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DIFFRACTION AND SCATTERING OF X-RAYS AND SYNCHROTRON RADIATION

Laue Diffraction of X-rays in GaAs at the Zero Value of the Real Part of the Structure Factor for Quasiforbidden Reflections

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Abstract—Specific features of the Laue diffraction of X-rays are considered for quasiforbidden reflections within the wavelength range between the *K*-absorption edges of Ga and As in a GaAs crystal. It is established that scattering for the 200 reflection is of the dynamical nature in the wavelength range where the real part of the structure factor F_{r200} is zero and diffraction is determined by the imaginary part of the atomic form factor $f''(\omega)$ alone. It is shown that the position of the minimum of the function $R_i = f(\lambda)$ is sensitive to the crystal nonstoichiometry predicted by the numerical calculations within the theory taking into account both the real and the imaginary parts of the structure factor. © *2000 MAIK "Nauka/Interperiodica".*

INTRODUCTION

The study of X-ray scattering at the wavelengths close to those of the *K*-absorption edge of Ga in GaAs crystals with the use of quasiforbidden Laue reflections has experimentally confirmed the dynamical character of the interaction between the X-ray radiation and the crystal lattice (Pendellösung intensity oscillations) and allowed the determination of some parameters of structural perfection of the samples such as the static Debye–Waller factor L, coefficient of intensity reduction due to diffuse scattering μ_{ds}, and the nonstoichiometry parameter [1–3]. Earlier [4, 5], it was shown that the intensity of the diffraction maximum in the vicinity of the *K*-absorption edge of the material should be described by the formulas of the dynamical theory of scattering, which takes into account the contributions of both real and imaginary parts of the atomic scattering function into the crystal polarizability χ. In the studies of X-ray diffraction for quasiforbidden reflections, of special interest is the situation where the real part of the structure factor for the *h*th reflection with due regard for the corrections for the atomic scattering function for anomalous dispersion, $f_0(\mathbf{h}) + f'(\omega)$, equals zero. The modified theory of X-ray scattering for this case established the existence of the Pendellösung fringes of intensity oscillations on the rocking curve for the Laue-diffraction [6]. It was also established that Pendellösung fringes are described by the imaginary part of the atomic scattering function alone. In the case of Bragg diffraction, narrow rocking curves were observed. The situation where $f_0(\mathbf{h}) + f'(\omega) = 0$ was observed experimentally in the measurements of integrated intensities in the vicinity of the *K*-absorption edge of Ge [7]. An attempt to use this wavelength range to control the nonstoichiometry parameter of crystals in the Bragg diffraction geometry was made in [8, 9], where the authors assumed that X-ray scattering is of purely kinematical character, i.e., that the total integrated reflectivity of a crystal consists of two components—those of coherent and diffuse scattering—and, according to [10], is independent of the structure defects [10].

Below, we describe the study of X-ray scattering for quasiforbidden reflections in real GaAs crystals in the case where the real part of the structure factor vanishes. We also investigated the effect of structure defects and crystal nonstoichiometry on the behavior of integrated intensities of the diffraction maxima in this wavelength range.

THEORETICAL CONSIDERATION

As is well known, the atomic scattering factor has the form

$$f = f_0(\mathbf{h}) + f'(\omega) + if''(\omega), \quad (1)$$

where $f_0(\mathbf{h})$ is the normal value of the atomic scattering factor for the h reflection, and $f'(\omega)$ and $f''(\omega)$ the frequency-dependent real and imaginary corrections to $f_0(\mathbf{h})$ caused by anomalous dispersion. These corrections are especially important in the vicinity of the *K*-absorption edges of the crystal components. It is seen from (1) that the condition $f_0(\mathbf{h}) + f'(\omega) = 0$ is fulfilled for reflections with large absolute values of the diffraction vector (since $f_0(\mathbf{h})$ decreases with an increase of $\mathbf{h}$) in the vicinity of the *K*-edge, where the corrections $f'(\omega)$ are especially pronounced. The case where the

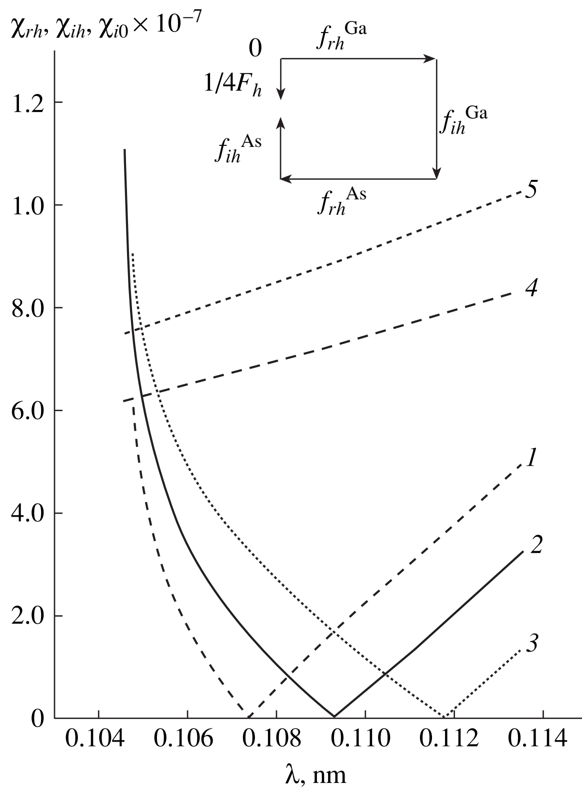

Fig. 1. Energy dependence of the real (curves *1–3*) and imaginary (curve *4*) parts of the Fourier coefficient of GaAs polarizability for crystals with various nonstoichiometries ($c_{Ga} - c_{As}$): (*1*) –0.03, (*2*) 0, (*3*) 0.03, (*5*) the value of χ_{i0}. 200 reflection. In the inset: the vector diagram of the contributions of the real and the imaginary parts of atomic Ga and As form factors to the structure factor of the quasiforbidden 200 reflection.

structure factor F_h is determined by the imaginary part of f alone was considered for a Ge crystal [7]. The situation where $F_{rh} \longrightarrow 0$ can readily be obtained for binary crystals, e.g., GaAs, by using the quasiforbidden 200 reflection and the wavelength range between the K-absorption edges of the crystal components ($F_{rh} \sim [c_{Ga}(f_{Ga} + f'_{Ga}) - c_{As}(f_{As} + f'_{As})] \longrightarrow 0$, where c_{Ga} and c_{As} are the atomic concentrations of Ga and As, respectively). For the reflections of the $h + k + l = 4n + 2$ type with $n = 0, 1, \ldots$ the atoms in the sphalerite-type lattice scatter the X-ray radiation in the counterphase. In this wavelength range, the imaginary part of the atomic scattering function, $f''(\omega)$, has quite a large value because of the jump in absorption at the K-absorption edge of gallium ($f''(\omega) \geq f_0(\mathbf{h}) + f'(\omega)$). Figure 1 shows the vector diagram of the structure factor of the 200 reflection for the wavelength $\lambda = 0.1093$ nm which shows that the resulting value, $1/4F_h$, is imaginary. Figure 1 also shows energy dependence of the Fourier coefficients of polarizability, $\chi_{rh} = -(e^2/mc^2)(\lambda^2/\pi V)F_{rh}$ and $\chi_{ih} = -(e^2/mc^2)(\lambda^2/\pi V)F_{ih}$, for the quasiforbidden 200 reflection in GaAs in the above indicated wavelength range. Here e^2/mc^2 is the classical radius of an electron, λ is the X-ray wavelength, and V is the unit-cell volume of the crystal, and $F_{ih} \sim f''(\omega)$. The dispersion corrections to the atomic form factor $f'(\omega)$ and $f''(\omega)$ were calculated by the method described in [11] with due regard for the oscillator strengths taken from [12]. It is seen that for the wavelength close to 0.109 nm, we have $\chi_{ih} \gg \chi_{rh}$. The integrated reflectivity of an ideal crystal for this case can be calculated by the formula of the modified theory [13] as

$$R_i = (\gamma_h/\gamma_0)^{1/2}(|\chi_h|/\sin 2\vartheta)\int R_i^y dy,$$

$$R_i^y = \exp(-\mu t')(1 - 2p\sin\delta) \quad (2)$$

$$\times [\sin^2((t/\Lambda)\mathrm{Re}M^{1/2}) + \sinh^2((t/\Lambda)\mathrm{Im}M^{1/2})],$$

where t is the crystal thickness; $t' = t\cos\vartheta\sin\beta/\gamma_0\gamma_h$; $\Lambda = \lambda(\gamma_0\gamma_h)^{1/2}/\pi|\chi_h|$ is the extinction length; γ_0 and γ_h are the directional cosines of the incident and reflected X-ray beams, respectively; ϑ is the Bragg angle; and β is the angle formed by the crystal surface and the reflecting planes. An important parameter entering (2) is the quantity M dependent on both χ_{rh} and χ_{ih}. All the remaining symbols correspond to those used in [13]. The influence of defects on the reflectivity can be estimated by the Molodkin formula [14], which takes into account the contribution of diffuse scattering from structure imperfections. To confirm the validity of these formulas for quasiforbidden reflections, we compared the reflectivities calculated by (2) and by formulas for an ideal crystal [15] with due regard for the contributions that come to χ_h from both real and imaginary components. The satisfactory agreement of these values allowed the numerical evaluation of the effect of structural imperfections on the reflectivity for quasiforbidden reflections at various values of the static Debye–Waller factors L for the case where $\chi_{rh} = 0$.

EXPERIMENTAL

The integrated intensities of the symmetric 200 Laue reflections were measured on a single-crystal spectrometer by the method suggested in [16] with the use of a standard perfect silicon crystal. The parameters of the X-ray tube (20 kV, 35 mA) provided the elimination of the parasitic effect of a harmonics multiple of $n\lambda/2$. We used a 35-μm-thick (100)-oriented GaAs sample with the dislocation density $\sim 10^3$ cm^{-2}. Special precautions were taken to avoid the elastic strains during sample positioning on a crystal holder. The energy resolution of the setup was characterized by the width of the spectral window $\Delta\omega$:

$$\Delta\omega = \omega\cot\vartheta\Delta\vartheta, \quad (3)$$

where ϑ is the Bragg angle, ω is the frequency of the X-ray radiation, and $\Delta\vartheta$ is the angular range of simultaneously recorded diffracted waves. With due regard

for the widths of two exit slits (50 μm), the resolution was 35 eV.

Since the integrated intensities of 200 Laue reflections in this wavelength range were close to the background intensity ($k_f = i_f/i_d = 2.0$, where i_f and i_d are the pulse densities in the intensity measurements of the background and the diffracted-beam intensity), the 1% measurement accuracy was attained by collecting the necessary number of pulses and multiple energy passages. The statistical error in the measurements of integrated intensities against the background was determined as

$$\varepsilon(i_d) = \frac{1}{\sqrt{i_d T}}\sqrt{1+2k_f}, \quad (4)$$

where T is the time of measuring the integrated intensity. In our case, $k_f = 2.0$, $\varepsilon \approx 0.01$, and the measurement time at each point for the normal law of error distribution ranged within 40–200 s (depending on the wavelength).

RESULTS AND DISCUSSION

Experimental points and the R_i curves calculated as functions of X-ray wavelength by (2) for crystals *1* (nonstoichiometric), *2* (ideal stoichiometric) at $\chi_{ih} = 0$, and *3* (ideal at $\chi_{ih} \neq 0$) are shown in Fig. 2. One can see that all the curves have the minima, with the extremum point E on the experimental curve being more clearly pronounced than the point T on the theoretical curve *3*. In the vicinity of the wavelengths where $\chi_{rh} = 0$, the experimental reflectivity R_i has the nonzero values and is determined by the value of χ_{ih}. The contribution of χ_{ih} to X-ray diffraction for the quasiforbidden 200 reflection is quite considerable in the whole wavelength range studied (Fig. 1) with the role of the real part χ_h being important only in the vicinity of the K-absorption edge of As ($\lambda_K^{As} = 0.1045$ nm). Therefore, neglecting the imaginary part ($\chi_{ih} \longrightarrow 0$) in the calculation of the parameters $b = \sqrt{2}\,|\chi_{ih}|/|\chi_{rh}|$ and $p = |\chi_{ih}||\chi_{rh}|/|\chi_h|^2$ determining M in (2) results in considerable discrepancies between the theory and the experiment in this wavelength range (see inset in Fig. 2 and curve *2*) everywhere except in the wavelength close to the K-absorption edge of As where the term of χ_{rh} becomes important. This fact shows that X-ray scattering at the point $\chi_{rh} = 0$ is described by the imaginary part of the structure factor alone. The positions of the reflectivity minima $R_i = f(\lambda)$ (both calculated and experimental) also seem to depend on the sample nonstoichiometry. Therefore, the positions of these minima slightly differ from the position of the point, where $\chi_{rh} = 0$.

In order to interpret the above discrepancies between the experimental and calculated integrated reflectivity for an ideal (stoichiometric) crystal (curve *3*), we studied the effect of nonstoichiometry on the energy position of the point $\chi_{rh} = 0$ and the run of $R_i = f(\lambda)$. The results of such calculations for the real part of the structure factor F_{rh} are shown in Fig. 1. The analysis of these data shows that the position of $F_{rh} = 0$ is sensitive to the variations in the chemical composition of the crystal. Indeed, calculated curve *1* in Fig. 2 with due regard for possible nonstoichiometry at the level $c_{Ga} - c_{As} = 0.01$ satisfactorily agrees with the experimental dependence $R_i = f(\lambda)$ and the position of its minimum. The variations in the component concentration at a level of 10^{17} cm^{-3} do not noticeably change the energy position of the structure factor minimum at the given width of the spectral window. The position of the reflectivity minimum is even less sensitive to such small variations in the chemical composition of the crystal.

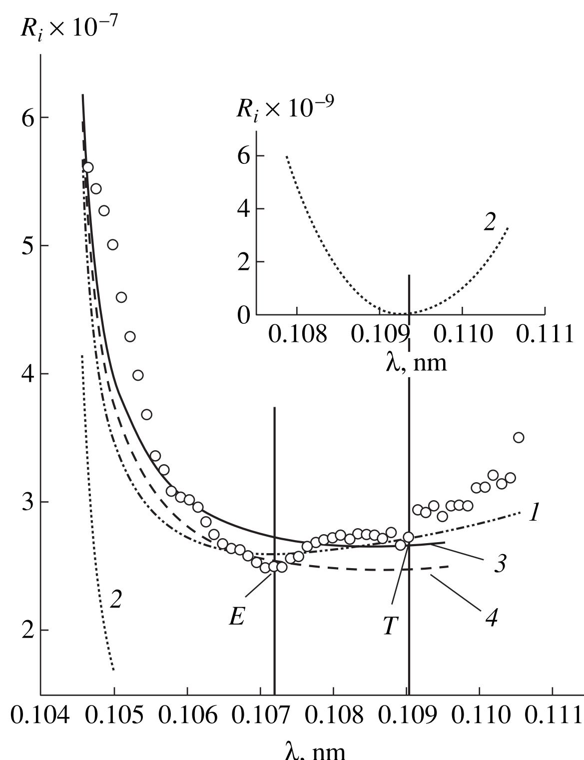

Fig. 2. Experimental reflectivity (dots) and reflectivity calculated by formula (2) as functions of the X-ray wavelength for a 35 μm-thick GaAs specimen; 200 reflection; (*1*) a nonstoichiometric crystal; (*2*) an ideal stoichiometric crystal with $\chi_{ih} = 0$; (*3*) an ideal stoichiometric crystal with $\chi_{ih} \neq 0$; (*4*) a real stoichiometric crystal with $\chi_{ih} \neq 0$, $L = 0.01$, and $\mu_{DS} = 30$ cm^{-1}. In the inset: curve *2* in the vicinity of the wavelength at which the reflectivity value calculated without the allowance for the contribution of χ_{ih} into diffraction is less by several orders of magnitude than its experimental value.

One more interesting fact established in the wavelength range studied is the effect of various structural defects on reflectivity. This problem is especially important because of obvious discrepancies between the theory (even with due regard for possible nonsto-

ichiometry) and the experiment in the wavelength range close to the *K*-absorption edge of As atoms (Fig. 2). The calculation of this effect at various values of the static Debye–Waller factors L and the coefficient of energy losses due to diffuse scattering μ_{DS} is illustrated by Fig. 2 (curve *4*). In the calculations, the values of the parameters L and μ_{DS} were assumed to be independent of the scattering parameters λ and χ_h. The calculations show that reflectivity is slightly sensitive to the variations in the parameter L but is strongly dependent on the coefficient μ_{DS}. This can be explained by the fact that a decrease in $\chi\exp(-L)$ cannot be compensated with an increase in reflectivity due to the diffuse-scattering component. Unlike the case of nonstoichiometry, structural defects described by the parameters L and μ_{DS} considerably influence the energy position of the minimum Laue intensity. This fact is very important for the separation of the effects caused by various factors on reflectivity of quasiforbidden reflections. The effect of λ on μ_{DS} and, thus, also on reflectivity of the quasiforbidden reflection in this wavelength range will be studied separately.

CONCLUSIONS

Thus, the study of X-ray dynamical scattering for the quasiforbidden 200 reflection for gallium arsenide crystals provided the establishment of the following facts.

In the wavelength range between the *K*-absorption edges of Ga and As, there is a weak reflectivity minimum (revealed experimentally), which is explained by the zero value of the real part of the structure factor in this range.

The experimental reflectivity values for quasiforbidden reflections cannot be adequately described by the scattering theory not taking into account the imaginary part of the Fourier coefficient of polarizability. The reflectivity at the wavelengths at which $\chi_{rh} \longrightarrow 0$ is described by the imaginary part of the atomic formfactor alone.

The position of the minimum on the energy dependence of reflectivity for the quasiforbidden reflection in this wavelength range is sensitive to crystal nonstoichiometry, but is independent of the parameters L and μ_{DS} of the structural perfection of the crystal.

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