

Dielectric permittivity in the x-ray region

A. V. Kolpakov, V. A. Bushuev, and R. N. Kuz'min

M. V. Lomonosov Moscow State University

Usp. Fiz. Nauk 126, 479–513 (November 1978)

The article reviews the current state of the study of the dielectric properties of a medium in the x-ray and γ -ray frequency regions. Particular attention is devoted to the systematic introduction of the dielectric permittivity tensor in the x-ray region, starting from the most fundamental property of crystals—spatial periodicity. The properties of the permittivity tensor are treated both in terms of the phenomenological theory and on the basis of specific microscopic models. The review includes theoretical and experimental investigations of the contributions of the electronic and nuclear subsystems to the permittivity of crystals. The polarizability of the electron subsystem is considered in the strong-coupling approximation and in terms of the Bloch formalism. Considerable space is devoted to discussion of the permittivity near isolated low-lying nuclear transitions in the presence of hyperfine interactions, where its properties are substantially more complicated and more interesting than those of the permittivity of the electron subsystem. A review is given of the principal phenomena of the polarization optics of Mössbauer radiation. The review analyzes the symmetry properties of the x-ray permittivity and establishes a number of useful relations regarding generalized symmetry operations.

PACS numbers: 77.20. + y

CONTENTS

| | |
|--|-----|
| 1. Introduction | 959 |
| 2. General properties of the permittivity of a crystal, not dependent on the model of the medium | 960 |
| a) Maxwell's equations and the material equation for a crystalline medium | 960 |
| b) Symmetry properties of the permittivity tensor | 961 |
| 3. Elementary classical introduction to the theory of dielectric permittivity and the principal optical phenomena in the x-ray and γ -ray regions | 962 |
| a) Polarizability of a medium consisting of classical oscillators. Electron and nuclear parts of the polarizability | 962 |
| b) Principal effects in x-ray optics | 963 |
| c) Principal phenomena in γ -ray optics | 964 |
| d) Polarizability of three-dimensionally periodic media | 966 |
| 4. Quantum-mechanical theory of x-ray and γ -ray dielectric permittivity | 966 |
| a) Basic equations of the theory | 966 |
| b) X-ray polarizability of the electron subsystem | 968 |
| c) The potential part of the electron contribution to crystal polarizability. Structure factor and atomic structure factor | 968 |
| d) Dispersion contribution to the electronic polarizability | 969 |
| e) Effect of inelastic scattering on polarizability | 970 |
| f) Nuclear polarizability of a crystal | 971 |
| 5. Application of the Bloch formalism to calculation of the electronic polarizability of crystals | 972 |
| a) Bloch representation of the wave functions of the electron subsystem | 972 |
| b) Current density in the representation of Bloch functions | 972 |
| c) Strong-coupling approximation | 973 |
| 6. Conclusion | 974 |
| References | 976 |

“The very existence of Bragg reflections of x rays shows that the generally accepted description of a medium—without inclusion of the spatial periodicity of the dielectric permittivity—is inapplicable.”

M. Lax

1. INTRODUCTION

The review is devoted to discussion of the current state of investigations of dielectric properties of a medium in the x-ray and γ -ray frequency regions. The discussion is carried out in terms of the unified formalism of the dielectric permittivity tensor (DPT, or depending on the context, DP). The review discusses

the problem of introducing the DPT $\epsilon_{ij}(\mathbf{k}, \mathbf{k}'; \omega)$ for crystalline media and describes its basic properties—symmetry, relation to the refractive index and scattering amplitude, representations in direct and reciprocal space, etc.

In the x-ray region the DPT was introduced in the 1930's by Laue,¹ Kohler,² and Molière³ in connection

with development of the dynamic theory of diffraction. Ewald showed in his early studies that the optical properties of spatially periodic media are in many respects quite unusual and require discussion in terms of the dielectric formalism. The periodic structure of crystals is responsible for the basic importance of spatial dispersion in x-ray optics; this appears at various levels, since it owes its origin both to the spatial location of the atoms and nuclei composing the crystal and to their individual and collective properties.

In recent years there has been a distinct trend toward intensive study of the optics of the x-ray and γ -ray regions, in both the theoretical and experimental areas. This trend has been reflected, in particular, in the construction of x-ray interferometers⁴ and resonators,⁵ attempts at more careful study of the coherent properties of radiation in this region,⁶ in searches for means of production of x-ray and γ -ray lasers,⁷ and the investigation of nonlinear effects.⁸ Investigators who previously worked in the longer-wavelength regions of the electromagnetic spectrum are now actively interested in x-ray and γ -ray optics.

Among the reasons for the increased interest in the optics of the x-ray region we shall mention two principal factors. First this interest is a development of studies in nuclear γ -ray resonance⁹ and Mössbauer radiation optics near the frequencies of nuclear γ transitions. Crystal optics in the vicinity of nuclear resonance frequencies presents significant interest, since in this case the crystal is a medium with a sharply expressed spatial and frequency dispersion and anisotropy. The results of a large number of experimental and theoretical studies (presented in terms of the amplitude or cross section for scattering) in nuclear γ -ray optics have been summarized in the reviews of Hannon and Trammell,^{10,11} Hannon, Carron, and Trammell,¹² and in the review of Belyakov,¹³ which is devoted to diffraction of nuclear γ rays in crystals containing nuclei of a Mössbauer isotope. The book by Baryshevskii¹⁴ discusses in the language of the refractive index optical phenomena arising in interaction of γ rays (and also neutrons and electrons) with crystals whose anisotropy is due to the polarization of nuclear moments. Second, in recent years, in addition to traditional x-ray tubes, Mössbauer isotopes and synchrotrons have begun to be used as radiation sources. X-ray synchrotron radiation already at the present time has substantially higher spectral and integral intensities than high-power x-ray tubes.¹⁵

The distinctive features of the dielectric properties of a medium in the x-ray region follow from the following simple circumstances: a) the wavelength of the radiation is $\leq a$, where a is the crystal lattice constant; b) $\lambda \leq r$, where r is the radius of the atom; however, already for elements with atom number $Z \geq 25$ we have the inequality $\lambda \gg a_0$, where $a_0 = \hbar^2/m_e^2 Z$ is the radius of the first Bohr orbit of the atom; c) usually there is the relation $\omega \sim \omega_i$ ($Z \geq 25$), where ω_i is one of the eigenfrequencies of the deepest atomic K or L levels; however, another relation $\omega \gg \omega_i^{\text{max}}$ is often used; d) all of the higher-lying electronic levels of the atom are oc-

cupied (except perhaps states near the edge of the continuum), so that transitions to these levels are forbidden as a result of the Pauli principle; e) if the crystal contains Mössbauer nuclei, then the nuclear subsystem at frequencies of γ transitions also contributes to the dielectric properties.

2. GENERAL PROPERTIES OF THE PERMITTIVITY OF A CRYSTAL, NOT DEPENDENT ON THE MODEL OF THE MEDIUM

The principal problem of x-ray optics is the study of the propagation and diffraction of plane monochromatic (normal) waves in crystals. The propagation of such waves is described by the Maxwell equations and the material equation establishing the relation between the perturbation and the response to it in terms of the DPT. In this section we consider the most general properties of the DPT, those which do not depend on a specific model of the crystal.

a) Maxwell's equations and the material equation for a crystalline medium

The macroscopic equations of the electromagnetic field in a nonmagnetic medium in the absence of extraneous currents and charges have the form¹⁶

$$\begin{cases} \text{rot } \mathbf{H} = \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}, & \text{rot } \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{H}}{\partial t}, \\ \text{div } \mathbf{D} = 0, & \text{div } \mathbf{H} = 0. \end{cases} \quad (2.1)$$

These equations are obtained from the exact macroscopic Maxwell equations by averaging the corresponding quantities over the ensemble. Averaging over physically infinitely small volume elements, which is ordinarily done in the visible region, is inapplicable in the x-ray region, since λ/a is of the order of unity.

The most general form of linear relation between the displacement $\mathbf{D}(\mathbf{r}, t)$ and the electric field $\mathbf{E}(\mathbf{r}', t')$ satisfying the principle of causality is the following integral equation¹⁷:

$$\mathbf{D}(\mathbf{r}, t) = \mathbf{E} + 4\pi\mathbf{P} = \int_{-\infty}^t dt' \int d\mathbf{r}' \hat{\epsilon}(\mathbf{r}, \mathbf{r}'; t, t') \mathbf{E}(\mathbf{r}', t'), \quad (2.2)$$

where \mathbf{P} is the polarization. All of the quantities entering into Eqs. (2.1) and (2.2) are real and are statistically averaged. At the same time they can vary with space and with time arbitrarily rapidly.

For stationary media the kernel of the integral operator (2.2) depends only on the difference $t - t'$. Then according to Eq. (2.2) the following connection exists between the Fourier components of the field and the induction:

$$D_i(\mathbf{k}, \omega) = \int d\mathbf{k}' \epsilon_{ij}(\mathbf{k}, \mathbf{k}'; \omega) E_j(\mathbf{k}', \omega), \quad (2.3)$$

where

$$\epsilon_{ij}(\mathbf{k}, \mathbf{k}'; \omega) = (2\pi)^{-3} \int d\mathbf{r} d\mathbf{r}' \epsilon_{ij}(\mathbf{r}, \mathbf{r}'; \omega) e^{-i\mathbf{k}\mathbf{r} + i\mathbf{k}'\mathbf{r}'}, \quad (2.4a)$$

$$\epsilon_{ij}(\mathbf{r}, \mathbf{r}'; \omega) = \int_0^\infty d\tau \epsilon_{ij}(\mathbf{r}, \mathbf{r}'; \tau) e^{i\omega\tau}. \quad (2.4b)$$

From the condition that the kernel $\hat{\epsilon}$ in Eq. (2.2) is real, it follows that

$$\epsilon_{ij}(\mathbf{k}, \mathbf{k}'; \omega)^* = \epsilon_{ij}(-\mathbf{k}^*, -\mathbf{k}'^*; -\omega^*). \quad (2.5)$$

Since the properties of a crystal are unchanged by a displacement by an arbitrary lattice vector \mathbf{R} , the kernel (2.4b) can be identically represented¹⁷ in the form of a sum over reciprocal-lattice vectors,

$$\varepsilon_{ij}(\mathbf{r}, \mathbf{r}'; \omega) = \varepsilon_{ij}(\mathbf{r} + \mathbf{R}, \mathbf{r}' + \mathbf{R}; \omega) = \sum_{\mathbf{H}} \varepsilon_{ij}^{(\mathbf{H})}(\mathbf{r} - \mathbf{r}'; \omega) e^{-i\mathbf{H}\mathbf{r}}, \quad (2.6)$$

where $\varepsilon_{ij}^{(\mathbf{H})}$ is the H component of the DPT. From Eqs. (2.4) and (2.6) it follows that

$$\varepsilon_{ij}(\mathbf{k}, \mathbf{k}'; \omega) = \sum_{\mathbf{H}} \varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) \delta(\mathbf{k}' - \mathbf{k} - \mathbf{H}), \quad (2.7a)$$

where

$$\varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) = \int d\rho \rho \varepsilon_{ij}^{(\mathbf{H})}(\rho, \omega) e^{-i\mathbf{k}\rho}, \quad (2.7b)$$

and therefore

$$D_i(\mathbf{k}, \omega) = \sum_{\mathbf{H}} \varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) E_j(\mathbf{k} + \mathbf{H}, \omega). \quad (2.8)$$

Equation (2.8) establishes a nonlocal relation in wave-vector space between the electric displacement with wave vector \mathbf{k} and the Fourier components of the field with all possible wave vectors $\mathbf{k}_{\mathbf{H}} \equiv \mathbf{k} + \mathbf{H}$, which is a consequence of the spatial periodicity of the crystal.

In the optical region Eq. (2.8) reduces to the simpler form $D_i(\mathbf{k}, \omega) = \varepsilon_{ij}(\mathbf{k}, \omega) E_j(\mathbf{k}, \omega)$, since in the long-wavelength approximation ($a \ll \lambda$) the crystal is spatially uniform.¹⁷

The presence in the displacement (2.8) of terms with $\mathbf{H} \neq 0$ leads to the diffraction phenomenon, which is of central importance for x-ray optics, consisting of the appearance of scattered waves with propagation directions differing from the primary direction \mathbf{k} by the reciprocal-lattice vectors \mathbf{H} . Under certain conditions, several terms $\mathbf{E}(\mathbf{k} + \mathbf{H}, \omega)$ in the sum (2.8) can be simultaneously of the same order of magnitude as the primary wave $\mathbf{E}(\mathbf{k}, \omega)$.

Substitution of the material equation (2.8) into Eq. (2.1) leads to a system of equations for the amplitudes of the normal waves in the crystal with allowance for anisotropy and spatial and time dispersion:

$$[n_{\mathbf{k}}^2 \hat{\pi}_{\mathbf{k}} - \hat{\varepsilon}^{(0)}(\mathbf{k}, \omega)] \mathbf{E}(\mathbf{k}, \omega) - \sum_{\mathbf{H} \neq 0} \hat{\varepsilon}^{(\mathbf{H})}(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k} + \mathbf{H}, \omega) = 0, \quad (2.9)$$

where $n_{\mathbf{k}}^2 \equiv c k_{\mathbf{H}} / \omega$, $\hat{\pi}_{\mathbf{k}} \equiv 1 - (\mathbf{k}_{\mathbf{H}} \mathbf{k}_{\mathbf{H}} / k_{\mathbf{H}}^2)$ is the operator of projection on the plane perpendicular to $\mathbf{k}_{\mathbf{H}}$.

Solution of the system of equations (2.9) with application of boundary conditions comprises the main problem of the dynamic theory of x-ray diffraction.¹⁸⁻²⁰

The case in which the amplitudes of the scattered waves $\mathbf{E}(\mathbf{k} + \mathbf{H}, \omega)$ in Eq. (2.8) are small in comparison with the amplitude of the primary wave $\mathbf{E}(\mathbf{k}, \omega)$ is called the kinematic approximation of diffraction.¹⁸ This approximation, as follows from solution of the Maxwell equations, is valid if the condition $\pi |\varepsilon^{(\mathbf{H})}| l / a \ll 1$, is satisfied, where l is the dimension of the crystal in the direction of propagation of the primary wave.

b) Symmetry properties of the permittivity tensor

On the basis of very general considerations we can derive a number of useful symmetry relations for the DPT.

The condition of periodicity (2.6) together with Eq. (2.5) leads to the relation $\varepsilon_{ij}^{(\mathbf{H})*}(\mathbf{k}, \omega) = \varepsilon_{ij}^{(-\mathbf{H})}(-\mathbf{k}^*, -\omega)$. If the crystal is centrally symmetric, i.e., if $\varepsilon_{ij}(\mathbf{r}, \mathbf{r}'; \omega) = \varepsilon_{ij}(-\mathbf{r}, -\mathbf{r}'; \omega)$, then it follows from this and from the relations (2.7) that

$$\varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) = \varepsilon_{ji}^{(-\mathbf{H})}(-\mathbf{k}, \omega). \quad (2.10)$$

The symmetry properties of the DP tensor with respect to permutations of the indices i and j follow from the fluctuation-dissipation theorem for distributed systems,²¹

$$\varepsilon_{ij}(\mathbf{r}, \mathbf{r}'; \omega) = \varepsilon_{ji}(\mathbf{r}', \mathbf{r}; \omega).$$

Thus, on interchange of the coordinates \mathbf{r} and \mathbf{r}' it is necessary simultaneously to interchange the indices i and j . Comparison of the last relation and (2.4) and (2.7) leads to an important symmetry property in wave-vector space:

$$\varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) = \varepsilon_{ji}^{(-\mathbf{H})}(-\mathbf{k} - \mathbf{H}, \omega). \quad (2.11)$$

For a centrally symmetric crystal it follows from (2.10) and (2.11) that

$$\varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) = \varepsilon_{ji}^{(-\mathbf{H})}(\mathbf{k} + \mathbf{H}, \omega). \quad (2.12)$$

If the relation (2.12) is satisfied, the crystal is nongyrotropic; however this does not prevent a crystal which is not centrally symmetric from being nongyrotropic. Equation (2.12) can be considered as a generalization of the ordinary definition of nongyrotropicity¹⁷ to the case of spatially periodic media ($\mathbf{H} \neq 0$). In an isotropic nongyrotropic medium Eq. (2.12) leads, in particular, to Friedel's law,¹⁸ well known in x-ray diffraction.

Let us consider some general symmetry relations for non-absorbing crystals. According to the fluctuation-dissipation theorem a medium is nonabsorbing if $\varepsilon_{ij}(\mathbf{r}, \mathbf{r}'; \omega) = \varepsilon_{ij}^*(\mathbf{r}', \mathbf{r}, \omega)$ (Ref. 21), i.e., if

$$\varepsilon_{ij}(\mathbf{k}, \mathbf{k}'; \omega) = \varepsilon_{ji}(-\mathbf{k}', -\mathbf{k}; -\omega); \quad (2.13)$$

here, \mathbf{k} , \mathbf{k}' , and ω are assumed real.

Then, as follows from (2.7) and (2.13), the following symmetry relation exists:¹⁹

$$\varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) = \varepsilon_{ji}^{(-\mathbf{H})}(-\mathbf{k} - \mathbf{H}; -\omega) = \varepsilon_{ji}^{(-\mathbf{H})*}(\mathbf{k} + \mathbf{H}, \omega). \quad (2.14)$$

Combination of Eqs. (2.11) and (2.14) leads to new symmetry properties:

$$\varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega) = \varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, -\omega) = \varepsilon_{ij}^{(-\mathbf{H})}(-\mathbf{k}, \omega). \quad (2.15)$$

From Eq. (2.14) we obtain Friedel's law for a non-absorbing medium. In a nongyrotropic nonabsorbing crystal the tensor $\varepsilon_{ij}^{(\mathbf{H})}(\mathbf{k}, \omega)$ according to Eqs. (2.10) and (2.15) is real.

Further symmetry relations can be obtained by breaking down the DPT $\varepsilon_{ij}^{(\mathbf{H})} = i\varepsilon_{ij,r}^{(\mathbf{H})} + i\varepsilon_{ij,i}^{(\mathbf{H})}$ into a real part (r) and an imaginary part (i), and also into two Hermitian tensors

¹⁹The realtion (2.14) for $\mathbf{H} = 0$ signifies the hermiticity of the tensor $\hat{\varepsilon}^{(0)}(\mathbf{k}, \omega)$. It can be considered as a more general definition of hermiticity for the tensor $\hat{\varepsilon}^{(\mathbf{H})}(\mathbf{k}, \omega)$.

$$\epsilon_{ij}^{(H)}(\mathbf{k}, \omega) = \epsilon_{ij}^{(H)(1)}(\mathbf{k}, \omega) + i\epsilon_{ij}^{(H)(2)}(\mathbf{k}, \omega), \quad (2.16a)$$

each of which in turn can be broken down into real and imaginary parts:

$$\epsilon_{ij}^{(H)(l)} = \epsilon_{ij,r}^{(H)(l)} + i\epsilon_{ij,i}^{(H)(l)} \quad (l=1, 2). \quad (2.16b)$$

A consequence of the hermiticity of the tensors $\epsilon_{ij}^{(H)(1)}$ and $\epsilon_{ij}^{(H)(2)}$ are the following symmetry relations:

$$\begin{aligned} \epsilon_{ij,r}^{(H)(1)}(\mathbf{k}, \omega) &= \epsilon_{ji,r}^{(H)(1)}(\mathbf{k} + \mathbf{H}, \omega), \\ \epsilon_{ij,i}^{(H)(1)}(\mathbf{k}, \omega) &= -\epsilon_{ji,i}^{(H)(1)}(\mathbf{k} + \mathbf{H}, \omega). \end{aligned} \quad (2.16c)$$

If the medium is nongyrotropic, i. e., if Eq. (2.12) is satisfied, then it follows from (2.16) that the Hermitian tensors in (2.16b) are real:

$$\begin{aligned} \epsilon_{ij,r}^{(H)(1)} = \epsilon_{ji,r}^{(H)(1)} = \epsilon_{ij,r}^{(H)}, \quad \epsilon_{ij,i}^{(H)(1)} = 0, \\ \epsilon_{ij,i}^{(H)(2)} = \epsilon_{ji,i}^{(H)(2)} = \epsilon_{ij,i}^{(H)}, \quad \epsilon_{ij,r}^{(H)(2)} = 0. \end{aligned}$$

In a nonabsorbing medium according to Eqs. (2.14) and (2.16) the second Hermitian tensor in (2.16a) vanishes but the tensor $\epsilon_{ij}^{(H)}$ in the general case remains complex, which is determined by the location of the atoms in the unit cell and by the existence of spatial dispersion. If in addition the crystal is nongyrotropic we again conclude that the tensor $\epsilon_{ij}^{(H)}(\mathbf{k}, \omega)$ is real.

The relations discussed above reflect the symmetry of the DPT with respect to permutation of the tensor indices and operations of spatial inversion. The DPT also has symmetry properties which follow from the symmetry of the crystal space. The DPT in direct space is proportional to the charge density $\rho(\mathbf{r})$ and consequently its symmetry is described by the 230 Fedorov space symmetry groups Φ . If at the location of an atom there is an arbitrary set of fields or interactions $\{V\}$, the symmetry of such a lattice turns out to be substantially nonclassical and is described by the generalized symmetry groups $G^{(p)}$, $G^{(q)}$, or $G^{(w)}$. The most general construction in the theory of groups of generalized symmetry is represented by the wreath product groups $G^{(w)}$, which are constructed as follows:²² $G^{(w)} = (P^{\otimes 1} \otimes \dots \otimes P^{\otimes n}) \otimes G$ where g_i are the transformation operators of geometrical space and the operators p_i of the group P describe local operations of change of a physical property (for example, rotation of a magnetic field vector or change of the angle between the magnetic field vector and the axes of the electric field gradient tensor), \otimes is a direct product, and \circledast is a semi-direct product.

In reciprocal space the wreath product groups $G^{(w)}$ in the general case correspond to the groups Φ . From consideration of the symmetry properties of reciprocal space it is also possible to obtain relations of the type (2.12) and (2.14). To the symmetry groups $G^{(p)}$, $G^{(q)}$, and $G^{(w)}$ of direct space, there correspond in reciprocal space the groups of twofold generalized symmetry $G^{(w_f w_m)} = [(P_f P_m)^{\otimes 1} \otimes \dots \otimes (P_f P_m)^{\otimes n}] \circledast G$, where P_f is the operation of change of the phase of the structure amplitude and P_m is the operation of change of its modulus.²³ In this case the following relation²³ is valid for the DPT:

$$\epsilon_{ij}^{(H)}(g^{(p1)}\mathbf{k}; g^{(p2)}\{V\}) = p_{p,m}(\hat{g}H_1, \{V\}) \epsilon_{ij}^{(H)}(\mathbf{k}; \{V\}),$$

according to which for a centrally symmetric crystal $\epsilon_{ij}^{(H)}(-\mathbf{k}; \{V\}) = \epsilon_{ij}^{(H)}(\mathbf{k}; \{-V\})$, and for a non-centrally-

symmetric crystal

$$\epsilon_{ij}^{(H)}(-\mathbf{k}; \{V\}) = \epsilon_{ij}^{(H)*}(\mathbf{k}; \{-V\}).$$

These relations are a generalization of the symmetry properties (2.11), (2.12), (2.14), and (2.15) to the case when an arbitrary set of fields $\{V\}$ is present.

We note, finally, that in reciprocal space there are statistical symmetry relations between the Fourier components of the DPT with various \mathbf{H} which are the consequence of statistical relations between the structure factors $F(\mathbf{H})$.²⁴

In the kernel of the integral operators in Eq. (2.2) it is convenient to separate the unit operator \hat{I} . Then the relation between the polarization $\mathbf{P}(\mathbf{r}, t)$ and the field $\mathbf{E}(\mathbf{r}', t')$ is established by means of the polarizability tensor²⁵ $\hat{\chi} = (\hat{\epsilon} - \hat{I})/4\pi$. According to Eqs. (2.4) and (2.7) the \mathbf{H} components of the DP tensors and of the polarizability are related as follows:

$$\epsilon_{ij}^{(H)}(\mathbf{k}, \omega) = \delta_{ij} \delta_{H0} + 4\pi \chi_{ij}^{(H)}(\mathbf{k}, \omega). \quad (2.17)$$

In view of the analyticity and boundedness of $\mathbf{E}(\mathbf{k}, \omega)$ and $\mathbf{D}(\mathbf{k}, \omega)$ the matrix $\hat{\chi}^{(H)}(\mathbf{k}, \omega)$ (2.17) as a function of the complex variable ω is also analytic and bounded. As a consequence of this, the real and imaginary parts of $\hat{\chi}^{(H)}$ satisfy the Kramers-Kronig relations²⁵

$$\text{Re}[e^{i(k'-k)R} \hat{\chi}^{(H)}(\mathbf{k}, \omega)] = \frac{2}{\pi} \int \frac{\omega' d\omega'}{\omega'^2 - \omega^2} \text{Im}[e^{i(k'-k)R} \hat{\chi}^{(H)}(\mathbf{k}, \omega')], \quad (2.18a)$$

which are valid for scattering at an arbitrary angle by a scatterer of radius R . In scattering at zero angle ($\mathbf{k}' = \mathbf{k}$), Eq. (2.18a) takes the form well known in the optics of the visible region

$$\text{Re} \hat{\chi}^{(0)}(\mathbf{k}, \omega) = \frac{2}{\pi} \int \frac{\omega' d\omega'}{\omega'^2 - \omega^2} \text{Im} \hat{\chi}^{(0)}(\mathbf{k}, \omega'). \quad (2.18b)$$

Equation (2.18b) is of particular interest in combination with the optical theorem,²⁶ which relates the imaginary part of $\hat{\chi}^{(0)}(\mathbf{k}, \omega)$ to the total absorption cross section $N\sigma_{\text{tot}}(\mathbf{k}, \omega)$ of the medium

$$\text{Im} \hat{\chi}^{(0)}(\mathbf{k}, \omega) = \frac{N\sigma_{\text{tot}}(\mathbf{k}, \omega)}{4\pi k}.$$

In the case of diffraction the argument of the exponential in (2.18a) is a multiple of 2π , since $\mathbf{k}' - \mathbf{k} = \mathbf{H}$ and R is an integral vector of the reciprocal lattice, so that we again arrive at a relation of the type (2.18b) but this time for the case of diffraction scattering, i. e., for $\hat{\chi}^{(H)}(\mathbf{k}, \omega)$.

3. ELEMENTARY CLASSICAL INTRODUCTION TO THE THEORY OF DIELECTRIC PERMITTIVITY AND THE PRINCIPAL OPTICAL PHENOMENA IN THE X-RAY AND γ -RAY REGIONS

a) Polarizability of a medium consisting of classical oscillators. Electron and nuclear parts of the polarizability

Let a medium consist of continuously distributed

²⁵Sometimes in the literature on the theory of x-ray scattering, the term polarizability is used to denote the quantity $4\pi\chi$ (see the book by Shpinel'¹⁸) or $4\pi\chi/\epsilon$ (see for example the books by James¹⁸ and Pinsker²⁰). The latter definition is applicable only in the case of isotropic media.

classical oscillators with eigenfrequencies ω_i and small damping constants γ_i . These oscillators may correspond to atomic electrons and (or) Mössbauer nuclei. The polarization of the medium is $P_i(\omega) = \chi_{ij}(\omega)E_j(\omega)$, where the tensor $\chi_{ij}(\omega)$ is the sum over all eigenfrequencies³⁾

$$\chi_{ij}(\omega) = \frac{Ne^2}{m} \sum_i \frac{g_{ij}(\omega_i)}{\omega_i^2 - \omega^2 - i\omega\gamma_i}; \quad (3.1)$$

here m is the mass of the electron (or proton), N is the charge density, and g_{ij} is the oscillator strength. Equation (3.1) is conveniently represented in a somewhat different form, separating a term which we shall subsequently call the potential or principal part of the polarizability:

$$\chi_{ij}(\omega) = -\frac{Ne^2}{m\omega^2} \delta_{ij} + \frac{Ne^2}{m\omega^2} \sum_i \frac{\omega_i^2 g_{ij}(\omega_i)}{\omega_i^2 - \omega^2 - i\omega\gamma_i}. \quad (3.2)$$

The second term in Eq. (3.2), the dispersion term, has an appreciable value only when $\omega \approx \omega_i$. If the frequency of the external action ω significantly exceeds the maximum frequency ω_i^{max} , then the contribution of the second term to the tensor χ_{ij} can be neglected. Here the DP takes the form $\epsilon(\omega) = 1 - (\omega_p^2/\omega^2)$, where $\omega_p = \sqrt{4\pi Ne^2/m}$ is the so-called plasma frequency, and becomes a scalar. Since in the x-ray region $\omega \gg \omega_p$, we have a permittivity $\epsilon < 1$.

An estimate of the value of the potential term in Eq. (3.2), for example, for Co⁵⁷ γ rays ($\lambda = 0.86 \text{ \AA}$) and a medium consisting of iron atoms ($N = N_{\text{at}}Z$, where $N_{\text{at}} = 8.47 \times 10^{22} \text{ cm}^{-3}$ and $Z = 26$) gives a value $\chi = -1.2 \times 10^{-6} < 0$. Therefore the phase velocity of x rays in the medium $c_p \approx c(1 - 2\pi\chi)$ is greater than the velocity of light in vacuum, and the group velocity $c_g \approx c(1 + 2\pi\chi)$ is less than c . Consequently in this frequency region the medium is optically less dense than a vacuum.

As we have noted above, the nuclear subsystem contributes to the polarizability only at frequencies near Mössbauer resonances. The principal feature of Mössbauer resonances is that their damping constants γ are extraordinarily small. Therefore the location of the spectral line of a nuclear γ transition on the frequency scale, its shape, and its other characteristics turn out to be very sensitive to conditions external to the nucleus. The external and internal crystalline magnetic fields and electric field gradients lead to Zeeman and quadrupole splittings of the nuclear levels,⁹ and the magnitude of the splitting can significantly exceed the natural line width γ . In this case it is necessary in Eq. (3.2) to sum over all hyperfine components l .

In the absence of splitting for $\omega \gg \omega_{l_{e1}}^{\text{max}}$ in the vicinity of a frequency ω_0 of a nuclear transition the polarizability of a medium is made up of two terms:

$$\chi(\omega) = \chi_{e1}(\omega) + \chi_{\text{nuc}}(\omega) = -N_{\text{at}}Zr_0\lambda^2 + (4\pi)^{-1}N_{\text{nuc}}c\sigma_0\gamma(\omega_0^2 - \omega^2 - i\omega\gamma)^{-1}, \quad (3.3)$$

where

³⁾For simplicity in Subsections a), b), and c) of this Section instead of $\chi_{ij}^{(0)}$ we shall write simply χ_{ij} .

$$\sigma_0 = \frac{\lambda^2}{2\pi} \frac{2I_2 + 1}{2I_1 + 1} \frac{f}{1 + \alpha}$$

is the maximum cross section for nuclear resonance absorption; I_1 and I_2 are the spins of the ground state and excited state of the nucleus; f is the recoil-free emission factor⁹; α is the internal conversion coefficient; $\lambda = \lambda/2\pi$; $r_0 = e^2/mc^2$.

The maximum value of the ratio $|\chi_{e1}/\text{Re}\chi_{\text{nuc}}|$ for the isotope Fe⁵⁷ is close to ten. Therefore in the frequency region $\omega < \omega_0$, in contrast to the usual situation, $\chi > 0$, the phase velocity is less than the velocity of light in vacuum. This leads in particular to the possibility of the Vavilov-Čerenkov effect in the x-ray frequency region.²⁷

The electron DP is a scalar and does not possess a spatial dispersion, if we do not consider small corrections near the eigenfrequencies ω_i , which can be appreciable only for heavy elements. The nuclear part of the DP, on the other hand, leads to an optics with spatial dispersion and in the presence of hyperfine splittings is a tensor.

b) Principal effects in x-ray optics

The most characteristic features of x-ray optics appear under conditions of diffraction. Since discussion of diffraction phenomena is beyond the scope of this review, we refer the reader to books¹⁸⁻²⁰ and to Belyakov's review¹³ on this subject. A detailed exposition of the early experiments on investigation of the refraction and reflection of x rays, which were historically the first experiments in x-ray optics, can be found in the well known books by James¹⁸ and Compton and Allison.²⁸ Study of the phenomenon of refraction can be carried out on the basis of the deviation from the Bragg law. Such measurements require great care as a result of the small value of the electronic polarizability χ , since the change in the grazing angle ψ in the transition of a wave from one medium to another is extremely insignificant: $\Delta\psi = 2\pi\chi \text{ctg}\psi$ is of the order of $10^{-6} - 10^{-4}$ radians.

Another method of investigating the refractive index is based on study of the phenomenon of total external reflection. This method, in contrast to the preceding one, must necessarily take into account absorption if $\omega \approx \omega_i$. Absorption smears out the limit at which total reflection appears and it turns out to be difficult to find the correct value of the critical angle ψ_c . Another feature of total external reflection is that the reflection coefficient

$$R = \left| \frac{1 - \sqrt{1 - \tau}}{1 + \sqrt{1 - \tau}} \right|^2, \quad (3.4)$$

where $\tau = 4\pi|\chi|/\psi^2$, is the same with accuracy to terms of the order $|\chi|^2$ for polarization parallel (π) and perpendicular (σ) to the plane of incidence. Therefore the reflected radiation does not possess a preferential polarization. A typical value of the critical angle is $\psi_c = (N\gamma_0/\pi)^{1/2}/\lambda \sim 10^{-3} - 10^{-4}$ radians.

In addition to direct measurement of the refractive index, one can turn to determination of the frequency

dependence of the absorption coefficient $\mu(\omega) = 4\pi(\omega/c)\chi''(\omega)$ of the medium, which is the basic problem of x-ray spectroscopy.²⁹ The total polarizability $\chi(\omega)$ of the medium can be established from spectroscopic data on absorption with the aid of the Kramers-Kronig relation (2.18b). The imaginary part χ'' of the polarizability receives contributions from various mechanisms reflecting the electron structure of solids, which manifests itself in the form of fine structure of the absorption edge.³⁰

In view of the fact that in the x-ray region of frequencies the electronic polarizability $\chi(\omega)$ is practically a scalar, and the spatial dispersion parameter (see Section 5) is small, the possibilities of x-ray optics without taking into account the periodic structure of the crystalline medium are limited by the factors discussed above. The optics of this region become considerably more interesting if we include in the discussion the nuclear subsystem of the crystal.

c) Principal phenomena in γ -ray optics

An important feature of the interaction of resonance γ rays with nuclei in the presence of hyperfine splitting is a polarization dependence of the absorption cross section. A study of the polarization dependences of the nuclear Zeeman effect in Fe⁵⁷ nuclei was carried out by Frauenfelder and his colleagues.³¹ However, Blume and Kistner³² first called attention to the fact that in such problems it is necessary to consider in more detail the propagation of the radiation over the thickness of the medium. In fact, a part of the radiation which is absorbed in the surface layer can then with a certain probability be again emitted in the forward direction. In the general case the re-emitted radiation has a polarization differing from that of the primary beam. As a result, as the combined wave propagates in the medium a change occurs in its polarization characteristics and in its interaction with the nuclei in each successive layer.⁴⁾

The real part of the polarizability tensor describes nuclear double refraction effects, the Faraday effect, the Cotton-Mouton effect, etc., while the imaginary part is responsible for the selective absorption of definite polarizations. Comparing the real and imaginary parts of $\chi(\mathbf{k}, \omega)$ (3.3), we can see that selective absorption is important near the exact resonance, but double-refraction effects appear most clearly at frequencies somewhat shifted from ω_0 .

For an analysis of nuclear effects of the rotation of the plane of polarization it is convenient to go over to the eigenwaves of a resonance medium, which in the general case are not orthogonal.³² The problem of finding the eigenwaves is closely related to the prob-

⁴⁾In the work cited³² and in a number of subsequent investigations by other workers the refraction coefficient matrix was introduced as a characteristic of the medium. It has dimensionality 2×2 and describes the change in the polarization of the wave in the plane perpendicular to the direction of propagation.

lem of diagonalization of the DPT matrix. This tensor in the general case is non-hermitian and its real and imaginary parts cannot simultaneously be reduced to diagonal form. However, it is sometimes possible to use symmetric representations in order to indicate conditions under which the DPT matrix is diagonalized. Housley, Grant, and Gonser³³ showed that diagonalization is possible when the radiation is propagated in certain high-symmetry directions in the crystal. Here, if a magnetically ordered crystal is considered, it is necessary to take into account also its magnetic symmetry.³⁴

The polarization vectors of the eigenwaves of the medium $\mathbf{e}_\mu(\mathbf{k}, \omega)$, from which it is possible to determine the polarization characteristics of the transmitted wave, are determined from solution of the eigenvalue problem¹¹ $\|\hat{\chi}\|\hat{\mathbf{e}} = \lambda\hat{\mathbf{e}}$, where $\hat{\mathbf{e}}$ is a column matrix of two elements. Solution of this equation gives the following eigenvalues and polarizations:

$$\lambda_\mu = \frac{1}{2}(\chi_{xx} + \chi_{yy}) \pm \sqrt{\frac{1}{4}(\chi_{xx} - \chi_{yy})^2 + \chi_{xy}\chi_{yx}} \quad (\mu = 1, 2), \quad (3.5)$$

$$\mathbf{e}_\mu(\mathbf{k}, \omega) = \alpha_\mu \left[\mathbf{e}_x + \frac{\mathbf{e}_y(\lambda_\mu - \chi_{xx})}{\chi_{xy}} \right],$$

$$1/\alpha_\mu = \sqrt{1 + \left| \frac{\lambda_\mu - \chi_{xx}}{\chi_{xy}} \right|^2}.$$

The explicit form of the polarization eigenvectors is somewhat arbitrary, since $\det \|\hat{\chi} - \lambda\| = 0$. It follows from Eq. (3.5) that the difference in the polarizabilities for eigenwaves is

$$\Delta\chi_{1,2} = \chi_1 - \chi_2 = \sqrt{(\chi_{xx} - \chi_{yy})^2 + 4\chi_{xy}\chi_{yx}}.$$

The real part of $\Delta\chi_{1,2}$ describes the double-refraction effect, and the imaginary part, as we have mentioned above, describes the selective absorption. Under appropriate conditions the imaginary part $\lambda_1(\mathbf{k}, \omega)$ according to Eq. (3.5) can turn out to be significantly larger than $\lambda_2(\mathbf{k}, \omega)$. Then the corresponding wave will be more strongly absorbed by the medium, which leads to an increase in the degree of polarization of the transmitted radiation.

The first experimental study of the nuclear Faraday effect was carried out in pure iron by Imber.³⁵ The angle of rotation of the polarization in his experiment was 14.3° (Fig. 1). Imbert's results showed, for example, that a more correct value of the internal conversion coefficient is $\alpha = 9.94$ instead of the previously

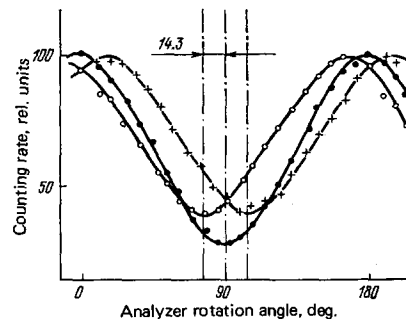


FIG. 1. Intensity of radiation transmitted through a dispersive medium as a function of analyzer rotation angle (from Ref. 35).

accepted value $\alpha = 15$ (at the present time it is considered that $\alpha = 9$). Housley and Gonser³⁶ also studied the Faraday effect in the partially inverse (mixed) ferromagnetic spinel magnesioferrite (MgFe_2O_4). The magnitude of the Faraday rotation in this compound consists of the sum of the rotations by the Fe^{57} nuclei in tetrahedral and octahedral positions. From data on the rotation angle the authors found the fraction of Mg atoms occupying tetrahedral sites. Subsequently Grant, Housley, and Gonser³⁷ used the results of the calculation of Ref. 36 for an experimental determination of the mean square displacement of the iron ion position in the sodium nitroprusside lattice, and also to obtain a more accurate value of the asymmetry parameter of the electric field gradient tensor at the iron nuclei in this compound. Gibb³⁸ studied the polarization dependence of the absorption in single crystals of iron ammonium sulfate for the purpose of determining correctly the parameters and orientation of the electric field gradient tensor. Gol'danskii, Makarov, Suzdalev, and Vinogradov³⁹ experimentally studied the polarization phenomena in the quadrupole splitting of the nuclear levels of Fe^{57} in uniaxial single crystals of siderite FeCO_3 . This enabled them to determine the anisotropy of the Mössbauer effect in this compound. Labushkin, Ivanov, and Chechin⁴⁰ observed and studied experimentally the double refraction of polarized γ rays in hematite single crystals below the Morin point where hematite is antiferromagnetic.

Alvazyan and Belyakov⁴¹ discussed the problem of searching for characteristic polarizations and the complex refractive indices corresponding to them for the allowed Zeeman spectrum in magnetically ordered crystals with various magnetic structures: ferromagnetic, weak ferromagnetic, antiferromagnetic, umbrella, and helicoidal structures. Subsequently Aivazyan⁴² extended these results to the case of arbitrary structure of the electric and magnetic fields at resonance nuclei. A general theoretical discussion of polarization effects within the framework of the S-matrix and density-matrix method has been given by Chrisman.⁴³

In paramagnetic crystals the problem of calculating the refractive index is substantially more complicated as the result of the phenomenon of electron relaxation.⁴⁴ In the intermediate case when the frequency of the electron paramagnetic relaxation is of the order of the reciprocal lifetime of the excited state of the nucleus, the splitting pattern of the nuclear levels becomes complex and depends on the electron populations of the atom. The fact that the dispersion dependence of the refractive index is very sensitive to the relaxation time and to the difference in the populations of electron levels allows one to investigate the nature of the paramagnetic state. Baryshevskii¹⁴ has given a general expression for the nuclear part of the current in the presence of a time-dependent low-frequency electromagnetic perturbation, which can be either an external radiofrequency field or a variable intracrystalline field under conditions of electron paramagnetic relaxation.

The phenomenon of total reflection of γ rays is particularly interesting in the presence of hyperfine split-

ting of the nuclear levels. The reflection coefficient for γ rays has the form (3.4), where $r = 4\pi |\chi_{e1} + \chi_{nuc}| / \psi^2$. Since both electrons and nuclei contribute to the polarizability, interference of the Rayleigh (electron) and resonance (nuclear) components occurs. Total reflection in the case of Zeeman hyperfine splitting of the nuclear levels has been investigated theoretically and experimentally by Bernstein *et al.*⁴⁵ The most interesting results obtained in these studies are the significant change of the polarization of radiation on reflection and the appearance of frequency-dependent phase shifts between the Rayleigh and resonance components. The interference between the electron and nuclear components of the reflected radiation can be constructive and destructive, when the scattered intensity is respectively greater or less than the sum of the intensities scattered individually by each channel. In this type of experiment it is possible to analyze not only the angular dependence of the integrated intensity of the reflected radiation (Fig. 2a), but also its frequency spectrum (Fig. 2b), which gives direct information on the dispersion of the electron and nuclear polarizabilities.

In writing out Eq. (3.1) for the nuclear polarizability we tacitly assumed that the dielectric properties of the medium can be obtained by summation of the independent contributions of individual nuclei. This assumption is in no way self-evident. In fact by means of multiple resonance scattering the excitation of any nucleus can be transferred by other nuclei. Conse-

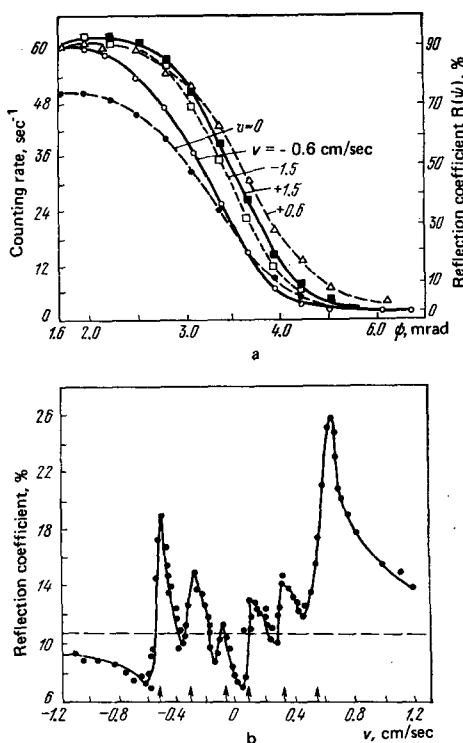


FIG. 2. a) Intensity of reflected radiation as a function of angle of incidence ψ for various rates of motion of radiation source (from Ref. 45); b) experimental dependence of reflection coefficient for a given angle of incidence $\psi = 4 \times 10^{-3}$ rad on rate of motion of source. The dashed line shows the electron contribution to the reflection coefficient (from Ref. 45).

quently, formation of a collected excited state of the entire system of nuclei as a whole is possible.⁴⁶

Taking into account collective effects leads to the appearance of a complex correction Γ to the transition frequency ω_0 and reduces for the most part to a certain renormalization of the decay constant (or line width of the resonance level: $\gamma' = \gamma + \text{Im}\Gamma$) of the nucleus, while the shift $\text{Re}\Gamma$ of the position of the resonance line can be assumed to be small. Therefore we can again discuss the medium as a set of individual oscillators, but now with somewhat changed properties. For an arbitrary (non-Bragg) direction in a single crystal allowance for collective effects leads to a change in the resonance width by some constant amount, while for a polycrystalline material this is a complicated function of the ratio λ/a , which can significantly change⁴⁷ the ordinary dispersion shape (3.1).

d) Polarizability of three-dimensionally periodic media

In elementary discussions it is usually assumed that the field \mathbf{E}' acting at each point of the medium is equal to the average field \mathbf{E} which enters into Maxwell's equations. In reality the local (effective) field at the location of a dipole differs from the average field [in cubic crystals, for example, we have $\mathbf{E}' = \mathbf{E} + (4\pi/3)\mathbf{P}$, where $4\pi\mathbf{P} = (\epsilon - 1)\mathbf{E}$]. In optics the inclusion of effective-field effects leads to the well known Lorenz-Lorentz formula.⁴⁸ However, in the x-ray region, where an important role is played by diffraction phenomena, determination of the polarization of the medium under the action of the effective field is only part of the problem, since the Lorenz-Lorentz formula does not indicate the nature of the propagation of wave fields in the crystalline medium. A rigorous discussion of this question, which forms the content of the Ewald-Oseen extinction theorem,⁴⁹ has been given for periodic media by Ewald.⁵⁰

In a medium, in addition to the primary incident wave, there are also secondary waves from the dipoles of the medium, which also exert an influence on the nature of the field which is established. The polarization at some point \mathbf{r}_0 of the medium is equal to

$$\mathbf{P}(\mathbf{r}_0) = \chi [\mathbf{E}_0(\mathbf{r}_0) + \sum_i \hat{F}(\mathbf{r}_0, \mathbf{r}_i) \mathbf{E}_i(\mathbf{r}_i)], \quad (3.6)$$

where \mathbf{E}_0 is the field of the incident wave, \mathbf{E}_i is the field produced by the i th dipole of the medium, the function $\hat{F}(\mathbf{r}_0, \mathbf{r}_i)$ describes the effect of all dipoles of the crystal on the dipole located at the point \mathbf{r}_0 , and the polarizability χ is defined as the sum of the polarizabilities α of the individual dipoles in a unit volume. Each term of the sum in Eq. (3.6) can be expressed in terms of a Hertz potential $F(\mathbf{r}, \mathbf{r}')\mathbf{P}(\mathbf{r}') = \text{rot rot P}(\mathbf{r}') \times \exp(i\omega R/c)R^{-1}$, where $R = |\mathbf{r} - \mathbf{r}'|$. Going over to a continuous distribution of dipoles in the medium instead of (3.6), it is possible to write down the integro-differential equation for determination of the polarization $\mathbf{P}(\mathbf{r})$:

$$\mathbf{P}(\mathbf{r}) = \chi \left[\mathbf{E}_0(\mathbf{r}) + \int (\text{rot rot P}(\mathbf{r}') e^{i(\omega/c)R} R^{-1}) d\mathbf{r}' \right]. \quad (3.7)$$

The integration must be carried out over the entire

volume of the medium, which is bounded by the surface Σ , except for a small volume near the point \mathbf{r}' of the medium, which is bounded by the surface σ . Subsequently σ must be made to approach zero. The operation of differentiation is carried out at the point \mathbf{r} of the medium. Equation (3.7) for determination of the polarization in the crystal is an exact result based on summation of the fields of the individual dipoles of the medium and on use of the microscopic Maxwell equations. Taking into account the translational-invariance properties of the crystal and also Eqs. (2.6)–(2.8), the integral equation (3.7) reduces to the equations for the Fourier amplitudes of the polarization $\mathbf{P}(\mathbf{k}_{\mathbf{H}}, \omega)$. Solution of these equations leads to the following result: for the amplitude with $\mathbf{H} = 0$ the integral is broken down into two parts, one of which extinguishes the primary wave in the medium [the first term in the right-hand side of (3.7)], so that from the boundary of the medium into its depth there is propagated only a dipole excitation wave with wave vector $k = (\omega/c)\sqrt{\epsilon}$ and phase velocity $c_p = c/\sqrt{\epsilon}$, and the polarizability of the medium and the permittivity, for example, in a cubic crystal, turn out to be related by the expression $(4\pi/3)\chi = (\epsilon - 1)/(\epsilon + 2)$, which coincides with the Lorenz-Lorentz result. All remaining waves with $\mathbf{H} \neq 0$ also are dipole waves in the medium with wave vectors $\mathbf{k}_{\mathbf{H}}$.

Since in the x-ray region the permittivity is very close in magnitude to unity, it is possible with a high degree of accuracy to use instead of the Lorenz-Lorentz formula the simpler relation $\epsilon - 1 = 4\pi\chi$, where $\chi = N\alpha$, which forms the basis for the discussion in the initial part of this section [see Eq. (3.1)]. In essence this means (see also Section 5) going over to the local interaction approximation, when in Eq. (2.2) it is possible to set $\hat{\epsilon}(\mathbf{r}, \mathbf{r}'; \omega) = \hat{\epsilon}(\mathbf{r}, \omega)\delta(\mathbf{r} - \mathbf{r}')$. The x-ray permittivity, in view of the spatial periodicity of the electron density $N = N(\mathbf{r})$ [see Eq. (3.1)], also is a spatially periodic function and therefore can be represented in the form of a Fourier series

$$\hat{\epsilon}(\mathbf{r}, \omega) = \sum_{\mathbf{H}} \hat{\epsilon}^{(\mathbf{H})}(\omega) \exp(i\mathbf{H}\mathbf{r}),$$

which leads to a system of equations for the amplitudes of the field $\mathbf{E}(\mathbf{k}_{\mathbf{H}}, \omega)$ (2.9).

Thus, in the x-ray region a crystal is characterized by a set of permittivities $\hat{\chi}^{(\mathbf{H})}$ corresponding to the various diffraction directions, which is one of the main features of optics in the x-ray frequency region.

4. QUANTUM-MECHANICAL THEORY OF X-RAY AND γ -RAY PERMITTIVITY

a) Basic equations of the theory

The quantum-mechanical theory of the dielectric properties of crystals for radiation in the x-ray frequency range was first treated by Kohler.² A detailed discussion of his approach can be found in the book by Von Laue.⁵¹ The response of a system of charges comprising a crystal to an external electromagnetic perturbation is usually treated in the quasiclassical approximation:

$$-i\hbar \frac{\partial \psi}{\partial t} = \hat{\mathcal{H}} \psi, \quad (4.1a)$$

$$\text{rot rot } \mathbf{E} + c^{-2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = -4\pi c^{-2} \frac{\partial \langle \mathbf{j} \rangle}{\partial t}. \quad (4.1b)$$

The current in the right-hand side of Eq. (4.1b) is the quantum-mechanical average over states of operators of the elementary currents induced by the external perturbation.

The complete Hamiltonian of the crystal $\hat{\mathcal{H}}$ in Eq. (4.1a) describes the electron and nuclear subsystems, and also the interaction between them. The individual terms of this Hamiltonian have the following form⁵²:

$$\begin{aligned} \hat{\mathcal{H}}_{el} &= \sum_s \frac{\hat{\mathbf{p}}_s^2}{2m_s} + \frac{1}{2} \sum_{s \neq s'} \frac{e^2}{|\mathbf{r}_s - \mathbf{r}_{s'}|} \quad \text{is the Hamiltonian of the electron subsystem of the crystal,} \\ \hat{\mathcal{H}}_{nuc} &= \sum_s \frac{\hat{\mathbf{P}}_s^2}{2M_s} + \frac{1}{2} \sum_{s \neq s'} V(\mathbf{R}_s - \mathbf{R}_{s'}) \quad \text{is the Hamiltonian of the nuclear subsystem of the crystal,} \\ \hat{\mathcal{H}}_{el-nuc} &= \sum_{s, s'} V(\mathbf{r}_s - \mathbf{R}_{s'}) \quad \text{is the Hamiltonian of the interaction of the } s\text{-th electron with nucleus } s', \\ \hat{\mathcal{H}}_{nuc-nuc} &= \frac{1}{2} \sum_{s \neq s'} V(\mathbf{R}_s - \mathbf{R}_{s'}) \quad \text{is the Hamiltonian of interaction of the nucleons in the nucleus.} \end{aligned}$$

In the Hamiltonian $\hat{\mathcal{H}}$ it is necessary also to include the term $\hat{\mathcal{H}}_\mu = \hat{\sigma}_s H(\mathbf{r}_s)$, which describes the interaction of the magnetic moment of the s -th charge σ_s with the magnetic field H at the point \mathbf{r}_s where this charge is located.⁵³

Not all terms of the complete Hamiltonian $\hat{\mathcal{H}}$ are equally significant in calculation of the permittivity.^{52,10,11} In the first stage of the solution it is not necessary to consider phonon vibrations in the crystal, which means that in the complete Hamiltonian we can neglect the term $\hat{\mathcal{H}}_{nuc}$. The intranuclear interactions, for example, in calculation of the electron contribution to the DP, also need not be taken into account. In addition, instead of the binary Coulomb interaction of the electrons with each other and with the nuclei it is possible to introduce an effective self-consistent potential $V_0(\mathbf{r})$ and to solve the one-electron problem. We note that, depending on whether we leave in the Hamiltonian the term $\sum_{s, s'} V(\mathbf{r}_s - \mathbf{R}_{s'})$ or $(1/2) \sum_{s \neq s'} (e^2/|\mathbf{r}_s - \mathbf{r}_{s'}|)$, there are introduced into the discussion either highly localized atomic electrons, or particles collectivized to a significant degree which belong to the valence band or the conduction band. If hyperfine interactions are present in the nuclei of the atoms in the crystal, they must be included in the Hamiltonian $\hat{\mathcal{H}}_{nuc-nuc}$.

In calculation of the DP, the electronic and nuclear subsystems can be treated as independent, so that the complete wave function of the system is broken up into the product of the wave functions of the electron subsystem, in which the coordinates of the nuclei enter as parameters, and those of the nuclear subsystem (adiabatic approximation). In terms of this approximation to the DP is the sum of two contributions—the electron part and the nuclear part, and therefore the further general discussion in this section refers in equal degree both to the electron and the nuclear subsystems.

The perturbation $\mathbf{E}(\mathbf{r}, t)$, which has the form of a plane monochromatic wave with wave vector \mathbf{k} and frequency ω , actually does have the form $\mathbf{E}(\mathbf{r}, t) = (1/2)\mathbf{E}^0[i(\mathbf{k}\mathbf{r} - \omega t)] + \text{c. c.}$, so that the vector potential is

$$\mathbf{A}(\mathbf{r}, t) = -\frac{ic}{2\omega} \mathbf{E}^0 \exp[i(\mathbf{k}\mathbf{r} - \omega t)] + \text{c. c.} \quad (4.2)$$

In the presence of the perturbation (4.2) the momenta of the charges must be generalized,⁵³ $\hat{\mathbf{p}}_s \rightarrow \hat{\mathbf{p}}_s - (e/c) \times \mathbf{A}(\mathbf{r}_s, t)$, and the appropriate substitution made in the Hamiltonian $\hat{\mathcal{H}}$. The addition to the Hamiltonian of the unperturbed system $\hat{\mathcal{H}}_0$ is

$$\hat{\mathcal{H}}'_{el} = - \sum_s^N \frac{e}{2mc} [\hat{\mathbf{p}}_s \mathbf{A}(\mathbf{r}_s, t) + \mathbf{A}(\mathbf{r}_s, t) \hat{\mathbf{p}}_s], \quad (4.3)$$

where N is the total number of electrons in a crystal of volume V and $\hat{\mathbf{p}}_s = -i\hbar\nabla_s$ is the momentum operator acting on the coordinates of the s -th charge. If we further take into account the zero-point oscillations of the field,⁵⁴ then as the vector potential $\mathbf{A}(\mathbf{r}_s, t)$ we must understand (see for example Ref. 55) the sum $\mathbf{A} + \mathbf{A}_0$ of the vector potential \mathbf{A} associated with the classical field and the quantum-mechanical operator for the potential $\hat{\mathbf{A}}_0$ corresponding to the quantum fluctuations:

$$\hat{\mathbf{A}}_0(\mathbf{r}) = \sum_{\mathbf{k}_1} [\hat{\mathbf{A}}(\mathbf{k}_1) e^{i\mathbf{k}_1\mathbf{r}} + \hat{\mathbf{A}}^*(\mathbf{k}_1) e^{-i\mathbf{k}_1\mathbf{r}}], \quad \hat{\mathbf{A}}(\mathbf{k}_1) = \left(\frac{2\pi\hbar c^2}{\omega_{\mathbf{k}_1} V} \right)^{1/2} \sum_{\lambda} e_{\lambda} \hat{a}_{\mathbf{k}_1, \lambda}, \quad (4.4)$$

where $\hat{a}_{\mathbf{k}_1, \lambda}$ is the annihilation operator of a photon with wave vector \mathbf{k}_1 and unit polarization vector e_{λ} .

In the first approximation with respect to the perturbation, the wave functions of the system of charges in the crystal have the following form:

$$\tilde{\psi}_n = \psi_n + \delta\psi_n, \quad (4.5)$$

where the addition to the unperturbed wave function ψ_n in the state n is

$$\delta\psi_n = \frac{i}{2\omega\hbar} \sum_{m \neq n} \left[\frac{\langle m | \mathbf{E}^0 \mathbf{J}(\mathbf{k}) | n \rangle}{\omega_{mn} - \omega} e^{-i\omega t} + \frac{\langle m | \mathbf{E}^0 \mathbf{J}(-\mathbf{k}) | n \rangle}{\omega_{mn} + \omega} e^{i\omega t} \right] \psi_m, \quad (4.6a)$$

$$\langle m | \mathbf{E}^0 \mathbf{J}(\mathbf{k}) | n \rangle = \int \psi_m^* \sum_s^N (\mathbf{E}^0 \hat{\mathbf{j}}^s(\mathbf{k})) \psi_n d\mathbf{r}_s, \quad (4.6b)$$

$$\hat{\mathbf{j}}^s(\mathbf{k}) = i\hbar \frac{e}{2m} (\nabla_s e^{i\mathbf{k}\mathbf{r}_s} + e^{i\mathbf{k}\mathbf{r}_s} \nabla_s). \quad (4.6c)$$

The frequency ω_{mn} corresponds to transition of the system from the state m to the state n . In Eq. (4.6a) we must understand by summation over the intermediate states m not only the sum over the discrete spectrum, but also integration over the continuum.

The unperturbed wave functions ψ_n are, in view of the symmetry of the medium, eigenfunctions of the operator of the translation \hat{T} by an arbitrary lattice vector \mathbf{R} . Consequently on action of the operator \hat{T} the wave function can acquire only a phase factor of the form $\exp(i\mathbf{H}_m \cdot \mathbf{r})$, where \mathbf{H}_m is an arbitrary vector of the reciprocal lattice. Therefore the matrix elements in Eq. (4.6b) acquire phase factors of the form $\mathbf{H}_m - \mathbf{H}_n \pm \mathbf{k}$, which must be equal to one of the reciprocal lattice vectors \mathbf{H} in order that the matrix elements also have translational symmetry.

The Fourier component of the symmetrized current-density operator in the general case has the form of a sum of individual contributions

$$\hat{\mathbf{j}} = \sum_{l=1}^4 \hat{\mathbf{j}}_l(\mathbf{k}, \omega), \quad (4.7)$$

where

$$\hat{\mathbf{j}}_1 = -\frac{e^2}{mc} \sum_s^N \mathbf{A}(\mathbf{r}_s, \omega) e^{i\mathbf{k}\mathbf{r}_s} \quad (4.8a)$$

is the potential part of the current;

$$\hat{j}_s = \frac{e}{2m} \sum_s^N (\hat{p}_s e^{ikr_s} + e^{ikr_s} \hat{p}_s) \quad (4.8b)$$

is the resonance part of the current;

$$\hat{j}_s = -\frac{e^2}{mc} \sum_s^N \hat{A}_0(r_s, \omega) e^{ikr_s} \quad (4.8c)$$

is the part of the current which takes into account the influence of elastic-scattering processes on the nature of the response of the medium, which is linear in the classical field $A(\mathbf{k}, \omega)$. Afanas'ev and Kagan⁵⁵ have taken into account the Compton and thermal-diffusion contributions to the linear polarizability;

$$\hat{k}(\mathbf{k}, \omega) = i \frac{e}{2m} \sum_s^N ((\hat{p}_s \hat{\sigma}^s) e^{ikr_s} - e^{ikr_s} (\hat{p}_s \hat{\sigma}^s)) \quad (4.8d)$$

is the magnetic part of the current. In view of their smallness we can drop the magnetic current for the electron subsystem,⁵⁶ and the potential current for the nuclear subsystem.

The average value of the current-density operator (4.7) in the state n with inclusion of (4.5) has the form

$$j^{(n)}(\mathbf{k}, \omega) = \langle \Psi_n | \hat{j}(\mathbf{k}, \omega) | \Psi_n \rangle + \sum_{l=2}^4 \langle \delta \Psi_n | \hat{j}(\mathbf{k}, \omega) | \Psi_n \rangle. \quad (4.9)$$

This current density obviously must then be averaged over all possible initial states n of the system, which is most convenient to carry out by means of the density-matrix formalism (see subsection b of Section V).

Further discussion requires use of a specific model, and therefore we proceed to a systematic discussion of the electron and nuclear contributions to the x-ray polarizability of a crystal.

b) X-ray polarizability of the electron subsystem

The connection between the polarization and the current in the Fourier representation is established by the well known relation $\mathbf{j} = -i\omega\mathbf{P}$. When translational symmetry of the crystal is taken into account in accordance with Eqs. (4.6), (4.8), and (4.9), the average polarization in the crystal in state n is

$$P_i(\mathbf{k}, \omega) = \sum_{\mathbf{H}} \chi_{ij}^{(H)}(\mathbf{k}, \omega) E_j(\mathbf{k} + \mathbf{H}, \omega), \quad (4.10)$$

where the polarizability tensor is

$$\chi_{ij}^{(H)}(\mathbf{k}, \omega) = -\frac{N e^2 \text{ nuc}}{V m \omega^2} [f(\mathbf{k}' - \mathbf{k}) + \Delta f_{ij}(\mathbf{k}', \mathbf{k}; \omega) + \Delta f_{ij}^{(NP)}(\mathbf{k}', \mathbf{k}; \omega)] \delta_{\mathbf{k}' - \mathbf{k}, \mathbf{H}}. \quad (4.11)$$

Here we have used the following notation:

$$f(\mathbf{k}' - \mathbf{k}) = \sum_n \langle n | e^{i(\mathbf{k}' - \mathbf{k})r_n} | n \rangle, \quad (4.11a)$$

$$\Delta f_{ij}(\mathbf{k}', \mathbf{k}; \omega) = \frac{\hbar}{m} \sum_{s, s' m \neq n} \sum \left[\frac{(\hat{B}_i^s(\mathbf{k}))_{mn} (\hat{B}_j^{s'}(-\mathbf{k}'))_{nm}}{\omega_m - \omega_n - \omega - (i\gamma/2)} + \frac{(\hat{B}_i^{s'}(-\mathbf{k}'))_{mn} (\hat{B}_j^s(\mathbf{k}))_{nm}}{\omega_m - \omega_n + \omega - (i\gamma/2)} \right], \quad \hat{B}_i^s = -\frac{im}{\hbar c} \hat{p}_i^s, \quad (4.11b)$$

$$\Delta f_{ij}^{(NP)}(\mathbf{k}', \mathbf{k}; \omega) = \frac{2\pi e^2}{mV} \sum_{m \neq n, s, k_1} \sum_{k_2} \frac{(e^{i(\mathbf{k}_1 - \mathbf{k})r_s})_{mn} (e^{i(\mathbf{k}' - \mathbf{k}_1)r_s})_{nm}}{(\omega - \omega_{mn} - \omega_{k_1} + (i\gamma/2)) (\hat{\pi}_{k_1})_{ij}}. \quad (4.11c)$$

Thus, the polarizability tensor (4.11) is the sum of the potential contribution (4.11a), the resonance contribution (4.11b), and the inelastic contribution (4.11c) to the dielectric properties of the crystal.

The various terms in Eq. (4.11) have different structures and therefore we shall discuss them individually below.

c) The potential part of the electron contribution to the crystal polarizability. Structure factor and atomic structure factor

According to Eq. (4.11a) the potential part of the polarizability is proportional to the sum of terms of the following form:

$$f_s(\mathbf{k}' - \mathbf{k}) = \int |\Psi_n(r_s)|^2 e^{i(\mathbf{k}' - \mathbf{k})r_s} dr_s, \quad (4.12)$$

where the integration is carried out over the volume of the unit cell v . The position vector of the s -th electron in the unit cell can be represented in the form of the sum $r_s = \mathbf{r}_1 + \mathbf{r}_{1,\alpha}$, where \mathbf{r}_1 is the position vector of the center of gravity of the l -th atom in the unit cell and $\mathbf{r}_{1,\alpha}$ is the position vector of the α -th electron in this atom. This representation, after substitution into (4.12) of the wave functions of the electrons in the atom (strong-coupling approximation) and summation over the electrons in the unit cell, leads to the following expression:

$$F(\mathbf{H}) = \sum_s f_s(\mathbf{k}' - \mathbf{k}) = \sum_l \left[\sum_{\alpha} f_{l,\alpha}(\mathbf{H}) \right] e^{i\mathbf{H}r_l}, \quad (4.13)$$

which is called the structure factor.¹⁸ The structure factor $F(\mathbf{H})$ takes into account the phase relations between the currents induced in different atoms within the unit cell. The intensities of the diffraction peaks in x-ray scattering in the kinematic approximation are proportional to the square of the modulus of the structure factor $|F(\mathbf{H})|^2$. The sum of the matrix elements (4.12) over all electrons of the l th atom is called¹⁸ the atomic structure factor $f_l(\mathbf{H})$:

$$f_l(\mathbf{H}) = \sum_{\alpha} f_{l,\alpha}(\mathbf{H}) = \int_{v_{at}} |\Psi_n(r)|^2 e^{i\mathbf{H}r} dr. \quad (4.14)$$

The main contribution to $f_l(\mathbf{H})$ according to Eq. (4.14) is obviously from the most high localized inner electrons.

Comparison of Eqs. (4.11a) and (4.13) leads to the well known relation for the crystal polarizability for $\omega \gg \omega_{mn}$:

$$\chi^{(H)} = -\frac{e^2}{m\omega^2} \frac{N e^2 \text{ nuc}}{V} F(\mathbf{H}). \quad (4.15)$$

Thus, in the case of a multiatomic crystal the potential or principal part of the polarizability is expressed in terms of the structure factor $F(\mathbf{H})$, and in the case of a monatomic crystal—in terms of the atomic structure factor $f(\mathbf{H})$. In the approximation used, the crystal response to an external perturbation is expressed in terms of the individual properties of its component atoms.⁵⁷ According to Eq. (4.15) the polarizability $\chi^{(H=0)}$ is proportional to the electron density averaged over the volume of the unit cell,

$$\chi^{(H=0)} = v^{-1} \int \chi(r) e^{i\mathbf{H}r} |_{\mathbf{H}=0} dr = -n_0 r_0 \lambda^2 = -(4\pi)^{-1} \frac{\omega_p^2}{\omega^2}.$$

The problem of calculating the atomic structure factor (4.14) reduces to the choice of an adequate model of the electron density distribution in the atom. A

summary of the experimental and theoretical investigations of this problem can be found in several books.^{18,28,51}

When the free-atom wave functions are used, the atomic structure factor (4.14) is real and spherically symmetric. Deformation of the valence electron shells as the result of chemical bonding effects leads to non-sphericity of the atomic structure factor. In this case, as follows from Eq. (4.14), it becomes complex. The effect of this deformation was observed experimentally for the first time by Renninger⁵⁸ (for more details see Ref. 59). It must be emphasized that the complex nature of the atomic structure factor arising in this way (like the complex nature of the structure factor in crystals which are not centrally symmetric) has nothing in common with the true photoelectric absorption of the radiation. Deformation of the electron shells of an atom can in principle be produced also by external actions. However, these actions, for example, that of electric fields, at the present time are not sufficiently intense to produce appreciable deformation of the inner electron shells.⁶⁰

d) Dispersion contribution to the electronic polarizability

The contribution of the dispersion terms (4.11b) to the polarizability becomes appreciable when the frequency of the external perturbation approaches one of the eigenfrequencies of the atom ω_{mn} . For the frequency region of interest to us these are the K and L electron shells for elements with intermediate atomic numbers and the M shell for the heaviest elements. A detailed discussion of the early theoretical and experimental investigations of dispersion corrections can be found in well known books.^{18,28,51} The results of more recent calculations are given in Ref. 61. A brief critical review of the current state of this field is given by Wagenfeld.⁶²

The dispersion part (4.11b) of the polarizability tensor is constructed with the wave functions belonging to the entire crystal. The spatial dispersion arising here will be investigated below (see Section 5). Here we limit ourselves to the strong-coupling approximation, which is completely satisfactory, especially for elements which are not too light. In this approximation the matrix elements in (4.11b) are substantially simplified, since the wavelengths of the radiation corresponding to frequencies close to the K absorption edge are significantly greater than the radius a_K of the region in which the K electrons are localized. In fact, $a_K/\lambda \approx \alpha Z$, where $\alpha = e^2/\hbar c = 1/137$ is the fine-structure constant. Therefore in the matrix elements of (4.11b) the exponentials are usually replaced by unity. In this approximation the dispersion part of the tensor $\hat{\chi}$, on the assumption of spherical symmetry of the K -electron wave functions, becomes scalar,

$$\chi^{(H)}(\omega) = -\frac{e^2}{m\omega^2} \frac{\hbar}{mV} \sum_s \sum_{m \neq n} \left[\frac{(\hat{B}^s(0))_{mn} (\hat{B}^s(0))_{mn}}{\omega_{mn} - \omega} + \frac{(\hat{B}^s(0))_{mn} (\hat{B}^s(0))_{mn}}{\omega_{mn} + \omega} \right]. \quad (4.16)$$

On the assumption of strong localization of the inner

electrons, the currents in the matrix elements of (4.16) refer to the same atom, which removes the double summation over the electrons in (4.11b). This means that the potential $\sum_{s,s'} V(r_s - R_{s'})$ in Eqs. (4.1a) is broken up into the sum of terms corresponding to each atom individually, and the equation thereby breaks up into a system of independent equations.

With the simplifying assumptions made above, the matrix elements in expression (4.16) are consequently equal to

$$\chi^{(H)}(\omega) = -\frac{e^2}{m\omega^2} \frac{N_{el-nuc}}{V} \frac{\hbar}{m} \sum_l \left[\frac{2(\omega_m^l - \omega_n^l)}{(\omega_m^l - \omega_n^l)^2 - \omega^2} |(\hat{B}^l(0))_{mn}|^2 \right] e^{iHr_l}. \quad (4.17)$$

The sum over l is a dispersion correction to the structure factor $F(H)$. Equation (3.2), which was obtained from elementary considerations, coincides with (4.17) if we set

$$g_{mn} = \frac{2\hbar}{m\omega_{mn}} |(\hat{B}(0))_{mn}|^2, \quad (4.18)$$

where g_{mn} is the oscillator strength.⁶³ Going over in Eq. (4.17) from summation over a discrete set of frequencies to integration over a continuous spectrum with inclusion of Eq. (4.18), we arrive at the well known expression obtained by Kronig⁶⁴ for the dispersion correction to the principal part of the atomic structure factor

$$\Delta f_K = \int_{\omega_K}^{\infty} d\omega' \frac{\omega'^2}{\omega'^2 - \omega^2} \frac{dg_K}{d\omega'}, \quad (4.19)$$

where $dg/d\omega'$ is the density of oscillator strengths. The integral g_K of the density of oscillator strengths over frequency must give the total number of electrons in the corresponding shell. Thus, for K electrons we should expect $g_K = 2$. In reality, as shown by Prins and by Williams,⁶⁵ $g_K \approx 1.3$. This is due to the fact that the filled discrete states in view of the Pauli principle do not contribute to the matrix elements of (4.18).

The dispersion correction to the atomic structure factor Δf_K , when damping in the denominators of (4.16) is taken into account, is in the general case of complex quantity: $\Delta f_K = \Delta f_K' + i\Delta f_K''$. The imaginary part $\Delta f_K''$ describes the photoelectric absorption of radiation. Far from the absorption edges the dispersion corrections $\Delta f_K'$ and $\Delta f_K''$ are numerically small in comparison with the atomic structure factor $f(H)$.

The problem of calculating the dispersion correction thus reduces to a search for the distribution function $dg/d\omega$. Kronig⁶⁴ and Kallmann and Mark⁶⁶ proposed a semiempirical method based on the relation between the atomic absorption coefficients $\mu_a(\omega)$ at frequency ω and the density of oscillators: $dg/d\omega = (mc/2\pi^2 e^2) \times \mu_a(\omega)$. The frequency dependence $\mu_a(\omega)$ can be found experimentally and can be represented by means of empirical formulas. In particular, for the K edge we have

$$\mu_a(\omega) = \begin{cases} 0, & \omega < \omega_K, \\ (\omega_K/\omega)^n \mu_a(\omega_K), & \omega > \omega_K, \end{cases}$$

where the exponent n is close to three.

The dispersion correction (4.11b) can be found⁶² from the experimental data on the frequency dependence of

the absorption coefficient by means of the optical theorem and the Kramers-Kronig relations.

The direct method of determining the dispersion correction, as was pointed out above, consists of calculating the matrix elements in (4.17). These calculations have been carried out by Sugiura⁶⁷ and by Hönl.⁶⁷

Inclusion of transitions to optical atomic levels and levels of x-ray exciton states leads to appearance of a fine structure of the absorption edge.^{29,30}

Replacement of the exponential by unity in the matrix elements of (4.11b) is approximate and, in principle, it is necessary to carry out a multipole expansion. The greatest contribution is from the dipole terms $\Delta f_D'' + i\Delta f_D''$ and the quadrupole terms $\Delta f_Q'' + i\Delta f_Q''$ of the expansion.^{18,68} Only on taking into account higher multipoles does the electronic polarizability become a tensor and a spatial dispersion appears. However, the contribution from the higher multipoles amounts to no more than a few percent according to the estimates of Wagenfeld⁶² and is within the accuracy of contemporary experimental measurements.

It is well known that the thermal vibrations of atoms in a crystal lattice change the intensity of the elastic diffraction peaks by a factor $\exp[-2W(\mathbf{k}' - \mathbf{k})]$, which is called the Debye-Waller factor.⁶⁹

The problem of taking into account phonon vibrations in the crystal polarizability consists of the analysis of matrix elements of the form

$$\langle n^0 | \exp[i(\mathbf{k}' - \mathbf{k}) \mathbf{u}_s] | n^0 \rangle_T \quad (4.20a)$$

for the potential part of the current, and of a sum of the form

$$\left\langle \sum_{\{n\}} \frac{\langle n^0 | \exp(i\mathbf{k}' \mathbf{u}_s) | n \rangle \langle n | \exp(-i\mathbf{k} \mathbf{u}_s) | n^0 \rangle}{\omega_{mn} - \omega + \sum_j (n_j - n_j^0) \omega_j + (i\gamma/2)} \right\rangle_T \quad (4.20b)$$

for the resonance part. The operation $\langle \dots \rangle_T$ indicates averaging over the initial states of the phonon system. The vector \mathbf{u}_s represents the displacement of the center of gravity of the s -th atom from the equilibrium position \mathbf{r}_s^0 and can be expanded in a series in normal modes of vibrations of the atoms in the lattice⁷⁰; $|n\rangle$ is the wave function of the phonon subsystem of the crystal, n^0 and n are the phonon occupation numbers in the initial and intermediate states, and j is the number of the vibration. In the harmonic approximation the averaging of the operator $\exp[i(\mathbf{k}' - \mathbf{k}) \mathbf{u}_s]$ in Eq. (4.20a) over the phonon occupation numbers n^0 leads to the following expression for the argument of the exponential in the Debye-Waller factor:

$$W(\mathbf{k}' - \mathbf{k}) = \frac{1}{2} \langle [(\mathbf{k}' - \mathbf{k}) \mathbf{u}_s]^2 \rangle_T = 8\pi^2 \left(\frac{\sin \theta}{\lambda} \right)^2 \langle u_{s,1}^2 \rangle, \quad (4.21)$$

where $u_{s,1}$ is the component of the displacement of the s -th atom from its equilibrium position in the direction of the reciprocal lattice vector \mathbf{H} .

Next for calculation of the mean square displacements $\langle u_{s,1}^2 \rangle$ it is necessary to resort to a specific model of the phonon spectrum of the crystal.⁶⁹

The complexity which arises in the attempt to carry

out a similar procedure with expression (4.20b) lies in the fact that it is impossible to carry out a summation over the intermediate states of the phonon system, as the result of presence of the phonon energy in the denominator. However, since for x-ray transitions $\gamma \sim 10^{16} \text{ sec}^{-1}$ while $\omega_j^{\text{max}} \sim 10^{13} \text{ sec}^{-1}$, during the time of the interaction in a resonance channel an atom cannot be displaced from its instantaneous position, and therefore the phonon sum in the denominator of (4.20b) can be neglected and this again leads to the Debye-Waller temperature factor (4.21).

Thus, the potential and resonance contributions to the polarizability, with inclusion of phonon vibrations, take the form

$$\chi^{(H)}(\mathbf{k}, \omega)_{(T)} = \chi^{(H)}(\mathbf{k}, \omega) \exp \left[-W \left(\frac{\sin \theta}{\lambda} \right) \right].$$

e) Effect of inelastic scattering on polarizability

In calculation of the polarizability, usually only first-order perturbation theory is used. Nevertheless, from simple physical considerations it is easy to understand that inelastic processes also must contribute to the polarizability. In fact, inelastic scattering, i.e., a process of the second order, leads to an additional non-dissipative damping μ_{NP} (extinction) of the incident wave, which is equivalent to an additional contribution to the imaginary part of the polarizability⁵⁾ and, in accordance with the Kramers-Kronig relations—also to the real part of χ .

Afanas'ev and Kagan⁵⁵ have shown that the contribution of Compton scattering and thermal-diffusion scattering (TDS) to the polarizability is determined by expression (4.11c), where in the latter case it is necessary to take into account phonon vibrations of the lattice. The vector potential \mathbf{A}_0 (4.4) characterizes the field of inelastic scattering with frequencies $\omega_{\mathbf{k}_1}$ shifted from the frequency of the external perturbation, and summation over \mathbf{k}_1 in (4.11c) corresponds to averaging of a linear (with respect to the classical vector potential \mathbf{A}) polarizability over all possible frequencies and directions of propagation of inelastically scattered waves.

Inclusion of Compton scattering and thermal-diffusion scattering leads⁵⁵ to a certain renormalization of the x-ray polarizabilities (the coefficients of the dynamical theory). Here the inelastic scattering by phonons should appear experimentally in the deviation of the temperature dependence of the structure factor from the Debye-Waller factor. In fact, Efimov⁷² and subsequently Baldwin *et al.*⁷³ and Sano *et al.*⁷⁴ have observed that the temperature behavior of the anomalous transmission coefficient differs from a Debye-Waller dependence. The contribution to absorption as the result of phonon scattering can amount to ~14% in silicon and germanium crystals for CuK_α or MoK_α radiation in first-order reflections.^{72,55,75}

Phenomenologically the influence of multiphoton elastic scattering processes on the polarizability can be

⁵⁾In the optical region near the long-wavelength edge of the exciton absorption band the addition μ_{NP} is dominant⁷¹ as the result of combination scattering by phonons.

explained⁷⁶ if nonlinear polarizabilities of the medium are introduced into the discussion. Then the total polarizability at the frequency of the incident radiation is equal to the sum of the ordinary linear polarizability χ and an additional nonlinear polarizability $\Delta\chi_{NP}$ which depends on the quadratic and cubic nonlinearities of the medium and is proportional to the intensity $I_{\mathbf{k}_1} = \mathbf{E}(\mathbf{k}_1)\mathbf{E}(\mathbf{k}_1)^*$ of the inelastically scattered wave $\mathbf{E}(\mathbf{k}_1)$. In the case of spontaneous inelastic scattering, when the number of photons scattered into a mode is much less than unity, it is possible to replace $I_{\mathbf{k}_1}$ by the intensity of vacuum fluctuations $2\pi\hbar\omega_{\mathbf{k}_1}\Omega^{-1}[1+N(\omega-\omega_{\mathbf{k}_1})]$, where $N(\omega) = [\exp(\hbar\omega/kT) - 1]^{-1}$. After averaging $\Delta\chi_{NP}$ over the final states \mathbf{k}_1 , this additional term acquires the meaning of a correction to the ordinary linear polarizability due to the influence of inelastic processes on the transmitted wave.

f) Nuclear polarizability of a crystal

For the nuclear subsystem the relation between the field and the polarization also is established, according to Eq. (4.10), in terms of the polarizability tensor $\chi_{ij}^{(n)}(\mathbf{k}, \omega)$. In the current-density operator of the nuclear subsystem it is necessary to retain two terms—the resonance term (4.8b) and the magnetic term (4.8d).

By analogy to the case of the electronic polarizability, the nuclear polarizability can be expressed in terms of the individual properties of the nuclei composing the crystal:

$$\chi_{ij}^{(n)}(\mathbf{k}, \omega) = -\frac{e^2}{M\omega^2} \frac{N}{V} f_{ij}(\mathbf{k}', \mathbf{k}; \omega), \quad (4.22)$$

where M is the proton mass and $f_{ij}(\mathbf{k}', \mathbf{k}; \omega)$ is the nuclear structure factor tensor determined by expression (4.11b). The total line width γ entering into the denominator of (4.11b) is made up of the radiation line width γ_r , which determines the probability of decay of an excited state of the nucleus by γ -ray emission, and the conversion width γ_α , which is related to the probability of transfer of excitation energy to an atomic electron. Nuclear Mössbauer transitions, as a rule, are transitions of pure $M1$ or $E2$ multipolarity or a mixture of them. The ground state $|m\rangle = |m_0 I_0\rangle$ and the excited state $|n\rangle = |m_1 I_1\rangle$ of the nucleus are characterized by total angular momenta I_0 and I_1 and magnetic quantum numbers m_0 and m_1 .

In the absence of hyperfine splitting, the resonance denominator in (4.11b) does not depend on the magnetic quantum number m and therefore it can be taken out from under the summation sign, after which it is only necessary to sum the expression

$$\sum_{m=n} \overline{(B_i(\mathbf{k}))_{mn} (B_j(-\mathbf{k}'))_{nm}}, \quad (4.23)$$

where the bar over the expression indicates averaging over the initial states. Calculation of the sum (4.23) (see Ref. 78) leads to the following result⁷⁷:

$$\begin{cases} b \left[(\mathbf{k} \cdot \mathbf{k}') \delta_{ij} + k_i k_j - \frac{3}{2} k_i k_j \right] & \text{for an } E2 \text{ transition,} \\ b \{ (\mathbf{k} \cdot \mathbf{k}') \delta_{ij} - k_i k_j \} & \text{for an } M1 \text{ transition,} \end{cases} \quad (4.24)$$

where

$$b = \frac{e^2}{4\omega_0^2} \frac{2I_2+1}{2I_1+1} \gamma_r \frac{M^2}{\hbar^2}.$$

For forward scattering ($\mathbf{k}' = \mathbf{k}$ and $\mathbf{k} = s\mathbf{k}$) we obtain from Eq. (4.24) respectively $b k^2 [\delta_{ij} - s_i s_j] + \frac{4}{3} s_i s_j$ and $b k^2 (\delta_{ij} - s_i s_j)$. The term $\delta_{ij} - s_i s_j$ is the operator of projection on a plane perpendicular to the vector $\mathbf{s} = \mathbf{k}/k$. Consequently, it is possible to introduce a transverse polarizability $\chi_\perp \propto b k^2 (\delta_{ij} - s_i s_j)$. The term $(4/3) b k^2 s_i s_j$ describes the response in a longitudinal field and is proportional to the longitudinal polarizability χ_\parallel , which is nonzero for an $E2$ transition. Thus, in the absence of hyperfine splitting, the medium possesses spatial dispersion, is isotropic, and nongyrotropic. Similarly discussion of the case $\mathbf{k}' = \mathbf{k} + \mathbf{H}$ shows that this remains true for an $M1$ transition, but for an $E2$ transition, since the vector \mathbf{H} distinguishes a certain direction in space, anisotropy arises.

If the nucleus is in an external field which lifts the degeneracy with respect to the magnitude and sign of the magnetic quantum number, it is necessary in the calculation of (4.11b) to carry out a multipole expansion of the current-density operators. This leads to the following expression for the nuclear polarizability:

$$\begin{aligned} \chi(\mathbf{k}, \mathbf{k}'; \omega) = & \frac{N}{V} \frac{2\pi}{k^2} \sum_{L'L''} \sum_{L\lambda} C(I_0 L' I; m_0 M m) \\ & \times C(I_0 L I; m_0 M m) \hat{Y}_{L'M'}^{(\lambda)*}(\mathbf{k}') \hat{Y}_{LM}^{(\lambda)}(\mathbf{k}) \\ & \times \sqrt{\gamma_r(L'\lambda') \gamma_r(L\lambda)} [\omega(j_n, m_0 + M) - \omega(j_0, m_0) - (i\gamma/2)]^{-1}, \end{aligned} \quad (4.25)$$

where $M = m - m_0$, $(L, 1) \equiv E(L)$ is the electric 2^L pole, and $(L, 0) \equiv M(L)$ is the magnetic 2^L pole. The quantities $\hat{Y}_{LM}^{(\lambda)}$ are vector spherical harmonics; the remaining designations are standard (see Ref. 79). Thus, nuclear polarizability is a tensor with an expressed spatial dispersion and according to Eq. (4.25) it is expressed in terms of the width γ_r for radiative nuclear γ transitions. A more detailed discussion of this question is given in the review of Hannon and Trammell¹¹ and in the work of Blume and Kistner.³² The expression (4.25) is valid in the limiting case of very slow (comparable with the lifetime of the excited state) electron paramagnetic relaxation. A general expression for the nuclear current in the presence of a time-dependent external perturbation is given in Ref. 14.

A number of studies^{11,32,36,37,41,45} devoted to the optics of Mössbauer radiation introduce the refractive index of the medium at the frequencies of nuclear transitions. Here, following Foldy⁸⁰ and Lax,⁵⁷ the refractive index is expressed in terms of the coherent probability amplitude for scattering of the radiation forward, i.e., the scattering tensor is introduced (see Ref. 78). The amplitude for scattering of a photon with wave vector \mathbf{k} and polarization \mathbf{e} into a photon \mathbf{k}' , \mathbf{e}' is given by the following expression:

$$f(\mathbf{k}', \mathbf{e}'; \mathbf{k}, \mathbf{e}) = e_i^* N_{ij}^s(\mathbf{k}, \mathbf{k}) e_j, \quad (4.26)$$

where N_{ij}^s is the scattering tensor of the s -th nucleus. In the representation of circular polarization $\eta(\pm) = \mp(\hat{\mathbf{e}}_\pm \pm \hat{\mathbf{e}}_\mp)/\sqrt{2}$ the spherical functions in Eq. (4.25) can be expressed in terms of the rotation D matrix

$$\begin{aligned} \hat{\eta}_{\mu'}^* \hat{Y}_{L'M'}^{(\lambda)*}(\mathbf{k}') \hat{Y}_{LM}^{(\lambda)}(\mathbf{k}) \hat{\eta}_\mu = & (8\pi)^{-1} (\mu')^{\lambda+1} (\mu)^{\lambda+1} \\ & \times \sqrt{(2L'+1)(2L+1)} D_{\mu'M'}^{(L')}(\hat{\mathbf{k}}', \hat{\mathbf{z}}) D_{\mu M}^{(L)}(\hat{\mathbf{k}}, \hat{\mathbf{z}}) \quad (\mu, \mu' = \pm 1). \end{aligned} \quad (4.27)$$

In particular, for a magnetic dipole transition, expressions (4.11b), (4.25), and (4.27) give the following result⁸¹:

$$\frac{2k_0}{\gamma_r} f_{m, m'} = \frac{1}{2I_1 + 1} \sum_{m_0} \sum_{\mu} \frac{3}{2} D_{\mu m'}^{(1)}(\hat{\mathbf{k}}', \hat{\mathbf{z}}) D_{\mu m}^{(1)}(\hat{\mathbf{k}}, \hat{\mathbf{z}}) \frac{C^2(I_1 I'; m_0 \mu)}{\omega - \omega(m_0, m_0 + \mu) + i(\gamma/2)}.$$

Blume and Kistner³² have calculated a general expression for the nuclear forward scattering amplitude in the presence of a magnetic field. Housley, Grant, and Gosner³³ have extended these results to the case of electric quadrupole interaction.

Inclusion of the effect of phonon vibrations on the polarizability of the nuclear subsystem leads to a somewhat different result than for the electron subsystem. In the case of interaction of γ rays with the nuclei in the crystal, between the times of the phonon vibrations and the lifetime of the excited state of the nucleus there exists ($\gamma \approx 10^7 \text{ sec}^{-1}$) the relation $\gamma \ll \omega_i^{\text{max}}$, which is the reverse of the case of resonance interaction with the electron subsystem (see Subsection d) of Section 4). In this case it is necessary to calculate systematically each term of the sum (4.20b). The greatest contribution is from the first phonon-free term of the sum

$$\langle (e^{-ik'u_n})_{(n_0)} (e^{ik'u_n})_{(n_0)} \rangle_T.$$

The result of averaging this expression over the initial states leads again to the exponential (the Lamb-Mössbauer factor) $\exp[-(W(\mathbf{k}) + W(\mathbf{k}'))]$, in which the temperature dependence of the argument agrees exactly with that of the Debye-Waller factor. A feature of the Lamb-Mössbauer factor is that while the Debye-Waller factor (4.21) depends on the difference $|\mathbf{k}' - \mathbf{k}| \propto \sin\theta/\lambda$, the Lamb-Mössbauer factor depends on the wave vectors \mathbf{k}' and \mathbf{k} separately.⁸¹

5. APPLICATION OF THE BLOCH FORMALISM TO CALCULATION OF THE ELECTRONIC POLARIZABILITY OF CRYSTALS

a) Bloch representation of the wave functions of the electron subsystem

A systematic approach to calculation of the polarizability of crystals obviously must rest on the notion that the behavior of the electrons in a solid has a Bloch nature. The bases for this approach were laid down by Adler⁸² and Wiser.⁸³ Bloembergen and his colleagues,⁸⁴ Cheng and Miller,⁸⁵ and Pine⁸⁶ have applied this method to calculation of polarizabilities of higher orders. The general formalism developed in Refs. 82–86 can be extended also to calculation of the polarizability at x-ray frequencies. The basis for this is the following consideration. In spite of the fact that the deformations of the outer electron shells arising in formation of a chemical bond are small, they are completely observable experimentally, since they are manifested as the result of the spatial periodicity of the crystals as independent effects, in particular, in the form of new diffraction peaks. Therefore for a rigorous description of the effect of chemical bonding on the dielectric permittivity it is necessary to go over from the strong-coupling approximation, which was used in Sections 4(c) and 4(d), to a description taking into ac-

count the effects of the band structure.

Following the work of Pine,⁸⁶ we note the principal aspects of calculation of the polarizability of a crystal. As we have already mentioned in Subsection a) of Section 4, one can introduce a one-electron self-consistent potential $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0(\mathbf{r}) + \hat{\mathcal{H}}'(\mathbf{r}, t)$, where $\hat{\mathcal{H}}_0 = (\mathbf{p}^2/2m) + V(\mathbf{r})$ is the periodic Bloch Hamiltonian, and the perturbation $\hat{\mathcal{H}}'$ is linear in the vector potential:

$$\hat{\mathcal{H}}'(\mathbf{r}, t) = -\frac{e}{2mc} [\mathbf{A}(\mathbf{r}, t) \mathbf{p} + \mathbf{p} \mathbf{A}(\mathbf{r}, t)]. \quad (5.1)$$

In expansion of all quantities in Fourier series it is convenient to separate in explicit form the wave vector \mathbf{q} reduced to the first band, so that any wave vector \mathbf{k} is equal to the sum of the vector \mathbf{q} and the reciprocal lattice vector \mathbf{Q} which supplements it. Then any function $V(\mathbf{r}, t)$ of the coordinates can be represented in the form of an expansion

$$V(\mathbf{r}, t) = \sum_{\mathbf{q}, \mathbf{Q}} V(\mathbf{q}, \mathbf{Q}; \omega) \exp[i(\mathbf{q} + \mathbf{Q})\mathbf{r} - i\omega t], \quad (5.2)$$

where

$$V(\mathbf{q}, \mathbf{Q}; \omega) = (2\pi\Omega)^{-1} \int_{\Omega} d\mathbf{r} \int dt V(\mathbf{r}, t) \exp[-i(\mathbf{q} + \mathbf{Q})\mathbf{r} + i\omega t].$$

The unperturbed Hamiltonian $\hat{\mathcal{H}}_0$ corresponds to a complete set of orthonormalized eigenfunctions $\varphi_{\mathbf{k}m}(\mathbf{r}) \equiv |\mathbf{k}, m\rangle$ and eigenvalues⁸⁷ $\epsilon_{\mathbf{k}m}$, where the wave vector \mathbf{k} of the electron lies in the first Brillouin zone and m is the number of the energy band. According to the Bloch theorem,⁸⁷ the wave function $\varphi_{\mathbf{k}m}(\mathbf{r})$ has the following form:

$$\varphi_{\mathbf{k}m}(\mathbf{r}) = \Omega^{-1/2} u_{\mathbf{k}m}(\mathbf{r}) e^{i\mathbf{k}\mathbf{r}},$$

where $u_{\mathbf{k}m}(\mathbf{r}) \equiv v^{-1/2} |k m\rangle$ and Ω is the normalization volume.

This approach permits inclusion into the discussion of partially collectivized electrons of the valence band and completely collectivized electrons of the conduction band.

The solution of the Schrödinger equation $\psi(\mathbf{r}, t)$ can be represented in the form of a series in the unperturbed wave functions, $\psi(\mathbf{r}, t) = \sum_{\mathbf{k}m} C_{\mathbf{k}m}(t) \varphi_{\mathbf{k}m}(\mathbf{r})$, whose coefficients determine the density matrix $\langle \mathbf{k}' m' | \rho | \mathbf{k} m \rangle = \langle C_{\mathbf{k}' m'} C_{\mathbf{k} m}^* \rangle$, where in the right-hand side of the equality we have carried out an averaging over the ensemble. The dynamics of the electron system is described by the Liouville equation for the density matrix ρ :

$$i\hbar \dot{\rho} = [\hat{\mathcal{H}}_0, \rho] + \frac{i\hbar}{\tau} (\rho_0 - \rho). \quad (5.3)$$

The density matrix $\rho(\mathbf{r}, t)$ can be represented in the form of a sum of an unperturbed part ρ_0 and a perturbed part $\rho_1(\mathbf{r}, t)$ linear in the vector potential $\mathbf{A}(\mathbf{r}, t)$. The matrix ρ_0 is diagonal in the Bloch representation and its eigenvalues are the Fermi-Dirac distribution function $n(\epsilon_{\mathbf{k}m}) = \{ \exp[(\epsilon_{\mathbf{k}m} - \epsilon_F)/kT] + 1 \}^{-1}$.

b) Current density in the representation of Bloch functions

The average over the ensemble of current-density

⁸¹Indication of the indices in $\epsilon_{\mathbf{k}m}$ should not lead to confusion with the dielectric permittivity $\epsilon_{ij}^{(E)}(\mathbf{k}, \omega)$.

operators is

$$j(\mathbf{r}, t) = \sum_{\mathbf{k}qmn} \left[\frac{e\hbar}{2mi} (\nabla \varphi_{\mathbf{k}m}^* (\nabla \varphi_{\mathbf{k}+q, n}) - (\nabla \varphi_{\mathbf{k}m}) \nabla \varphi_{\mathbf{k}+q, n}) - \frac{e^2}{mc} \mathbf{A}(\mathbf{r}, t) \varphi_{\mathbf{k}m}^* \varphi_{\mathbf{k}+q, n} \right] (\mathbf{k}+q, n | \rho(\mathbf{r}, t) | \mathbf{k}m). \quad (5.4)$$

The periodicity of the functions $u_{\mathbf{k}m}(\mathbf{r})$ and $\nabla u_{\mathbf{k}m}(\mathbf{r})$ permits rather simply going over in Eq. (5.4) to the Fourier components of the current:

$$j(\mathbf{q}, \mathbf{Q}; \omega) = \sum_{\mathbf{k}mn} \left(\left[\frac{e}{m} (\mathbf{k}m | \left\{ p + \hbar \left[\mathbf{k} + \frac{1}{2} (\mathbf{q} + \mathbf{Q}) \right] \right\} e^{-i\mathbf{Q}r} | \mathbf{k}+q, n \right) (\mathbf{k}+q, n | \rho_1(\mathbf{r}, \omega) | \mathbf{k}m) \right] - \left[\sum_{\mathbf{q}'\mathbf{Q}'} \frac{e^2}{mc} \mathbf{A}(\mathbf{q}-\mathbf{q}', \mathbf{Q}-\mathbf{Q}'; \omega') \times (\mathbf{k}m | e^{-i\mathbf{Q}'r} | \mathbf{k}+\mathbf{q}', n) (\mathbf{k}+\mathbf{q}', n | \rho_0(\mathbf{r}; \omega-\omega') | \mathbf{k}m) \right] \right), \quad (5.5)$$

where $\mathbf{k}m | \mathbf{B} | \mathbf{k}'m' = v^{-1} \int_V d\mathbf{r} u_{\mathbf{k}m}^* \mathbf{B}(\mathbf{r}) u_{\mathbf{k}'m'}$ is the reduced matrix element. The expression for ρ_1 is determined from Eq. (5.3) and has the following form:

$$\langle \mathbf{k}+q, n | \rho_1(\mathbf{r}, \omega) | \mathbf{k}m \rangle = -\frac{e}{2mc} R(\mathbf{k}+\mathbf{q}, n; \mathbf{k}, m; \omega) \langle \mathbf{k}+q, n | \mathbf{A}(\mathbf{r}, \omega) \mathbf{p} + \mathbf{p} \mathbf{A}(\mathbf{r}, \omega) | \mathbf{k}m \rangle, \quad (5.6)$$

$$R(\mathbf{k}', m'; \mathbf{k}, m; \omega) = \frac{n(\varepsilon_{\mathbf{k}'m'}) - n(\varepsilon_{\mathbf{k}m})}{\varepsilon_{\mathbf{k}'m'} - \varepsilon_{\mathbf{k}m} - \hbar\omega - (i\hbar/\tau)}. \quad (5.6')$$

In terms of the scheme of reduced bands the relation between the polarization and the field is established in terms of the polarizability tensor, which depends explicitly on the reciprocal lattice vectors \mathbf{Q} and \mathbf{Q}' :

$$P_i(\mathbf{q}, \mathbf{Q}; \omega) = \sum_j \chi_{ij}^{Q, Q'}(\mathbf{q}, \omega) E_j(\mathbf{q}, \mathbf{Q}'; \omega), \quad (5.7)$$

where according to Eqs. (5.5) and (5.6) we have

$$\chi_{ij}^{Q, Q'}(\mathbf{q}, \omega) = -\frac{e^2}{m\omega^2 Q} \sum_{\mathbf{k}mn} \left[n(\varepsilon_{\mathbf{k}m}) (\mathbf{k}m | e^{-i(\mathbf{Q}-\mathbf{Q}')r} | \mathbf{k}n) \delta_{mn} \delta_{ij} + m^{-1} R(\mathbf{k}+\mathbf{q}, n; \mathbf{k}, m; \omega) (\mathbf{k}m | \left\{ p + \hbar \left[\mathbf{k} + \frac{1}{2} (\mathbf{q} + \mathbf{Q}) \right] \right\}_i e^{-i\mathbf{Q}r} | \mathbf{k}+q, n) \times (\mathbf{k}+q, n | e^{i\mathbf{Q}'r} \left\{ p + \hbar \left[\mathbf{k} + \frac{1}{2} (\mathbf{q} + \mathbf{Q}') \right] \right\}_j | \mathbf{k}m) \right]. \quad (5.8)$$

In contrast to the definition of the polarizability tensor $\chi_{ij}^{(H)}(\mathbf{k}, \omega)$ in Eq. (2.8), the polarizability (5.8) also can be considered to be a tensor in the indices \mathbf{Q} and \mathbf{Q}' .

From comparison of the definitions (2.8), (2.17), and (5.7) we obtain the following relations between the tensors $\hat{\chi}^{(H)}(\mathbf{k}, \omega)$ and $\hat{\chi}^{Q, Q'}(\mathbf{q}, \omega)$:

$$\chi_{ij}^{(H)}(\mathbf{k}, \omega) = \chi_{ij}^{Q, Q+H}(\mathbf{k}-\mathbf{Q}, \omega) \quad (5.9)$$

and reciprocally

$$\chi_{ij}^{Q, Q'}(\mathbf{q}, \omega) = \chi_{ij}^{Q'+Q}(\mathbf{q}+\mathbf{Q}, \omega),$$

where $\mathbf{k} = \mathbf{q} + \mathbf{Q}$.

The symmetry properties of the tensor $\chi_{ij}^{Q, Q'}$, which do not depend on the model of the medium, follow from the general relations for the tensor $\chi_{ij}^{(H)}$, which were obtained previously in Section 2(b). For example, the condition of hermiticity of the tensor $\chi_{ij}^{(H)}(\mathbf{k}, \omega)$ leads to the relation

$$\chi_{ij}^{Q, Q'}(\mathbf{q}, \omega) = [\chi_{ji}^{Q', Q}(\mathbf{q}, \omega)]^*,$$

which shows that the tensor $\chi_{ij}^{Q, Q'}$ is Hermitian simultaneously in the indices i and j and in the indices \mathbf{Q} and \mathbf{Q}' .

The rigorous calculation of the polarizability (5.8) requires a knowledge for each specific crystal structure of the Bloch functions, which usually are calculated

numerically with use of various pseudopotential models. The general expression (5.8) for the electronic polarizability includes the limiting cases of free and strongly bound electrons which are frequently used in the theory.

An exhaustive discussion of the permittivity of a degenerate free-electron gas, whose properties are extremely interesting in the ultraviolet frequency region ($\omega \lesssim \omega_p \approx 10^{16}$ Hz) is given in Pines' well known book.⁵²

c) Strong-coupling approximation

In the strong-coupling approximation the wave function of the Hamiltonian $\hat{\mathcal{H}}_0$ can be represented in the form of a superposition of eigenfunctions of free atoms. Strictly speaking, the strong-coupling approximation in this formulation cannot lead to an exact solution of the one-electron Schrödinger equation, since the wave functions of the bound states in an atom do not form a complete system of functions. This results from the overlapping of the electron orbitals of neighboring atoms in the lattice. This difficulty can be avoided by constructing an orthogonal system of localized Wannier functions.⁵³ The approximation involved is justified only for electrons located in sufficiently deep atomic shells whose orbit radius is small compared to the interatomic distance. However, it is just these electrons which give the principal contribution to the polarizability at x-ray frequencies [see Subsections c) and d) of Section 4].

Suppose that the unit cell of a crystal contains n_s atoms with coordinates \mathbf{r}_s . Then the Bloch function is

$$\varphi_{\mathbf{q}n} = N^{-1/2} \sum_s e^{i\mathbf{q}r_s} \psi_n(\mathbf{r} - \mathbf{r}_s - \mathbf{r}_s), \quad (5.10)$$

$$\int \psi_n^*(\mathbf{r} - \mathbf{r}_l - \mathbf{r}_s) \psi_{m'}(\mathbf{r} - \mathbf{r}_l' - \mathbf{r}_s) d\mathbf{r} = \delta_{mn'} \delta_{ll'} \delta_{ss'},$$

where φ_m is the wave function of an electron in state m with energy ε_m and \mathbf{r}_i is the position vector of the unit cell. We designate the matrix elements with respect to the atomic functions ψ_m as follows:

$$[\mathbf{B}(\mathbf{r})]_{mn} = \int_{\Omega} \psi_m^*(\mathbf{r}) \hat{\mathbf{B}}(\mathbf{r}) \psi_n(\mathbf{r}) d\mathbf{r}.$$

Using the definition (5.10) and going over in Eq. (5.8) from the matrix elements $\mathbf{k}m | \mathbf{B} | \mathbf{k}'m'$ to $[\mathbf{B}]_{mm'}$, with allowance for Eq. (5.9) we obtain for the polarizability the following expression:

$$\chi_{ij}^{(H)}(\mathbf{k}, \omega) = -\frac{Ne^2}{m\omega^2} F_{ij}(\mathbf{H}, \mathbf{k}; \omega), \quad (5.11)$$

where F_{ij} is the tensor of the structure factor

$$F_{ij}(\mathbf{H}, \mathbf{k}; \omega) = \sum_s e^{i\mathbf{H}r_s} f_{ij}^{(s)}(\mathbf{H}, \mathbf{k}; \omega),$$

$$f_{ij}^{(s)}(\mathbf{H}, \mathbf{k}; \omega) = f_{ij}^{(s)}(\mathbf{H}) \delta_{ij} + \Delta f_{ij}^{(s)}(\mathbf{H}, \mathbf{k}; \omega),$$

$f_{ij}^{(s)}$ is the atomic structure factor of the s -th atom, $f_0^{(s)}(\mathbf{H}) = \sum_m n(\varepsilon_m) [e^{i\mathbf{H}r}]_{mm}^{(s)}$ is the potential part of the atomic structure factor, and $\Delta f_{ij}^{(s)}$ is the dispersion correction, which has the form

$$\Delta f_{ij}(\mathbf{H}, \mathbf{k}; \omega) = m^{-1} \sum_{mn} R_{mn} [B_j(-\mathbf{k})]_{mn} [B_j(\mathbf{k}+\mathbf{H})]_{nm}, \quad (5.12)$$

where

$$\mathbf{B}(\mathbf{k}) \equiv e^{i\mathbf{k}r} \left(\mathbf{p} + \frac{\hbar\mathbf{k}}{2} \right), \quad R_{mn} \equiv \frac{n(\varepsilon_m) - n(\varepsilon_n)}{\hbar[\omega_{mn} - \omega - (i/\tau)]}.$$

The appearance of the term $\hbar\mathbf{k}/2$ in the expression for

the operator $B(\mathbf{k})$ in Eq. (5.12) [compare with Eq. (4.11b)] is due to the representation of the Bloch wave function (5.10) in the form of a superposition of localized atomic wave functions.

On the basis of the specific form of the polarizability (5.11) it is easy to check the validity of the general symmetry relations (2.12), (2.13), and (2.14). According to Eqs. (5.11) and (5.12), spatial-dispersion effects are included only in Δf_{ij} .

The magnitude of this term is significant only for radiation with frequency $\omega \approx \omega_{mn}$. As follows from Eq. (5.12) and the expression for $B(\mathbf{k})$, the spatial-dispersion parameter is $\hbar k/p \approx Z/137$, where $p \approx \omega_{mn} m a_0$. In the case of light atoms, consequently, the spatial dispersion is expressed weakly and can be neglected. Then the expression (5.12) takes the simpler form

$$\Delta f_{ij}(\omega) = \sum_{m \neq n} n(\epsilon_m) \frac{\omega_{mn}^2 g_{ij}^{mn}}{\omega_{mn}^2 - \omega^2 - (2i\omega/\tau)}$$

and is a generalization of the expressions (4.17) and (4.19) given previously for the dispersion correction.

6. CONCLUSION

In the present review we have devoted the principal attention to a systematic introduction of the permittivity tensor in the x-ray frequency region and to a description of its properties, based on use of the most fundamental properties of crystals—spatial periodicity. Study of the dielectric properties of a medium in the x-ray region permits description of its crystalline structure, electronic structure, and the structure of the internal electric and magnetic fields, in the same way that investigation of the propagation and scattering of light provides information on the atomic and molecular structure of matter.

Review of the current state of research on the dielectric properties of media in the x-ray and γ -ray regions shows that along with the substantial experimental and theoretical achievements, there exist problems of a fundamental nature which are still awaiting solution. The dielectric formalism gives a universal description of the properties of a medium interacting with an electromagnetic field, and therefore progress in study of dielectric properties must be accompanied by further development of this technique. The fact that a need for this has not existed for many years reflects only the correspondence of the stage of development achieved in x-ray experiments to the strong-coupling approximation.⁷⁾ Since there were no radiation sources with high

⁷⁾As we have already noted above, in the strong-coupling approximation the overlap of the outer electron shells is not taken into account and for this reason the effects of chemical bonding are left aside. Nevertheless, in the presence of anisotropic chemical bonds it is necessary to use general tensor expressions for the dispersion part of the permittivity tensor (4.11b) and (5.8). Goal-oriented experimental study of the effect of anisotropy of chemical bonds on the dielectric properties of the medium, carried out in Ref. 96, has shown that in the x-ray frequency region anisotropy of absorption and rotation of polarization are observed.

coherence properties and the external perturbations were not sufficiently strong, it appeared that we were in no position to study the finer effects or to affect in a desired manner the dielectric properties of a medium in this region. At the present time there is every basis to expect significant progress in this field. Below we shall mention some of the most promising directions of development of theory and experiment in the field of interaction of x rays and γ rays with matter.

a) From the experimental point of view it is necessary first of all to have sources of x rays and isotopic γ rays which would have spectral characteristics (both in frequency and in space) better by 2–4 orders of magnitude than existing ones.⁸⁾ As an illustration of what can be accomplished by an increase in the coherence properties of the radiation, we mention that the well known Darwin curve of reflection from a perfect crystal, predicted by the dynamical theory of diffraction many years before its experimental verification, was observed for the first time in pure form only with use of the extraordinarily monochromatic radiation of the Mössbauer isotope Co^{57} . Apparently the most realistic means of obtaining radiation with substantially better coherence properties at the present time is the use of synchrotron radiation, which already is being applied successfully to investigations in solid-state physics. At the present time in a number of countries including the Soviet Union there are being built and put into service synchrotrons with characteristics which satisfy the condition imposed above. In addition to high luminosity, synchrotron radiation has a high degree of polarization, which is particularly important in study of the anisotropy of crystals, and the possibility of smooth tuning of the frequency over a wide range makes it an irreplaceable means of investigation of frequency-dispersion effects. The properties and possibilities of application of synchrotron radiation in the x-ray region are analyzed in the review by Kulipanov and Skriskii.⁸⁹

b) Recently there has been a distinct tendency toward investigation of dielectric properties of crystals in the x-ray region in the presence of an active external perturbation of the medium, which obviously is a new stage in the development of this field of physics. The external perturbations which can be used include constant and variable electromagnetic fields, laser radiation, ultrasound, charged-particle beams, and so forth.

A number of recent publications have discussed the effect of several of the perturbations mentioned on the process of dynamical diffraction⁹⁰ and photoelectric absorption.⁹¹ Entin⁹⁰ has experimentally observed a resonance in the integrated intensity of reflection of Laue-diffracted x rays in a silicon plate placed in the field of an ultrasonic wave which modulates the electron density of the crystal and in this way affects its dielectric permittivity.

One of the possible mechanisms of affecting the diel-

⁸⁾This would immediately revive interest in such old problems as the "magnetic" scattering of x rays in magnetically ordered crystals.

electric properties of a medium in the x-ray frequency region by irradiating it by a laser wave consists of a change in the wave functions of the electrons in the final state. Here the dependence of the absorption cross section on the parameters of the laser radiation permits the information to be obtained on the structure of partially occupied and completely free electron states of the medium. According to the estimates of Tzoar and Jain,⁹¹ the imaginary part of the permittivity, which is responsible for the absorption, can be increased by an order of magnitude with the laser powers achievable at the present time.

In Subsection d) of Section 4 we noted that occupied electron states of atoms do not contribute to the dispersion terms. However, under the action of sufficiently intense electron or x-ray irradiation these states can be vacated, and this should lead to appearance of additional resonance contributions to the permittivity. The possibility of using this effect for generation of coherent x rays on passage of charged particles through a crystal has been discussed theoretically by Akhmanov and Grishanin.⁹²

c) In the area of development of the theory it is appropriate to point out the following aspects. First, further systematic development and use of the apparatus of the theory of the dielectric formalism is necessary in treatment of the entire complex of problems of interaction of x rays and γ rays with a medium. From the material presented in the review it follows that the current theory is not free of approximations which limit its applicability. The theory of the permittivity tensor is constructed on the approximation of an unbounded crystal, which for a certain number of problems can be considered quite good. However, from the fundamental point of view it is unsatisfactory, since real crystals are bounded, contain as a rule a significant number of defects, have a mosaic structure, and only very crudely can be treated as ideal. All this indicates the need of constructing a theory of the permittivity tensor of real crystals. We can indicate the following path of construction of such a theory.

In the Darwin model, real crystals are an assemblage of small crystals slightly disoriented and (or) displaced with respect to each other. For a bounded single crystal Eq. (2.7a) goes over into a similar equation where instead of the δ function arising from the assumption of an unbounded crystal the function $\delta_V = (2\pi)^{-3} \int_V \exp[i(\mathbf{k}' - \mathbf{k} - \mathbf{H}(\mathbf{r})\mathbf{r})\mathbf{r}]d\mathbf{r}$ appears. The dependence of \mathbf{H} on \mathbf{r} corresponds to taking into account local distortion of the crystal lattice.

On integration within the bounds of volume within which the crystal can be considered sufficiently perfect (the coherence volume), δ_V is none other than the Laue interference function with a finite width $\Delta k \sim l^{-1}$, where l is the characteristic parameter of the coherence region. Here the diagram of the directivity of the δ_V function for each block of the mosaic has its own direction, which differs somewhat from the others. Integration over the region of strong distortions (interblock boundaries, etc.) leads to the condition $\mathbf{k}' = \mathbf{k}$, which corresponds to a diffraction-free propagation of

the radiation forward. On the other hand, the irregularity of the interblock boundaries leads to partial or complete loss of the phase relations between the different blocks of the mosaic. In other words, the correlation between currents induced in different coherence volumes of the medium is destroyed, and as a result the total current, and consequently also the permittivity, acquire a dependence on the coordinates with a characteristic scale of the order of the linear dimensions of the coherence volume.

One of the possible approaches to treatment of the dielectric properties of a crystal with defects is the approach developed by Afanas'ev and Kohn⁹³ in construction of a dynamical theory of Bragg diffraction in such crystals. The authors considered defects as special regions where the regular periodicity of the crystal is destroyed, and introduced a polarizability which depends on the local degree of distortion of the crystal lattice.

Thus, the apparatus of dielectric permittivity, which is a generally accepted and the most effective instrument for conducting research in the optical region of the spectrum, can be systematically developed also in x-ray and γ -ray optics with incorporation of the specific features of these regions. On the basis of the general features pertaining to the dielectric approach to these regions of the spectrum, it is possible to formulate the problem of developing new directions of research in the x-ray region, similar to the directions which have already received development in optics. One of these directions, in particular, may be the search for additional waves in the x-ray region, for example, for narrow Mössbauer or x-ray exciton lines.

The productiveness of this type of analogy has already been demonstrated in description of elastic scattering of x rays in crystals.⁹⁴ In treating the diffraction of visible light in certain types of liquid crystals, Belyakov and Dmitrienko,⁹⁵ on the other hand, used the method of the dynamical theory of diffraction of x rays, proceeding from the fact that in this case the parameter a/λ also can be of order unity.

Another fundamental experimental and theoretical problem which is being stimulated by already existing experimental studies is that of constructing the x-ray permittivity of nonequilibrium media, examples of which are a high-temperature plasma and a solid-state plasma at high excitation levels, and also the active medium of an x-ray or γ -ray laser.

We can therefore see that the development of coherent x-ray and γ -ray optics at the present time depends on improvement of experimental accuracy, increase of the spectral intensity of radiation sources, and further detailed development of the theory of dielectric properties of crystals (and not only of crystals) in this range of wavelengths.

In conclusion the authors express their sincere indebtedness to M. A. Andreeva, S. F. Borisova, E. N. Ovchinnikova, and Yu. P. Khapachev for numerous helpful discussions of the problems outlined in this review.

- ¹M. von Laue, *Ergeb. exakt. Naturwiss.* **10**, 133 (1931).
- ²M. Kohler, *Sitzungsber. Preuss. Akad. Wiss. Kl, Math. und Phys.*, 1935, p. 334.
- ³G. Molière, *Ann. Phys. (Berlin)* **35**, 272, 279 (1939); **36**, 265 (1939).
- ⁴U. Bonse and M. Hart, *Z Phys.* **194**, 1 (1966). M. Hart, *Proc. Roy. Soc. Ser. A* **346**, 1 (1975). U. Bonse, W. Graeff, and G. Materlink, *Rev. Phys. Appl.* **11**, 83 (1976). F. O. Éíramdzhyan, K. G. Truni, and P. A. Bezirganyan, *Uch. zap. Erev. un-ta. Estestv. nauki (Scholarly Writings of Erevan University Natural Sciences)* No. 3 (217), 49, 1974.
- ⁵L. A. Rivlin, *Voprosy radioélektron. Ser. 1, No. 6*, 60 (1962). A. V. Kolpakov, A. V. Kuz'min, and V. M. Ryaboy, *J. Appl. Phys.* **41**, 3549 (1970). R. A. Fisher, *Appl. Phys. Lett.* **24**, 598 (1974). A. Yariv, *Appl. Phys. Lett.* **25**, 105 (1974). G. A. Lyakhov, *Kvantovaya Élektron. (Moscow)* **3**, 838 (1976) [*Sov. J. Quantum Electron.* **6**, 456 (1976)].
- ⁶A. V. Kolpakov, B. Sh. Galyamov, and R. N. Kuz'min, *Vestn. Mosk. un-ta. Ser. Fiz.-astron. (Moscow University Physics Bulletin)* No. 3, 369 (1973). V. L. Indenbom, *Kristallografiya* **21**, 479 (1976) [*Sov. Phys. Crystallogr.* **21**, 264 (1976)].
- ⁷A. G. Molchanov, *Usp. Fiz. Nauk* **106**, 165 (1972) [*Sov. Phys. Uspekhi* **15**, 124 (1972)]. V. A. Bushuev and R. N. Kuz'min, *Usp. Fiz. Nauk* **114**, 677 (1974) [*Sov. Phys. Uspekhi* **17**, 942 (1975)]. G. C. Baldwin and R. V. Kokhlov, *Physics Today*, February 1975, p. 32. V. A. Bushuev and R. N. Kuz'min, *Problemy sozdaniya lazerov rentgenovskogo diapazona dlín voln. (Problems of Producing Lasers in the X-ray Wavelength Region)*, Moscow, Znanie, 1976.
- ⁸I. Freund and B. F. Levine, *Phys. Rev. Lett.* **23**, 854 (1969); **25**, 1241 (1970). P. Eisenberger and S. L. McCall, *Phys. Rev. Lett.* **26**, 684 (1971). S. S. Jha and J. W. F. Woo, *Nuovo Cimento B10*, 229 (1972). I. V. Krasnov and N. Ya. Shaparev, *Kvantovaya Élektron. (Moscow)* **2**, 2604 (1975) [*Sov. J. Quantum Electron.* **5**, 1420 (1975)].
- ⁹V. S. Shpínel', *Rezonans gamma-lucheí v kristallakh (Gamma-Ray Resonance in Crystals)*, Moscow, Nauka, 1969.
- ¹⁰J. P. Hannon and G. T. Trammell, *Phys. Rev.* **169**, 315 (1968).
- ¹¹J. P. Hannon and G. T. Trammell, *Phys. Rev.* **186**, 306 (1969).
- ¹²J. P. Hannon, N. J. Carron, and G. T. Trammell, *Phys. Rev.* **B9**, 2791, 2810 (1974).
- ¹³V. A. Belyakov, *Usp. Fiz. Nauk* **115**, 553 (1975) [*Sov. Phys. Uspekhi* **18**, 267 (1975)].
- ¹⁴V. G. Baryshevskii, *Yadernaya optika polarizovannykh sred (Nuclear Optics of Polarized Media)*, Minsk, Belorussian University, 1976.
- ¹⁵G. Rosenbaum, H. C. Holmes, and J. Witz, *Nature* **230**, No. 5294, 434 (1971). M. Hart, *J. Appl. Crystallogr.* **8**, 436 (1975). J. Bordas, A. M. Galazer, and H. Hauser, *Phil. Mag.* **32**, 471 (1975).
- ¹⁶L. D. Landau and E. M. Lifshits, *Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media)*, Moscow, Gostekhizdat, 1957.
- ¹⁷V. M. Agranovich and V. L. Ginzburg, *Kristallografiya s uchétom prostranstvennoí dispersii i teoriya éksitonov (Crystal Optics With Inclusion of Spatial Dispersion and the Theory of Excitons)*, Moscow, Nauka, 1965.
- ¹⁸R. W. James, *The Optical Principles of Diffraction of X Rays*, G. Bell and Sons, Ltd., London, 1948. Russ. transl., IL, 1950.
- ¹⁹V. I. Iveronova and G. P. Revkevich, *Teoriya rasseyaniya rentgenovskikh lucheí (Theory of Scattering of X Rays)*, Moscow, Moscow University, 1972.
- ²⁰Z. G. Pinsker, *Dinamicheskoe rasseyanie rentgenovskikh lucheí v ideal'nykh kristallakh (Dynamical Scattering of X Rays in Ideal Crystals)*, Moscow, Nauka, 1974.
- ²¹M. L. Levin and S. M. Rytov, *Teoriya ravnovesnykh teplovykh fluktuatsii v élektrodinamike (Theory of Equilibrium Thermal Fluctuations in Electrodynamics)*, Moscow, Nauka, 1967.
- ²²V. A. Koptsik and I. N. Kotsev, *JINR Communications*, Dubna, R 4-8067, R 4-8086, 1974. V. A. Koptsik, *Kristall und Technik* **10**, 231 (1975).
- ²³A. V. Kolpakov, E. N. Ovchinnikova, and R. N. Kuz'min, *Kristallografiya* **20**, 233 (1975); **21**, 901 (1977) [*Sov. Phys. Crystallogr.* **20**, 135 (1975); **22**, 517 (1977)].
- ²⁴M. J. Buerger, *Crystal Structure Analysis*, 1960. Vector Space, 1959. Wiley, N. Y., Russ. transl., IL, 1961.
- ²⁵H. M. Nussenzveig, *Causality and Dispersion Relations*, Academic Press, 1972. Russ. transl., Mir, 1976.
- ²⁶R. G. Newton, *Scattering Theory of Waves and Particles*, N. Y., McGraw-Hill, 1966. Russ. transl., Mir, 1969.
- ²⁷A. V. Kolpakov, *Yad. Fiz.* **16**, 1003 (1972) [*Sov. J. Nucl. Phys.* **16**, 554 (1973)].
- ²⁸A. H. Compton and S. K. Allison, *X Rays in Theory and Experiment*, N. Y., D. Van Nostrand, Second Edition, 1935. Russ. transl., Moscow-Leningrad, Gostekhizdat, 1941.
- ²⁹M. A. Blokhin, *Fizika rentgenovskikh lucheí (Physics of X Rays)*, Moscow, Gostekhizdat, 1957.
- ³⁰G. D. Mahan, *Phys. Rev.* **163**, 612 (1967) P. Nozières and C. T. de Dominicis, *Phys. Rev.* **178**, 1097 (1969). G. A. Ausman, Jr., and A. J. Glick, *Phys. Rev.* **183**, 687 (1969). H. Petersen, *Phys. Rev. Lett.* **35**, 1363 (1975). A. A. Maiste, A. M. E. Saar, and M. A. Élango, *Fiz. Tverd. Tela (Leningrad)* **16**, 1720 (1974) [*Sov. Phys. Solid State* **16**, 1118 (1974)]. K. I. Karakhanyan, E. M. Kazaryan, and P. A. Bezirganyan, *Fiz. Tverd. Tela (Leningrad)* **18**, 511 (1976) [*Sov. Phys. Solid State* **18**, 295 (1976)].
- ³¹H. Frauenfelder, D. E. Nagle, R. D. Taylor, D. R. F. Cochran, and W. M. Visscher, *Phys. Rev.* **126**, 1065 (1962).
- ³²M. Blume and O. C. Kistner, *Phys. Rev.* **171**, 417 (1968).
- ³³R. M. Housley, R. W. Grant, and U. Gonser, *Phys. Rev.* **178**, 514 (1969).
- ³⁴Yu. A. Izyumov and R. P. Ozerov, *Magnitnaya neitronografiya (Magnetic Neutronography)* Moscow, Nauka, 1966.
- ³⁵P. Imbert, *Phys. Lett.* **8**, 95 (1964); *J. Phys. (Paris)* **27**, 429 (1966).
- ³⁶R. M. Housley and U. Gonser, *Phys. Rev.* **171**, 480 (1968).
- ³⁷R. W. Grant, R. M. Housley, and U. Gonser, *Phys. Rev.* **178**, 523 (1969).
- ³⁸T. C. Gibb, *J. Phys.* **C7**, 10001 (1974).
- ³⁹V. I. Gol'danskii, E. F. Makarov, I. P. Suzdalev, and I. A. Vinogradov, *Zh. Eksp. Teor. Fiz.* **58**, 760 (1970) [*Sov. Phys. JETP* **31**, 407 (1970)].
- ⁴⁰V. G. Labushkin, S. N. Ivanov, and G. V. Chechin, *Pis'ma Zh. Eksp. Teor. Fiz.* **20**, 349 (1974) [*JETP Lett.* **20**, 157 (1974)].
- ⁴¹Yu. M. Áivazyán and V. A. Belyakov, *Fiz. Tverd. Tela (Leningrad)* **13**, 968 (1971) [*Sov. Phys. Solid State* **13**, 808 (1971)].
- ⁴²Yu. M. Áivazyán, *Izv. AN Arm. SSR Ser. "Fizika"* **9**, 374 (1974).
- ⁴³B. L. Chrisman, *Nucl. Phys.* **A186**, 264 (1972).
- ⁴⁴A. V. Mitin, *Phys. Stat. Sol.* **B53**, 93 (1972). F. Hartman-Boutron, *J. Phys. (Paris)* **37**, 537, 549 (1976). D. Barb and M. Rogalski, *J. Phys. (Paris)* **37**, 17 (1976).
- ⁴⁵S. Bernstein and E. C. Campbell, *Phys. Rev.* **132**, 1625 (1963). S. Bernstein, E. C. Campbell, and C. W. Nestor, Jr., *J. Phys. and Chem. Sol.* **26**, 883 (1965).
- ⁴⁶A. M. Afanas'ev and Yu. Kagan, *Pis'ma Zh. Eksp. Teor. Fiz.* **2**, 130 (1965) [*JETP Lett.* **2**, 81 (1965)]. Yu. Kagan and A. M. Afanas'ev, *Zh. Eksp. Teor. Fiz.* **50**, 271 (1966) [*Sov. Phys. JETP* **23**, 178 (1966)]. A. M. Afanas'ev and Yu. Kagan, *Zh. Eksp. Teor. Fiz.* **52**, 191 (1967) [*Sov. Phys. JETP* **25**, 124 (1967)].
- ⁴⁷Fam Zui Khien, *Zh. Eksp. Teor. Fiz.* **59**, 201 (1970); **61**, 35 (1971) [*Sov. Phys. JETP* **32**, 111 (1971); **34**, 191 (1972)].
- ⁴⁸V. M. Agranovich, *Usp. Fiz. Nauk* **112**, 143 (1974) [*Sov.*

- Phys. Uspekhi 17, 103 (1974)].
- ⁴⁹M. Born and E. Wolf, Principles of Optics, Pergamon, N. Y., 1959, Russ. Transl., Moscow, Nauka, 1970.
- ⁵⁰P. P. Ewald, Ann. d. Phys. 49, 1 (1916).
- ⁵¹M. von Laue, Röntgenstrahlinterferenzen, Frankfurt/M.: 1960.
- ⁵²D. Pines, Elementary Excitations in Solids, Benjamin, 1963. Russ. transl., Mir, 1965.
- ⁵³L. D. Landau and E. M. Lifshits, Kvantovaya mekhanika (Quantum Mechanics), Moscow, Fizmatgiz, 1963.
- ⁵⁴W. Heitler, The Quantum Theory of Radiation, London, Oxford Univ. Press, 3rd ed., 1954. Russ. transl., IL, 1956.
- ⁵⁵A. M. Afanas'ev and Yu. Kagan, Acta Crystallogr. A 24, 163 (1967).
- ⁵⁶D. T. Keating, Phys. Rev. 178, 732 (1969). S. S. Jha and W. F. Woo, Nuovo Cimento B 10, 229 (1972).
- ⁵⁷M. Lax, Rev. Mod. Phys. 23, 287 (1951).
- ⁵⁸M. Renninger, Z. tech. Phys. 16, 440 (1935).
- ⁵⁹S. T. Konobeevskii, Usp. Fiz. Nauk 44, 21 (1951). B. Downson and P. Sanger, Proc. Roy. Soc. A301, 195 (1967). D. J. Stukel and R. N. Euwema, Phys. Rev. B1, 1635 (1970). C. M. Bertoni, V. Bortolani, C. Calandra, and F. Nizzoli, J. Phys. C6, 3612 (1973). Yu. A. Rozenberg and L. I. Kleshchinskii, Fiz. Tverd. Tela (Leningrad) 18, 2687 (1976) [Sov. Phys. Solid State 18, 1567 (1976)].
- ⁶⁰A. D. Buckingham, Acta Crystallogr. 17, 960 (1964).
- ⁶¹L. R. Saravia and S. Caticha-Ellis, Acta Crystallogr. 20, 927 (1966). D. T. Cromer, Acta Crystallogr. 18, 17 (1965). D. T. Cromer and D. Liberman, J. Chem. Phys. 53, 1891 (1970).
- ⁶²H. Wagenfeld, in: Anomalous Scattering: Proceedings of an Inter-Congress Conference, Madrid, Spain 22-26 April, 1974, Copenhagen, 1975.
- ⁶³A. Sommerfeld, Atombau und Spektrallinien, Braunschweig, 1939, Vol. 2. Russ. transl. Gostekhizdat, 1956.
- ⁶⁴R. L. de Kronig, J. Opt. Soc. Am. 12, 547 (1926).
- ⁶⁵J. A. Prins, Z. Phys. 47, 479 (1928). E. J. Williams, Proc. Roy. Soc. A143, 358 (1934).
- ⁶⁶H. Kallmann and H. Mark, Naturwissenschaften 14, 648 (1926); Ann. Phys. (Berlin) 82, 585 (1927).
- ⁶⁷Y. Sugiura, J. Phys. et Radium 8, 113 (1927). H. Hönl, Z. Phys. 84, 1 (1933).
- ⁶⁸H. Wagenfeld, Phys. Rev. 144, 216 (1966).
- ⁶⁹A. A. Maradudin, E. W. Montroll, and G. H. Weiss, Theory of Lattice Dynamics in the Harmonic Approximation, 2nd ed., Solid State Phys., Suppl. 3, 1971, Academic Press. Russ. transl., 1st ed., Mir, 1965.
- ⁷⁰M. Born and K. Huang, Dynamical Theory of Crystal Lattices, Oxford University Press, N. Y., 1954. Russ. transl., IL, 1958.
- ⁷¹V. M. Agranovich and Yu. V. Konobeev, Fiz. Tverd. Tela (Leningrad) 3, 360 (1961) [Sov. Phys. Solid State 3, 260 (1961)].
- ⁷²O. N. Efimov, Phys. Stat. Sol. 22, 297 (1967).
- ⁷³T. O. Baldwin, F. Young, and A. Merlini, Phys. Rev. 163, 591 (1967).
- ⁷⁴H. Sano, K. Ohtaka, and Y.-H. Ohtsuki, J. Phys. Soc. Japan 27, 1254 (1969).
- ⁷⁵W. Kohn, Kristallografiya 15, 20 (1970) [Sov. Phys. Crystallogr. 15, 14 (1970)].
- ⁷⁶V. A. Bushuev and R. N. Kuz'min, Zh. Tekh. Fiz. 44, 2568 (1974) [Sov. Phys. Tech. Phys. 19, 1590 (1975)].
- ⁷⁷Yu. Kagan and A. M. Afanas'ev, Zh. Eksp. Teor. Fiz. 49, 1504 (1965) [Sov. Phys. JETP 22, 1032 (1966)].
- ⁷⁸V. B. Berestetskii, E. M. Lifshits, and L. P. Pitaevskii, Relyativistskaya kvantovaya teoriya, Part 1, Moscow, Nauka, 1968. Engl. transl., Relativistic Quantum Theory, Addison-Wesley, 1971.
- ⁷⁹A. I. Akhiezer and V. B. Berestetskii, Kvantovaya élektrodinamika, Moscow, Nauka, 1969. English transl. of an earlier edition, Quantum Electrodynamics, New York, Wiley, 1965.
- ⁸⁰L. L. Foldy, Phys. Rev. 67, 107 (1945).
- ⁸¹G. T. Trammell, Phys. Rev. 126, 1045 (1962).
- ⁸²S. L. Adler, Phys. Rev. 126, 413 (1962).
- ⁸³N. Wisner, Phys. Rev. 129, 62 (1963).
- ⁸⁴J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962).
- ⁸⁵H. Cheng and P. B. Miller, Phys. Rev. 134, A683 (1964).
- ⁸⁶A. S. Pine, Phys. Rev. 139, A901 (1965).
- ⁸⁷C. Kittel, Quantum Theory of Solids, Wiley, N. Y., 1963. Russ. transl., Moscow, Nauka, 1967.
- ⁸⁸J. Calloway, Theory of Energy Band Structure, Russ. transl., Mir, 1969.
- ⁸⁹G. N. Kulipanov and A. N. Skrin'skii, Usp. Fiz. Nauk 122, 369 (1977) [Sov. Phys. Uspekhi 20, 559 (1977)].
- ⁹⁰I. R. Entin, Pis'ma Zh. Eksp. Teor. Fiz. 26, 392 (1977) [JETP Lett. 26, 269 (1977)].
- ⁹¹N. Tzoar and M. Jain, Optics Comm. 19, 417 (1976). M. Jain and N. Tzoar, Phys. Rev. A15, 147 (1977).
- ⁹²S. A. Akhmanov and B. A. Grishanin, Pis'ma Zh. Eksp. Teor. Fiz. 23, 562 (1976) [JETP Lett. 23, 515 (1976)].
- ⁹³A. M. Afanas'ev and V. G. Kohn, Acta Cryst. A 27, 421 (1971).
- ⁹⁴V. A. Bushuev and R. N. Kuz'min, Usp. Fiz. Nauk 122, 81 (1977) [Sov. Phys. Uspekhi 20, 406 (1977)].
- ⁹⁵V. A. Belyakov and V. E. Dmitrienko, Fiz. Tverd. Tela (Leningrad) 18, 2880 (1976) [Sov. Phys. Solid State 18, 1681 (1976)].
- ⁹⁶G. G. Cohen and M. Kuriyama, Phys. Rev. Lett. 40, 957 (1978).

Translated by Clark S. Robinson