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SURFACE, THIN FILMS

QUANTUM SIZE EFFECT DURING NORMAL INCIDENCE OF A BEAM OF MEDIUM ENERGY ELECTRONS ON A GROWING HETEROEPITAXIAL FILM

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Abstract. The reflection coefficient of medium-energy electrons (about 10 KeV) was calculated for their normal incidence on a thin growing single-crystal film. It is shown that in this case a quantum size effect occurs, which manifests itself in harmonic oscillations of the intensity of the reflected beam. The amplitude and period of oscillations depend on the thickness of the growing film and the energy of the incident electrons. A graphic illustration of the results obtained is provided.

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INTRODUCTION

Oscillations of the specular and diffracted beam intensity in the diffraction pattern obtained from a growing single-crystal film were first observed in [1–3]. The oscillation period was equal to the deposition time of one monomolecular Ga + As layer, equal to $c/2$, where c is the GaAs lattice period. In [2], it was assumed that these oscillations were associated with a periodic change in the surface roughness of the growing crystal, caused by the formation of two-dimensional nuclei on the atomically smooth surface, their growth and unification into a continuous monolayer, the formation of nuclei of the next layer, etc. In subsequent years, a series of works were published in which this phenomenon was studied and used to investigate the mechanism and kinetics of epitaxial layer growth [4–8].

However, in [9] it was shown that, along with scattering from the film surface, in this experimental situation there should be a reflection of electrons from the volume of the growing heteroepitaxial film, which leads to a modulation of the intensity of short-wave oscillations caused by scattering on the surface, and long-wave oscillations caused by the quantum size effect in the volume of the single-crystal film.

The quantum size effect of electrons in thin metal and semiconductor films has been well studied both theoretically and experimentally [10]. Initially, when modeling the particle scattering process, the electrostatic potential in the film was chosen in the form of a rectangular well. Since the experimental observation of the effect requires that the electron wavelength be comparable to or greater than the inhomogeneities on the film surface, it was necessary to take the particle energy of the order of units of electron volts and less. In this case, the electron wavelength was of the order of units of angstroms, which is comparable to the surface roughness. At higher energies, the inhomogeneities in the film significantly exceed the particle wavelength. Thus, the rectangular well model prohibited the experimental observation of the effect at high electron energies.

The situation changes when analyzing the scattering of electrons on a single-crystal film, since Bloch waves are formed in it, the length of which varies from the period of the crystal lattice to the film thickness in each allowed zone of the one-dimensional crystal, which is significantly greater than the film inhomogeneity. The scattering of waves and particles on a one-dimensional limited periodic potential of a single-crystal film was first theoretically investigated in [11–18]. In [19], an expression was obtained for the one-dimensional quantum size effect taking into account the average internal potential of the lattice of the single-crystal film. This extremely capacious formula has been repeatedly used for the theoretical consideration of various models for the manifestation of this effect.

In [20], the quantum size effect was investigated taking into account the inelastic absorption of electrons in the film and a comparison of the theory with experiment was made. In [9, 20], an experimental situation was described in which a beam of electrons falls on a growing heteroepitaxial film at a small grazing angle and the intensity of the diffraction pattern is recorded.

In contrast, in [21] the situation of normal incidence of electrons on a thin single-crystal film is theoretically investigated. In this case, epitaxial growth of the film does not occur, and the energy of the incident electrons varies in a small range near the value of ~10 keV. Calculations show the appearance of oscillations of the intensity of reflected electrons caused by the quantum size effect. Previously, this effect at such high energies was not theoretically studied and was not observed experimentally. Its manifestation with such a large amplitude of oscillations is due to the fact that it is a single-crystal film that is being studied. In it, the passing electrons propagate in the form of Bloch waves and therefore the quantum size effect appears on them in the form of oscillations of the intensity of reflected electrons of large amplitude.

In [22], the same model of the effect was theoretically investigated as in [21], but the energy of the incident particles was taken to be ~100 keV. It was shown that even at such high energies, the oscillations of the intensity of the reflected electrons have a significant amplitude.

In this work, unlike [21, 22], the normal incidence of an electron beam of medium (~10 keV) energy on a growing heteroepitaxial film is considered. However, the argument in the formulas is no longer the electron energy, but the thickness of the growing film. It turns out that, as in the case of a grazing incidence of particles [9], a significant manifestation of the quantum size effect takes place.

SELECTION OF MODEL AND CALCULATION FORMULAS

Fig. 1 shows a diagram of the potential energy acquired by an electron falling normally on a single-crystal film. Analytically, it is given by the following formula:

$$V(x) = -V_0 + \frac{\hbar^2}{2m} cU \sum_{n=1}^{N} \delta\left(x - c(n-1)\right),$$

$$\text{for } -\frac{c}{2} \le x \le cN - \frac{c}{2}, \tag{1}$$

$$V(x) = 0, \textit{for } x < -\frac{c}{2}, x > cN - \frac{c}{2}.$$

Here c is the period of the one-dimensional lattice, N is the number of periods in it, i.e. the number of planes in the film parallel to its surface, $V_0 > 0$ is the height of the potential step at the film boundaries, $\delta(x)$ is the Dirac delta function, $U = 2\pi y/c^2$ is the "power" of the δ-function potential, y is the dimensionless parameter of the model, $\hbar$ is the Planck's constant, m is the electron mass, x is the coordinate along the axis perpendicular to the film surface.

Solving the stationary Schrödinger equation with this potential, we obtain the following expressions for the transmission coefficients T and reflection R [19]:

$$T = tt^* = \frac{1}{1 + \frac{1}{4}\left(\frac{\lambda}{k}\tan\frac{kc}{2}\cot\frac{\mu c}{2} - \frac{k}{\lambda}\cot\frac{kc}{2}\tan\frac{\mu c}{2}\right)^2 \sin^2 \mu c N} \tag{2}$$

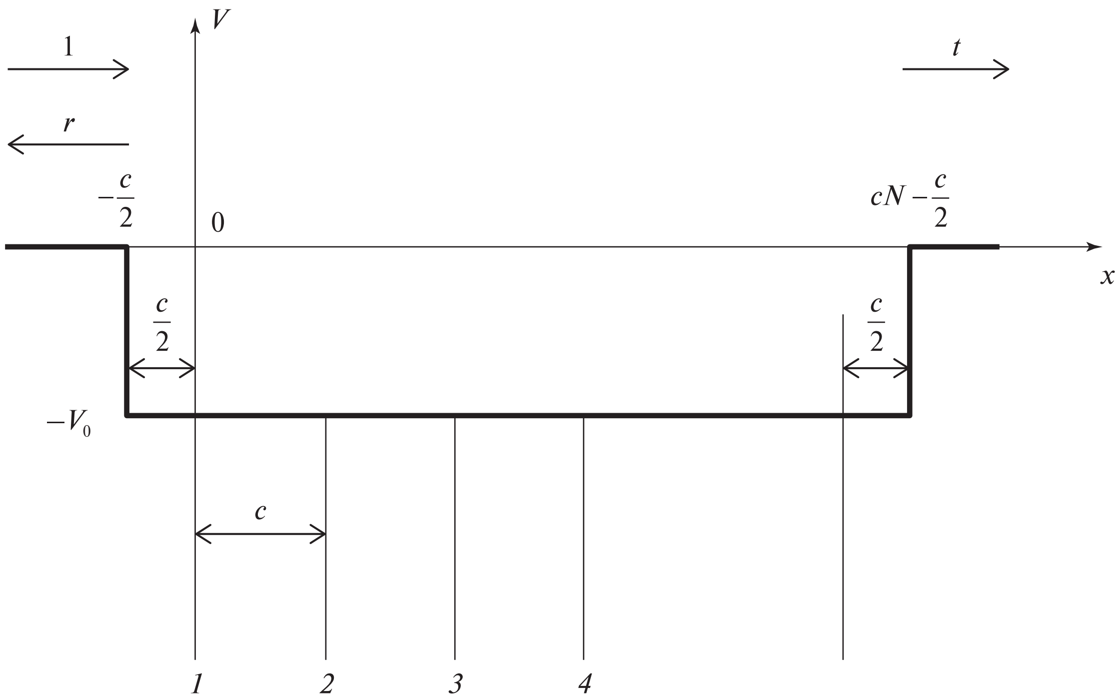

Fig. 1. Scheme of the potential energy of an electron in a film: V_0 is the value of the average internal energy, N is the number of monolayers in the film parallel to the surface, c is the period of the one-dimensional lattice, t is the amplitude of the transmitted wave, r is the amplitude of the reflected wave.

where the modulus of the Bloch wave vector μ is determined from the dispersion equation:

$$\cos\mu c = \cos kc + \frac{cU}{2k}\sin kc. \quad (3)$$

Here k is the modulus of the electron wave vector in the region with potential energy $-V_0$, $\lambda = \pi\sqrt{0.0268E_1}$, $k = \pi\sqrt{0.0268(E_1 + V_0)}$, where $E_1 = \left(\sin^2\left(\frac{\alpha\pi}{180}\right)\right)E$ is the normal component of the electron energy E of the incident beam (specified in eV), V_0 is the magnitude of the potential jump at the film boundaries, also specified in electron volts. The grazing angle α is measured in degrees, the moduli of the wave vectors λ and k are obtained in inverse angstroms. The reflection coefficient $R = 1 - T$.

Note that the factor 0.0268 in the expressions for λ and k given above is a dimensional coefficient whose numerical value is 0.0268 and whose dimension is [$Å^{-2} \cdot eV^{-1}$].

Detailed recommendations for the computational algorithm in the case where the right-hand side of the dispersion equation (3) becomes greater than one are given in [19].

Formula (2) takes on a particularly simple form when the average internal potential is not taken into account, i.e. when $V_0 = 0$. Then:

$$T = \frac{1}{1 + \left(\frac{cU}{2k}\right)^2 \frac{\sin^2 \mu cN}{\sin^2 \mu c}}. \quad (4)$$

DISCUSSION OF RESULTS

Let us consider the dependences of the reflection coefficient R on the beam electron energy $R(E)$ for a film with a thickness of $cN = 200$ Å. We will use them to select the particle energy for further study during film growth. In Fig. 2, the dependences $R(E)$ are shown for $c = 5$ Å, $y = -1$, $V_0 = 10$ eV. These values were selected based on the following considerations. The lattice period $c = 5$ Å corresponds to the average value of interplanar distances of planes with small crystallographic indices in most crystals. The value of the parameter $y = -1$ leads to a forbidden band between the valence and conduction bands of the crystal of the order of units of electron volts, which corresponds to the real situation. $V_0 = 10$ eV is a typical average value of the internal energy of electrons in crystals.

In Fig. 2a, sharp intense peaks are visible. They are explained by diffraction on a one-dimensional grating and are described by a modified Wulff-Bragg formula. The distance between the peaks is

$$\Delta E_n \approx \frac{2\sqrt{E_n}}{\sqrt{0.0268c^2}}, \quad (5)$$

where E_n is the energy corresponding to the n-th peak. The derivation of this formula is given in [22]. In Fig. 2b, on an enlarged scale, we see that the reflection intensity noticeably oscillates between the peaks. These oscillations are one of the manifestations of the quantum size effect and become especially clear with an even greater increase in scale (Fig. 2c).

In Fig. 2c we select the energy value $E = 10520$ eV, corresponding to the position of one of the diffraction peaks, and substitute it into formula (1) as a parameter. After that, we calculate $R(N)$ at this energy value, which corresponds in the experiment to recording the reflection intensity depending on the thickness of the

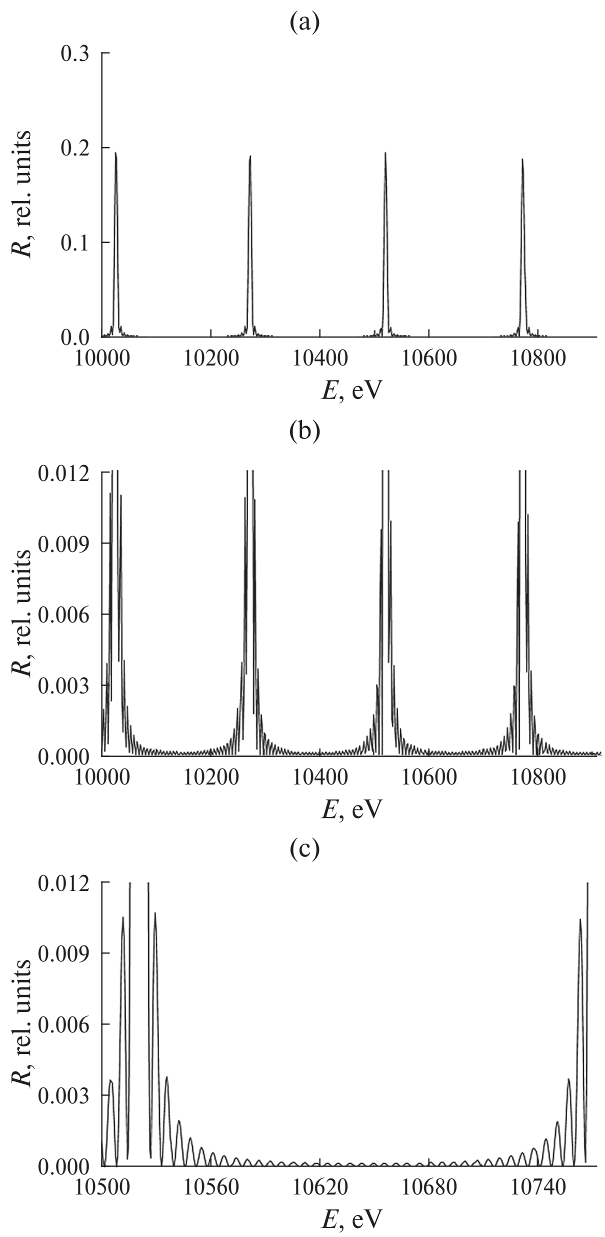

Fig. 2. Graphs of the R(E) dependence in different scales for $c = 5$ Å, $y = -1$, $V_0 = 10$ eV.

growing heteroepitaxial film at a given energy of the beam electrons. Fig. 3a shows this dependence. It is evident that the intensity of the diffraction peaks is equal to unity approximately at a thickness of Nc = 1000 Å. There are no oscillations corresponding to the quantum size effect. They appear if we shift slightly in energy (Fig. 2c) to the right. The value of the oscillation envelope at the energy E = 10524 eV in this figure corresponds to the oscillation amplitude in Fig. 3b, which shows the dependence $R(N)$ at the beam energy E = 10524 eV. It is evident that the oscillations have a significant amplitude and a period of 350 Å. That is, these are long-wave oscillations similar to those that appear during a grazing incidence of a beam [9]. With an even greater shift in energy to the right E = 10528 eV (Fig. 2c), we obtain oscillations with a smaller amplitude and period (Fig. 3c).

Next, we calculate the intensity of the reflected beam taking into account the inelastic absorption and scattering of electrons in the film. To do this, we multiply $R(N)$ by the empirical factor

$$S = \exp\left(-æcN\right),$$

where the attenuation coefficient æ is 0.01, which is typical for a beam energy of E = 10 keV. The attenuation coefficient is the sum of the absorption coefficient and the scattering coefficient. We obtain an approximate empirical formula:

$$I = I_1 R\left(N\right) S\left(N\right),$$

I_1 is set equal to one. Note that the last formula is approximate and only qualitatively reflects the experimental situation. However, since the calculations are model ones, we consider this choice justified. The results are shown in Fig. 4. Case (a) corresponds

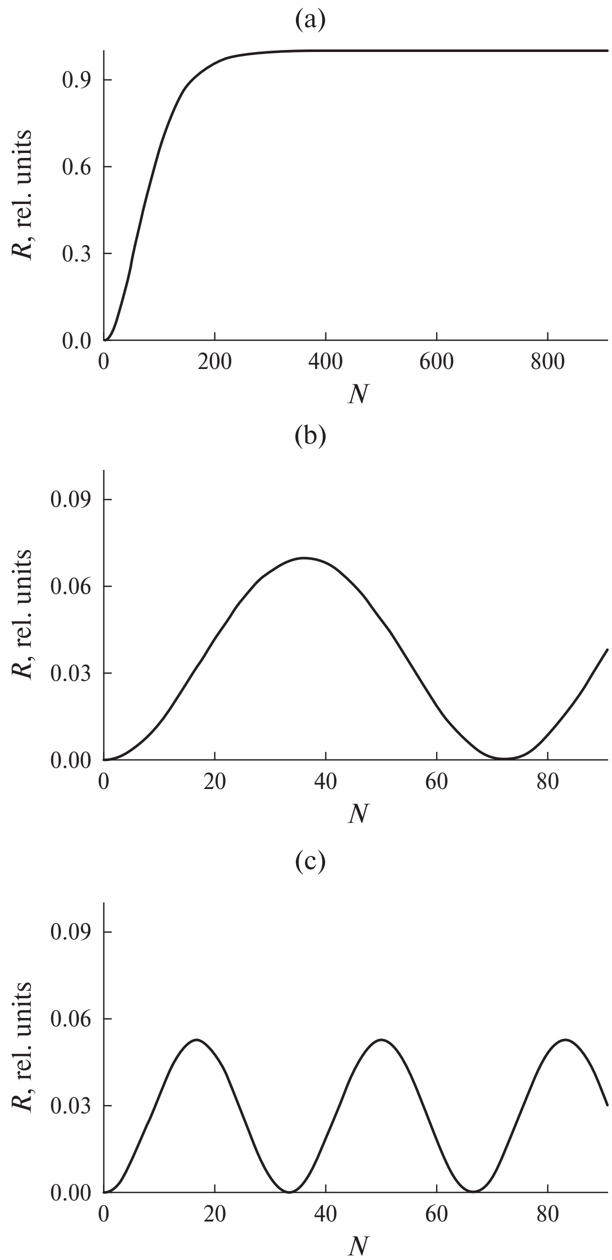

Fig. 3. Graph of the $R(N)$ dependence for three beam energy values: a – the energy value lies in the forbidden zone, b, c – in the allowed zone.

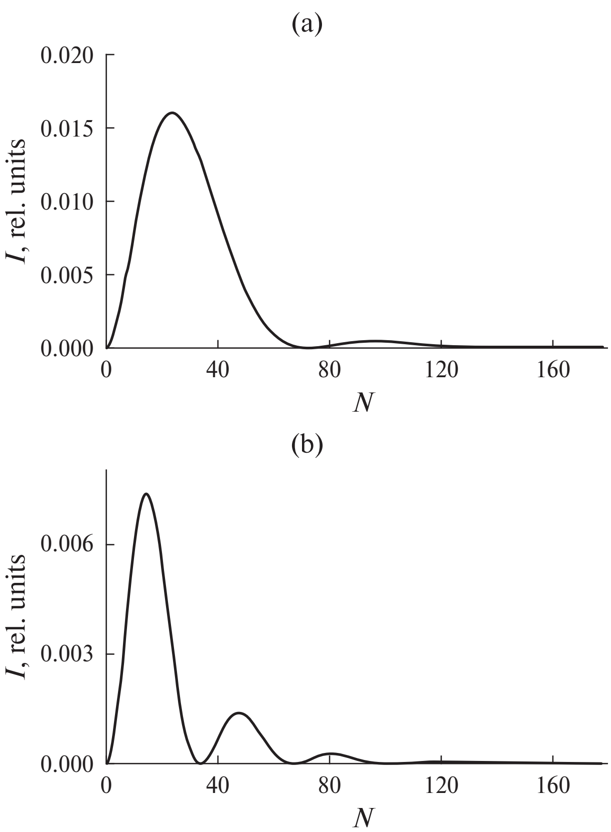

Fig. 4. Dependence of the reflected beam intensity on the thickness of the growing film, taking into account the attenuation caused by the absorption and scattering of electrons, for two energy values.

to the beam energy $E = 10524$ eV, and variant (b) – $E = 10528$ eV. It is evident that, compared to the situation of grazing incidence of the beam [20], the decrease in the reflection intensity is not as sharp. This is due to the fact that the attenuation coefficient in the case of normal incidence is smaller.

Until now, the influence of film roughness on the reflection of electrons from its surface has not been taken into account. At normal beam incidence, this phenomenon has not been studied either theoretically or experimentally. Let us assume that the formula for the reflection intensity is similar to the case of grazing beam incidence [20, 23]:

$$I = \frac{1}{2}I_0 + I_1 R(N) S(N) \cos(2\pi N - \varphi). \quad (6)$$

The results of calculations using formula (6) are shown in Fig. 5, where $I_0 = 1$. It is evident that short-wave oscillations caused by scattering from the surface are modulated by long-wave oscillations caused by the quantum size effect in the film volume. A decrease in their intensity is also observed due to the absorption and scattering of electrons. The reflection curve exhibits similar behavior in theoretical calculations [20] and is observed experimentally [24] in the case of grazing incidence of the beam.

CONSIDERATIONS ON THE EXPERIMENTAL OBSERVATION OF THE EFFECT

Theoretical calculations were performed for a one-dimensional potential model. We believe that such a model reflects the main characteristics of electron scattering by a growing film. In a three-dimensional picture, it is impossible to obtain an analytical expression for describing the quantum size effect. When a beam of medium and high energy electrons is reflected from a single crystal, either diffraction reflections or Kikuchi bands, or both are often observed [25, 26]. Therefore, when adjusting the energy of the beam reflected from a single-crystal substrate, it is necessary to choose a value lying within the allowed zone. In this case, its intensity is low (Fig. 2), so the background on the reflection curve is also low. The electron energy should also correspond to a value lying within the allowed one-dimensional zone and the growing single-crystal film, since only then can long-wave oscillations characteristic of the quantum size effect be observed. In this case, the reflection intensity is low (Fig. 2). Therefore, it is not possible to roughly estimate the oscillations. Equipment that records them is required.

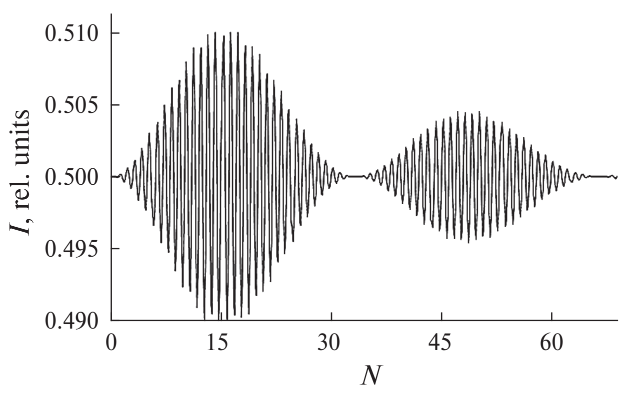

Fig. 5. Short-wave oscillations modulated by long-wave oscillations, taking into account the weakening of the beam intensity.

ON THE INTENSITY OF BRAGG REFLEXES

In [27], a formula was derived for the intensity of Bragg reflections that appear when varying the energy of electrons falling normally on a single-crystal film (Fig. 2). Below, we will repeat this derivation in more detail, and also continue to transform the previously obtained formula to a final simple expression.

It should be noted that the appearance of narrow resonance peaks is not at all associated with the quantum size effect, but is due to the well-known Wulff-Bragg formula:

$$2d \sin\alpha = n\lambda. \quad (7)$$

At normal incidence, the sine of the grazing angle is equal to one and the parameter in the resonance condition remains the wavelength of the radiation, which is determined by its energy. Since in this case the interplanar distance is $d = c$, and $\lambda = 2\pi/k$, the Wulff-Bragg formula is written as

$$kc = \pi n. \quad (8)$$

From it, formula (5) for the distances between peaks is derived in [22].

So, let us continue deriving the formula for the intensity of the Bragg peaks. Since their position is determined by condition (8) and at the same time $\mu c = kc + \pi m$, where m is an integer (formula (3)), then, substituting it into formula (4) and differentiating twice according to L'Hôpital's rule the numerator and denominator of the fraction in the denominator of the right-hand side of equation (4), under condition (8) we obtain:

$$T = \frac{1}{1 + \left(\frac{cU}{2k}\right)^2 N^2}. \quad (9)$$

From here:

$$R = 1 - T = \frac{\left(\frac{cU}{2k}\right)^2 N^2}{1 + \left(\frac{cU}{2k}\right)^2 N^2}. \quad (10)$$

In [27] a conclusion was made about the independence of the resonance peak intensity from its number n. This is an incorrect conclusion. The fact is that in (10) k depends on the energy E, i.e. on n. Let us show this. Let us substitute (8) into (10). Then, since

$$U = \frac{2\pi y}{c^2},$$

we obtain:

$$R = \frac{1}{\dfrac{1}{\dfrac{\left(c^2UN\right)^2}{\left(2\pi n\right)^2}} + 1} = \frac{1}{\dfrac{n^2}{N^2}y^2 + 1}. \qquad (11)$$

It is evident that the intensity depends on N, i.e. on the number of reflecting planes in the film, in other words, on its thickness, depends on y – the scattering capacity of an individual crystallographic plane, and also on the number of the resonance peak n.

Let us give some examples for the values $y = -1$, $N = 80$. The intensity of the first peak ($n = 1$) is almost equal to one, the intensity of the Nth peak ($n = N = 80$) is equal to $R = ½$. Then, as the number n increases, the intensity decreases monotonically (11).

CONCLUSION

It is shown that despite the fact that the electron energy is relatively high (10 keV), a quantum size effect is manifested in the growing single-crystal film at its normal incidence. It is expressed in harmonic oscillations of the reflected beam intensity depending on the thickness of the growing single-crystal film. An approximate formula is obtained that takes into account the weakening of the reflected beam intensity due to inelastic absorption and scattering of electrons, and the reflection coefficient is calculated using it.

Reflection curves are calculated in the case of short-wave oscillations caused by reflection from the surface of a growing single-crystal film. It is shown that they are modulated by long-wave oscillations determined by the quantum size effect.

In addition, it is noted that when the beam energy is varied, narrow resonance peaks are observed on the reflection curve. A formula is derived that determines their intensity, and an expression for the intervals between them is given.

Conditions conducive to experimental observation of the effect have been identified.

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CONFLICT OF INTERESTS

The author of this work declares that he has no conflicts of interest.

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