Asymmetry of the scattering of polarized electrons by polycrystalline gold

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Experimental and theoretical investigations were made of the asymmetry of the scattering of polarized electrons on the surface of polycrystalline gold. Atomic data were used to calculate this asymmetry for multiple collisions. The asymmetry of the scattering into a wide solid angle, set by a collector in a quasispherical analyzer, was determined using a beam of polarized electrons of energies E_p from 100 to 500 eV incident at angles of 0–32°. The good agreement between the calculated and experimental results confirmed the validity of the adopted model.

1. INTRODUCTION

Investigations carried out in the last decade have established that participation of the spin states in the interaction of electrons with a solid can provide additional information on the scattering mechanisms, characteristics of the potential barrier at a solid–vacuum interface, and physicochemical properties of the surface (in particular, the magnetization).[1-3]

The progress in this topic has been largely due to the construction of highly efficient sources of polarized electrons based on photoemission from III-V semiconductor compounds.

The present paper reports the results of a calculation and of measurements of the scattering asymmetry exhibited by elastic scattering of polarized electrons of $E_p \leqslant 500$ eV energy from the surface of polycrystalline gold.

The process of the scattering of slow electrons by atoms is influenced by the spin–orbit interaction in the course of the motion of electrons in the screened Coulomb field of the nucleus.[1] When a totally polarized beam of electrons ($P_0 = 1$) is scattered, the process exhibits an asymmetry

$$A=(I^{+}-I^{-})/(I^{+}+I^{-}), \tag{1}$$

where I^{+} and I^{-} are the intensities of the electron beams scattered through symmetric angles θ and $-\theta$, and the magnitude of this asymmetry is maximal in the case when $\mathbf{P}_0$ is directed either along or opposite to the normal $\mathbf{n}$ to the scattering plane.[1]

2. INFLUENCE OF MULTIPLE INTERACTIONS

An analysis of the scattering of electrons of moderate energies by the surface of a solid is impossible without allowance for the multiple scattering of electrons on target atoms. Several models have been suggested to allow for multiple interactions.[4,5] It is reported that the experimental results can be explained satisfactorily by calculations of the intensity of electrons scattered elastically from disordered Hg, Au, and Ag targets in the energy range $E_p = 0.1$–2 keV.

In the present study we calculated the asymmetry in the kinematic approximation using a scheme proposed in Ref. 4 to determine the intensity of a beam of elastically scattered electrons. We made the following assumptions.

1. Elastic scattering of electrons on single atoms represents independent events and the differential cross sections for single scattering are the same for atoms in isolated and aggregate states. In view of the absence of experimental results, we used theoretical values of the differential cross sections and asymmetries of elastic scattering by free atoms.[6,7] (The polarizations P acquired in the course of inelastic scattering of an unpolarized electron beam are given in Ref. 6. However, a comparison of the results in Refs. 6 and 7, and a general analysis of the problem[1] demonstrate that in the case of single collisions we have $P = A$.)

2. The intensity of a beam of electrons that have not lost the primary energy E_p in the course of motion in matter decreases continuously and is governed by an attenuation coefficient.

The following comments should be made about these assumptions. The use of the differential scattering cross sections of isolated atoms in the calculation of the intensity of a single interaction of electrons with ionic cores in a solid is fully justified. Scattering through a large angle (backward scattering) is characterized by a small impact parameter, i.e., it occurs at distances close to the center of an atom. Naturally, the values of the potentials of a free atom and an ionic core in a solid should be similar at such distances. Nevertheless, this assumption is not well satisfied when the differential cross sections represent the scattering through small angles which occur in the expression for calculating the intensity in the case of multiple interactions. However, as shown in Ref. 4, the calculated intensities of elastically and multiply scattered electrons are not very sensitive to details of the dependences of the low-angle scattering cross sections because of the dominant contribution of single scattering.

As far as the assumption 2 is concerned, the good agreement between the experimental and theoretical values of the intensity of elastically scattered electrons is used in Ref. 4 to draw the conclusion that the main mechanisms governing the scattering process are included in the adopted model.

The asymmetry of the elastic scattering (through an angle θ) of polarized electrons in a plane parallel to the specular reflection plane of the beam can generally be found from the relationship

$$A(\theta)=\sum_{n=1}^{N} A_n(\theta) I_n(\theta) \Big/ \sum_{n=1}^{N} I_n(\theta), \tag{2}$$

where A_n is the asymmetry for n-fold scattering of polarized electrons and I_n is the intensity of n-fold scattered electrons.

In the case of single scattering, $A_1(\theta)$ is the asymmetry for the scattering by a single atom and

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$$I_1(\theta) = I_0 \int_0^\infty \exp\left(-\frac{z\sigma}{\cos\alpha}\right)\frac{\rho\, dz}{\cos\alpha}\frac{d\sigma(\theta)}{d\Omega}\exp\left(-\frac{z\sigma}{\cos\gamma}\right) = I_0 \frac{\rho\,(d\sigma(\theta)/d\Omega)}{\sigma(1+\cos\alpha/\cos\gamma)}, \quad (3)$$

where I_0 is the intensity of the primary beam; α and γ are the angles of incidence and reflection measured from the normal to the target surface; ρ is the atomic density; $d\sigma(\theta)/d\Omega$ is the differential scattering cross section; $\sigma = \rho(\sigma_e + \sigma_i)$ is the attenuation coefficient; σ_e and σ_i are the integral cross sections for elastic and inelastic processes calculated per one atom; z is the running coordinate directed along the normal to the surface and into the target.

It is shown in Ref. 4 that in the case of double interaction the intensity obtained in the approximation of small-angle scattering is

$$I_2(\theta_3) \approx I_0 \frac{\rho^2}{2\sigma^2(1+\cos\alpha/\cos\gamma)}\int_{\Omega_2}\frac{d\sigma(\theta_1)}{d\Omega}\frac{d\sigma(\theta_2)}{d\Omega}d\Omega_2, \quad (4)$$

where θ_3 is the angle between the direction of the incident and scattered beams, whereas θ_1 and θ_2 are the large and small intermediate scattering angles. Similarly, the intensity of electrons that have suffered n-fold interaction is

$$I_n(\theta_3) = I_0 \frac{\rho^n}{n(1+\cos\alpha/\cos\gamma)\sigma^n} \times \int_{\Omega_2}\cdots\int_{\Omega_n}\frac{d\sigma(\theta_1)}{d\Omega}\cdots\frac{d\sigma(\theta_n)}{d\Omega}d\Omega_2\ldots d\Omega_n. \quad (5)$$

The small-angle approximation is justified because the differential cross section for the scattering through angles less than 20° exceeds considerably the cross section for the scattering for the other angles. Figure 1 shows schematically double scattering for a fixed value of a small angle θ_2. (In this figure the angles θ_1 and θ_2 are shown for the backward scattering. This is allowed for in the calculation program.) We can see that in the scattering through an angle θ_3 an electron should experience two interactions: 1) reflection through a large angle θ_1, the value of which is determined by the azimuthal angle φ when θ_3 and θ_2 are fixed; 2) scattering through a small angle θ_2.

The sequence in which the scattering events occur (first through a large angle and then through a small one or vice versa) is of no importance. It is clear from Fig. 1 that when the angles θ_1, θ_2, and θ_3 are related by trigonometric expressions, we can write down the integrand explicitly.

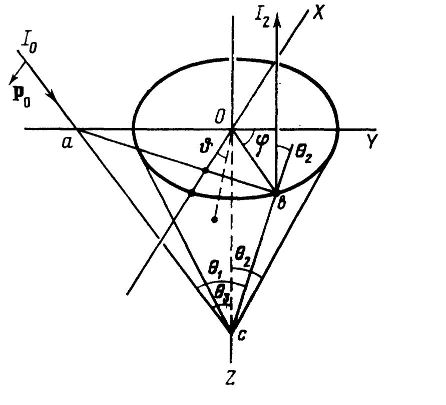

FIG. 1. Schematic representation of double scattering of a beam of polarized electrons. The notation is explained in text.

We calculated the value of $d\sigma_n(\theta_3)/d\Omega$ in order to compare the probabilities of the scattering through an angle θ_3 in interactions of different multiplicity. For example, in the case of double scattering this quantity is given by

$$\frac{d\sigma_2(\theta_3)}{d\Omega} = \int_{\Omega_2}\frac{d\sigma(\theta_1)}{d\Omega}\frac{d\sigma(\theta_2)}{d\Omega}d\Omega_2 \Big/ \int_{\Omega_2}\frac{d\sigma(\theta_2)}{d\Omega}d\Omega_2, \quad (6)$$

where the integral in the denominator is $\approx \sigma_e$.

The asymmetry in the course of double scattering is calculated from

$$A_2(\theta_3) = \int_{\Omega_2}\frac{d\sigma(\theta_2)}{d\Omega}\frac{d\sigma(\theta_1)}{d\Omega}A_1(\theta_1)\cos\vartheta\, d\Omega_2 \Big/ \int_{\Omega_2}\frac{d\sigma(\theta_2)}{d\Omega}\frac{d\sigma(\theta_1)}{d\Omega}d\Omega_2, \quad (7)$$

where the factor $\cos\vartheta$ allows for intermediate scattering in a plane nonparallel to the specular reflection plane of the beam. Here, ϑ (Fig. 1) is the angle between the normal n and the plane $a0c$ of the scattering through an angle θ_3, on the one hand, and the normal to the plane abc of the scattering through the large angle θ_1. (The direction of polarization of the incident beam is parallel to the vector $\mathbf{n}$.) In the notation adopted in Fig. 1, we have

$$\cos\vartheta = \left[1 + \left(\frac{\sin\theta_2\sin\varphi}{\cos\theta_2\sin\theta_3 + \sin\theta_2\cos\theta_3\cos\varphi}\right)^2\right]^{-1/2}. \quad (8)$$

In the calculation of $A_2(\theta_3)$ an allowance is made for the contribution of the asymmetry which appears only in the case of scattering through a large angle θ_1. The asymmetry in the scattering of polarized electrons through small angles is ignored, because in this case the values of $A_1(\theta)$ are small.[6,7] These calculations also ignore the change in the direction of polarization in the forward scattering case. As shown in Ref. 1, when a beam with the polarization $\mathbf{P}_0$ is scattered, the magnitude and the new direction of the polarization $\mathbf{P}$ after the collision depends on the ratio of the amplitudes of the forward scattering (f) and the scattering accompanied by spin flip (g). According to Refs. 6 and 7, in the case of small angles we have $g \ll f$ and, consequently, $\mathbf{P} \approx \mathbf{P}_0$.

Before calculation of the scattering cross sections and asymmetries the range of integration is usually selected after an analysis of the dependence of the integral in the denominator of Eq. (6) on the angle of the cone representing scattering through the small angle θ_2. The angle of the onset of saturation of this dependence governs the range of the angle of θ_2. For example, in calculation of the cross sections and asymmetries of the scattering of electrons of energy $E_p = 500$ eV we used the range of small angles up to 20°.

Calculations of $A_n(\theta)$ and $d\sigma_n(\theta)/d\Omega$ were made by numerical integration of Eqs. (6) and (7) on a computer for double scattering and for scattering of higher multiplicity of $E_p = 500$ eV electrons and the results are presented in Figs. 2 and 3.

The asymmetries and the cross sections for triple, quadruple, etc. scattering processes were calculated as follows. According to the approximation of small-angle scattering, in the case of multiple backward scattering only one of the scat-

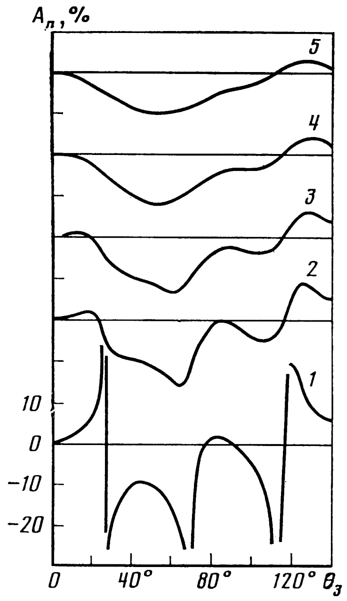

FIG. 2. Dependences of the asymmetry or the scattering angle for $E_p = 500$ eV. The numbers alongside the curves give the scattering multiplicity. The angle $\theta_3 = 0$ corresponds to backward scattering.

tering angles is large and the others are small. This is justified by the very rapid fall of the cross section on increase in the scattering angle. Therefore, in the case of triple scattering, we substituted in Eqs. (6) and (7) not the tabulated asymmetries and cross sections for single scattering $A_1(\theta_1)$ and $d\sigma_1(\theta_1)/d\Omega$, but the calculated values of $A_2(\theta_3)$ and $d\sigma_2(\theta_3)/d\Omega$. Following exactly the same procedure in dealing with quadrupole scattering, we substituted in Eqs. (6) and (7) the values of $A_3(\theta_3)$ and $d\sigma_3(\theta_3)/d\Omega$. The cross sections for the scattering through a small angle $d\sigma(\theta_2)/d\Omega$ were constant in calculations of the interactions of any multiplicity.

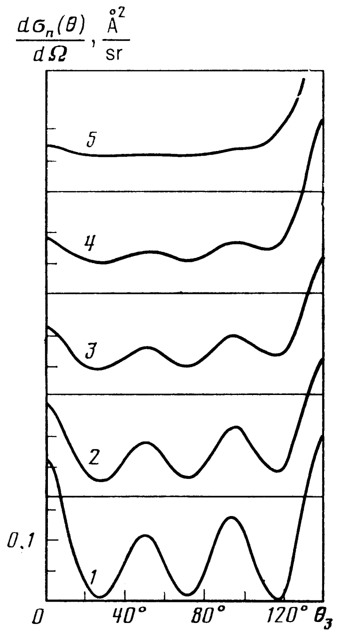

FIG. 3. Differential scattering cross sections of $E_p = 500$ eV The numbers alongside the curves give the scattering multiplicity. The angle $\theta_3 = 0$ corresponds to backward scattering.

Typical results of the calculations presented in Fig. 3 showed that an increase in scattering multiplicity results in rapid smoothing out of all the singularities typical of the differential cross section for single scattering. For $n > 3$ and relatively large values of θ the angular distributions become nearly isotropic. In the case of the scattering asymmetry (Fig. 2), in spite of the fact that the singularities of the dependence $A_n(\theta_3)$ are also smoothed out on increase in the multiplicity, the absolute values of $A_n(\theta_3)$ remain sufficiently large for some angles θ_3. Hence, in the case of multiple collisions there are such scattering directions for which a fairly strong asymmetry is combined with a high scattering intensity.

3. ELASTIC SCATTERING ASYMMETRY

We investigated experimentally the angular dependences of the asymmetry in elastic scattering of polarized electrons by the surface of polycrystalline gold. This investigation was carried out using an M-24 ultrahigh vacuum unit[8] at a residual pressure of less than 10^{-11} Torr. In these experiments we used a source of polarized electrons, based on photoemission from $GaAs_{0.64}P_{0.36}$ solid solutions.[8,9] The polarization P_0 of a beam from a source was $35 \pm 2\%$. The asymmetry was separated by modulation of the direction of polarization of the primary beam of electrons by reversing the sign of the circular polarization of light at a frequency of 31 kHz.

In these experiments we determined the dependence of the current representing elastically scattered electrons reaching a collector of a four-grid quasispherical analyzer (collection angle 104°) on the angle of incidence α of a beam on a target. (We used the same symbol I for the current as for the intensity of the electron beam.) We measured simultaneously a signal ΔI resulting from reversal of the direction of polarization of the incident beam. This signal was recorded by the method of lock-in detection at the frequency of modulation of the direction of polarization. The asymmetry resulting from such scattering was [see Eq. (1)]

$$A = \frac{1}{P_0}\frac{I^+ - I^-}{I^+ + I^-} = \frac{1}{2P_0}\frac{\Delta I}{I}, \tag{9}$$

where I^+ and I^- are the currents to a collector corresponding to different directions of polarization of the incident beam. The coefficient 1/2 in Eq. (9) was used because the measured current I was the average of the currents I^+ and I^-.

The target was a thick film of gold ($\gtrsim 100$ Å) evaporated on a beryllium substrate under ultrahigh vacuum conditions directly in the M-24 unit. A polycrystalline beryllium substrate was in turn formed by evaporation on a target of Au or W. It is known[10] that this deposition method ensures formation of smooth and continuous polycrystalline films. Moreover, an analysis of our films by the low-energy electron diffraction method demonstrated complete absence of crystallographic ordering of the Be and Au surfaces.

During the initial stage of the investigation we determined also the angular dependence of the asymmetry of the scattering of polarized electrons by a polycrystalline foil 50 μm thick. These measurements showed that the asymmetry did not agree with the results obtained for an evaporated film. Moreover, the dependence $A(\alpha)$ had a number of sin-

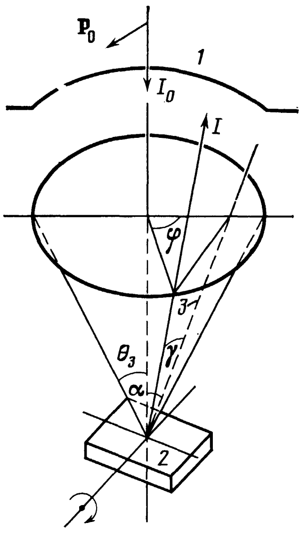

FIG. 4. Schematic representation of the scattering of polarized electrons along the direction of a collector 1 in a four-grid quasispherical analyzer; 2 is the target and 3 is the normal to the surface.

gularities which were clearly of diffraction origin. An x-ray structure analysis of the foil confirmed the hypothesis that it exhibited long-range crystallinity.

The thickness of an evaporated film was measured both with a quartz balance and by the method of Auger electron spectroscopy. The difference in the thicknesses determined by the two methods was less than 10%. This result confirmed the layer (and not island) nature of film growth. The Auger spectrum of a thick gold film showed no peaks of the main accidental impurities S, C, or O.

In comparing the calculated and experimental results we first found the asymmetry of the scattering of polarized electrons by a solid in a wide solid angle equal to the angle of collection by the four-grid analyzer. Figure 4 shows the case of electron reflection for some fixed scattering angle θ_3. The primary electron beam with the P_0 polarization was incident on a target at an angle α relative to the normal to the surface. Clearly, the actual angle γ of emergence of electrons depended on the azimuthal angle φ. In the case of such scattering the quantity $A(\alpha)$ can be found from the expression

$$\sum_{n=1}^{N}\sum_{m=1}^{M} A_n^m \cos\varphi\, I_n^m\, \Delta\Omega_m \Big/ \sum_{n=1}^{N}\sum_{m=1}^{M} I_n^m\, \Delta\Omega_m, \tag{10}$$

where I_n^m and A_n^m are the current and the asymmetry of the scattering into an element $\Delta\Omega_m$ of a wide solid angle. The factor $\cos\varphi$ allows for the scattering of electrons in a plane nonparallel to the specular reflection plane of the beam.

An analysis of the expression for the intensity in the case of multiple scattering of electrons [Eq. (5)] made it possible to simplify the procedure of calculation of the quantity $A(\alpha)$ in Eq. (10). We represented Eq. (5) as consisting of two factors. One of them $(\sim\sigma_y^{n-1}\rho^n/n\sigma^n)$ is proportional to the probability of emergence of electrons in vacuum as a result of n-fold scattering, whereas the second $\{\sim[(1-\cos\alpha/\cos\gamma)^{-1}d\sigma_n(\theta_3)/d\Omega]\}$ determines the scattering cross section for the n-fold interaction allowing for the angles of incidence and emission of electrons from a solid. In calculation of the intensity of thre electrons scattered into a wide solid angle it was found that in the case of the n-fold scattering process it is governed primarily by the first factor and is practically independent of the second. This is not surprising because the dependences of the scattering cross sections of different multiplicity differ mainly in respect of the depth of modulation (Fig. 3). Therefore, the values of the integrals of these functions, calculated for a wide range of angles, are practically identical. We used this conclusion to calculate independently the asymmetry of the scattering of electrons for processes of different multiplicity in a wide solid angle.

We also allowed for surface losses which appear when an electron crosses the target–vacuum interface. The dependence of the probability of the surface losses on the angle of incidence (or emergence) for gold is given by the expression[5]

$$W(E_p,\alpha)=2{,}7/E_p^{1/2}\cos\alpha. \tag{11}$$

Therefore, the final expression for the scattering asymmetry of polarized electrons of different multiplicity in a wide solid angle is

$$A_n(\alpha)=\sum_{m=1}^{M}A_n(\theta_3)\cos\varphi[1-W(E_p,\gamma)]\left(1-\frac{\cos\alpha}{\cos\gamma}\right)^{-1}\frac{d\sigma_n(\theta_3)}{d\Omega}\Delta\Omega_m\Big/\sum_{m=1}^{M}[1-W(E_p,\gamma)]\left(1-\frac{\cos\alpha}{\cos\gamma}\right)^{-1}\frac{d\sigma_n(\theta_3)}{d\Omega}\Delta\Omega_m. \tag{12}$$

The calculations were carried out numerically on a computer in the range of angles θ_3 from 12 to 52°, set by the actual details of the analyzer. Use was made of the values of $A_n(\theta_3)$ and $d\sigma_n(\theta_3)/d\Omega$ shown in Figs. 2 and 3.

The results of calculations of $A_n(\alpha)$ for two electron energies E_p are presented in Fig. 5. We can see that: 1) the asymmetry of the scattering of polarized electrons rises monotonically on increase in the angle of incidence of elec-

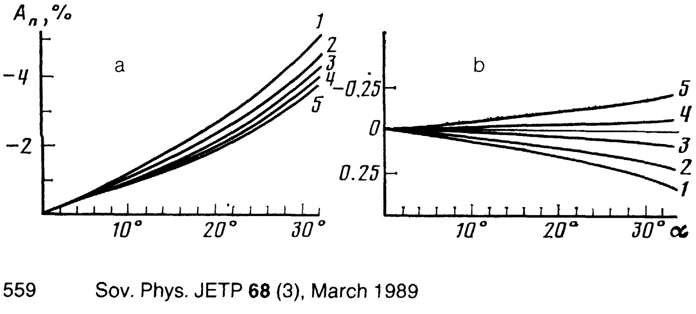

FIG. 5. Dependences of the asymmetry on the angle of incidence α for different multiplicities of the scattering of polarized electrons into a collector of a four-grid analyzer: a) $E_p = 100$ eV; b) $E_p = 250$ eV. The numbers alongside the curves give the scattering multiplicity.

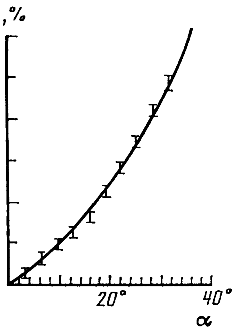

FIG. 6. Experimental dependence of the scattering asymmetry of polarized electrons of $E_p = 100$ eV energy on the angle of incidence on the surface of polycrystalline gold. The continuous curve is calculated by the method described in the present paper.

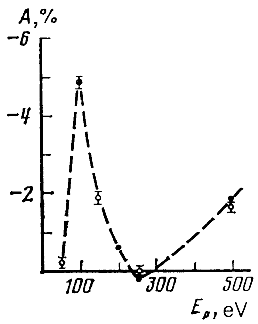

FIG. 7. Experimental (O) and calculated (●) values of the scattering asymmetry for different energies of a primary polarized beam of electrons. The angles of incidence was $\alpha = 32°$. The dashed curve is drawn through for clarity.

trons on the target; 2) an increase in the scattering multiplicity reduces the scattering asymmetry of polarized electrons reflected into a wide solid angle. However, even for the fivefold scattering the value of $A_n(\alpha)$ remains considerable.

The final calculation of $A(\alpha)$ was made using

$$A(\alpha)=\sum_{n=1}^{5} A_n(\alpha)\frac{\sigma_e^{n-1}\rho^n}{n\sigma^n}\bigg/\sum_{n=1}^{5}\frac{\sigma_e^{n-1}\rho^n}{n\sigma^n}. \qquad (13)$$

Since the probability of n-fold scattering decreases rapidly with n, we retained only the first five terms of the series. In these calculations we used the attenuation coefficient σ obtained in Ref. 5 by comparing the experimental and calculated intensities of elastically scattered electrons.

Equation (13) makes it possible to estimate also the contribution of the single interaction to the total scattering asymmetry. The results of this calculation showed that the contribution in question depends very weakly on the electron energy E_p and amounts to $\approx 60\%$ for the investigated substance.

A typical dependence of the calculated and experimental values of the scattering asymmetry on the angle of incidence for a fixed electron energy E_p is plotted in Fig. 6. (In comparing the results of calculations and experiments we allowed for the contact potential between the photocathode and the target, and also for the internal potential of the target.)

Since the experimental and calculated $A(\alpha)$ curves were monotonic dependences with their origin at the center of the coordinate system and this was true of all the electron energies, we were able to plot in Fig. 7 a summary graph containing the results of experiments and calculations for a fixed angle of incidence $\alpha = 32°$. Clearly, the experimental results agreed well with the calculations carried out by the method described above.

4. CONCLUSIONS

Our results thus demonstrate that the proposed scattering model is valid. According to the model, the asymmetry due to the interaction of polarized electrons with a polycrystalline solid is due to the scattering by an atomic-like potential of ionic cores in the target. The specific influence of a solid reduces to the following.

1. In view of the high atomic density, the elastically scattered electrons experience not only single but multiple scattering by ionic cores in a solid and this alters the symmetry.

2. The existence of a large inelastic scattering cross section results in rapid reduction in the intensity of a beam of elastically scattered electrons, which determines finally the dominant role of the asymmetry due to single scattering.

3. When an electron crosses the target–vacuum interface, it loses energy and this reduces the intensity of a beam of elastically scattered electrons which in its turn affects the asymmetry of the scattering in a wide solid angle.

In conclusion, the authors regard it as their pleasant duty to thank Yu. P. Yashin and V. N. Yakovlev for their help in the experiments.

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