E3-2001-224

V. L. Aksenov, Yu. V. Nikitenko, V. V. Proglyado,
E. A. Raitman*, V. N. Gavrilov*

NEUTRON REFLECTION FROM AN ULTRASONICALLY EXCITED LAYERED STRUCTURES

Submitted to «Surface. Roentgen, Synchrotron
and Neutron Investigations»

---

*Physics and Power Engineering Institute, LV-1006 Riga, Latvia

1. Introduction

Layered synthetic nanostructures are finding ever-widening application in microelectronics, e.g., to produce data storage devices, magnetic elements for audio- and video-recording heads, thermal or magnetic sensors, etc. This is because layered nonostructures have unusual magnetic, elecrophysical, thermal, etc. properties that are not observed in continuous materials. The properties of the structures are determined, to a large extent, by the interfaces that often occupy a quite considerable volume of the structure. At the same time, the interfaces determine the conditions of transmission through the structure of different types of radiation such as neutrons or sound. This explains why the study of the transmission of neutron or sonic waves through the structures and of their interactions in the interior of the structures is also an efficient way of investigation of the structures themselves.

The effect of ultrasonic waves (USW) on the scattering of neutrons or X-rays in mono-crystals is studied in many works [1-5]. In [6] the diffraction of neutrons from the surface of the acoustic wave at glancing angles of incidence is studied. In [7] the spin-echo method is applied to measure the spectrum of the neutrons scattered on USW. In addition, the problem of UCN storage made the authors of [8,9] to investigate theoretically the effect of sonic vibrations on the total reflection of UCN neutrons.

This article presents the first results on the glancing reflection of neutrons from layered structures when transversal and longitudinal standing acoustic waves are excited in the structures.

The reflection of neutrons with a glancing angle of several milliradians appears to be an efficient tool. Actually, one energy quantum of the acoustic wave with a frequency of say 100 MHz is 413.6 neV and the wave vector $k = 1.26\times10^{-5}$ Å^{-1} (for the velocity of sound in medium 5000 m/s). These small energy and momentum transfers «sensed» by the neutron are comparable with its energy and wave vector in the direction perpendicular to the interface in the structure. Thus, they can be easily determined by measuring a change in an absolute value of the reflection coefficient or by observing the arising off-specular reflection.

2. Scheme of measurements

Figure 1 illustrates the measurement schematically. The neutron falls at the glancing angle θ_0 and is reflected at the glancing angle θ. A longitudinal acoustic wave LAW(acoustic wave vector q is aligned with the wave amplitude) or a transverse acoustic wave TAW(wave amplitude is perpendicular to the acoustic wave vector) is used. At the same time, the wave amplitude is always parallel to the perpendicular component of the neutron wave vector k_z. To excite the LAW, the piezotransformer P1 is on the surface of the substrate and to excite a TAW, the transformer P2 is fixed on a lateral side of a sample.

The reflection of neutrons from the layered structure is determined by k_z, the wave vector component perpendicular to the interface. In this connection, at neutron scattering on excited oscillations of importance is the momentum transfer δk_z(the transfer of the wave vector) and the neutron scattering probability $W(\delta k_z)$ in this direction.

3. Reflection from the wave resonator

Let us investigate the layered structure glass/Cu(100nm)/Ti(150nm)/Fe(12nm)/Gd(3nm)/Ti(50nm)/Cu(10nm) placed in the external magnetic field H inclined to the sample surface. The structure is excited with longitudinal sound. The expected change of the perpendicular component of the wave vector is small, on the order of 10^{-5}Å^{-1}. In the titanium layer of the structure a field of standing neutron waves is formed at k_z smaller than its critical value in the copper layer, $k_{lim}= 9.1\times10^{-3}$Å^{-1} [8]. For certain k_z the antinodes of the neutron standing wave lie either on the iron or gadolinium layer. In the first case, there is an increase in the flux of neutrons that experience a transition from one to the other neutron spin state (spin-flip) in the magnetic field. As a result, the reflection coefficient of spin-flip neutrons increases and that of nonspin-flip neutrons decreases. In the second case when the antinode of the standing wave lies on the gadolinium layer, neutron absorption increases and the neutron reflection coefficient falls. The discussed layered structure is a measurer of the neutron wave phase φ that the neutron acquires as it moves from the absorbing layer to the neutron reflector (Cu(100nm) layer) and backwards:

$$\varphi = 2L\, k_z= 2L(k_{0z}^{2}-\alpha U)^{1/2}\,, \qquad (1)$$

where L is the reflector to absorbing layer distance, k_{0z} is the perpendicular component of the wave vector of the incident neutron, U is the potential of the titanium layer, $\alpha=m(2\pi/h)^2=2.41\times10^{-7}$ Å^{-2} neV^{-1}, m is the neutron mass and h is the Planck constant.

For the momentum transfer in the Z-axis direction $\delta\, k_z$ as the neutron flies the distance $2L-L_1$, the phase shift is:

$$\varphi(L_1) = (2L-L_1)\, k_z+L_1(k_z\pm \delta\, k_z). \qquad (2)$$

Equating (1) to (2) we have the wave vector of the neutron flying the distance 2L equal to:

$$k_z'(L_1) = ((k_{0z}')^2 - u)^{1/2} = [(2L-L_1)\, k_z + L_1(k_z \pm \delta\, k_z)]\ /2L \qquad (3)$$

Changes in L_1 within the limits 0÷2L are equivalent to changes in the wavelength within the limits $\lambda_{min(max)}=2\pi\sin(\theta_0)/[\ (k_z \pm \delta\ k_z)^2 + U/\alpha]^{1/2}$. As a result, we obtain the following dependence of the reflection coefficient on the ultrasonic wave-excited structure:

$$R_{exc}(\lambda)=W \int R(\lambda')d\lambda' / \int d\lambda' + (1-W)\, R(\lambda), \qquad (4)$$

where $R(\lambda)$ is the reflection coefficient from the nonexcited structure, W is the probability of neutron scattering on medium vibrations in the titanium layer, the integration limits being λ_{min} and λ_{max}.

Figures 2a,b show the results of measurements of the reflection coefficient without acoustic excitation, $R^+(\lambda)$, and of the ratio of reflection coefficient at acoustic excitation with the frequency 90 MHz to that without acoustic excitation, $\eta = R_{exc}^+(\lambda)/R^+(\lambda)$. Fitting experimental η to that calculated by Eq. (4) we obtain $\delta\ k_z$ and W that depend on the neutron wavelength and whose values for the frequency 30 MHz are given in Table 1. It is seen from the Table that the ensemble average wave vector transfer lies within the limits $(4\div7)\times10^{-5}$ Å^{-1}. The experimental data obtained for the frequency 90MHz give the average momentum transfer 1.5×10^{-4} Å^{-1} already.

4. Reflection from the periodic layered structure

Figure 3a shows the $R(\lambda)$ dependence curves of the layered structure Zr(10nm)/[^{57}Fe(1.6nm)/Cr(1.7nm)]×26/Cr(50nm)/glass with a repetition period of 32.84 Å. The black squares are for the structure excited with 150 MHz transverse sound and the white circles stand for the structure without acoustic excitation. Transverse sound with $\varphi = 0$ is used. It is seen that the effect of sound is analogous to quasielastic neutron scattering. Figures 3 b, c, d present the ration η of the reflection coefficient from the excited structure to that from nonexcited one for the frequencies 30, 90 and 150 MHz. It is seen that the effect gets stronger as the frequency increases. According to the experimental data, the mean square wave vector transfer $\delta k_z = 6.9\times10^{-4}\text{Å}^{-1}$ for 150 MHz. At the same time, the perpendicular component of the wave vector is 0.096Å^{-1} at Bragg reflection of the first order (λ=2.77 Å). Let us make estimates assuming that neutron scattering on a gridding made of density changes in the medium due to formation of standing acoustic waves. If sound is in the direction $\varphi = 0$, the change in the wave vector is:

$$\Delta k_z(\varphi=0) = [(k\times\sin(\theta_i))^2 + 2n\alpha\, E - (ns\, q)^2 - 2nsq\, k\cos(\theta_i)]^{1/2} - k\times\sin(\theta_i), \qquad (5)$$

where n = ±1, ±2 .. is the order of scattering, s = ± corresponds absorption (birth) of an acoustic quantum, E = hf , $q=2\pi f\times u^{-1}$, f is the frequency of the acoustic wave, and u is the velocity of sound in the titanium layer.

From Eq. (5) it follows that $\Delta k_z \approx 9.4\times10^{-4}$ Å^{-1}. Since δk_z corresponds to the ensemble average neutron wave vector transfer, using δk_z and Δk_z we obtain the scattering probability $W = \delta k_z / \Delta k_z \approx 0.73$.

5. Neutron channeling

Figures 4a,b present the neutron reflection coefficient for the structures glass/Cu(100nm)/Ti(150nm)/Cu(30nm) and glass/Cu(100nm)/Ti(150nm)/Cu(30nm) with a 1 cm long Gd_2O_3 absorbing layer on the central surface part of the Cu(30nm) layer. It shows the absorption and propagation of neutrons in the wave-guide whose walls are the copper layers. It is seen that for the first structure at $\chi = \lambda/\theta = 0.75$; 0.9; 1.6; 3.45 there are observed minimums due to resonance-enhanced density of neutrons in the titanium layer at these values of χ (four resonance modes). For the second structure, one can see maximums at the same values of $\chi = 0.75$, 1.6, which points also to an increase in the neutron density in the titanium film for the given values of the parameter χ.

Figures 5a,b,c present the wavelength dependence of the ratio η between the reflection coefficients of ultrasonically excited and ultrasonically nonexcited structure for the reflection angle interval $\theta_1 \div \theta_2 = 0.14 \div 1.23$mrad and different acoustic frequencies. This interval of angles corresponds to energy transfer in the neutron medium. From Fig. 5b it is seen that the dependence $\eta(\lambda)$ has a peak at $\lambda = 3.75$ Å for the frequency 90 MHz. Let us determine the momentum transfer that corresponds to neutron reflection at angles in the interval $\theta_1 \div \theta_2$. The momentum transfer in the direction perpendicular to the interface boundaries is:

$$\Delta k_z = 9\alpha^{1/2} (\theta_0 - \theta)\lambda^{-1}, \qquad (6)$$

where the angle is in mrad and the wavelength is in angstroms. For the known $\theta_0 = 2.6$ mrad and average value of angle $\theta = 0.96$ mrad we obtain $\Delta k_z = -2.0\times10^{-3}$ Å^{-1}. Using this value of momentum transfer and its average value determined in section 3 of this article, 1.5×10^{-4} Å^{-1}, we obtain the scattering probability equal to 7.5 % for the frequency 90 MHz. It is a little higher than the probability obtained for the frequency 30 MHz. Further we assume that the picture of neutron reflection from the excited structure looks as follows. Neutrons are channeled at resonance wavelengths and the excited medium changes the k_z mode of the neutrons. To the initial mode with the wavelength 3.75 Å there corresponds $\eta \approx 3.75/2.6 = 1.45$ which is close to $\eta_3 = 1.6$ of the third mode (Fig. 4a,b). A reasonable question is then: what resonance mode does the neutron change to? To answer it, we use the relation:

$$\Delta k_z = 9\alpha^{1/2} \times \theta_0 (\lambda_i^{-1} - \lambda_f^{-1}) \qquad (7)$$

From Eq. (7) it follows that the final mode has the wavelength $\lambda_f = 9.5$ Å or $\eta = 3.65$ which is close to $\eta_4 = 3.45$ of the fourth mode of the structure (Fig. 4a).

Thus, in the acoustically excited structure there are observed neutron transitions from one resonance mode to another at neutron channeling.

6. Total neutron reflection from glass

Figures 6 a,b,c,d show the angular dependence ΔR(θ) of changes in the neutron reflection coefficient on glass being excited or not excited with sound for the neutron wavelengths 2, 4 , 6, and 8Å. Figures 6 a,b present the ΔR curves for longitudinal sound with the frequencies 30 and 90MHz, respectively. It is seen that there exists an asymmetry that is most strong for the frequency 90MHz. This is evidence for the fact that the probability of momentum transfer in the direction from surface into the interior of the glass is higher than in the backward direction. It is also seen that the dependence on the wavelength is weakly expressed. At the same time, for the wavelength 8Å one can see periodic dependence on the glancing angle (with a step of an order of 0.7 mrad). Figures 6c,d show two ΔR(θ) curves for transverse sound with 49.6 and 149.9 MHz, respectively. It is seen that the ΔR (θ) maximum value is five times larger for transverse than for longitudinal sound. The dependence on the neutron wavelength is also seen, namely, to larger wavelengths there correspond larger scattering angles. From Fig. 6d it is seen that the scattering angle of neutrons with the wavelength larger than 6 Å exceeds 9 mrad. Figures 7a,b show the curves of the wavelength dependence ΔR(λ) of the difference of neutron reflection coefficients from transverse sound-excited and transverse sound-nonexcited glass over different glancing angle intervals for the frequencies 49.6 and 149.9 MHz, respectively. These data confirm the conclusions made on the basis of the data shown in Fig. 6. Figure 7a presents three curves for the glancing angle intervals 0÷1.96 mrad (from sample plane to specular beam – curve 1), 2.44÷14.8 mrad (from specular beam in the direction of larger angles - curve 2) and 1.96÷2.44 mrad (specular beam - curve 3). All the curves demonstrate an increase in the ultrasonic effect in the vicinity of the critical wavelength. It is also seen that if the intensity of the specular beam decreases, the intensities of two off-specular beams increase. Saturation is observed as the wavelength grows. Fig. 7b shows analogous dependence for the frequency 149.9 MHz. In contrast to the curves in Fig. 7a the curves in Fig. 7b bend so that ΔR(λ) gets negative even in the areas of off-specular reflection. This may be explained if assume that diffusely scattered neutrons in the absence of ultrasonic excitation get larger energy at ultrasonic excitation with the frequency 149.9 MHz than 49.6 MHz. Interesting to note also that the intensity of the specular beam decreases more significantly at 149.9 than 49.6 MHz ultrasonic excitation. Also, it should be noted that scattering increases in the vicinity of the critical wavelength for which the antinode of the standing wave is on the glass surface (the effect is the strongest at 150MHz).

Let us make estimates assuming that the scattering of neutrons is on a gridding made of periodic density changes due to formation of the acoustic standing wave. From Fig. 7d it follows that the scattering angle in vacuum Δθ is 1 mrad for the wavelength 4Å and it is as large as 2.5 mrad for the wavelength 6Å. For the scattering angle Δθ at φ=0 we have:

$$\Delta\theta =[(k_i^2+2n\alpha E)- (nsq+ k_i\cos(\theta_i))^2]^{1/2}/ k_i - \theta_i \qquad (8)$$

Table 1. The momentum transfer $\delta K_z(Å^{-1})$ and the scattering probability W(%) over the wavelength intervals $\lambda_{min} \div \lambda_{max}$ for sonic wave with frequency 30 MHz.

$\lambda_{min} \div \lambda_{max}$, Å	$\delta K_z(Å^{-1})$	W(%)
3.0÷4.0	**8.5×10^{-4}**	**6**
4.0÷4.8	**1.0×10^{-3}**	**7**
4.8÷8.6	**2.1×10^{-4}**	**20**

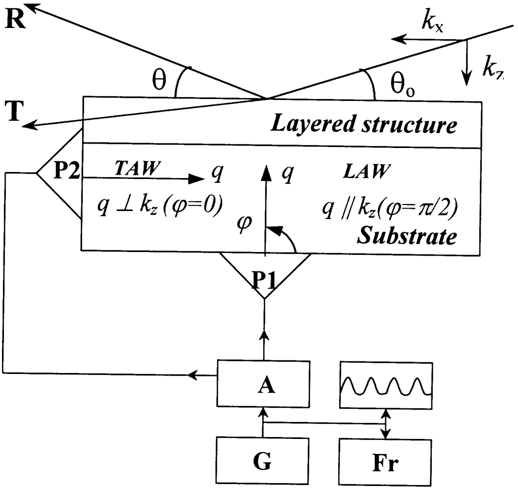

Fig. 1. The schematic of the reflectometric experiment with ultrasonic excitation of the sample.

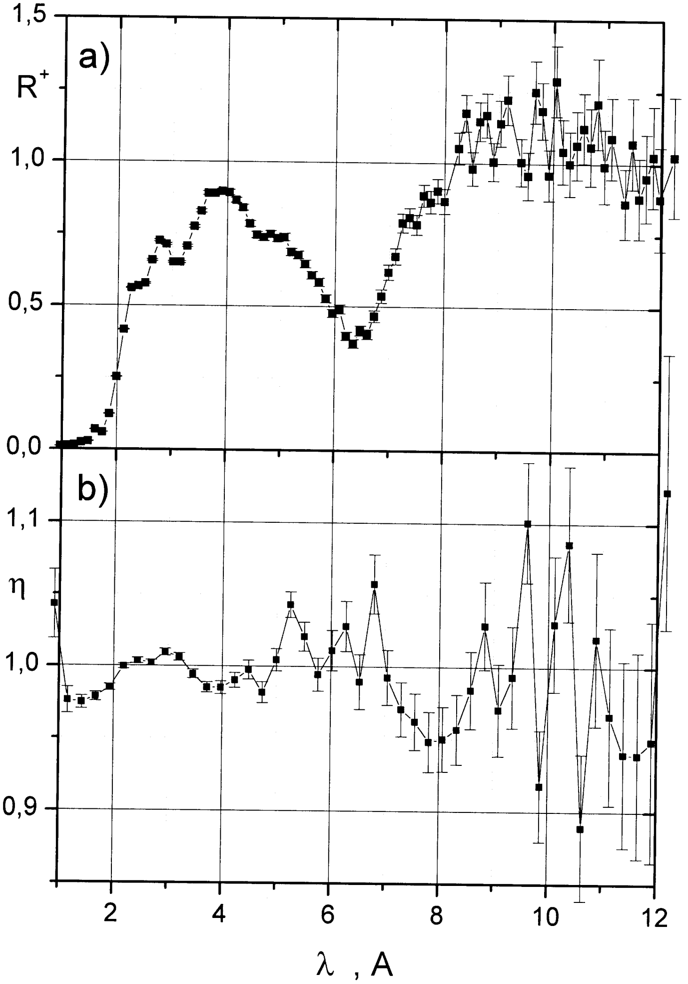

Fig. 2. a) The wavelength dependence of the neutron reflection coefficient without acoustic excitation of the structure $R^{+}(\lambda)$; b) the wavelength dependence η (λ) of the ratio of the neutron reflection coefficient $R_{exc}{}^{+}(\lambda)$ with 90MHz acoustic excitation to the reflection coefficient $R^{+}(\lambda)$ without acoustic excitation of the structure.

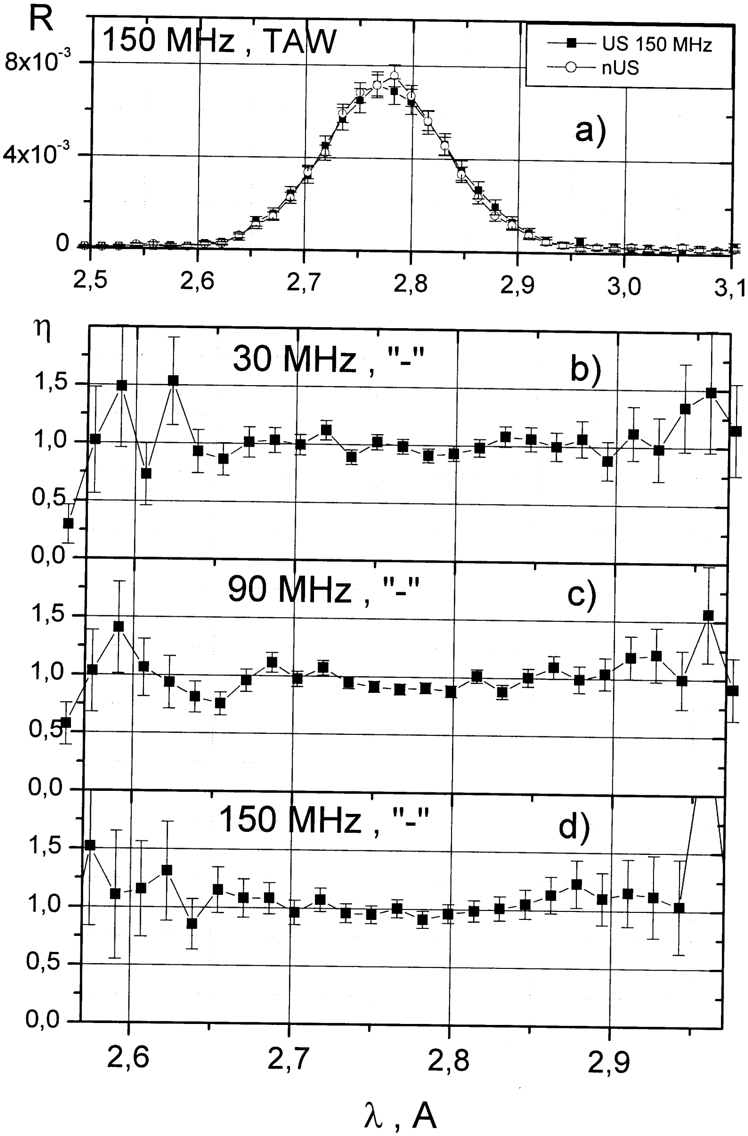

Fig. 3. a) The wavelength dependence of the reflection coefficient from the layered structure Fe/Cr without acoustic excitation and with 150MHz acoustic excitation. b),c),d)-The wavelength dependence of the ratio between the reflection coefficient with and without acoustic excitation.

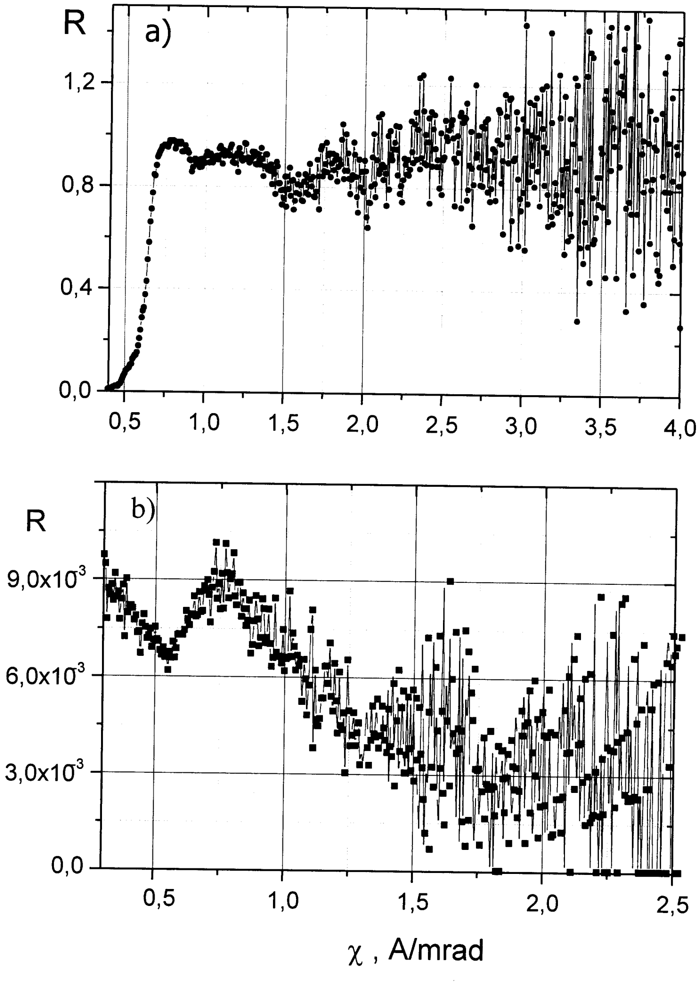

Fig. 4. The wavelength dependence of the neutron reflection coefficient: a) for the structure glass/Cu(100nm)/Ti(150nm)/Cu(30nm); b) for the structure glass/Cu(100nm)/Ti(150nm)/Cu(30nm) with a Gd_2O_3 absorbing layer 1cm long on the surface of the layer Cu(30nm) in its center.

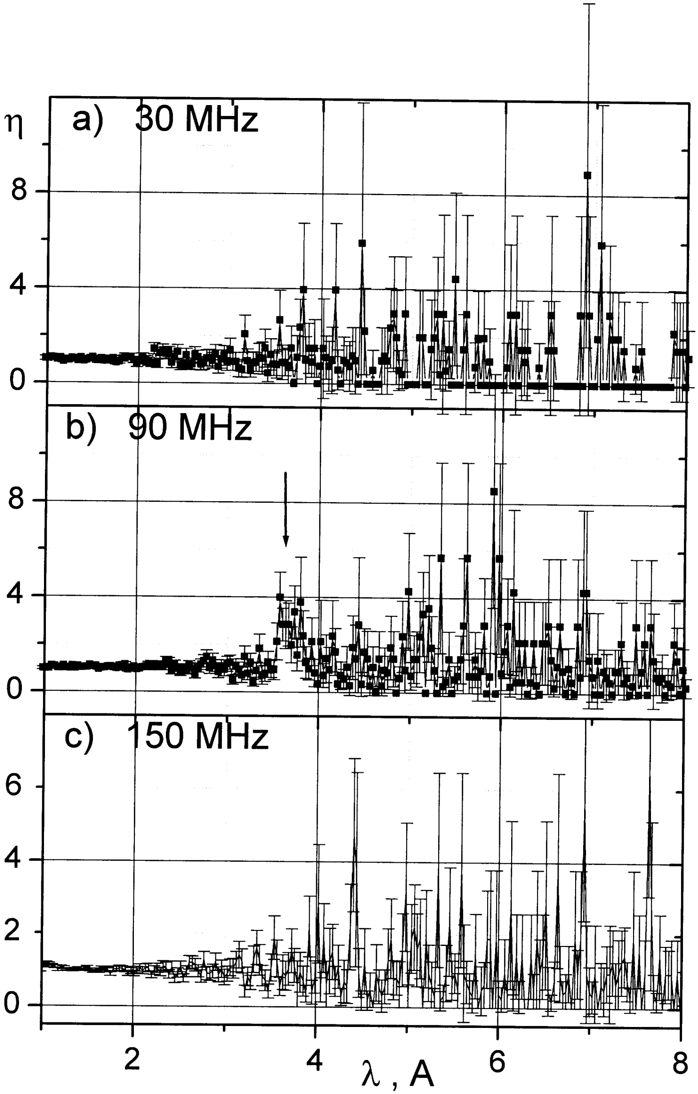

Fig. 5. The wavelength dependence of the ratio η of reflection coefficients from the ultrasonically excited and nonexcited structure for the reflection angle interval $\theta_1 \div \theta_2 = 0.14 \div 1.23$мрад: a) f=30 MHz; b) f=90 MHz; c) f=150 MHz.

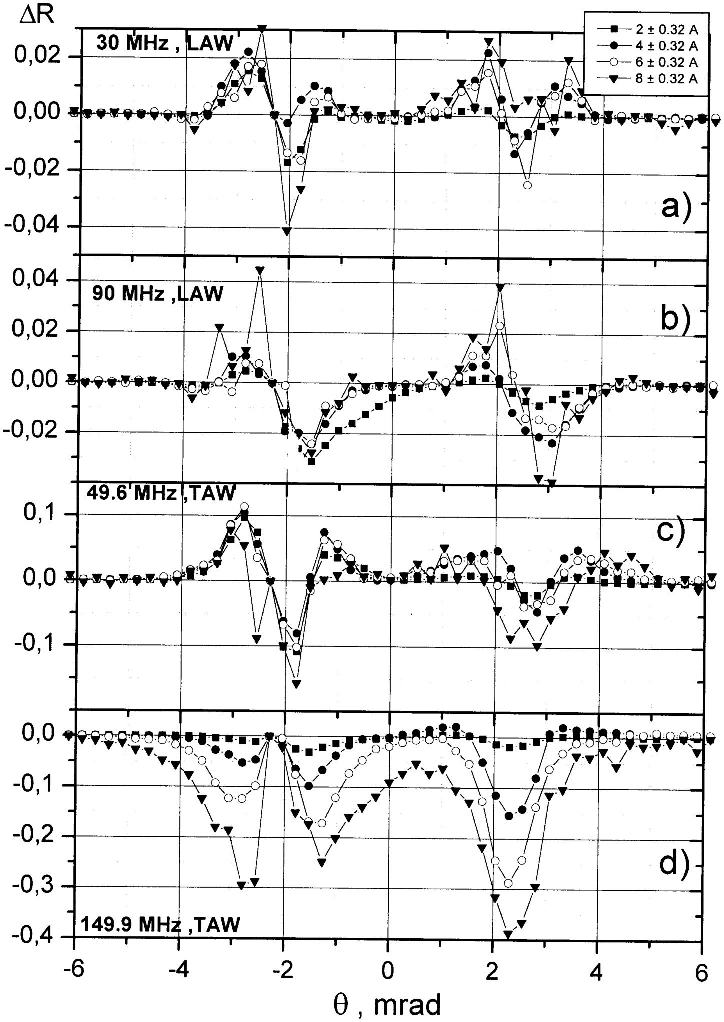

Fig. 6. The angular dependence of changes in the reflection coefficient ΔR(θ) from the acoustically excited glass for the wavelengths 2, 4 , 6 and 8Å: a) longitudinal sound, f = 30MHz; b) longitudinal sound, f = 90MHz; c) transverse sound, f = 49.6MHz; d) transverse sound, f= 149.9 MHz.

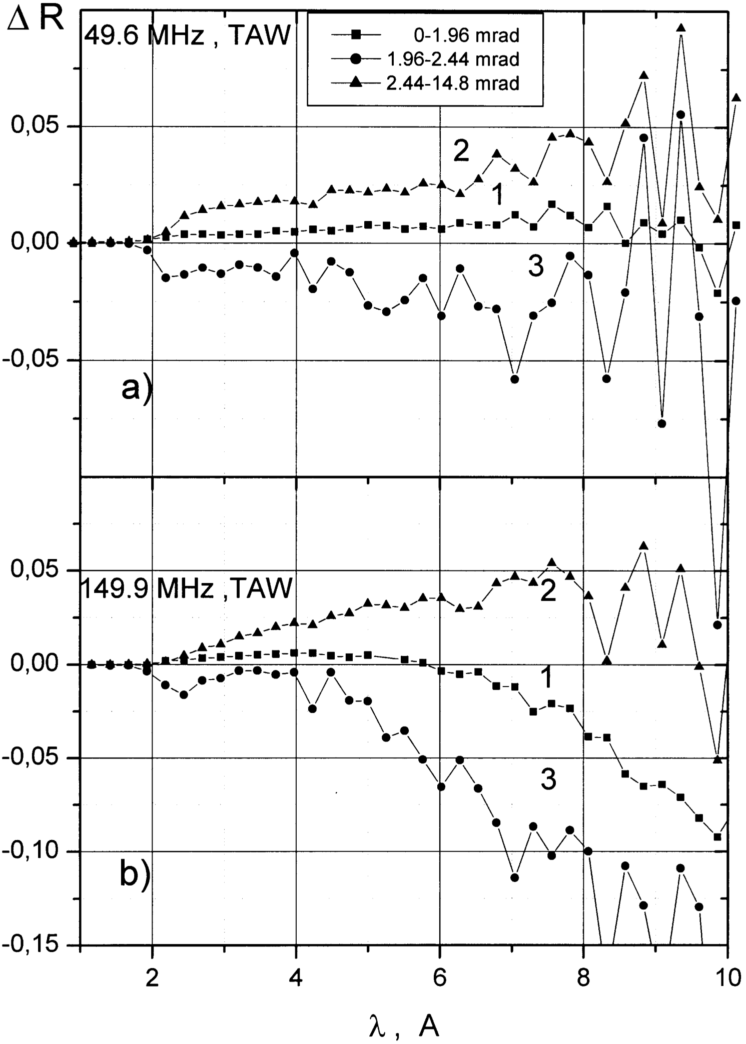

Fig. 7. The wavelength dependence of the difference ΔR(λ) between reflection coefficients from transverse sound-excited glass over different glancing angle intervals: a) f = 49.6 MHz; b) f = 149.9 MHz.

From Eq. (8) it is not clear how to obtain the experimental value of $\delta\Delta\theta$ (λ_1=4Å, λ_2=6Å) = $\Delta\theta$(6Å)- $\Delta\theta$(4Å) = 1.5 mrad. It seems likely that the basic effect of scattering is due to surface curvature. In support of the statement speaks the fact that the scattering angle does not actually depend on either the manner of excitation or, which is most important, neutron wavelength. This statement is also not in contradiction with the fact that reflection increases starting from the critical wavelength.

Conclusion

It is experimentally shown that in the glancing geometry of neutron reflection from various ultrasonic wave-excited layered structures, there is observed the scattering on neutrons accompanied with momentum transfer in the direction perpendicular to the interface boundaries in the structure. The momentum transfer is within the limits $2.1\times10^{-4} \div 2\times10^{-3}$Å^{-1} and the probability of scattering varies from 6 to 73% for different structures. The aim of further investigations is a comparison of the experimental data and the results of model calculations.

References

1. W.J. Spencer and G.T. Pearman, Adv.X-ray Anal. **13** (1970) 507.
2. P. Mikula, P. Lukas and J. Kulda, Acta Cryst. **A48** (1992) 72.
3. R. Kohler, W. Mohling and H. Peibst, Phys. Status Solidi (b) **61** (1974) 439.
4. E.M. Iolin, E.V. Zolotoiabko, E.A. Raitman, B.V. Kuvaldin, ZhETF **91** (1986) 2132.
5. E.M. Iolin, E.A. Raitman, V.N. Gavrilov, B.V. Kuvaldin, ZhETF **94** (1988) 218.
6. W.A. Hamilton, A.G. Klein, P.A. Timmins et al., Phys. Rev. Lett. **58** (1987) 2770.
7. E. Iolin, B. Farago, E. Raitman, F. Mezei et al., Physica B **241-243** (1998) 1213.
8. A.S. Gerasimov, V.K. Ignatovich, M.V. Kazarnovskii, **P4 – 6940**, Dubna, 1973.
9. V.K. Ignatovich, **P4 – 8687**, Dubna, 1975.
10. V.L. Aksenov, Yu.V. Nikitenko, Physica B **297** (2001) 101.

Received by Publishing Department
on October 22, 2001.

Аксенов В. Л. и др. E3-2001-224

Отражение нейтронов от слоистой структуры, возбужденной ультразвуковой волной

Исследованы режимы полного и брэгговского отражения поляризованных нейтронов от слоистой структуры в зависимости от амплитуды и частоты возбуждаемых в структуре продольных и поперечных ультразвуковых волн. Обнаружены вызванные звуковой волной незеркальное отражение нейтронов и сдвиг «пучностей» и «узлов» нейтронного волнового поля.

Работа выполнена в Лаборатории нейтронной физики им. И. М. Франка ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2001

Aksenov V. L. et al. E3-2001-224

Neutron Reflection from an Ultrasonically Excited Layered Structures

The regimes of total and Bragg reflection of polarized neutrons from a layered structure are investigated as a function of the amplitude and frequency of transverse and longitudinal ultrasonic waves excited in the structure. The off-specular reflection of neutrons and the shifting of the nodes and antinodes of the neutron wave field are observed.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2001

Макет Т. Е. Попеко

Подписано в печать 19.12.2001
Формат 60 × 90/16. Офсетная печать. Уч.-изд. л. 2,81
Тираж 300. Заказ 53019. Цена 3 р. 40 к.

Издательский отдел Объединенного института ядерных исследований
Дубна Московской области