Purely nuclear diffraction of γ radiation in a resonant multilayer mirror

A. I. Chumakov and G. V. Smirnov
I. V. Kurchatov Institute of Atomic Energy, 123182, Moscow

S. S. Andreev, N. N. Salashchenko, and S. I. Shinkarev
Institute of Applied Physics, Russian Academy of Sciences, 603600, Nizhniĭ Novgorod

(Submitted 28 February 1992)
Pis'ma Zh. Eksp. Teor. Fiz. **55**, No. 9, 495–499 (10 May 1992)

A multilayer structure described by the formula $[^{57}\mathrm{Fe}(22)/\mathrm{Sc}(11)/\mathrm{Fe}(22)/\mathrm{Sc}(11)]\cdot 25$ has been grown by magnetron sputtering. A purely nuclear reflection of γ radiation occurs in this structure, because of a difference between the periods of the variations in the electron density and the density of resonant nuclei. The energy and angular distributions of the γ-ray diffraction in this purely nuclear reflection have been studied.

The resonant diffraction of γ radiation in synthetic multilayer structures is attracting interest because it is stimulating further development of the physics of coherent interactions of radiation with matter. It is also of interest in connection with the promising outlook for the development of optical systems for producing γ-ray beams with a very narrow energy spread at synchrotron-radiation sources. The ability to tap radiation with a very narrow energy spread from a synchrotron spectrum is pertinent to holography and interferometry. It is also pertinent to research on the temporal characteristics of resonant nuclear fluorescence,[1] which is a rapidly developing field. One problem here is to single out a portion of the radiation near the energy of a nuclear transition with an energy width several hundred times the natural width of the nuclear level. For the isotope ^{57}Fe ($\Gamma_0 = 4.7\times 10^{-9}$ eV), for example, this width is about 1 μeV. The impressive technological progress in the fabrication of synthetic multilayer structures in recent years[2–4] has brought us to the point that this problem can be solved with the help of such structures.

An attractive feature of the use of synthetic multilayer structures is that it is possible to arrange a large interplanar distance in the superlattice and thus to achieve small diffraction angles. As the Bragg angle is reduced to 1°, the angular width of the reflection increases to several arc minutes, i.e., to a level much larger than the characteristic values of the divergence of synchrotron-radiation beams. Under these conditions, the intensification of the radiation channel for nuclear scattering reaches a peak.[5] It thus becomes possible, under conditions corresponding to a collective interaction of the nuclear system with the radiation (e.g., under diffraction conditions), to increase the width of the nuclear resonance by a large factor. Calculations of the energy spectra of the γ-ray diffraction in resonant synthetic multilayer structures have shown[6] that reflection can be achieved in an energy band on the order of 1 μeV. Another interesting aspect of synthetic multilayer structures is the fairly wide latitude in the choice of structural elements, so that it becomes possible to develop nuclear mirrors for the resonant energies of a number of isotopes. Finally, a desired hyperfine structure of the nuclear levels can be constructed during the fabrication of resonant synthetic multilayer structures, by selecting the appropriate surroundings for the resonant isotope.

The scattering of nuclear γ radiation in a resonant mirror has been studied[7] for the particular synthetic multilayer structure $[^{57}\mathrm{Fe}(20)/\mathrm{Sc}(33)]\cdot 20$ {here and below, the notation $[A(d_A)/B(d_B)]\cdot N$ means that the synthetic multilayer structure contains N layers of elements A and B, with respective thicknesses d_A and d_B, expressed in angstroms}. We studied the energy spectrum of the structurally allowed reflection, which resulted from the scattering of γ radiation by both the nuclear and electronic systems of the sample.

Our purpose in the present study was to achieve purely nuclear diffraction of γ radiation in a synthetic multilayer structure. For this purpose we fabricated a structure based on a given pair of elements, but with an alternating isotopic composition of iron: $[^{57}\mathrm{Fe}(22)/\mathrm{Sc}(11)/\mathrm{Fe}(22)/\mathrm{Sc}(11)]\times 25$. The period of the variation in the density of the resonant ^{57}Fe nuclei in this structure is twice the period of the variation in the electron density. In this situation we would expect the appearance of a Bragg reflection of a purely nuclear nature. Similar studies have been carried out on the structure $[^{57}\mathrm{Fe}_5\mathrm{B}_4\mathrm{C}(39)/^{56}\mathrm{Fe}_5\mathrm{B}_4\mathrm{C}(48)]\times 10$.

The sample was fabricated by magnetron sputtering in argon at a pressure of 6.0×10^{-3} torr. Fifty layers of Fe and Sc, with respective thicknesses of 22.8 and 10.9 Å, were deposited alternately on a glass substrate with dimensions of 60×30 mm. The Fe was sputtered under dc conditions, and the Sc in an rf discharge. The Fe layers were alternately layers of enriched iron (95%) and of natural-abundance iron. The deviations from the nominal technical conditions during the sputtering were less than 1%. Particular attention was paid to the problem of keeping the thicknesses of the enriched iron and natural-abundance iron equal, since deviations from a uniform thickness might lead to an unwanted periodicity (66 Å) in the electron density. The characteristics of the structure were monitored on an x-ray diffractometer with Cu $K\alpha$ radiation, by a procedure which has been tested previously.

The hyperfine structure of the nuclear levels was studied by conversion-electron Mössbauer spectroscopy. A flowing-gas electron proportional detector was used.[10] The structure of the nuclear levels (Fig. 1) turned out to be similar to that which had been found previously[7] for the structure $[^{57}\mathrm{Fe}(20)/\mathrm{Sc}(33)]\cdot 20$. The spectrum has a single, asymmetric peak with a width of about $8.3\Gamma_0$ ($\Gamma_0 = 0.097$ mm/c). When these results are combined with earlier results demonstrating that the iron layers are nonmagnetic and amorphous,[7] we are led to the conclusion that the spectrum is a superposition of several unsplit doublets, as would be characteristic of the amorphous alloy α-$\mathrm{Fe}_{90}\mathrm{Sc}_{10}$ (Ref. 11). Model-based calculations of the spectrum have shown that the width of the distribution of hyperfine parameters is small, and the result of the superposition can be described accurately by a single asymmetric doublet. The shape of the Mössbauer diffraction spectra discussed below supports this idea.

The angular and energy characteristics of the reflection of the nuclear γ radiation were measured on a two-crystal Mössbauer diffractometer. A ^{57}Co(Cr) Mössbauer source was used. The Fe (110) monochromator gave the beam an angular collimation

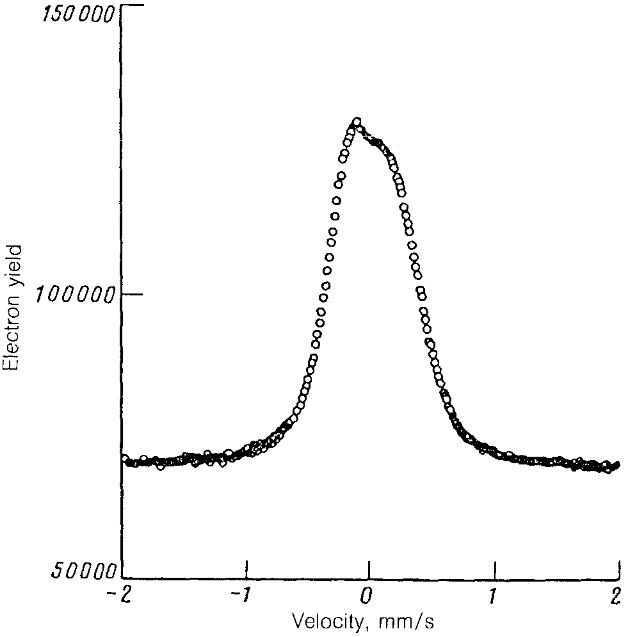

FIG. 1. Conversion-electron Mössbauer spectrum of the multilayer structure.

$\simeq 15''$. Away from the resonance (Fig. 2a), the angular curve of the reflection of γ radiation from the structure has a trace of total external reflection. This trace disappears at an angle of incidence $\theta \simeq 15'$. There is also a Bragg peak of the first order of reflection, with $\theta_{\rm Br} \simeq 46'$ and a width $\sim 2.1'$. This peak corresponds to an interplanar distance of 33 Å. In contrast, the rocking curve measured near the resonance (the velocity of the source was varied uniformly over the range ± 2.0 mm/s; Fig. 2b) has an additional reflection with a width $\sim 3.2'$ at an angle of incidence half as large, $\theta_{\rm Br} \simeq 24'$. This reflection corresponds to the period of the variation in the density of ^{57}Fe nuclei, i.e., 66 Å. The additional reflection peak is thus a consequence of purely nuclear scattering. (Nuclear reflections in a synthetic multilayer structure have recently been observed by neutron diffraction.[2,12])

The ratio of the measured intensities in the purely nuclear peak and the structurally allowed peak (Fig. 2b) does not correspond to the ratio of reflection coefficients, since the transverse dimension of the γ-ray beam (0.6 mm) was relatively large, and at the angular position corresponding to purely nuclear scattering some of the radiation did not strike the sample. The reflection coefficients calculated with this circumstance in mind are 10% and 8% for the purely nuclear peak and the structurally allowed peak, respectively.

The low intensity of the γ radiation rules out accurate estimates of the extent to which electron scattering is suppressed in the purely nuclear peak. Possible sources of an electron component are total external reflection and differences between the thicknesses of the ^{57}Fe and natural-abundance Fe layers. To resolve this question we mea-

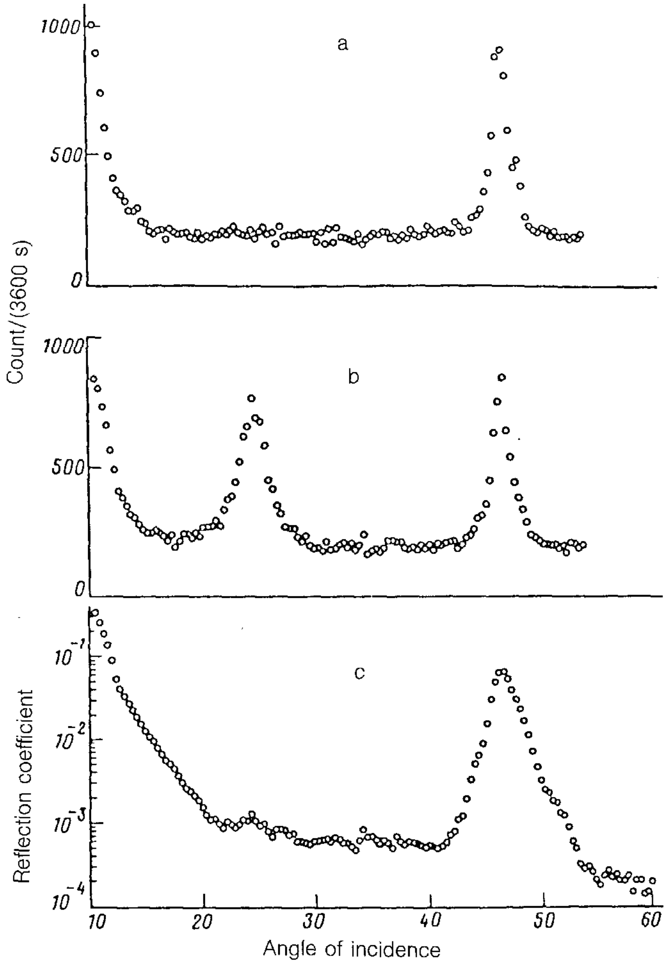

FIG. 2. a,b—Angular distributions of the intensity of the scattered γ radiation (a) away from the resonance and (b) at the resonance; c—reflection coefficient of 14.4-keV x radiation.

sured the angular distribution of the reflection of 14.4-keV x radiation, taken from the bremsstrahlung spectrum of an x-ray tube (Fig. 2c). The angular divergence of this radiation was 1′. The intensity of the total external reflection decreases rapidly with increasing angle of incidence. At the angular position $\theta \simeq 20'$, the reflection coefficient is no more than 10^{-3}. As the angle of incidence is reduced further, diffuse scattering becomes predominant in the measured intensity. At $\theta \simeq 24'$, there is a slight peak which can be attributed to differences in the thicknesses of the ^{57}Fe and natural-abundance Fe layers. The extent to which the electron scattering is suppressed in the purely nuclear reflection is no worse than 10^{-3}. A comparison of this figure with the intensity of the peak of purely nuclear reflection yields 10^2 as the ratio of the coefficients of nuclear and electron scattering.

Figure 3 shows the energy spectra of the purely nuclear diffraction of γ radiation at several angular positions near the Bragg peak. Each spectrum has two lines, corresponding to source velocities $\sim \pm 0.5$ mm/s, but the components of the overall intensity corresponding to these lines depend in different ways on the angle of incidence. The peak on the left is predominant at smaller angles of incidence, while that on the right is predominant at larger angles. This behavior supports the suggestion (made above) that the hyperfine structure of the nuclear levels is approximately a single doublet. The fact that the peaks do not appear simultaneously reflects the difference between the Bragg angles for the resonances on the left and on the right. A correction

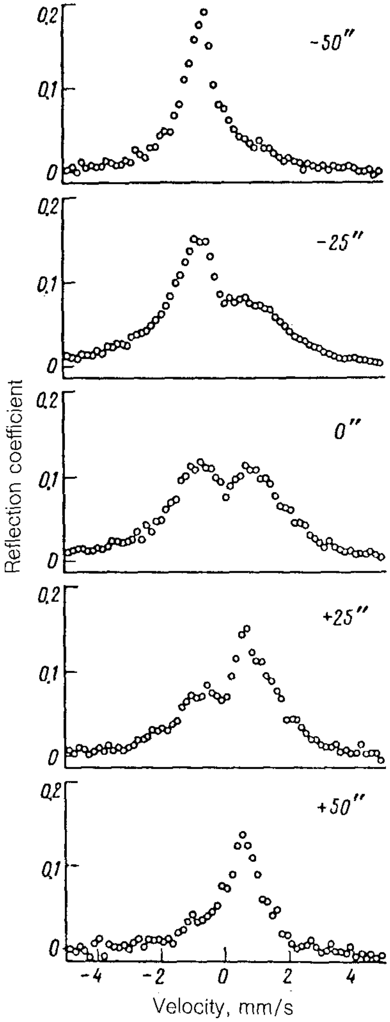

FIG. 3. Mössbauer spectra of the diffraction of the γ radiation in the purely nuclear reflection for various angular positions.

is made to the Bragg angle for the refraction of the radiation as it enters the mirror. At small reflection angles, this refraction reaches several arc minutes. The refractive indices for the left and right peaks are different, since the resonances are close along the energy scale. The dispersion increments in the refractive index are large, and opposite in sign for the left and right peaks. Consequently, the radiation at the corresponding energies is refracted in different ways as it enters the sample. The Bragg angle is thus shifted to the left or right of the exact value.

The width of the Mössbauer reflection spectrum exactly at the Bragg position (the middle spectrum in Fig. 3) reaches $40\Gamma_0$ ($\Delta E \simeq 0.2\ \mu$eV), or five times the width of the nuclear resonance in the isolated nucleus, measured by conversion-electron Mössbauer spectroscopy (Fig. 1). This broadening is evidence that collective effects play an important role in shaping the reflected wave. Of particular importance is the intensification of the radiation channel for nuclear scattering. The distance between the peaks in the energy spectrum is greater than the splitting of the quadrupole doublet. In the gap between these peaks we see a minimum, which is not found in the spectrum of conversion electrons. The spectrum has this shape because the scattering of the radiation by two resonances occurs in a destructive fashion in the gap between the resonances, while it occurs in a constructive fashion in the wings of the resonances.

This study shows that resonant synthetic multilayer structures hold promise for filtering monochromatic beams of resonant radiation from a synchrotron-radiation spectrum. In the structure studied here, $[^{57}\mathrm{Fe}(22)/\mathrm{Sc}(11)/\mathrm{Fe}(22)/\mathrm{Sc}(11)]\cdot 25$, a purely nuclear reflection of γ radiation occurs in an energy band at 0.2 μeV. The average reflection coefficient is 10%. The level of electron scattering is no greater than 0.1%.

From energy spectra of the diffraction measured for various angles (Fig. 3) we find a complete and detailed picture of the coherent interaction of γ radiation with the nuclear structure of the sample. The large angular width of the reflection makes these measurements comparatively easy in comparison with those on ordinary crystals. This circumstance, combined with the wide latitude in choosing the resonant isotope in the structure and the hyperfine structure of the nuclear levels, highlights resonant multilayer structures as entities of a unique type for studying fundamental questions in the collective interaction of nuclear radiation with matter. On the other hand, the sensitivity of the Mössbauer radiation to the magnetic order in the structure means that nuclear diffraction can be harnessed as a method for studying the properties of multilayer structures.

We wish to thank E. V. Mel'nikov and M. A. Volkov for fabricating the targets of enriched iron and A. S. Subbotin for assistance in the x-ray measurements.

[1] R. Rüffer, E. Gerdau, and M. Grote *et al.*, Hyperfine Interact. **61**, 1279 (1990).
[2] T. Shinjo, Surf. Sci. Rep. **21**, 49 (1991).
[3] A. Barthélémy, A. Fert, M. N. Baibich *et al.*, J. Appl. Phys. **67**, 5908 (1990).
[4] Yu. Ya. Platonov, N. N. Salashchenko, and L. A. Shmaenok, Izv. Akad. Nauk SSSR, Ser. Fiz. **7**, 1322 (1991).
[5] A. M. Afanas'ev and Yu. Kagan, Zh. Eksp. Teor. Fiz. **48**, 327 (1965) [Sov. Phys. JETP **21**, 215 (1965)].

[6] A. I. Chumakov and G. V. Smirnov, Pis'ma Zh. Eksp. Teor. Fiz. **53**, 258 (1991) [JETP Lett. **53**, 271 (1991)].

[7] A. I. Chumakov, G. V. Smirnov, S. S. Andreev *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **54**, 220 (1991) [JETP Lett. **54**, 216 (1991)].

[8] R. Röhlsberger, E. Witthoff, E. Lüken, and E. Gerdau, in *Physics of X-ray Multilayer Structures, Technical Digest Series*, Vol. 7, Wyoming, March 1992.

[9] A. G. Achsachaljan, A. A. Fraerman, N. I. Polushkin *et al.*, Thin Solid Films **203**, 317 (1991).

[10] A. I. Chumakov, G. V. Smirnov, and A. B. Dubrovin, Nucl. Instrum. Methods **216**, 505 (1983).

[11] D. H. Ryan, J. O. Ström-Olsen, W. B. Muir, *et al.*, Phys. Rev. B **40**, 11208 (1989).

[12] N. Hosoito, S. Araki, K. Mibu, and T. Shinjo, J. Phys. Soc. Jpn. **59**, 1925 (1990).

Translated by D. Parsons