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Standing waves and reflectivity from ultrathin layer

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We have analytically found that the reflectivity amplitude from one thin layer in the presence of the other reflecting layers is modulated by the squared amplitude of the radiation field but not by the first order amplitude as it was supposed in the paper of R. Röhlsberger (Hyperfine Interactions **123/124**, 455 (1999)) for the delayed nuclear resonance reflectivity. Hence, a larger sensitivity to the standing waves is expected for the reflectivity signal from selected groups of atoms or nuclei compared with the secondary radiation emission.

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Standing waves are created when incident wave is coherently reflected from crystals or multilayers (diffraction, total external reflection for X-ray and neutrons). Since long time this effect have been effectively used for investigation of the depth position of selected group of atoms by means of secondary radiation (fluorescent signal, photoelectrons) [1,2].

Nowadays it has become possible to select the contribution from the selected group of atoms *in reflectivity signal itself* – by investigation of the spectrum of reflectivity or by a selection of the scattering channel (e.g. nuclear resonant scattering is selected from the electronic scattering by time gating [3]). Since the first experiment, where the peak near the critical angle of the total external reflection in delayed nuclear resonant reflectivity was observed, the idea of the influence of the standing waves on the reflectivity signal itself has been discussed [4 – 7]. Investigations of the shape of the time spectra of Bragg reflectivity and its variation with angle [8, 9] also led the authors to the conclusion that the energy or time dependence of standing waves inside a periodical resonant multilayer influences the reflectivity energy or time spectrum [10]. Several experiments with resonant ^{57}Fe layers embedded at different depths in thin magnetic films have been performed in which the enhancement of the reflectivity signal due to the standing wave creation near the critical angle was used [11, 12]. The influence of standing waves on the reflectivity signal is even more essential in the new type of measurements recently performed by R.Röhlsberger et at. [13] where ^{57}Fe layers are placed inside a wave guide acting structure. In all such measurements the reflectivity signal was investigated but not the secondary radiation. All authors in [4 – 13] imply that a connection between standing waves and delayed nuclear resonant reflectivity exists, but no analytical expression were derived with the exception of [7], where such expression was suggested, but the wrong one (as we show here).

Here we have got analytical expression which describes the "standing wave influence" on the reflectivity signal from ultrathin layer in the presence of the other reflecting layers. It is essential that the reflectivity can not be considered as the secondary radiation emission, because at reflectivity measurements the detected radiation is coherent with the incident radiation and it obeys the laws of propagation in layered medium.

The most understandable result can be obtained from the kinematical approximation for reflectivity [14] when we simply add the reflectivity amplitudes from individual layers with an appropriate phase factor:

$$R = \sum_n r_n e^{2i\varphi_n}. \tag{1}$$

The reflectivity from a thin layer r_n is described by the well-known expression [15]

$$r = i\frac{\lambda}{\sin\vartheta}\rho f, \tag{2}$$

where λ is the wavelength of radiation, ϑ is the glancing angle, ρ is the surface density of scattering centers and f is the forward scattering amplitude by a single scattering center.

At a first sight the contribution from each layer to the reflectivity in (1) is independent from the others. But let us consider the contribution to the total reflectivity R^{tot} from one additional layer on top of a multilayer or other substrate with the reflectivity R. The result will be the following:

$$R^{\mathrm{tot}} = R\exp\left(iQd + i\frac{2\lambda\rho}{\sin\vartheta}f\right) + r = R' + r(1 + 2R'), \tag{3}$$

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where $Q = (4\pi/\lambda)\sin\vartheta$, d is the thickness of the layer. In (3) we also took into account the phase and refraction corrections of the total reflectivity caused by the added layer through the phase factor $2i\varphi$ (the used approximation is valid for the angular region beyond the critical angle ϑ_c of the total external reflection):

$$
\begin{gathered}
2\varphi = \frac{4\pi}{\lambda}\eta d, \\
\eta = \sqrt{\sin^2\vartheta + \frac{\lambda^2}{\pi}\rho_v f} \cong \sin\vartheta + \frac{\lambda^2}{2\pi\sin\vartheta}\rho_v f,
\end{gathered} \tag{4}
$$

and used the determination $\rho = \rho_v d$ (ρ_v is the volume density of the scattering centers). R' in (3) is the reflectivity R at the distance d from the top of the multilayer (relative the incident wave at the same height):

$$
R' = Re^{iQd}. \tag{5}
$$

For very thin layers we can neglect this phase factor. However (5) can be generalized for the case when the added layer is placed at some distance H from the surface. In this case d in (5) is replaced by H. It is this phase factor which determines the angular or depth variations of the standing waves. The above consideration can easily be repeated for the thin layer placed inside the multilayer.

For illustration we consider a resonant ^{57}Fe layer on the top of nonresonant superstructure [Si/W]·20. The calculated spectrum of Mössbauer reflectivity $R^{\text{tot}}(\omega)$ is determined by $r(\omega)$ and by the position H of the resonant layer relative the surface of the superstructure (through the phase shift between the amplitudes of scattering R' and $r(\omega)$). The standing wave influence appears through the complex factor $(1 + 2R')$ modulating $r(\omega)$ – Fig.1. This factor enhances the contrast of the spectrum when ^{57}Fe layer is placed in the antinode of standing wave (Fig.1a) and suppresses it in the node (Fig.1b).

So in the kinematical approximation (when the reflectivity R is small enough) the change of the single layer scattering amplitude caused by the other layers in multilayer is described by the factor $(1 + 2R')$. This result already shows up the difference of the reflectivity and the secondary radiation emission, because the amplitude of the last one is proportional to the amplitude of the radiation field $(1 + R')$.

For angles close to the critical angle the kinematical approximation in the reflectivity theory is not valid. We should use here the exact Parratt recursive formula for reflectivity amplitude:

$$
R_j = \frac{r_j + R_{j+1}e^{2i\varphi_j}}{1 + r_j R_{j+1}e^{2i\varphi_j}}. \tag{6}
$$

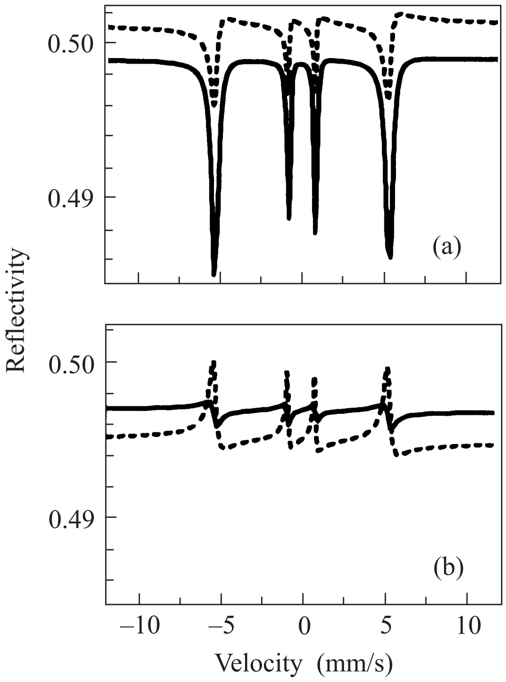

Fig.1. Calculated Mössbauer spectra of reflectivity at the exact Bragg angle 14.75 mrad from a thin ^{57}Fe layer placed at the distance $H = D/2$ (a) and $H = 0$ (b) from the surface of a multilayer with period $D = 3.0$ nm (solid lines). Dash lines correspond to the pure interference of $r(\omega)$ and R without the standing wave factor $(1 + R)^2$

Here r_j is the Fresnel reflectivity amplitude, R_{j+1} the multiple reflectivity amplitude at the previous boundary, the expression for the phase shift φ_j is given by (4), but the approximate formula for the square root can not be used now.

For calculation of the reflectivity from a thin layer by (6) we suppose that $2i\varphi$ is small. In order to simplify the calculations we insert an artificial vacuum buffer layer, with zero thickness, between the tested layer and substrate, although the algebra can be carried out also without this layer. Neglecting $(2i\varphi)^2$ term we obtain

$$
R^{\text{tot}} = \frac{r_{01} + \widetilde{R}}{1 + r_{01}\widetilde{R}} + \frac{\widetilde{R}(1 - r_{01}^2)}{(1 + r_{01}\widetilde{R})^2} 2i\varphi_1 \tag{7}
$$

and

$$
\widetilde{R} = \frac{r_{01} + R}{1 + r_{10}R} \tag{8}
$$

where r_{01} is the Fresnel reflectivity amplitude from the tested layer and R is the reflectivity amplitude from the substrate. And finally we get:

$$
R^{\text{tot}} = R(1 + iQd) + (1 + R)^2 r \tag{9}
$$

were r is again determined by (2).

The obtained result is surprisingly simple and clear. The layer reflectivity amplitude r in the presence of the other reflecting layers is modulated by the "squared standing wave" $(1 + R)^2$, because the standing wave structure of the radiation field reveals itself for the incident (absorption process) as well as for reflected wave (emission process) as well. The kinematical approach obtained above (3) was true because when R is small than $(1 + R)^2 \approx 1 + 2R$.

The obtained formula (9) differs from the analogous expression (4.1) presented in [7]:

$$R^{\rm tot} = R + (1 + R)(r - r_S) \qquad (10)$$

(we rewrite it here in our notations, r_S is determined by (2) but with the substrate parameters). The expression (10) from [7] implies that the reflected from a thin layer amplitude is proportional to the amplitude of the total radiation field $(1 + R)$ (the unessential term r_S can be effectively combined with R) just as in the case of the secondary radiation emission. The correct expression (9) contains this amplitude squared $(1 + R)^2$.

If we again suppose that the investigated thin layer is the resonant layer ($r = r(\omega)$), then the delayed nuclear resonance reflectivity is calculated by the Fourier transform of (9) which gives

$$|R^{\rm tot}(t)|^2 = |(1 + R)^2|^2 |r(t)|^2. \qquad (11)$$

In Fig.2 we plot the result of the calculations by formula (9) and (10) (formula (4.2) from [7]). We see that (10) does not give the right behavior of the delayed nuclear resonance reflectivity in any case.

Of course calculations can be done correctly by the main formula (6), but the importance of the presented expression (9) is that it clarifies the physical understanding of how the reflectivity from a thin layer is changed in the presence of the other reflecting substance. It also directly shows the role of the standing waves in the reflectivity process. The illustrations here were given for the case of nuclear resonant reflectivity experiments, but the result refers also to any kind of reflectivity measurements, e.g. magnetic X-ray scattering, neutron reflectivity and so on.

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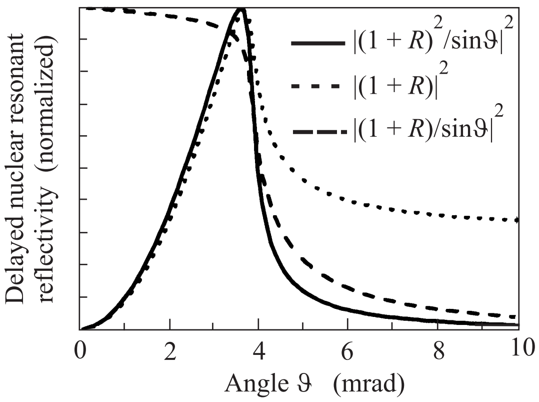

Fig.2. Calculated by means of Parratt formula (6) the integral nuclear resonant reflectivity from thin ^{57}Fe layer on the top of nonresonant ^{56}Fe substrate (solid line), which gives exactly the same result as (9). Notice that the factor $1/\sin\vartheta$ appears from (2). Dotted line is the dependence suggested by Röhlsberger [7] if we neglect the $1/\sin\vartheta$ factor in r (the well known dependence for secondary radiation emission [2]), dashed line – if we do not neglect this factor (no peak at the critical angle ~ 3.7 mrad appears)

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