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Depth of Formation of Specular Reflection of X-Rays and Neutrons and its Relationship with the Group Delay Time

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Abstract. The problem of the connection of the group delay time (GDT) with the depth in the medium at which the formation of specular reflection of X-rays and neutrons from a semi-infinite homogeneous medium with an ideally sharp interface in the regions up to the threshold of the total external reflection (TER) and above this threshold is discussed. The article has a discussion character and is based on materials that are not included in the review [1].

Keywords: neutrons, X-rays, total external reflection, group delay time

1. Introduction

Using the example of reflection of a neutron pulse (wave packet) from two- and three-layer planar structures in a review [1] it was shown that due to some time spent on penetrating radiation deep into the medium and back, the reflection of the pulse occurs with some delay in time with respect to the incident pulse. To a first approximation, this delay is determined by the so-called group delay time (GDT), equal to the derivative in energy from the phase of the amplitude reflection coefficient. The concept of group delay time was introduced into scientific use by Eisenbud, Bohm, and Wigner [2-4] as a measure of the interaction time in quantum mechanics.

In this paper, a seemingly simpler case (in comparison with [1]) is considered, namely, specular reflection of $X-$rays and neutrons from a semi-infinite homogeneous medium. As is known, the amplitude of specular reflection and the distribution of the field (or wave function) in a medium in the entire range of angles and incidence energies is described by exact Fresnel formulas. Fresnel formulas give exact expressions for the reflection amplitude, field structure, and depth of radiation penetration into the medium, both below the threshold of total external reflection (TER) and above this threshold, but they do not give any answer about the thickness of the near-surface layer in which the reflected wave is formed.

The article discusses the problem of the connection between the GDT and the depth at which the formation of specular reflection of $X-$rays and neutrons from a semi-infinite homogeneous medium with an ideally sharp interface in regions up to the total external reflection threshold and above this threshold occurs. Calculations show that in the region below the threshold of the TER this formation depth is exactly equal to the penetration depth of the exponentially decaying evanescent wave. However, in the region above the threshold of TER, where the penetration depth of radiation increases significantly, the GDT and the depth of reflection formation, on the contrary, are greatly reduced. Moreover, the GDT increases with increasing absorption in the medium, while the penetration depth decreases.

It will be shown below that various attempts to determine the relationship between the depth of reflection formation and the GDT lead to some contradictions both in the physics of the phenomenon and in the specific numerical values of this depth, especially in the region above the threshold of the total external reflection. Moreover, a number of questions have not yet received an exhaustive explanation. Our proposed method for estimating the depth of reflection formation based on the first Born approximation in the scattering theory gives highly overestimated results in comparison with direct calculations of the GDT and the reflection time of wave packets. In conclusion, the possible causes of such a sharp discrepancy are briefly discussed.

2. Calculation method

Along with the Goos–Hänchen effect [5, 6], which consists in the longitudinal displacement of the reflected wave beam during its oblique incidence on the surface of the medium (see also [7]), there is a phenomenon of time delay of the reflected pulse with amplitude $A_R(t)$ relative to the incident pulse $\Psi_{in}(t) = A_{in}(t)\exp(-i\omega_0 t)$ with slowly varying complex-valued amplitude $A_{in}(t)$ (see, for example, the review [1] and references therein). In the framework of the spectral approach, it is easy to show that the slowly varying amplitude of the reflected pulse $A_R(t)$ is described by the integral relation

$$A_R(t) = \int_{-\infty}^{\infty} R(\omega) A_{in}(\Omega) e^{-i\Omega t} d\Omega \,, \tag{1}$$

where

$$A_{in}(\Omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} A_{in}(t) e^{i\Omega t} dt \tag{2}$$

is the spectral amplitude of the incident pulse, $R(\omega) = |R(\omega)|\exp(i\varphi)$ is the amplitude reflection coefficient of a plane monochromatic wave with a frequency ω from an arbitrary structure uniform in a plane perpendicular to its surface, $\varphi(\omega)$ is the reflection phase, $\Omega = \omega - \omega_0$ is the frequency offset, ω_0 is the central radiation frequency of the incident pulse with the spectrum $A_{in}(\Omega)$.

Consider the case when the modulus of the reflection coefficient $|R(\omega)|$ varies rather weakly within the spectrum $A_{in}(\Omega)$. We represent the reflection phase as decomposition $\varphi(\omega_0+\Omega)\approx\varphi(\omega_0)+(d\varphi/d\omega)\times\Omega$ and substitute this expression into the integral (1). As a result, up to insignificant phase factors, we obtain that the reflected pulse coincides in shape with the incident pulse, but it time shifted by τ:

$$A_R(t)\approx|R(\omega_0)|A_{in}(t-\tau), \tag{3}$$

where the value τ is called the group delay time and is determined by the ratio

$$\tau=\frac{d\varphi}{d\omega}=\hbar\frac{d\varphi}{dE}. \tag{4}$$

If the derivative of the phase in relation (4) is positive, then the pulse is reflected with some positive time delay $\tau>0$, which is quite natural, since the pulse spends some time on the forward and backward propagation in the medium.

If the derivative (4) is negative, then we arrive at a physically impossible result $\tau<0$, namely, the pulse is reflected (or begins to be reflected) even before the incident pulse falls on the surface of the medium. Obviously, such a paradox arose due to the oversimplification of the general formula (1) as a result of the expansion of the reflection phase $\varphi(\omega)$ in a series accurate to the first term in Ω. In reality, the spectral dependences of the functions $R(\omega)$ and $A_{in}(\Omega)$ under the sign of the integral in (1) must be taken into account precisely. For this reason, the GDT τ (4) is only an approximate evaluation of the time shift of the reflected pulse.

In a number of examples, it was shown in [1] that, when neutron pulses are reflected from layered structures of finite thickness, the GDT τ can be either positive or negative, which, however, does not contrary to the principle of causality. It turns out that in the case $\tau<0$ the shape of the reflected pulse is distorted in such a way that the position of its maximum is actually slightly ahead of the maximum of the incident pulse, but the condition $A_R(t)<A_{in}(t)$ is fulfilled for all times $t<0$.

In this paper, we consider the Fresnel (mirror) reflection of a wave from a homogeneous amorphous semi-infinite medium. The wave functions (or fields) of the incident, reflected, and transmitted radiation have the following form: $\Psi_{in}=A_{in}\exp(i\mathbf{kr})$, $\Psi_R=A_R\exp(i\mathbf{k}_R\mathbf{r})$ and $\Psi_T=A_T\exp(i\mathbf{qr})$, respectively. In this case, for the complex amplitude reflection coefficient $R(\omega)=A_R/A_{in}$ there is the following well-known exact solution:

$$R(\omega)=\frac{k_z-q_z}{k_z+q_z}. \tag{5}$$

where k_z and q_z are the values of the *z*-projections of the wave vectors of the incident plane monochromatic wave in vacuum and in the medium, respectively (the *z* axis is directed deep into the medium perpendicular to the interface).

For X-ray radiation, the *z*-projection $k_z = k\sin\theta$, where $k = \omega / c = 2\pi / \lambda$ is the magnitude of the wave vector in vacuum, ω is the frequency, λ is the wavelength, θ is the gliding angle with respect to the surface, $q_z = q'_z + iq''_z = k(\sin^2\theta + \chi)^{1/2}$ is the *z*-projection of the wave vector in the medium in the region $z > 0$, $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$ is the complex polarizability of the medium, and the real value $\chi'(\omega) < 0$ is negative.

For neutrons, by virtue of the potential dispersion law $q^2 = k^2 - k_b^2$, one can restrict oneself to the case of normal incidence. Here $q = q' + iq''$, $k(\omega) = MV / \hbar$ is the wave number of neutrons in vacuum, M is the neutron mass, V is the velocity of incident neutrons with energy $E = \hbar\omega = MV^2 / 2$, k_b is the boundary (critical) value of the wave number of neutrons in the medium, which is determined from the relation $k_b^2 = 4\pi Nb = 2MU / \hbar^2$, where N is the bulk density of nuclei, $b = b' - ib''$ is the complex length of coherent scattering, $U = U' + iU''$ is the effective complex potential of the medium. The neutron reflection coefficient can also be represented in the following form, equivalent to expression (5):

$$R(E) = \frac{\sqrt{E} - \sqrt{E - U}}{\sqrt{E} + \sqrt{E - U}} = |R| e^{i\varphi}. \tag{6}$$

The shape of the specular reflection curve $|R(E)|^2$, phase $\varphi(E)$, and GDT $\tau(E)$ substantially depends on the energy of the incident neutron. In accordance with this, we will distinguish two cases: 1) sub-barrier reflection, when $k \le k'_b$ ($E \le U'$), and 2) over-barrier reflection, for which $k > k'_b$ ($E > U'$).

The reflection phase is determined from the general relation $\mathrm{tg}\varphi = R'' / R'$. In the case of $E \le U'$ and $U' << U'$ the phase $\varphi = -\mathrm{arctg}(x)$, where $x = 2\sqrt{E(U' - E)} / (2E - U')$. To calculate the GDT τ (4), we use the following simple relations: $\tau = \hbar(d\varphi / dE) = \hbar(d\varphi / dx)(dx / dE)$ and $d\varphi / dx = -d[\mathrm{arctg}(x)] / dx = -1/(1 + x^2)$. Taking into account these relations, after a series of mathematical transformations, we obtain the following final expression for the GDT (see also relation (12) in [1]):

$$\tau = \frac{\hbar}{\sqrt{E(U' - E)}}. \tag{7}$$

From formula (7) it follows that the GDT is maximum at energies $E \to 0$ and $E \to U'$. In a real situation, of course, it is necessary to take into account the absorption, i.e. imaginary part of the potential U'', which leads to the disappearance of the divergence, which is done on a computer by numerically differentiating the reflection phase $\varphi(E)$.

3. Results and Discussion

The Fig. 1 shows the dependencies of the reflection phase, the reflection coefficient modulus, and its real and imaginary parts on the neutron energy. It should be noted that on the scale adopted here it may seem that the phase of reflection is zero at energy $E > U'$ above the threshold of total external reflection (TER). In reality, this phase is simply very small, since in this region the phase is determined by the imaginary part of the potential $U'' << U'$ (see also formula (9) below).

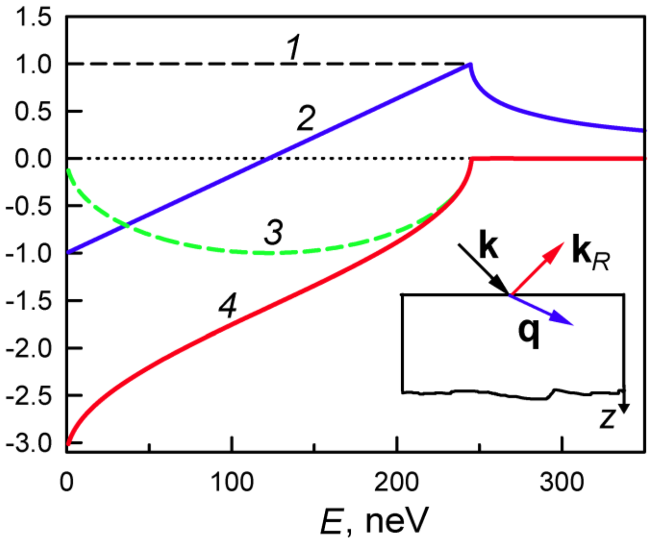

Fig. 1. The module of the amplitude reflection coefficient $|R|$ (curve *1*), the real R' (curve *2*) and imaginary R'' (curve *3*) parts, as well as the reflection phase φ (curve *4*) depending on the energy of neutrons incident on a semi-infinite nickel medium. The inset shows the location and orientation of the wave vectors of the incident, reflected and transmitted waves.

The depth of radiation penetration into the medium is determined by the ratio $L_z = 1/\mathrm{Im}(q_z)$. In the region below of the threshold of TER for neutrons, i.e. at energy $E < U'$, the value L_z is equal to the attenuation depth of the evanescent wave: $L_z = 1/(k_b'^2 - k^2)^{1/2}$. From the most general intuitive considerations, we can assume that the delay time is equal to $\tau_z = 2L_z / V$. Here, the lower index z indicates that this delay time is obtained from the assumption of the double passage of neutrons (deep into the medium and up to the interface) of the near-surface layer with an effective thickness equal to the penetration depth L_z.

The most surprising thing is that this estimated time τ_z exactly coincides with the GDT obtained from the exact relation $\tau = d\varphi / d\omega$ (1) (see curves *2* and *3* in Fig. 2). Indeed, taking into account the relations $E = \hbar^2 k^2 / 2M$, $U' = \hbar^2 k_b'^2 / 2M$, and also $\hbar k = MV$ and $L_z = 1/\mathrm{Im}(q_z)$ from formula (7), it is easy to obtain that

$$\tau = \frac{2M}{\hbar k\sqrt{k_b'^2 - k^2}} = \frac{2}{V\,\mathrm{Im}(q_z)} = \frac{2L_z}{V} \equiv \tau_z \,. \tag{8}$$

From this we can conclude that the depth of reflection formation L_R coincides with the depth of penetration into the medium, i.e. $L_R = L_z$. The reasons for this coincidence are completely incomprehensible, since there is no backward wave $\sim \exp(-iq_z z)$ in a semi-infinite medium (see the inset in Fig. 1), and the assumption that the neutron velocity is equal V in the medium and in the vacuum, and even in the field of total external reflection, does not correspond to reality. In the field of TER, the *z*-projection of the wave vector is purely imaginary and it is not entirely correct to talk about the neutron velocity.

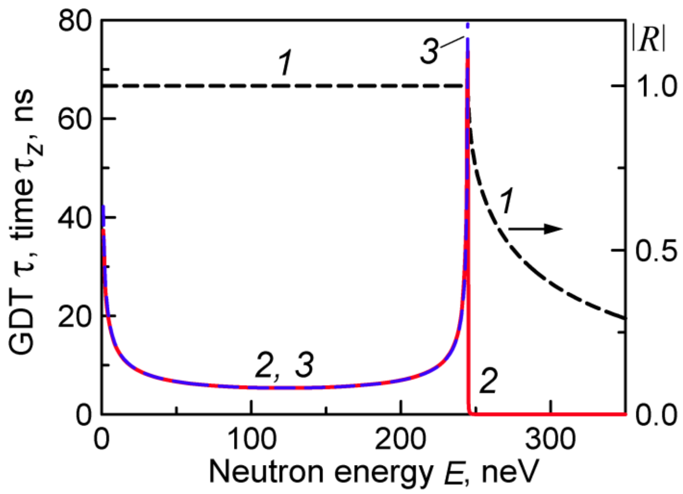

Fig. 2. The dependence of the reflection coefficient modulus $|R|$ (curve *1*, right scale), GDT τ (curve *2*) and the estimated time $\tau_z = 2L_z / V$ (curve *3*) on the neutron energy E, when they are reflected from a semi-infinite nickel medium ($U'_{\mathrm{Ni}} = 245\,\mathrm{neV}$). Curve *3* is shown in the energy region up to 245.1 neV, since then it increases very sharply.

Nevertheless, rigorous calculations show that the delay time τ_z of the reflected pulse $A_R(t)$ is in very good agreement with the GDT calculated by the formula $\tau = d\varphi / d\omega$ (see curves *2* and *3* in Fig. 2 in the region $E < U'$). At this scale in Fig. 2, it may seem that in the region above the threshold of total external reflection the GDT $\tau = 0$, but this is not so, although indeed the group delay time is very short (see below for more details).

Similar results are also valid for X-ray radiation, for which in the region of total external reflection ($\theta < \theta_c = |\chi'|^{1/2}$) the group delay time is determined by the ratio

$\tau = 2\theta / [\omega(\theta_c^2 - \theta^2)^{1/2}]$, and the depth of mirror reflection formation is equal to $L_z = \tau c / (2\sin\theta)$.

In the region above the total external reflection threshold, the penetration depth L_z increases significantly, while the GDT and, consequently, the depth of reflection formation $L_R = \tau n V / 2$, on the contrary, decrease significantly, where $n = [1 - (k_b / k)^2]^{1/2}$ is the refractive index (see Tables 1 and 2). Here, the lower index R means that the depth of formation L_R in the region above the TER threshold is determined according to the GDT τ, i.e. derivative (4) of the reflection coefficient phase R. Moreover, with a decrease in absorption, i.e. with a decrease in the imaginary parts b'' for neutrons and χ'' for X-rays, the depth L_z of radiation penetration into the medium increases, while the GDT decreases:

$$\tau = \frac{M k_b'^2}{\hbar k (k^2 - k_b'^2)^{3/2}} \frac{b''}{b'}, \quad (E > U') \tag{9}$$

$$\tau = 2 \frac{\chi''(\omega_0)}{\chi'(\omega_0)} \frac{\omega_0^2}{\omega^3}, \quad (\theta > \theta_c). \tag{10}$$

It can be seen from Tables 1 and 2 that in the region above the TER threshold, group delay times τ are much shorter than the period T of incident waves, and the corresponding depths of mirror reflection formation are generally comparable or even much smaller than atomic sizes. In this regard, the interpretation of the results obtained above within the framework of existing concepts based on the Fresnel formula, encounters certain difficulties.

Table 1. Group delay time τ and the depth of formation of neutron reflection L_R from nickel depending on the energy E of incident neutrons ($U' = 245$ neV, $U'' = 0.024$ neV, period $T \approx 8$ ns).

Energy E, neV	**200**	**240**	**244**	**250**	**270**
GDT τ, ns	6.9	18.9	41.8	0.04	0.004
Depth L_R, Å	215	642	1430	0.22	0.04

Table 2. Group delay time τ and depth L_R of formation of reflection of X-ray CuK_α-radiation ($\lambda = 1.54$ Å) from silicon depending on the angle of incidence θ (critical angle of TER $\theta_c = 13.4$ arc.min, wave period $T = 0.5$ as).

Angle θ, arc.min	**10**	**13**	**14**	**20**	**30**
GDT τ, as	0.18	0.64	0.08	0.007	0.005
Depth L_R, Å	95	264	28	1.8	0.8

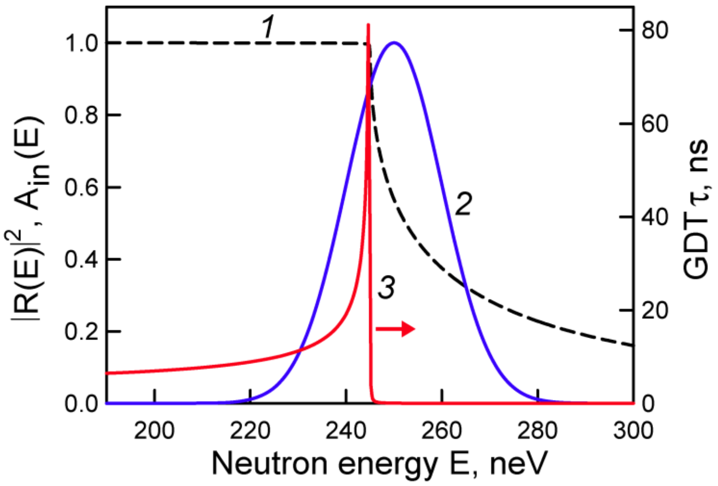

Fig. 3. *1* - reflection coefficient $|R(E)|^2$, *2* - spectrum of the incident neutron pulse $A_{in}(E)$ with central energy $E_{in} = 250$ neV and spectral width $\Delta E = 10$ neV (pulse duration $\tau_{in} = 66$ ns), *3* - dependence of the group delay time $\tau = \hbar(d\varphi / dE)$ with a narrow sharp peak at energy $E = U'$ on energy (right scale), $U'_{\mathrm{Ni}} = 245$ neV

It is interesting to compare the real time shift Δt of the reflected pulse, calculated according to the exact formula (1), with the value of the GDT τ (4). It must be borne in mind here that the calculation results depend not only on the central neutron energy E_{in}, which can be both above and below the threshold of total external reflection, but also on the duration of the incident pulse τ_{in}. This is due to the fact that the pulse has a finite spectral width $\Delta E = \hbar / \tau_{in}$, therefore, the "tail" of the spectrum $A_{in}(E)$ can penetrate into the total reflection region, in which the group delay time τ is relatively large, even if the energy of the incident pulse $E_{in} > U'$ is higher than the threshold (see Fig. 3 and Fig. 4).

For comparison, we note that in the region up to the threshold of total external reflection, if $E_{in} = 220$ neV and $\Delta E = 10$ neV, then the real time shift $\Delta t = 9.5$ ns, that is only a little more than the GDT $\tau = 8.9$ ns. However, if $E_{in} = 240$ neV and $\Delta E = 10$ neV, then the real time shift $\Delta t = 13.8$ ns, which is less than the GDT $\tau = 18.9$ ns, is precisely because the "tail" of the spectrum penetrate into the region above the TER threshold. With increasing pulse duration, i.e. with a decrease in the width of its spectrum by 10 times, the values Δt and τ are practically compared ($\Delta t = 32.9$ ns and $\tau = 30.4$ ns at energy $E_{in} = 243$ neV, spectral width $\Delta E = 1$ neV, and the pulse duration $\tau_{in} = 660$ ns). In the case of a relatively short pulse, even at energy $E_{in} = 270$ neV $> U'_{\mathrm{Ni}}$=245 neV and $\Delta E = 10$ neV, a real time delay is equal to $\Delta t = 0.38$ ns, which is almost two orders of magnitude larger than the GDT $\tau = 0.004$ ns, despite the very small amplitude of the spectrum $A_{in}(E)$ in the region below the TER threshold.

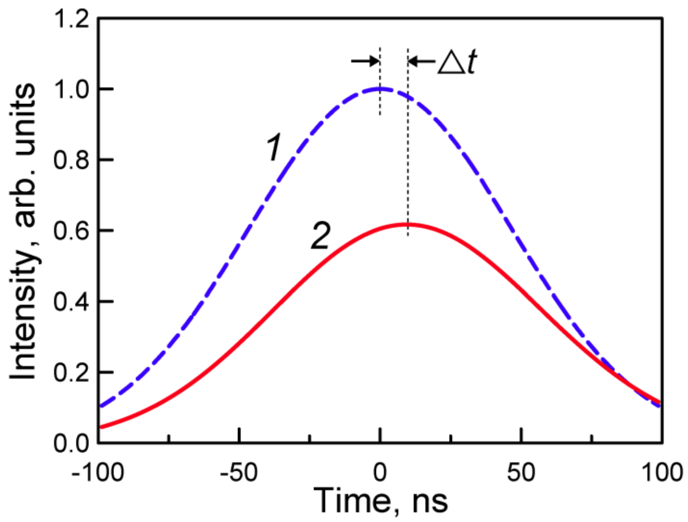

Fig. 4. The intensity of the incident (curve *1*) and reflected (curve *2*) pulses. The duration of the incident pulse $\tau_{in} = 66$ ns, the group delay time at the energy $E_{in} = 250$ neV is equal to $\tau = 0.044$ ns, the real time shift $\Delta t = 9.5$ ns, which is much more than the GDT τ (4)

It is clear that to exclude the influence of the "tails" of the spectrum $A_{in}(E)$, especially in the region below the TER threshold in the case when $E_{in} > U'$, it is necessary to use sufficiently long pulses. However, this raises the problem of determining extremely small time delays against the background of such long pulses.

4. Conclusions

Calculations of the group delay time τ and the depth of reflection formation $L_R \approx 1/(2k_z)$ within the framework of the first Born approximation of the scattering theory in the kinematic region above the TER threshold, where $q_z \approx k_z$, give highly overestimated results compared to the depth L_R obtained both from the GDT (4), (8)-(10), and with the calculation of the reflection time based on rigorous calculations of the amplitudes of the reflected pulses $A_R(t)$ (1).

It is possible that the role of a backward wave in a medium is played by a nonradiative wave of delayed excitation in a dispersive medium characterized by a spectral refractive index $n(\omega)$. On the other hand, it is not entirely clear whether the macroscopic "bulk" and "structureless" concepts of polarizability $\chi(\omega)$, scattering length b, and effective potential U can be transferred to microscopic regions with thicknesses much smaller than the wavelength and atomic sizes. It is possible that our a priori idea of the relationship between the reflection time, the depth of formation of the reflected pulse and the velocity of wave propagation in matter may also be incorrect.

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Conflict of Interest

The authors have no conflict of interest.

Author Contributions

Both authors equally participated in the statement of the problem, in the conduct of mathematical and computer calculations, in discussions of the results obtained and in writing the text.

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