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The influence of interface roughness on Mössbauer filtration of synchrotron radiation at an isotope interface

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The influence of interface irregularities on synchrotron radiation Mössbauer filtration at an isotope interface is examined theoretically. Calculations are performed for a random-roughness interface model for a ^{56}Fe/^{57}Fe isotope interface. The analysis shows that, despite a drop in the efficiency in the real structures compared with the idealized structures, the efficiency of Mössbauer filtration of synchrotron radiation remains high enough to ensure a successful experiment. In particular it was found that, as well as a regular saw-like profile of the external interface, a randomly rough profile of the external interface may also be used in the Mössbauer filtration experiment if the roughness parameters satisfy certain demands.

Keywords: Mössbauer filtration; isotope interfaces.

1. Introduction

Progress in the application of synchrotron radiation combined with the Mössbauer effect has produced a new type of spectroscopy with a unique energy resolution, limited only by the line energy width (of the order of 10^{-8} eV for the most popular Mössbauer isotope, ^{57}Fe). First experiments in this field have resulted, for example, in a direct measurement of phonon spectra (excitation spectra) in condensed matter, both in crystal (Seto *et al.*, 1995) and fluid phases (Zhang *et al.*, 1995), and also in the measurement of inelastic scattering spectra in gases (Chumakov, Smirnov *et al.*, 1996). These results demonstrate the great potential of the method. This paper is concerned with Mössbauer monochromatization (filtration) at an isotope interface. This has been considered previously in terms of an X-ray wavelength standard (Belyakov & Zhadenov, 1994, 1995). The influence of the imperfections (roughness) of material interfaces is examined. Such imperfections, which inevitably exist in a real experiment, can also be artificially created in samples to optimize the Mössbauer synchrotron radiation filtration.

Filtration at an isotope interface (Belyakov & Zhadenov, 1994, 1995) is one of several methods of Mössbauer synchrotron radiation filtration currently under development. Other methods use purely nuclear diffraction reflections (Chechin *et al.*, 1983; Gerdau *et al.*, 1985), grazing-incidence anti-reflection films (GIAR films) (Hannon *et al.*, 1979; Hannon, Hung *et al.*, 1985*a*,*b*; Hannon, Trammell *et al.*, 1985*a*,*b*) and artificially grown periodical structures with large period values and modulated isotopic composition (Chumakov *et al.* 1993; Toellner *et al.*, 1995). The general challenge to all approaches is the necessity of eliminating the background contribution due to non-resonant scattering of the synchrotron radiation beam in the sample. Therefore, as a rule, experiments are carried out with temporal filtration of the beams (Belyakov, 1992). Temporal filtration is based on the time-delayed technique, *i.e.* the filtered synchrotron radiation beam is registered with some time delay with respect to the synchrotron radiation impulse. This approach allows the separation of the slow processes of nuclear resonant scattering, which are characterized by Mössbauer-level lifetimes, from the fast processes of the initial synchrotron radiation beam scattering, which are unconnected with the nuclear scattering. The improvement of the methods of synchrotron radiation pre-monochromatization (decreasing the linewidth to as low as ~1–2 meV) (Chumakov, Metge *et al.*, 1996) may allow a future series of experiments to be carried out without employment of the time-delay approach (Smirnov *et al.*, 1997). However, at present, the time-delayed technique has to be used.

2. Dielectrical permittivity in the conditions of Mössbauer scattering

Consider total reflection at a flat ^{56}Fe/^{57}Fe isotope interface. The reflection in the range of the critical angles of incidence has been calculated (Belyakov & Zhadenov, 1994, 1995) using the Fresnel formulae (Born & Wolf, 1959; Stratton, 1941; Rölsberger *et al.*, 1992) with allowance made for the complex refraction index in the vicinity of Mössbauer resonance.

As is well known, the dielectrical permittivity ε of a medium is determined by

$$\varepsilon = 1 + N\lambda^2 f/\pi,$$

where f is the forward-scattering amplitude, λ is the wavelength and N is the scatterer density. From the side of the ^{56}Fe isotope, $f = f_r$ where

$$f_r = -r_e = -e^2/mc^2$$

and r_e is the classical radius of the electron. For isotope ^{57}Fe, $f = f_r + f_m$ where f_m is the forward Mössbauer scattering amplitude. For an unsplit Mössbauer line, f_m has the form

$$f_m = -[p\lambda(2j'+1)\Gamma_i f^M]/\{8\pi(2j+1)[E - E_m + (i\Gamma/2)]\},$$

where j and j' are the spins of the ground and Mössbauer levels, respectively, Γ and Γ_i are the total and radiation widths of the Mössbauer level, respectively, E_m and E are the energies of the Mössbauer transition and synchrotron radiation quanta, respectively, f^M is the Lamb–Mössbauer factor and p is the Mössbauer isotope abundance.

For the numerical calculations we use $(f^M)^2 = 0.7$. The jump of ε at the isotope interface, and consequently synchrotron radiation reflection, occurs for synchrotron radiation quanta with energy close to the Mössbauer transition energy E_m. Strong reflection takes place for incidence angles not larger than the critical angle of total reflection φ_c, determined by

$$\cos\varphi_c = \{[1 + N\lambda^2(f_r + f_m)/\pi]/(1 + n\lambda^2 f_r/\pi)\}^{1/2}$$

$$\varphi = (-N\lambda^2 f_m/\pi)^{1/2}.$$

It follows that for a synchrotron radiation beam incident at the interface from the ^{56}Fe side, the condition of total reflection is valid only if the synchrotron radiation photon energy is higher than the Mössbauer transition energy.

3. Roughness of the isotope interface

The technique for the preparation of the isotope interface utilizes the modern methods of growing layered structures (see, for example, Chumakov, 1993), whereby alternate evaporation of layers with the required element (isotope) composition onto the interface is carried out. Naturally, as this takes place, the isotope interface cannot be absolutely smooth, but contains random deviations from an ideal plane. Only in the most favourable case is the deviation of the interface from the ideal plane on an atomic scale and, in general, significant boundary roughness is to be expected. Here we analyse the influence of roughness of an isotope interface with a random profile (see Fig. 1) on the efficiency of synchrotron radiation Mössbauer filtration.

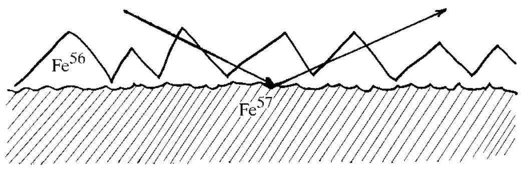

Figure 1
The idealized scheme considered for the roughness structure for synchrotron radiation Mössbauer filtration.

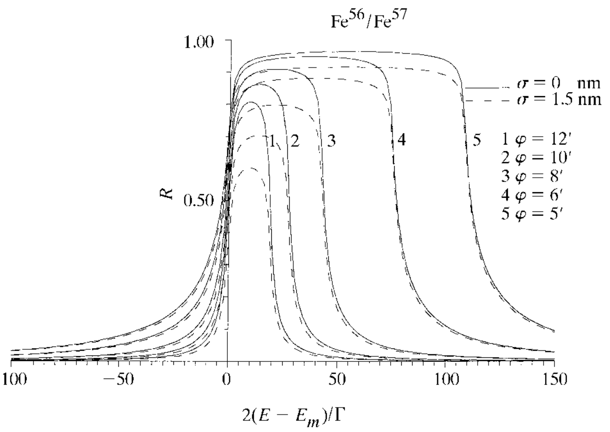

Figure 2
The energy dependence of synchrotron radiation reflection at an isotope interface for several values of grazing angle φ and a roughness parameter σ = 1.5 nm.

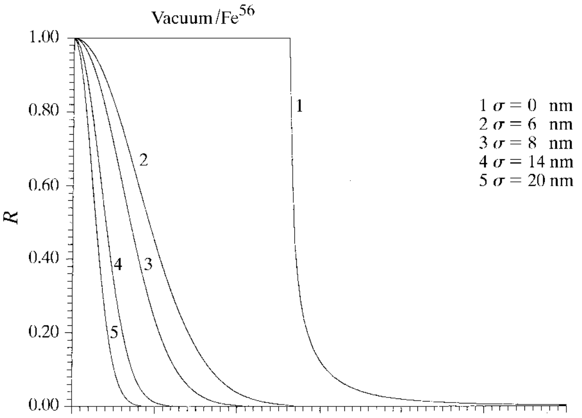

Figure 3
The angular dependence of the synchrotron radiation specular reflection coefficient, at a rough external boundary, for different values of the roughness parameter σ.

The influence of the surface roughness on the X-ray scattering (see, for example, Beckman & Spizzichino, 1963; Eastman, 1978; Nevot & Crose, 1980; Pynn, 1992; de Boer, 1994) causes the specular reflection coefficient to be decreased compared with the case of an ideal flat interface. Moreover, diffuse reflection takes place.

Fig. 2 shows the calculated energy dependence of the synchrotron radiation reflection coefficient at the ^{56}Fe/^{57}Fe isotope interface close to the total reflection region, for several grazing angles at a root-mean-square roughness σ = 1.5 nm. As would be expected, the influence of roughness on the reflectivity decreases as the grazing angle decreases and is generally not very strong in the grazing-angle range acceptable for synchrotron radiation filtration. For σ = 1.5 nm and a grazing angle $\varphi = 12'$ the decrease of reflectivity is approximately 10%.

4. Roughness of the external boundary

In the calculation of synchrotron radiation filtration at a structure with a saw-like external boundary profile, an idealized situation has been assumed, *i.e.* a rigorously regular saw-like profile. This gives an angular beam separation, reflected at the isotope and external boundaries (Belyakov & Zhadenov, 1994, 1995). For optimal synchrotron radiation filtration the angle at the base of an individual tooth has a range of less than $10'$ and the tooth height has a range 0.01–0.05 µm (Belyakov & Zhadenov, 1994, 1995).

Evaluation of the influence of relief irregularities (Belyakov & Semenov, 1997) can be most simply estimated in a random roughness model of the interface, with the root-mean-square parameter σ set close to the optimal tooth height. The corresponding calculation shows (see Fig. 3) that a range of roughness parameters σ is found, in which the specular reflection coefficient at an external surface is very strongly suppressed (by a factor of 10^3) compared with the reflectivity at an ideal plane.

However, a totally irregular relief structure generates diffuse scattering, which is a further non-resonant background source. In the wavelength, grazing-angle and roughness-parameter ranges considered, the ratio of the intensity of diffusely scattered radiation to the intensity of specular reflection does not exceed 1% (Sinha *et al.*, 1988).

Fig. 4 shows calculated curves of the angular dependence of the integral, with respect to energy, of the reflection coefficient at an

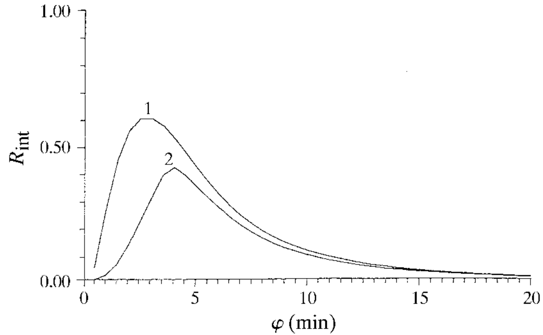

Figure 4
The angular dependence of the integral, with respect to energy (plot 1, $10^5\Gamma$; plot 2, $10^2\Gamma$), of the reflection coefficient at the isotope interface, accounting for the reduction of beam intensity as a result of non-resonant scattering at the external boundary and from absorption in the ^{56}Fe layer.

isotope interface. The numerical values correspond to the spectral density of the incident beam, which is equal to 10^{-2} quanta s^{-1} in the energy interval Γ.

When the roughness parameter is 10 nm, comparison of plots 1 and 2 in Fig. 4 shows that the bulk of the intensity of the filtered synchrotron radiation, at grazing angles greater than $4'$, is concentrated into the region $\sim 10^2\Gamma$ close to the resonance.

The calculations performed on synchrotron radiation Mössbauer filtration at an isotope interface, with allowance made for the irregularity of the boundaries, show that these structures might satisfy the requirements for synchrotron radiation filtration.

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References

Beckman, P. & Spizzichino, A. (1963). *The Scattering of Electromagnetic Waves from Rough Surfaces*. London: Pergamon.

Belyakov, V. A. (1992). *Diffraction Optics of Complex-Structured Periodic Media*, ch. 10. New York: Springer.

Belyakov, V. A. & Semenov, S. V. (1997). *Izmer. Tekh.* 5–9.

Belyakov, V. A. & Zhadenov, I. V. (1994). *Izmer. Tekh.* pp. 55–57; *Phys. Lett. A*, **189**, 248–252.

Belyakov, V. A. & Zhadenov, I. V. (1995). *Nucl. Instrum. Methods A*, **359**, 195–199.

Boer, D. K. G. de (1994). *Phys. Rev. B*, **49**, 5817–5820.

Born, M. & Wolf, E. (1959). *Principles of Optics*. London: Pergamon.

Chechin, A. I., Andronova, N. V., Zalepukhin, M. V., Artem'ev, A. N. & Stepanov, E. P. (1983). *Pis'ma Zh. Eksp. Teor. Fiz.* **37**, 531–534.

Chumakov, A. I., Baron, A. Q. R., Rüffer, R., Grünsteudel, H., Grünsteudel, H. F. & Meyer, A. (1996). *Phys. Rev. Lett.* **76**, 4258–4261.

Chumakov, A. I., Metge, J., Baron, A. Q. R., Grünsteudel, H., Grünsteudel, H. F., Rüffer, R. & Ishikawa, T. (1996). *Nucl. Instrum. Methods A*, **383**, 642–644.

Chumakov, A. I., Smirnov, G. V., Baron, A. Q. R., Arthur, J., Brown, D. E., Ruby, S. L., Brown, G. S. & Salaschenko, N. N. (1993). *Phys. Rev. Lett.* **71**, 2489–2492.

Eastman, J. M. (1978). *Phys. Thin Films*, **10**, 167–226.

Gerdau, E., Rüffer, R., Winkler, H., Tolksdorf, W., Klages, C. P. & Hannon, J. P. (1985). *Phys. Rev. Lett.* **54**, 835–838.

Hannon, J. P., Hung, N. V., Trammell, G. T., Gerdau, E., Mueller, M., Rüffer, R. & Winkler, H. (1985*a*). *Phys. Rev. B*, **32**, 5068–5080.

Hannon, J. P., Hung, N. V., Trammell, G. T., Gerdau, E., Mueller, M., Rüffer, R. & Winkler, H. (1985*b*). *Phys. Rev. B*, **32**, 5081–5092.

Hannon, J. P., Trammell, G. T., Gerdau, E., Mueller, M., Rüffer, R. & Winkler, H. (1985*a*). *Phys. Rev. B*, **32**, 6363–6373.

Hannon, J. P., Trammell, G. T., Gerdau, E., Mueller, M., Rüffer, R. & Winkler, H. (1985*b*). *Phys. Rev. B*, **32**, 6374–6384.

Hannon, J. P., Trammell, G. T., Mueller, M., Gerdau, E., Winkler, H. & Rüffer, R. (1979). *Phys. Rev. Lett.* **43**, 636–639.

Nevot, L. & Crose, P. (1980). *Rev. Phys. Appl.* **15**, 761–779.

Pynn, R. (1992). *Phys. Rev. B*, **45**, 602–612.

Rölsberger, R., Gerdau, E., Harsdorff, M., Leupold, D., Lüken, E., Metge, J., Rüffer, R., Rüter, H. H., Sturhahn, W. & Witthoff, E. (1992). *Europhys. Lett.* **18**, 561–564.

Seto, M., Yoda, Y., Kikuta, S., Zhang, X. W. & Ando, M. (1995). *Phys. Rev. Lett.* **74**, 3828–3831.

Sinha, S. K., Sirota, E. B., Garoff, S. & Stanley, H. B. (1988). *Phys. Rev. B*, **38**, 2297–2311.

Smirnov, G. V., van Bürck, U., Chumakov, A. I., Baron, A. Q. R. & Rüffer, R. (1997). *Phys. Rev. B*, **55**, 5811–5815.

Stratton, J. A. (1941). *Electromagnetic Theory*. New York/London: McGraw-Hill.

Toellner, T. S., Sturhahn, W., Rölsberger, R., Alp, E. E., Sowers, C. H. & Fullerton, E. E. (1995). *Phys. Rev. Lett.* **74**, 3475–3478.

Zhang, X. W., Yoda, Y., Seto, M., Maeda, Yu., Ando, M. & Kikuta, S. (1995). *Jpn. J. Appl. Phys.* **34**, L330–L333.