Nuclear-resonance monochromator for synchrotron radiation using a multilayer ^{57}Fe–^{56}Fe structure

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(Submitted 8 February 1991)
Pis'ma Zh. Eksp. Teor. Fiz. **53**, No. 5, 258–262 (10 March 1991)

The parameters of a multilayer ^{57}Fe–^{56}Fe structure optimized in terms of the ability to reflect γ radiation near an energy of 14.4 keV are calculated. The structure consists of 67 layers of the isotopes ^{57}Fe and ^{56}Fe, with respective thicknesses of 30 and 15 Å. The bandwidth of the reflected spectrum of synchrotron radiation is 0.7 μeV. The reception angle of the mirror is 3′.

In a new application of synchrotron radiation, it is used to excite low-lying nuclear levels, and the nuclear fluorescence which arises as a result is studied.[1–4] In one approach, the nuclear target excited by the synchrotron radiation serves as a source of recoilless γ radiation. If the spectral characteristics of a source of this type are similar to those of a radioactive source of Mössbauer radiation (a single line with a natural width Γ_0),[5,6] one can say that the Mössbauer γ rays are filtered out of the synchrotron radiation and that there are related possibilities for developing conventional Mössbauer spectroscopy.

A more promising version of nuclear-resonance spectroscopy, and a natural one in view of the nature of synchrotron radiation, is to study the nuclear fluorescence of a sample excited by synchrotron-radiation pulses as a function of the time. The quantum intensity beats observed in a time dependence of this sort would provide comprehensive information about the hyperfine splitting of the nuclear levels, since the temporal spectrum of the fluorescence is the Fourier transform of the frequency spectrum which is measured in conventional Mössbauer spectroscopy.[7] The universal applicability of temporal Mössbauer spectroscopy with respect to the structure of the samples has been demonstrated in measurements of the temporal spectrum of the γ radiation scattered in the direction of the primary beam.[8]

In the temporal version of Mössbauer spectroscopy, the energy range of the γ rays must be wide enough to span all possible values of the resonant energies for the excitation of the given isotope in any absorber. In the case of the isotope ^{57}Fe, for example, this spectral range must be greater than the maximum value of the hyperfine splitting, on the order of $200\Gamma_0$ or about 1 μeV. It would be pointless to broaden the energy range further, since the only result would be to overload the detector with nonresonant radiation.

At present, the best results in terms of a preliminary monochromatization of synchrotron radiation are achieved with "butterfly" monochromators with large Bragg angles.[2] The energy range of the radiation in a beam of this sort, however, is more than three orders of magnitude greater than the optimum value of 1 μeV; the effect is to seriously complicate the operation of the detection system. In addition, that monochromatization method reduces the angle in which the radiation is received to a

fraction of an arc second, so there is a serious loss of intensity of the resonant radiation.

In this letter we analyze another method for monochromatizing synchrotron radiation. This method makes it possible to single out the radiation in an energy interval of about 0.7 μeV with an angular aperture of a few arc minutes. In the coherent scattering of γ radiation by nuclei, the frequency range of the reflection broadens substantially near the Bragg angle.[9] For ordinary crystals, however, the small angular width of the rocking curve—a few arc seconds—makes it possible to achieve a significant broadening of the resonance line only in a small angular interval. A suggestion for overcoming this limitation is to use artificial multilayer structures with a large lattice constant. It would then become possible to substantially broaden the angular aperture of the monochromator and to simultaneously achieve a large coherent broadening of the resonance lines. The substantial progress which has been achieved in the development of such structures[10,11] makes the idea of developing a monochromator of this type a completely definite possibility.

The multilayer structure which we are proposing consists of layers of the isotopes ^{57}Fe and ^{56}Fe, deposited alternately on a substrate in thicknesses on the order of tens of angstroms. From the standpoint of scattering by the electron shells of atoms, a sample of this sort has no periodicity along its thickness. In contrast, the nuclear-resonance radiation, which interacts exclusively with the isotope ^{57}Fe, perceives the structure as a one-dimensional crystal. All orders of the reflection of radiation from such a crystal are purely nuclear, and the coherently scattered beam consists exclusively of resonant γ rays. The possibility of developing structure of this sort was discussed previously in Refs. 12 and 13.

Analysis shows that the properties of a multilayer resonant mirror are determined by three basic parameters: the lattice constant d, the ratio (β) of the thickness of the layer of the resonant isotope (^{57}Fe) to the lattice constant, and the thickness of the structure, t. Since increasing the period of the artificial lattice reduces the Bragg angle and (as is easily shown) broadens the angular interval of reflection, the lattice constant d must be chosen as large as possible. An upper limit is set on this constant by total external reflection of the radiation. In the case of a nonresonant scattering, a reflection coefficient on the order of 10^{-3} is realized at a glancing angle of about 37′. If we take 10^{-3} as the limiting permissible level of electron scattering, we conclude that the Bragg angle should be no greater than 37′. This situation corresponds to a period of 45 Å for the monochromator structure (the shift of the rocking curve away from the exact Bragg position toward larger angles due to refraction, amounting to about 4′, is taken into account).

Figure 1 shows the results of an analysis of the reflection coefficient of the monochromator as a function of the thickness of the structure and the extent to which the period of the lattice is filled with the resonant isotope, β. The reflection coefficient of the monochromator, which is plotted along the ordinate here, is the reflection coefficient, as a percentage, averaged over an energy interval of $\pm 100\Gamma_0$. A thickness of 0.2 μm at the given Bragg angle (37′) corresponds to the electron absorption length. We see that an increase in the thickness beyond 0.3 μm causes no substantial increase in the reflection coefficient. The mirror thus needs no more than 70 layers for the partic-

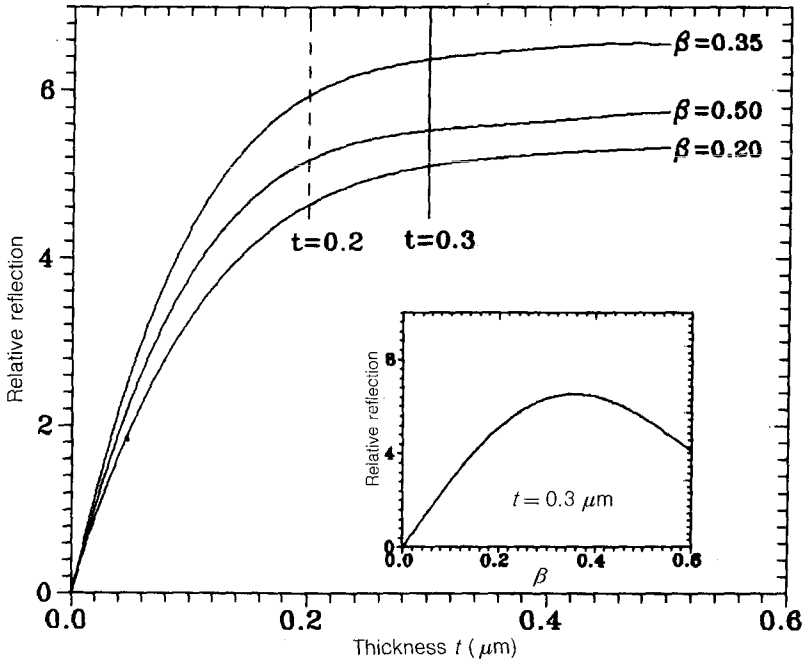

FIG. 1. Reflection coefficient of the monochromator (as explained in the text proper) versus the thickness of the multilayer structure, t, for various values of β, which is the extent to which the structure is filled with the resonant isotope.

ular lattice constant selected. The inset in Fig. 1 shows the reflection coefficient of the monochromator versus the coefficient β for a structure with a thickness of 0.3 μm. This curve has a peak, since at small values of β the density of resonant nuclei in the sample is small, while at large β the lattice has a small structure factor, so the resonant absorption of γ rays by the nuclei outweighs the Bragg reflection of the γ rays. The optimum value of the parameter β is 0.33; i.e., the optimum filling of the structure with the resonant isotope is one-third. The optimum structure thus consists of 67 layers of the iron isotopes ^{57}Fe and ^{56}Fe, deposited alternately in thicknesses of 15 and 30 Å, respectively.

The frequency reflection spectrum of the monochromator depends strongly on the direction in which the sample is magnetized. Let us assume that the magnetic properties of the monochromator are precisely the same as those an ordinary iron sample. We assume that the synchrotron radiation is scattered in the horizontal plane. In this case the radiation incident on the mirror has the π polarization. We assume that the vector magnetic field at the nuclei lies in the plane of the mirror and is perpendicular to the scattering plane. This situation corresponds to the maximum scattering of specifically the π-polarized component of the radiation. For the orientation of the magnetic field selected, the radiation causes a maximal excitation of transitions with $\Delta m = 0$ and does not excite other transitions.

Possible imperfections of the layered structure, which can be described by a devi-

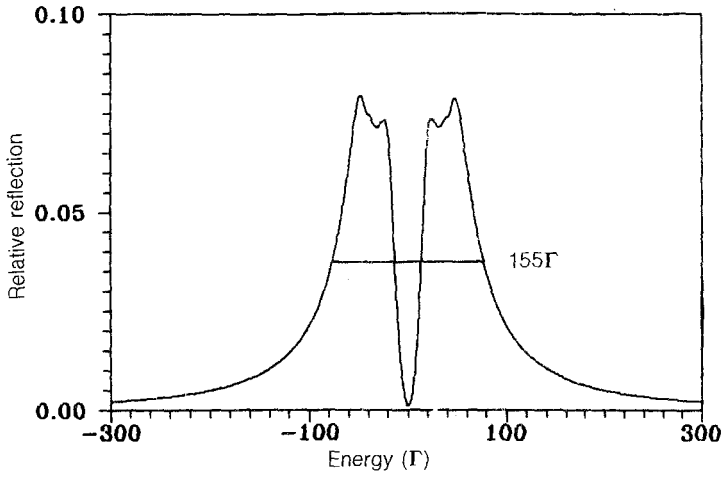

FIG. 2. Reflection frequency spectrum of the monochromator.

ation from a perfectly planar arrangement and by a roughness of the substrate, was included in the calculations. The first of these imperfections would tend to average the angular distributions over the angular interval characterizing the deviation from a planar configuration. The second of these imperfections would cause neighboring layers to penetrate into each other; this factor could be dealt with by introducing something akin to a Debye–Waller factor. To calculate characteristics of the monochromator, we selected 5 Å as a typical roughness size and 10″ as a typical deviation from a

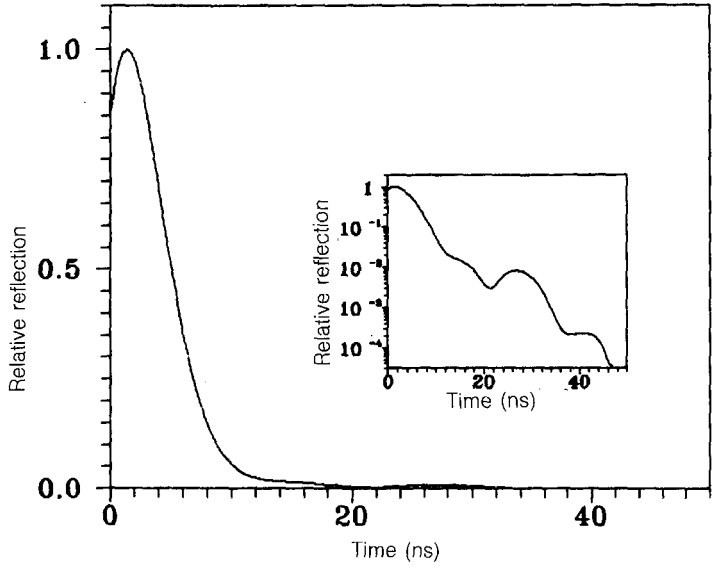

FIG. 3. Temporal response of the monochromator (the inset shows the same results in logarithmic scale).

perfectly planar configuration. These imperfections were taken into consideration in the optimization of the structure which was described above.

The rocking curve of the monochromator was calculated for the optimum parameter values of the structure and for the expected level of imperfections. The peak value of the reflection coefficient (after an averaging over an energy interval of $\pm 100\Gamma_0$) turns out to be about 6.5%, which is worse than the maximum possible value by a factor of about 2. The width of the rocking curve is about 3′. The reflection frequency spectrum (Fig. 2) has two broad lines, separated by a minimum. The reason why there is no overlap of the lines is a destructive interference of the scattering of the synchrotron radiation in the intermediate region between the two resonances. The total width of the reflection frequency spectrum is $155\Gamma_0$, greater than the distance between the outermost lines of the hyperfine structure of iron ($\sim 110\Gamma_0$). The frequency range characterizing the reflection capability of this monochromator is thus wide enough that the radiation reflected from the monochromator could be used to excite resonant transitions in most iron compounds.

The broad reflection energy spectrum means that the response of the monochromator is short in duration, ~ 5 ns in the case at hand (Fig. 3). This duration is shorter than the smallest period of the quantum intensity beats, which is ~ 8 ns for an iron sample. It might be possible to improve the characteristics of the monochromator further (to make the frequency spectrum broader, to reduce the response time, and to eliminate the minimum at the center of the energy distribution) by varying the orientation of the crystal and the magnetization direction.

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Translated by D. Parsons