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THE INVESTIGATION OF THE SPECTRAL DEPENDENCE OF "FORBIDDEN" REFLECTIONS PARAMETERS FOR SILICON AND GERMANIUM USING SYNCHROTRON RADIATION

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During the last years the synchrotron radiation (SR) generated by orbiting electrons in accelerators or storage rings is widely used in X-ray investigations, due to high photon flux density, spectrum continuity and almost total polarization of the radiation [1].

These characteristics of SR stimulated the works on studying the fine structure of absorption edges (EXAFS), X-ray anomalous scattering, Compton and small-angle scattering, topography, etc. [2].

Besides, there is a great interest in the diffractometrical investigations using SR, in particular, the investigation on determination of the integral reflection coefficients and structural factors of the "forbidden" and "quasi-forbidden" reflections, which contain the important information about electron behaviour in solids, lattice dynamics and ordering processes.

The necessity in such kind of investigations is caused by the fact that the exact determination of the (222) structure factors for the crystal with diamond lattice is essential part of the analysis of the electron spatial distributions, and its deviation from the spherical symmetry due to the chemical bonds (see, e.g. [3-5]). On the other hand, as soon as the extinction depth for (222) reflection is two orders of magnitude higher than that for the (111) reflection, the measurement of the $\bar{R}(222)$ in large wavelength range gives a chance to create a new method for determination of the defects distribution in the crystal.

This work represents the results of the measurement of spectral dependence of the "forbidden" reflex (222) integral coefficient $\bar{R}_{222}(\lambda)$ for perfect monocrystals Si and Ge in the wavelength range 0.56 A ÷ 1.62 Å using SR beam of the Yerevan accelerator [6].

The perfect monocrystals of Si and Ge with dislocation density less than 10 cm^{-2} were investigated. The reflecting surface of the samples was parallel to the crystallographical planes (111). The broken layer arising during the surface processing was taken away by standard means.

A schematical representation of the experimental facility as well as its basic elements and beam axis mutual disposition are shown in Fig.1. The SR beam formed by two collimators K_1 and K_2 located respectively at 17.5 m and 25.2 m from the point of the beam generation and had the transverse dimensions 2 x 1 mm^2. The beam divergency was $3.8 \cdot 10^{-4}$ and $2.4 \cdot 10^{-4}$, respectively in the horizontal and vertical planes. The measurement was performed by a two-crystal spectrometer in the antiparallel position. Although the rocking curve in this case was essentially broader than that in the parallel disposition of crystals, the spectral band of diffracted beam was considerably narrower, which is very important in SR research. In accordance with the graphical interpretation of the two-crystal spectrometer theory [7] for symmetric reflection, in the case of usage of SR, the relation between the measured value of intensities J_{222}, J_{333} and the integral reflection coefficients $\bar{R}_{222}$, $\bar{R}_{333}$ is determined by the formula (see [8]):

$$\frac{J_{222}(\lambda)}{J_{333}(\lambda)} = \frac{\bar{R}_{222}(\lambda)\cdot(1+\mathrm{ctg}\,\theta_M\,\mathrm{tg}\,\theta_{333})}{\bar{R}_{333}(\lambda)\cdot(1+\mathrm{ctg}\,\theta_M\,\mathrm{tg}\,\theta_{222})} \qquad (1)$$

where θ_M is the incidence angle of the SR beam for the monochromator-crystal, θ_{222} and θ_{333} are the angles of the second crystal (the sample) relative to the direction of the monochromatic beam.

The intensity of the diffracted beam was measured by a NaJ(Tl) scintillator which had the best resolution in the given wavelength range [9]. This scintillator was connected to PM-85 which is known as conventional detector BDS-1 of the DRON-2 diffractometer.

The exclusion of the errors in J_{222} and J_{333} measurements arising due to the presence of harmonics (see Fig.2a diagram) of the monochromatic SR beam was done by a one-channel analyzer. The width of the channel and discrimination threshold was determined using (222) reflection.

The reduction of the harmonics intensity up to the background level is shown in Fig.2b.

The multiple scattering contribution (the Renninger effect [10]) to the measured intensity J_{222} was avoided by corresponding choice of the crystal rotation angle around the axis normal to the reflecting surface, at which the multiple reflection peaks were absent. The small vertical divergency of the SR beam caused rather small width of the multiple scattering peaks, which allowed to choose the working point at the distance not less than 10 mean halfwidths from the nearest peaks. Approximating the multiple scattering peaks shape by the Gauss curve one may affirm that the contribution of these peaks tails to the measured intensity does not exceed 0.02%. A typical Renninger diagram obtained by rotating the crystal in the azimuthal plane around the scattering vector corresponding to reflection (222), is plotted in Fig.3 (Si, λ = 0.56 Å).

Using the SR beams of the electron accelerators the most essential limitation of the measurement accuracy is the fluctuation of the particles current value. In this case the number of the pulses detected as J_{222}, J_{333} and J_{backgr} was related to the number of the electrons accelerated by the synchrotron. The measurement of the number of accelerated electrons was done by a special detector [11].

To exclude errors arising from the nonlinearity of the scintillation counting characteristics at the J_{333} measurements the J_{333} intensity was reduced by aluminium plates to the level of the J_{222} value. For restoration of the true, undistorted value of J_{333} the absorption linear coefficients for Al in the wavelength range $0.56\text{Å} \div 1.55\text{Å}$ were measured using (222) reflection, which allowed to avoid errors due to the presence of $\lambda/2$ and $\lambda/3$ harmonics in the monochromatized radiation. The absence of $\lambda/3$ harmonic after reflecting from the second crystal is connected with the fact that the corresponding wavelength and angle θ_{222} do not satisfy Bregg condition, and the intensity of the spectral component of $\lambda/2$ - wavelength according to calculations even for the longwave edge ($\lambda = 1.55\text{Å}$) of the range is 65 times reduced with respect to the intensity of (222) reflection for λ. Moreover, the utilization of the one-channel analyzer still reduces $\sim 10^2$ times the contribution of the harmonic component with $\lambda/2$ wavelength. Following these measurements the dependence $\mu(\lambda)_{Al}$ in the mentioned wavelength interval is described by the power function with the exponent 2.865 ± 0.014.

In Table I the measurement results for J_{222}, J_{backgr} as well as the restored by the measured values of $\mu(\lambda)_{Al}$ intensity of J_{333} for Si and Ge are given. The values of the $\bar{R}_{222}(\lambda)$ calculated by (1) using the experimental data in Table I and corresponding values of the $F(222)$ obtained by iteration over the dynamic theory formulae are presented in Table II. The calculation of the integral reflection (333) coefficients with account of the absorption was done using the formulae of the dynamic theory [13, 14]. The formfactors were taken from [12, 15], the dispersion corrections from [16] and absorption coefficients of Si and Ge from [17]. In these calculations the polarization properties of the SR beam were obtained by the formulae

$$\bar{R}_{hk\ell}(\lambda) = \frac{1}{1+KC}\bar{R}^{\sigma}_{hk\ell}(\lambda) + \frac{KC}{1+KC}\bar{R}^{\pi}_{hk\ell}(\lambda), \qquad (2)$$

where

$$C = \frac{\bar{R}^{\pi}_{111}(\lambda)}{\bar{R}^{\sigma}_{111}(\lambda)}, \qquad (3)$$

$$K = \frac{J^{\pi}_{o}(\lambda)}{J^{\sigma}_{o}(\lambda)} \qquad (4)$$

The values of the J^{σ}_{o} and J^{π}_{o} of SR for the given λ and energy of the accelerated electrons E were determined using the polarization $P(\lambda, E)$ calculated by formulae evaluated in [18].

Taking into account the fact that the energy of the electrons varies during an acceleration cycle the value of the polarization $P(\lambda, E)$ was determined by the formula

$$P(E,\lambda) = \frac{\sum P(E_i, \lambda)\alpha_i}{\sum \alpha_i} \qquad (5)$$

where α_i is the ratio of the power radiated by electrons at the energy E_i to the power radiated at $E_{max} = 4.5$ GeV [19].

Using the value $F_{Si}(222) = 1.433$ which was obtained by averaging the experimental data in Table II the values of $\bar{R}_{222}(\lambda)$ were calculated both by dynamic and kinematic theories; the calculation results are given in the corresponding columns of Table II. The values of the $\bar{R}_{222}(\lambda)$ calculated by the both theories and presented in Table II notably differ on the shortwave edge of the used interval. This difference reduces by increasing λ . Such a behaviour is caused by the fact that the characteristic length of the photoelectrical absorption at the long λ decreases much more rapidly than the extinction depth, and the scattering process becomes kinematical.

The solid lines in Fig.4 present the calculations of $\bar{R}_{222}(\lambda)$ for Si using dynamic (a) and kinematic (b) theories for the value $F(222)= 1.433$. The results of the $\bar{R}_{222}(\lambda)$ calculation for Ge are presented by the corresponding curve (c) ibidem. The experimentally determined values of this parameter are presented here by points. These results show that the experimental values of $\bar{R}_{222}(\lambda)$ are close to their absolute quantities calculated by the dynamic theory, and at the same time their dependence on λ repeats the shape of the curve calculated by the kinematic theory. This phenomenon may be explained by the fact that at $\lambda \sim 0.5$ Å the values of the characteristic photoabsorption length, extinction depth and the mean size of the coherent scattering region became close to each other, and the structure imperfectiveness of the investigated samples displays in the scattering process. It is not excluded that besides the dislocation fields the point defects act an important role at small λ .

At the same time, assuming the dynamic theory validity when describing the X-ray diffraction in the studied crystals, one may alternatively explain the obtained results consisting in the fact that the structure factor (222) reflection is wavelength-dependent.

The spectral dependence of F(222) for Si obtained under this assumption is shown in Fig.5. The earlier published values of F(222) (see [3, 4] and references therein) plotted here by crosses are in good agreement with the results of this paper. The dependence of F(222) on λ experimentally obtained in this work contradicts the modern theoretical conceptions because this reflection is due to the valence electrons, while the responsibility

for the absorption (the wavelength-dependent corrections for the anomalous scattering) lies almost on K-electrons only. However, it is known that the spherically symmetrical part of the electron distribution of atom in a crystal differs from that in free state [4, 12], and there are no convincing proofs that the spherically symmetrical part of the electron distribution is responsible for only the valence electrons. Moreover, the investigations of the "aluminium problem" have shown that for the agreement of experimental and theoretical formfactors in this material one should take into account the change of space distribution of inner electrons in the crystal as compared with the free atom [20, 21].

To solve these problems the further experimental and theoretical investigations are, of course, necessary. From the experimental point of view, to expand the spectral interval the variety of the samples and studying the other "forbidden" reflections are required. In such kind of experiments the use of SR beams permits to simplify the solution of a number of principle problems, and allows to obtain interesting information unavailable earlier.

Table I

	λ (Å)	$J_{222} \times 10^{-3}$	$J_{backgr.} \times 10^{-3}$	$J_{333} \times 10^{-3}$
Si	0,579	33,37 ± 0,38	0,77 ± 0,02	506,59 ± I,23
	0,893	42,I4 ± 0,33	0,64 ± 0,0I	II62,67 ± 6,98
	I,085	38,54 ± 0,3	0,53 ± 0,0I	II38,9I ± 4,78
	I,299	II,65 ± 0,03	0,20 ± 0,0I	285,35 ± 2,4I
	I,55I	6,4 ± 0,I	0,34 ± 0,02	I74,77 ± I,I6
Ge	I,I72	I2,I2 ± 0,09	0,98 ± 0,0I	3660,38 ± 30,88
	I,420	9,82 ± 0,04	I,33 ± 0,04	2395,52 ± 23,I0
	I,62I	6,I2 ± 0,II	I,25 ± 0,0I	I559,I7 ± II,53

Table II

	λ (Å)	$\bar{R}_{222}(\lambda)$ exp.	$\bar{R}_{222}(\lambda)$ dyn.th.	$\bar{R}_{222}(\lambda)$ kin.th.	$\bar{R}_{333}(\lambda)$ dyn.th.	$\Gamma(222)$ exp.
Si	0,579	$I,I96 \cdot I0^{-7}$	$I,096 \cdot I0^{-7}$	$I,887 \cdot I0^{-7}$	$0,256 \cdot I0^{-5}$	I,523
	0,893	$0,79I \cdot I0^{-7}$	$0,874 \cdot I0^{-7}$	$I,067 \cdot I0^{-7}$	$0,3I0 \cdot I0^{-5}$	I,325
	I,085	$0,670 \cdot I0^{-7}$	$0,694 \cdot I0^{-7}$	$0,747 \cdot I0^{-7}$	$0,290 \cdot I0^{-5}$	I,405
	I,299	$0,526 \cdot I0^{-7}$	$0,520 \cdot I0^{-7}$	$0,540 \cdot I0^{-7}$	$0,I98 \cdot I0^{-5}$	I,442
	I,55I	$0,358 \cdot I0^{-7}$	$0,330 \cdot I0^{-7}$	$0,338 \cdot I0^{-7}$	$0,I7I \cdot I0^{-5}$	I,473
Ge	I,I72	$I,350 \cdot I0^{-8}$	–	$I,350 \cdot I0^{-8}$	$0,643 \cdot I0^{-5}$	I,I08
	I,420	$0,805 \cdot I0^{-8}$	–	$0,892 \cdot I0^{-8}$	$0,349 \cdot I0^{-5}$	I,052
	I,62I	$0,696 \cdot I0^{-8}$	–	$0,629 \cdot I0^{-8}$	$0,369 \cdot I0^{-5}$	I,I65

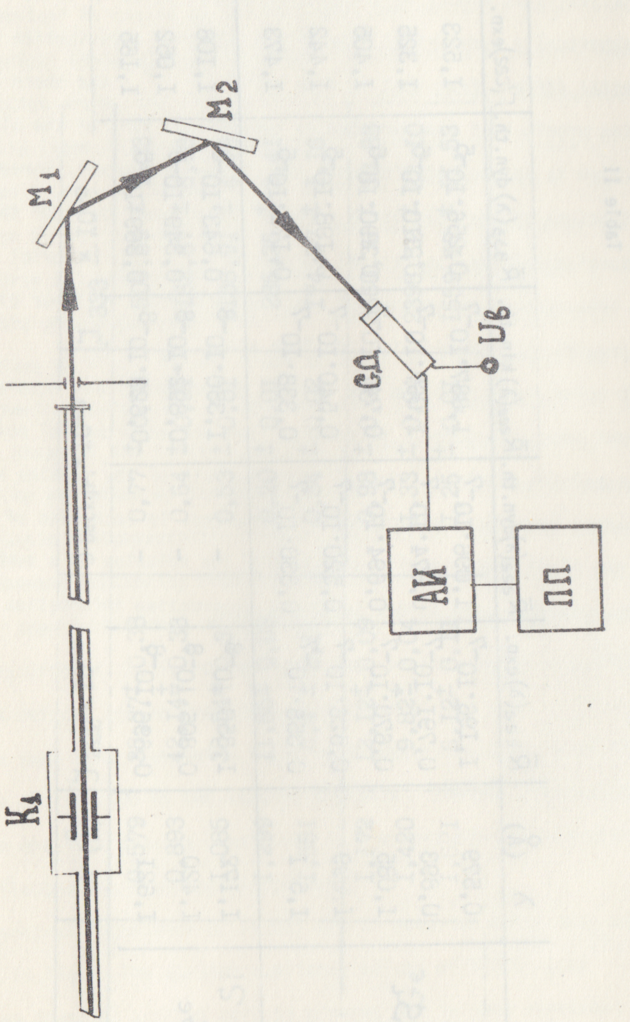


Fig.1

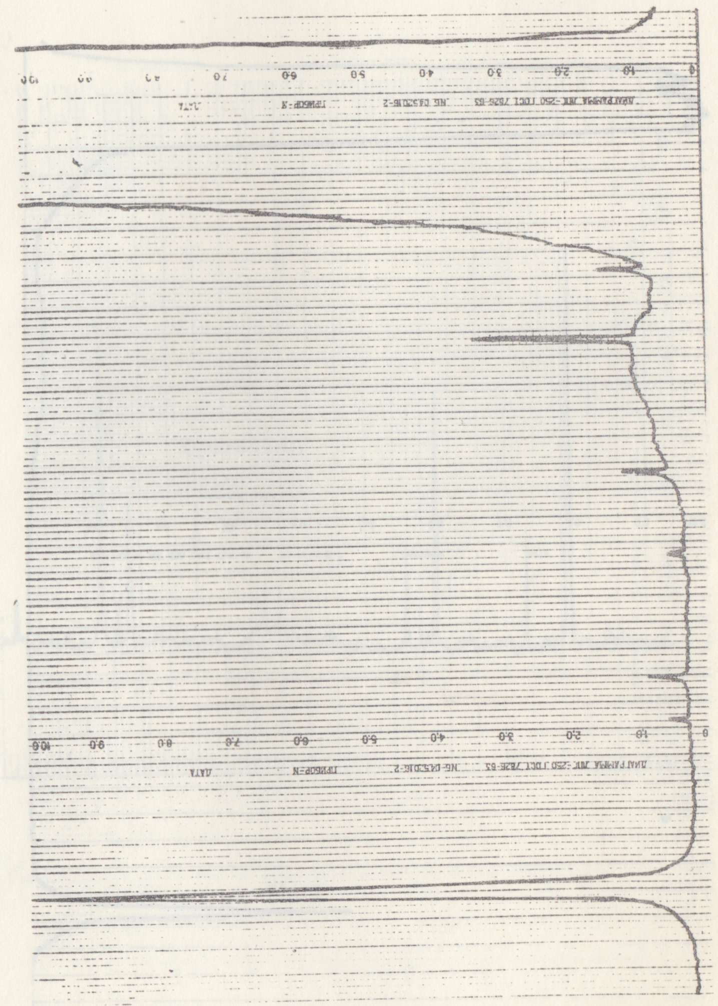


Fig.2a

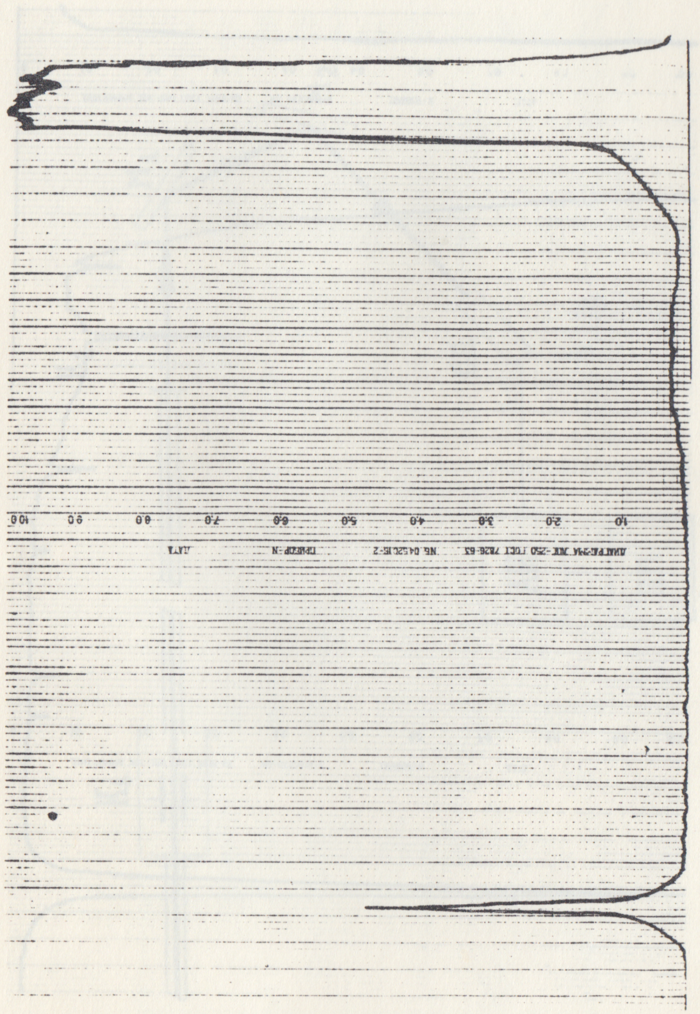


Fig.2b

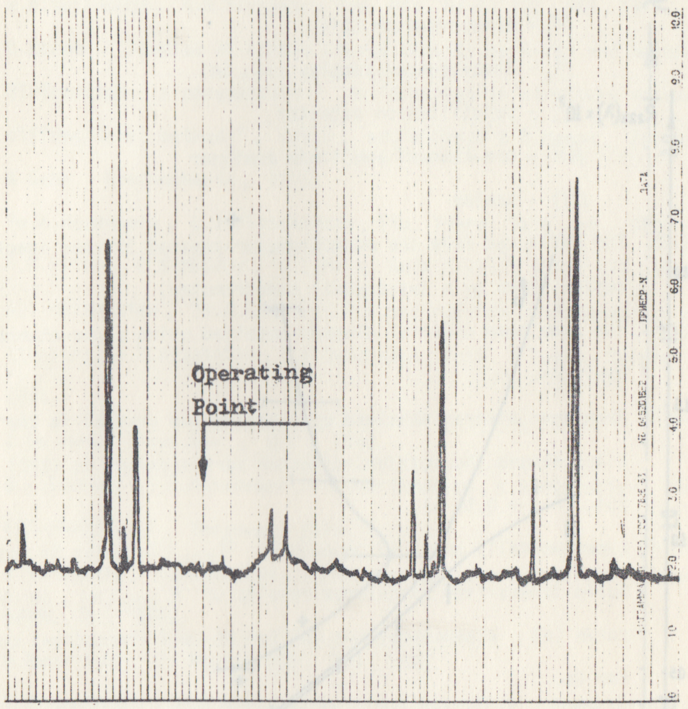
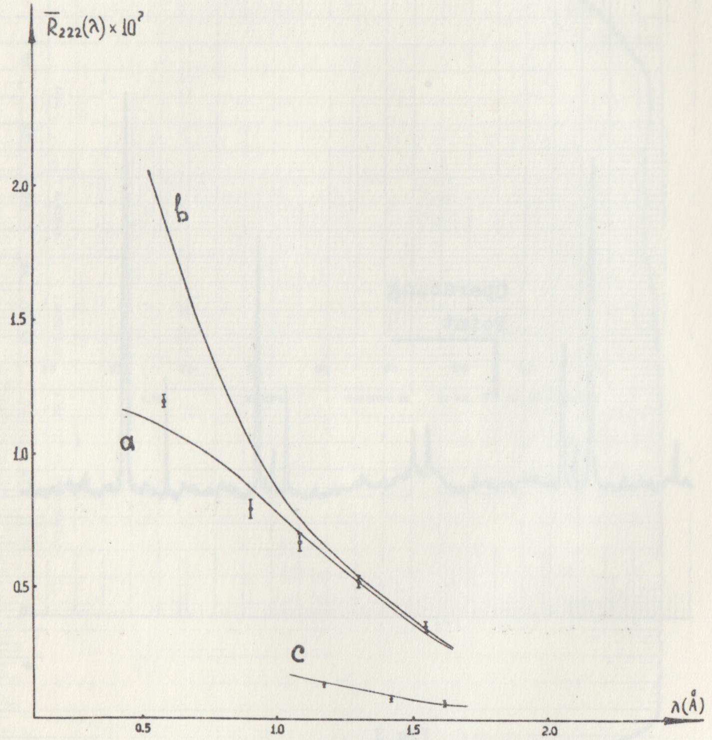
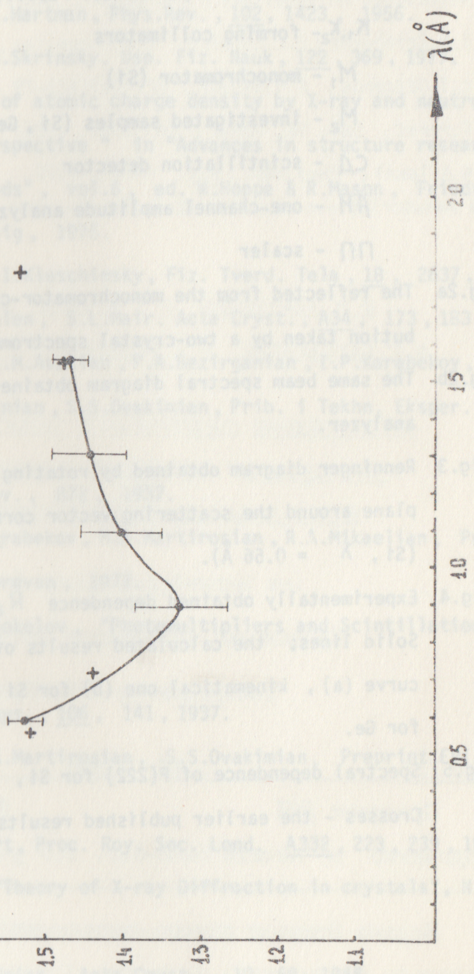


Fig.3



$\overline{R}_{222}(\lambda)\times10^7$
2.0
1.5
1.0
0.5
a
b
c
0.5
1.0
1.5
2.0
λ(Å)

Fig.4



λ(Å)
F_{222}(Si)
2.0
1.5
1.0
0.5
1.6
1.5
1.4
1.3
1.2
1.1

Fig.5

FIGURE CAPTIONS

Fig.1 The sketch of the experimental facility:

K_1, K_2 - forming collimators

M_1 - monochromator (Si)

M_2 - investigated samples (Si, Ge)

СД - scintillation detector

АИ - one-channel amplitude analyzer

ПП - scaler

Fig.2a The reflected from the monochromator-crystal SR beam spectral distribution taken by a two-crystal spectrometer.

Fig.2b The same beam spectral diagram obtained using a one-channel amplitude analyzer.

Fig.3 Renninger diagram obtained by rotating the crystal in the azimuthal plane around the scattering vector corresponding to (222) reflection (Si, $\lambda = 0.56$ Å).

Fig.4 Experimentally obtained dependence $\bar{R}_{222}(\lambda)$ for Si and Ge (dots) Solid lines; the calculated results of $\bar{R}_{222}(\lambda)$ on dynamic curve (a), kinematical one (b) for Si and kinematical curve (c) for Ge.

Fig.5 Spectral dependence of F(222) for Si, obtained in this work (dots). Crosses - the earlier published results of other authors.

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ИССЛЕДОВАНИЕ СПЕКТРАЛЬНОЙ ЗАВИСИМОСТИ ПАРАМЕТРОВ "ЗАПРЕЩЕННЫХ" ОТРАЖЕНИЙ КРЕМНИЯ И ГЕРМАНИЯ С ИСПОЛЬЗОВАНИЕМ СИНХРОТРОННОГО ИЗЛУЧЕНИЯ

(на английском языке)

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