STUDY OF ANGULAR DEPENDENCES OF REFLECTION ELECTRON ENERGY LOSS SPECTRA OF Ge BY PRINCIPAL COMPONENT ANALYSIS

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Angular dependences of reflection electron energy loss (REEL) spectra of polycrystalline germanium for primary electron energies of 300 and 500 eV and under various experimental geometry conditions have been explored. It has been found that within the range of electron scattering angles of 94—146°, the REEL spectra of Ge can be described by linear combinations of three components. The first component contains the single surface plasmon peak, the second component — the single bulk plasmon peak, and the third component — none of them. The relative intensities of the REEL spectrum components depend differently on the electron incidence, reflection, and scattering angles. The strongest dependence on the electron scattering angle was observed for the bulk plasmon peak. In the scattering angle range of 94–146°, its intensity can vary by a factor of 2.8, whereas the intensity of the surface plasmon peak varies no more than by a factor of 1.3. These features of spectra should be taken into account both in quantitative REEL spectroscopy and when developing relevant physical models.

Electron energy loss spectroscopy is widely used for electron structure studies of surface layers of solids [1–3]. Its "reflection" variant is the most convenient for practice, since the fabrication of very thin specimens transparent for electrons is not needed. In real experiments, various geometric setups are exploited, which differ by the angle between the target surface and the primary electron beam (α), by the angle between the target surface and the direction of registration (β), and by the electron scattering angle θ. As early as in [4], it was shown that there are specific dependences of the REEL spectrum shapes on the angles α and β, but no attention was paid to their probable dependence on the electron scattering angle θ. We have shown that θ notably affects the REEL spectra of Mg, Al, Ge, and In, even at α and β constant [5]. But a strong overlapping of certain REEL spectrum components might modify substantially the dependences obtained in this work for the peak intensities of surface and bulk plasmons, normalized by the peak intensity of the elastically scattered electrons, on the scattering angle θ.

In the previous works [6, 7], we have proposed a new approach to the analysis of the REEL spectrum modifications depending on the geometric conditions of experiment. Its specific feature consists in a substitution of the conventional analysis of a separate spectrum by an analysis of an ensemble of experimental spectra obtained at various angles of electron incidence, reflection, and scattering. Such a spectral analysis does not include any suggestion about the number, shapes, and positions of spectral components. The analysis of an ensemble of experimental REEL spectra makes it possible to determine the spectral components and to present each spectrum as a linear combination of the latter.

Taking advantage of this approach, we have analyzed the REEL spectra of Al at the primary electron energies $E_p = 300 \div 700$ eV [6], and of In at $E_p = 300$ eV [7]. The angular dependences of the ratio of the peak intensities for surface and bulk plasmons were established. The peak intensity of the bulk plasmon in In occurred to depend on the scattering angle θ stronger than on the angles α and β, whereas in Al, the dependence of the spectrum shape on the angle θ in the investigated range of energy E_p was not observed. The aim of this article was to apply our method of analyzing REEL spectra to both the consideration of those of Ge and the determination of the dependence of the component peak intensities on the geometry of a setup, in particular, on the electron scattering angle.

The specimen for investigation was produced by sputtering a polycrystalline film of Ge on a substrate of polished Si *in situ* in a vacuum chamber with a base pressure of $\approx 10^{-10}$ Torr. The element composition of the target was controlled by means of electron Auger spectroscopy. The REEL spectra of Ge were measured using a Hughes—Rojansky electron energy analyzer with an energy resolution of $\approx 0.2\%$. The setup scheme and the procedure of the spectrum calibration were described in the previous work [7]. The measurements were carried out at the energy of primary electrons $E_p = 300$ and 500 eV. The scheme of electron scattering is outlined in Fig. 1, where it is shown how the angles α, β, and θ were reckoned. An electron gun and the specimen can rotate independently around two mutually perpendicular axes, which makes it possible to vary not

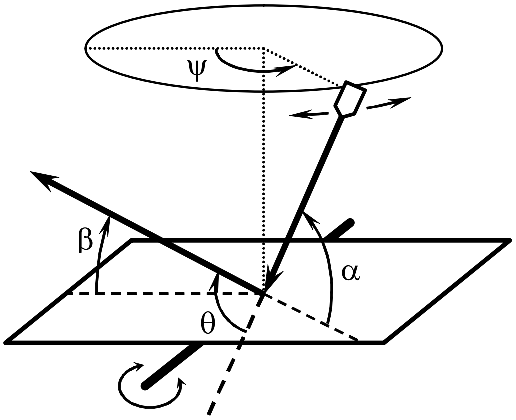

Fig. 1. A geometric setup of experiment

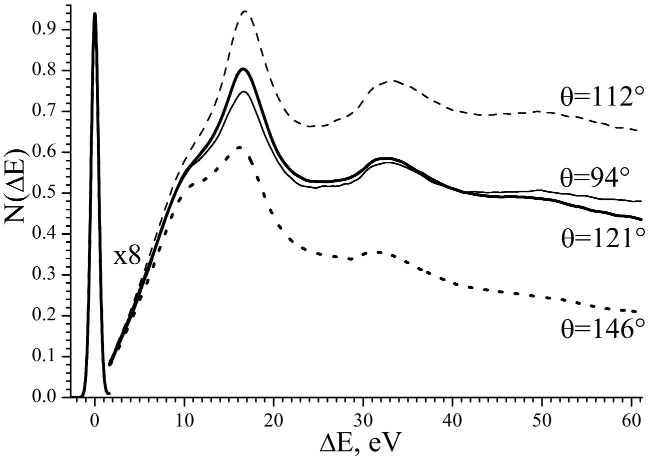

Fig. 2. The REEL spectra of Ge at constant $\varphi = \varphi_0 = 3.11$ ($\beta = 30°$, $\alpha = 64°$), $E_p = 300$ eV, and for various θ

only the angles α and β but also an azimuthal angle ψ that determines the electron scattering angle

$$\theta = \pi - \arccos\left(\sin\alpha_0 \sin\beta_0 + \cos\alpha_0 \cos\beta_0 \cos\psi\right), \qquad (1)$$

where $\alpha_0 = 64°$ and $\beta_0 = 30°$. To characterize the dependences concerned on the angles α and β, we used, as in the previous works [6–8], the angle parameter $\varphi = \sin^{-1}\alpha + \sin^{-1}\beta$.

The REEL spectra of Ge were measured at six positions of an electron gun, i.e. at six angles ψ that corresponded to the electron scattering angles θ's in the range of 94–146°, and at five different incident angles of primary electrons α that corresponded to five values of the parameter φ in the interval from $\varphi_{\min}(\theta)$ at $\alpha = \beta$ to $\varphi_{\max}(\theta)$ at $\beta = 10°$. All the measured spectra were calibrated, reduced to the same instrument function, which was a Gaussian possessing the width $H = 1$ eV at half of the height, and normalized to the unit area of the peak of elastically reflected electrons, as was done in [7]. An ensemble $\{N_i(\Delta E)\}$ of REEL spectra, treated in such a manner, of Ge was used for further analysis. In Fig. 2, the REEL spectra for Ge are shown, as an example, for $E_p = 300$ eV, $\varphi = \varphi_0 = 3.11$, and several values of θ. It is seen that the peak intensities of the bulk and surface plasmons depend nonmonotonically on the scattering angle θ, as it took place also for In [7].

A quantitative analysis of the angular dependences of REEL spectra was carried out making use of principal component analysis [9–11], analogously to what has been described in detail in our previous work [7]. At first, the inelastic parts of the REEL spectra of Ge, $N_i(\Delta E)$, in the energy loss interval of $0 < \Delta E < 60$ eV, which stood for the principal components of the input function set, were determined for every energy of primary electrons. The REEL spectra were measured at 5 values of the parameter φ for each of the 6 scattering angles θ's. All the input spectra $\{N_i(\Delta E),\ i = 1, 2, \ldots, n;\ n = 30\}$ were discretized with a steady step of 0.02 eV. It was found that the whole ensemble of the spectra $\{N_i(\Delta E),\ i = 1..n\}$ can be described, as in the In case [7], making use of the first three principal components, whose contribution relations define the shape of each spectrum. The linear combinations of those principal components determine the components: S, V, and C_3. The first component, $S(\Delta E)$, includes the surface plasmon peak and does not include the bulk plasmon one. The second component, $V(\Delta E)$, includes the bulk plasmon peak and does not include the surface plasmon one. The third component, $C_3(\Delta E)$, was determined in such a manner to include neither the surface peak nor the bulk one. The absence of the relevant plasmon peak features from the components was monitored by their differentiation.

The S-, V-, and C_3-components of Ge, obtained at $E_p = 300$ eV, are depicted in Fig. 3. Their linear combination

$$N_i(\Delta E) = k_i^s S(\Delta E) + k_i^v V(\Delta E) + k_i^3 C_3(\Delta E), \qquad (2)$$

where k_i^s, k_i^v, and k_i^3 are the weight factor of each component contribution to the spectrum $N_i(\Delta E)$, can describe every spectrum of the input ensemble $\{N_i(\Delta E)\}$. From Fig. 3, one can see that the C_3-component is almost free of peculiarities of the surface

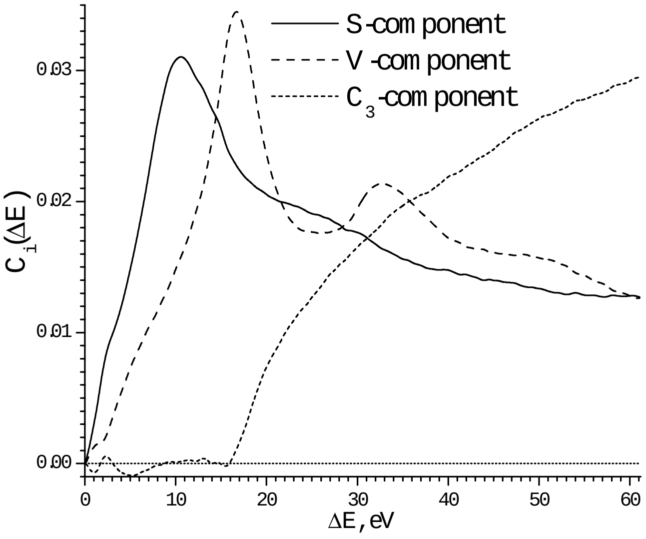

Fig. 3. The components of REEL spectra of Ge

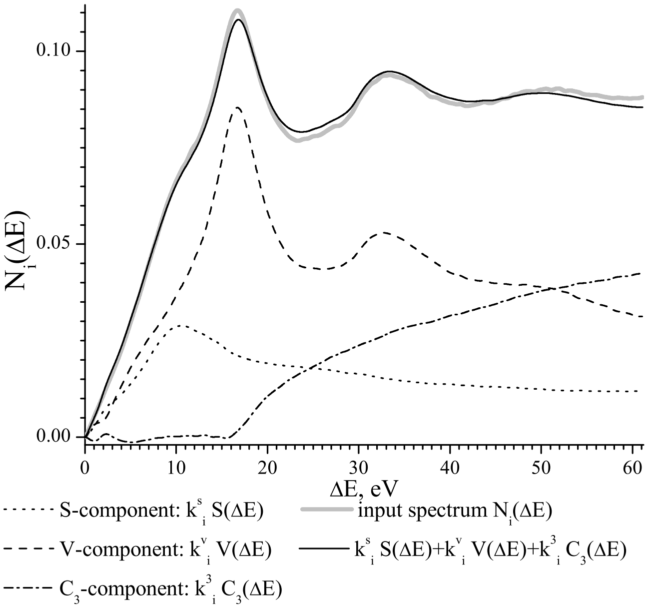

Fig. 4. The REEL spectrum of Ge with the maximal contribution of the third component (E_p = 300 eV, θ = 104°, β = 50°). S-component $k_i^s S(\Delta E)$ – dotted curve, V-component $k_i^v V(\Delta E)$ – dashed curve, C_3-component $k_i^3 C_3(\Delta E)$ – dash-dotted curve, input spectrum $N_i(\Delta E)$ – gray curve, a three-component linear combination $k_i^s S(\Delta E) + k_i^v V(\Delta E) + k_i^3 C_3(\Delta E)$ – solid curve

or bulk plasmon origin. A contribution of the third component to the range of the plasmon single peaks is illustrated in Fig. 4. Here, the shape of the spectrum from the ensemble $\{N_i(\Delta E)\}$ is shown, for which the contribution of the C_3-component is maximal, i.e. the spectrum with the maximal value of k_i^3. It is seen that even for this spectrum, the contribution of the third component in the excitation range of single plasmons can be neglected. Assuming that the surface plasmon peak is totally contained in the S-component of the spectrum, and the bulk plasmon peak in the V-component, the coefficients k_i^s and k_i^v, which, according to (2), determine the contribution of those components into the spectrum $N_i(\Delta E)$, can be used for plotting the relative angle dependences of single plasmon peak intensities. Nevertheless, for plotting the relative dependences of the peak intensities, we used the values, which estimated the intensities of the plasmon peaks in the $N_i(\Delta E)$ spectrum, rather than the values of the coefficients k_i^s and k_i^v themselves. For the evaluation of the former, we used the areas under the curves describing the shapes of S- and V-component in the energy loss intervals of 0–18 and 0–24 eV, respectively:

$$I_i^s = k_i^s \int_0^{18} S(\Delta E) d(\Delta E), \tag{3}$$

$$I_i^v = k_i^v \int_0^{24} V(\Delta E) d(\Delta E). \tag{4}$$

Since the spectra $\{N_i(\Delta E)\}$ are normalized by the peak intensity of elastically reflected electrons, the values of I_i^s and I_i^v are also normalized. Such an estimation of the peak intensities is rather rough. The values of those intensities should be further subjected to refinement using, e.g., the bulk and surface loss functions for the approximation of the shapes of the plasmon peaks. But the estimation procedure for those intensities does not affect their angular dependences.

In Fig. 5, the dependences of I^s and I^v on the electron scattering angle θ are shown for E_p = 300 and 500 eV, and at the constant value of the angle parameter $\varphi = \varphi_0 \equiv \sin^{-1}\alpha_0 + \sin^{-1}\beta_0 = 3.11$. One can see from the figure, that, in contrast to the results of [5], the dependences $I^s(\theta)$ and $I^v(\theta)$ are different. The dependences $I^v(\theta)$ have a clear maximum, whose position shifts towards lesser θ's with E_p increasing. Those dependences are similar to the changes of the Ge REEL spectra ordinate at the point corresponding to the peak position, obtained in [5]. But it occurred that the intensity variations of the bulk plasmon peak by more than a factor of 2.5 at varying θ, are substantially

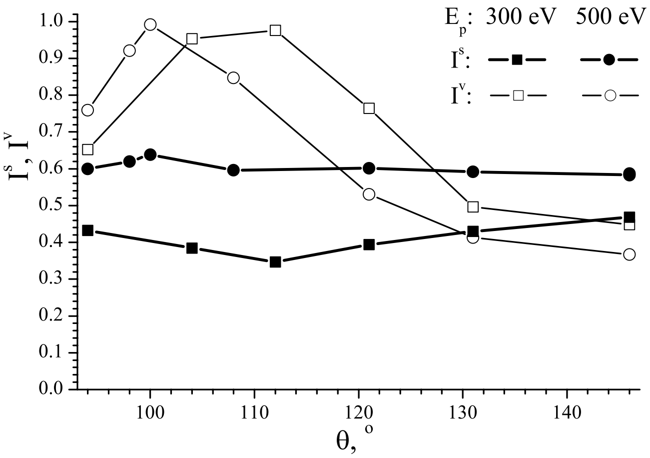

Fig. 5. Dependences of the values I^s, I^v on the scattering angle θ at $\varphi = \varphi_0$ and for various E_p

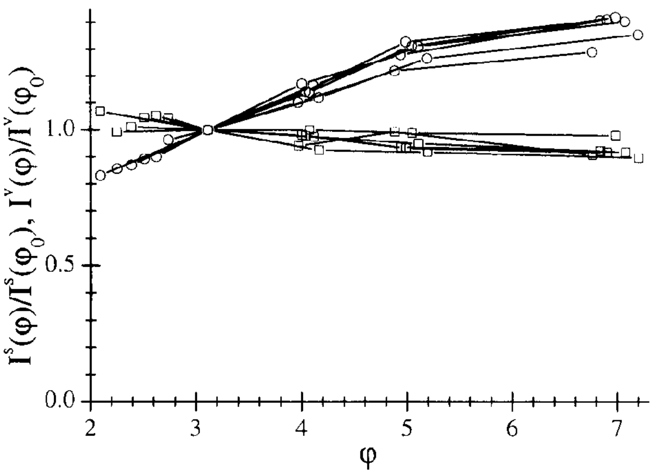

Fig. 6. Relative dependences of the values $I^s$and I^v on the angle parameter φ for $E_p = 300$ eV and at various scattering angles θ

greater than the variations of the relevant spectrum ordinate by a factor of 1.3. The intensity of the surface plasmon peak is practically independent of the angle θ, which contradicts to the dependence established in [5]. Those deviations in the dependences on θ are due to the essential peak overlapping in the REEL spectrum of Ge, as was noted in [5].

In Fig. 6, the dependences $I^s(\varphi)/I^s(\varphi_0)$ and $I^v(\varphi)/I^v(\varphi_0)$ for $E_p = 300$ eV and various scattering angles θ are shown. They are seen to depend weakly on θ. The deviations from the averaged values are not greater than 5%. At $E_p = 500$ eV, the dependences under consideration are similar. The value of I^s and I^v grows and diminishes, respectively, with the increasing angle parameter φ, which does not contradict a conventional picture [4, 8]. A comparison of the dependences $I^s(\theta)$ and $I^v(\theta)$ with $I^s(\varphi)$ and $I^v(\varphi)$ evidences for that the variation of the scattering angle θ affects the shape of the REEL spectrum stronger than the variation of the angle parameter φ, i.e. of the angles α and β.

Thus, the analysis of the REEL spectra of Ge, carried out above for primary electron energies of 300 and 500 eV and various angles of electron incidence, registration, and scattering, showed that those spectra can be described by a linear combination of three components. The relative angle dependences of the intensities of single surface and bulk plasmons have been obtained. The profiles of the dependences $I^s(\varphi)$ and $I^v(\varphi)$ are shown to be almost identical at each value of the scattering angle θ from the investigated interval of 94 – 146°. The intensity of the bulk plasmon peak depends stronger on the scattering angle at the constant value of the angle parameter $\varphi = \varphi_0 = 3.11$, than that of the surface plasmon peak. The intensity of the bulk plasmon peak can vary almost by a factor of 3, whereas for the surface one this factor is substantially smaller.

An essential influence of the scattering angle on the intensity of the bulk plasmon peak in REEL spectra has to be taken into account when analyzing electron spectra measured in various geometric configurations of experiment, as well as when developing physical models of interaction between electrons of intermediate energies and solids.

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АНАЛІЗ КУТОВИХ ЗАЛЕЖНОСТЕЙ СПЕКТРІВ ХАРАКТЕРИСТИЧНИХ ВТРАТ ЕНЕРГІЇ ЕЛЕКТРОНІВ Ge МЕТОДОМ ГОЛОВНИХ КОМПОНЕНТІВ

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Резюме

Досліджено кутову залежність спектрів характеристичних втрат енергії електронів (ХВЕЕ) на відбиття від немонокристалічного германію при енергіях первинних електронів 300 і 500 еВ та в різних геометричних умовах експерименту. Встановлено, що спектри ХВЕЕ Ge у діапазоні кутів розсіяння електронів 94—146° можуть бути описані лінійною комбінацією трьох компонентів. Один із них містить пік однократного поверхневого плазмона, другий – пік однократного об'ємного плазмона, а третій не містить піків ні об'ємного, ні поверхневого плазмонів. Відносні інтенсивності компонентів спектра ХВЕЕ по-різному залежать від кутів падіння, виходу та розсіяння електронів. Найсильніша залежність від кута розсіяння електронів спостерігалась для об'ємного плазмона. В інтервалі кутів розсіяння 94—146° його інтенсивність може змінюватись в 2,8 раза, тоді як інтенсивність піка поверхневого плазмона змінюється не більше ніж у 1,3 раза. Ці особливості спектрів повинні враховуватися як у кількісній спектроскопії ХВЕЕ на відбиття, так і при побудові відповідних фізичних моделей.