an infinite number of additional degrees of freedom. Such system is known to behave as dissipative ones even when energy of the system is conserved [16]. The decoherence (entropy increasing), accompanied by depopulation of the non-diagonal cells of density matrix, occurs in such systems. It means that during the diffraction, states of an X-ray photon with different momentum directions become incoherent. Thus, in order to describe further diffraction it is necessary to add probabilities instead amplitudes.

The routine method of description of opened quantum systems represents the introduction of the complex non-Hermitian Hamiltonian [16]. In this method, incoherent diffuse scattering can be considered as an additional reaction channel. At the same time, coherent diffraction can be described with the usual single-channel scattering theory, which introduces imaginary corrections to the scattering potential. That allows one to take into account dissipation of the coherent field energy through the incoherent diffuse scattering channel. Note that this approach directly introduces breakdown of T invariance (reciprocity principle), since the reaction channel "diffuse scattering $\rightarrow$ coherent diffraction" is postulated to be impossible and, consequently, no time reversal process is possible. The breakdown of T invariance can also be illustrated by the following argumentation. The incident X-ray field can be described with some density matrix, ρ_0. In accordance with the general principles of statistical quantum physics, the initial quantum state of the incident X-ray field can be described with some value of entropy [17]:

$$S_0 = -k_{\mathrm{B}} \sum_n \rho_{0,n} \ln \rho_{0,n} = -k_{\mathrm{B}} \operatorname{Tr}(\rho_0 \ln \rho_0) ,$$

where k_{B} is the Boltzmann constant and $\rho_{0,n}$ is the occupation number of the state $|n\rangle$ of the Hamiltonian $H_0 = -\Delta + V_0$. The entropy is evident to increase during the scattering, $S_1 > S_0$. This implies violation of T invariance, otherwise the time reversal process must cause entropy decreasing, which is impossible.

From the point of view of the basic channel of coherent diffraction, the introduction of imaginary corrections to the scattering potential causes violation of the reciprocity theorem, too. Even in case, when it is possible to ignore the dynamical nature of diffraction, T invariance is violated if energy dissipation of coherent X-ray wave depends on its direction of propagation [8]. Therefore, the T invariance violation must be especially evident for the angles of propagation, for which the dissipation discussed is high. In the case of a multilayer and MG, these angles of propagation are Bragg angles. Indeed, behavior of macro-roughness is completely conformal [18], which causes resonant increase of the diffuse scattering cross-section at Bragg angles.

The MG studied was prepared from a W/Si X-ray multilayer mirror by the holographic lithography technique with ion-beam etching [13, 19]: the grating period was about 0.8 μm and the multilayer period was about 3 nm. X-ray diffraction measurements were performed using SR from the VEPP-3 storage ring at the wavelength λ=0.154 nm. A channel-cut Si(111) crystal was used as the monochromator ($\Delta\lambda/\lambda \approx 2\times10^{-4}$) and a single-crystal Ge(111) was used as the secondary crystal collimator. The measurements were performed in the vertical plane so that the crystal monochromator, MG and the secondary crystal collimator were placed in the (+,+,+) geometry. The vertical component of the spatial coherence was estimated to have a value of about 5 μm. Characterizing the sample by X-ray diffraction, we have found that the measured integrated reflectivities of the diffraction orders (~ 35, 51 and 30% for −1, 0 and +1 orders, respectively) correspond well (~80–90%) to the theory [20], which assumes the perfect structure of the MG (Fig. 1(a)). High quality of the sample can be illustrated by the fact that intermixing of the different diffraction orders was smaller than 0.1%.

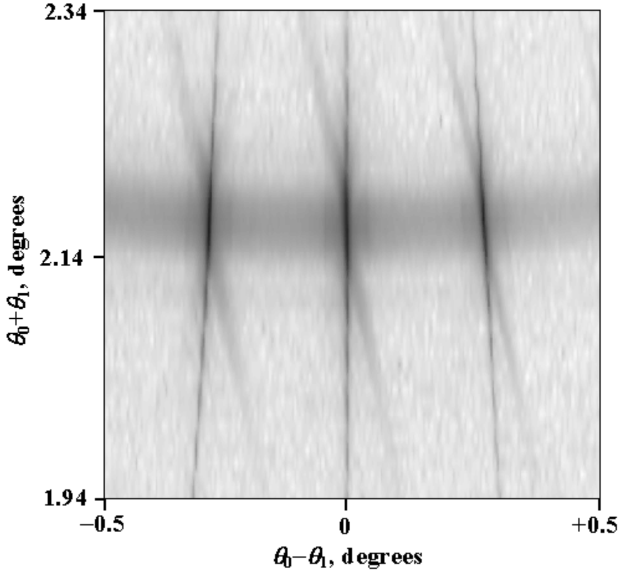

Fig. 2. The diffraction map of 0 and ±1 orders of reflections from the MG studied. The intensity is shown on the logarithmic scale. The dynamical range of measurements was about 10^5. $\theta_0 - \theta_1$ ($\propto q_x$) (θ_0 and θ_1 are the incoming and outgoing angles relative to the multilayer planes, respectively, $\boldsymbol{q}$ is the momentum transfer) is plotted parallel to the horizontal axis. The total diffraction angle, $\theta_0 + \theta_1$ ($\propto q_z$), is plotted parallel to the vertical axis.

In order to study diffuse scattering from a sample we have performed measurements, varying the incoming and outgoing diffraction angles. The obtained diffraction map of 0 and ±1 diffraction orders is shown in Fig. 2. This map was obtained as a set of transverse scans (ω-scans, $\theta_0 + \theta_1 = \text{const}$, where θ_0 and θ_1 are the incoming and outgoing angles relative to the multilayer planes). The vertical streak in the center of the map corresponds to specular and quasi-specular scattering, which are attributed with the 0-order of diffraction from the MG. The slightly inclined vertical streaks on the flanks of the map represent the same scattering attributed to the ±1 diffraction orders. The horizontal halo in the map represents the well-known quasi-Bragg diffuse scattering caused by conformal behaviour of roughness [18, 21]. Absence in the map of the intensity symmetry relative to the specular 0-order reflection (the vertical streak in the center) indicates violation of the reciprocity theorem. This asymmetry reveals itself with the existence of streaks across the Bragg points inclined to the same side. These features arise at incoming angles that correspond to the maximum reflectivity from the MG. We have observed the same features in the case of multilayer mirrors [7, 22]. The mechanism of their appearance is provided by dynamical diffraction [23] and can be explained in the framework of the conventional theory [24, 25]. Nevertheless, in accordance with the reciprocity principle, they must arise symmetrically relative to exchange of the incoming and outgoing angles, which was not observed. Note that this effect can not be explained by roughness anisotropy in the lateral directions, because sample rotation around the z axis through 180° does not cause any essential changes in the map. Finally, angular spread of the features discussed allows us to conclude that their appearance is provided by large-scale imperfections (≥ a few microns).

Thus, the experimental data obtained demonstrate an evident breakdown of the reciprocity principle. This effect indicates the high decay rate of the coherent X-ray field through the diffuse scattering channel, which dominates over the coherent diffraction.

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