Mössbauer filtration of synchrotron radiation

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This paper reviews the first experiments on Mössbauer filtration of synchrotron radiation (that is, experiments on separating, out of the continuous energy spectrum of the synchrotron radiation, a beam of extremely highly monochromatized radiation with a relative linewidth of the order of 10^{-12} at an energy of 14.4 keV) by coherent nuclear resonance scattering of the synchrotron radiation by the nuclei of a Mössbauer isotope. The corresponding theoretical investigations as well as possible variations of Mössbauer filtration of synchrotron radiation are discussed. The combination of the advantages of synchrotron radiation and Mössbauer spectroscopy, which is characterized by a uniquely high energy resolution, opens up new prospects for the use of synchrotron radiation in basic and applied investigations. This outlook refers first of all to the nuclear physics of solids and in particular to the topic of nuclear excitons in a crystal, to studies of the magnetic and electric field structure in crystals, metrological applications of synchrotron radiation, etc. The possibility of implementing new Mössbauer experiments with the use of synchrotron radiation is discusssed.

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1. INTRODUCTION

In 1983 a paper was published[1] on Mössbauer investigations with the use of synchrotron radiation, carried out in the Soviet Union at the VÉPP-3 storage ring in Novosibirsk (Institute of Nuclear Physics, Siberian branch of the Academy of Sciences of the USSR). In 1985 a similar investigation[2,3] was carried out in the Federal Republic of Germany at the synchrotron radiation source in Hamburg (HASYLAB). These investigations demonstrate the effectiveness of using synchrotron radiation in yet another promising area of research and they are milestones in a number of Mössbauer investigations with synchrotron radiation.[3–5] The essence of these works is that they have succeeded in extracting (filtering) out of the continuous synchrotron radiation spectrum an extremely highly monochromatized and tightly collimated beam of gamma radiation of rather high intensity. The physical principles underlying the filtration process are the properties of Mössbauer diffraction (diffraction of the radiation when it is scattered resonantly by the nuclei of the crystal through the Mössbauer transition); this diffraction makes it possible to separate spatially and energetically the radiation that has undergone coherent resonance scattering. The degree of monochromatization $\Delta E/E$ and the energy E of the beam are determined by the corresponding parameters of the Fe^{57} Mössbauer transition used in both experiments, i.e., $E = 14.4$ keV and $\Delta E/E \sim 3 \cdot 10^{-13}$. Its collimation is determined by the sharp directionality of the synchrotron radiation and the low divergence of the beams of radiation diffracted by perfect crystals. All these factors have made it possible, especially in the work reported in Ref. 2, to generate the highest beam brightness ever attained (by comparison with diffracted beams using ordinary Mössbauer sources). In combination with the pulsed time structure that the beam derives from the synchrotron radiation source and its controllable polarization characteristics, these beams open up new possibilities in experimental gamma-ray optics.

Because the works cited above are at the forefront of the present stage of the efforts carried out by a number of groups of investigators in various countries, and because these investigations are based both on interesting physical effects and on sophisticated modern experimental technology, it seems advisable, before discussing these experiments in detail, to dwell for a time on the physical concepts underlying this field and to present a short history of its development.

2. MÖSSBAUER DIFFRACTION OF GAMMA RADIATION

Shortly after the discovery of the Mössbauer effect a number of investigators pointed out that when Mössbauer radiation is scattered coherently from a crystal it undergoes diffraction in the same way as x rays (see references to the original papers in Ref. 8). The diffraction of Mössbauer radiation differs significantly from x-ray diffraction in that,

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along with scattering by electrons, the Mössbauer radiation also undergoes resonance scattering at the nuclei if the crystal contains the appropriate Mössbauer isotope.

The existence of resonance nuclear scattering introduces a qualitative difference into the diffraction of Mössbauer radiation (Mössbauer diffraction) as compared to x-ray diffraction, and therefore the study of this radiation provides a great deal of information about the crystal that cannot be obtained by the x-ray method. In particular, when the diffraction occurs in a crystal that has an ordered magnetic structure[7,8] [or in a crystal in which the intracrystalline electric field gradients (crystal field gradients) have a complicated structure,[9] or both at the same time[10]], there exist (purely nuclear) magnetic diffraction peaks which are formed solely as a result of nuclear resonance scattering of the Mössbauer radiation.

The physical reason for the existence of purely nuclear diffraction peaks in Mössbauer scattering is quite transparent, and it is associated with the dependence of the Mössbauer coherent scattering amplitude on the magnetic field and the crystal field gradient acting on the nucleus. Under conditions of scattering by electrons (Rayleigh scattering) and resonance nuclear scattering the total structure factor for the scattering of the gamma rays can be written in the form

$$F_\tau = F_\tau^{\mathrm{R}} + F_\tau^{\mathrm{N}}, \tag{1}$$

where τ is a reciprocal lattice vector of the crystal (in the general case, where the periodic structure of the magnetic and electric fields in the crystal are taken into account), F_τ^{R} is the well known x-ray structure factor, and F_τ^{N} is the structure factor associated with the nuclear resonance scattering. The quantity F_τ^{N} depends resonantly on the gamma-ray energy (see Ref. 6) and is given by the expression

$$F_\tau^{\mathrm{N}}(\mathbf{k},\ \mathbf{e};\ \mathbf{k}',\ \mathbf{e}') = \sum_q f^q(\mathbf{k},\ \mathbf{e};\ \mathbf{k}',\ \mathbf{e}')_{\mathrm{coh}} \exp\left[i(\mathbf{k}-\mathbf{k}')\,\mathbf{r}_q\right], \tag{2}$$

where the summation is taken within the unit cell of the crystal only over the Mössbauer nuclei, $\mathbf{k}$, $\mathbf{e}$, $\mathbf{k}'$, and $\mathbf{e}'$ are the wave vectors and the polarization vectors of the primary and scattered photons, f_{coh} and $\mathbf{r}$ are the coherent amplitude of scattering by an isolated nucleus and the position vector of the Mössbauer nucleus, and the index q enumerates the nuclei in the unit cell.

The dependence of the Mössbauer scattering amplitude on the magnetic field and on the crystal field gradients leads to the following situation: in structures where the crystallographic unit cell differs (either in size or in symmetry) from the unit cell of the magnetic structure or the crystal field gradient structure or both, there will exist purely nuclear diffraction peaks that are forbidden by the space group of the crystal (Fig. 1). In terms of the structure factors, this means that reflections exist in which the nuclear structure factor $F_\tau^{\mathrm{N}} \neq 0$ while the Rayleigh structure factor $F_\tau^{\mathrm{R}} \equiv 0$ by reason of symmetry.

It follows from this discussion that the intensity and polarization of the radiation diffracted in the purely nuclear reflections depend on the details of the structure of the crystal field gradients and the magnetic fields in the crystal, and in particular, depend on the orientation of these fields. For

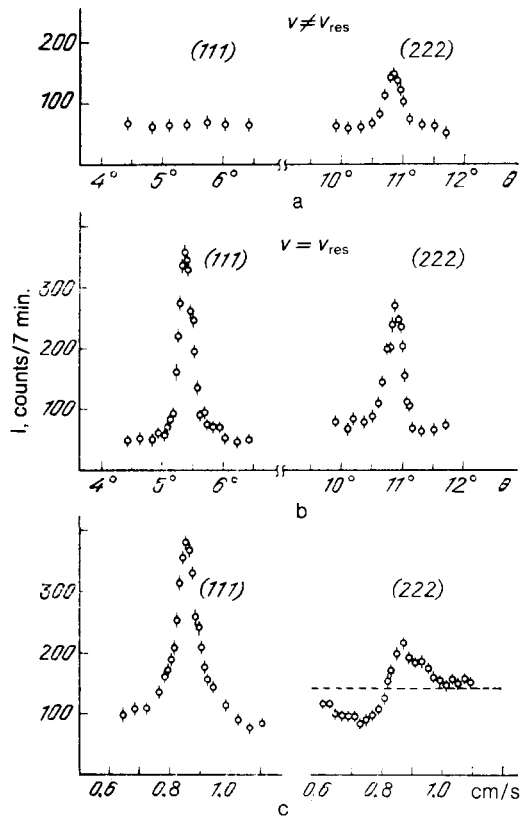

FIG. 1. Magnetic diffraction peaks in scattering from a hematite crystal.[20] The peak is absent if there is no nuclear scattering (a), and is observed if there is nuclear resonance scattering (b). Part c of this figure shows the energy dependence of the intensity of the magnetic (111) and crystallographic (222) reflections.

instance, in the case of a dipole Mössbauer transition, the dependence of the intensity of the nuclear reflection on the orientation of the magnetic field in an antiferromagnet has the form[11]:

$$I(\mathbf{k},\ \mathbf{k}',\ \mathbf{H}) = A\left|\sum_q \exp\left[i(\mathbf{k}-\mathbf{k}')\,\mathbf{r}_q\right]\right|^2 \times (\cos^2\theta + \cos^2\theta' + \sin^2\theta \sin^2\theta' \sin^2\phi), \tag{3}$$

where θ and θ' are, respectively, the angles that the directions of the incident and scattered photons make with the antiferromagnetic axis. The factor A contains the resonance dependence on the energy, and it depends on the parameters of the Mössbauer transition; the quantity ϕ is the difference in the azimuthal angles of the vectors $\mathbf{k}$ and $\mathbf{k}'$, around the antiferromagnetic axis, and the summation in (3) is taken over the nuclei in the unit cell that are located on sites with the same magnetic field direction, either $\mathbf{H}$ or $-\mathbf{H}$.

For the case of an M1 multipole transition and an unpolarized beam incident on an antiferromagnetic substance, the dependence on the orientation of the magnetic fields of the polarization of the radiation scattered into the nuclear diffraction peaks is described by the Stokes parameters, given by the following relations (in terms of the linear polarization vectors, one of which lies in the H-k′ plane and the other normal to it):

$$\xi_1 = \sin^2\theta \cos\theta' \sin 2\phi \times [\cos^2\theta + \cos^2\theta' + \sin^2\theta \sin^2\theta' \sin^2\phi]^{-1}, \quad \xi_2 = 0,$$

$$\xi_3 = [\cos^2\theta' - \cos^2\theta - \sin^2\theta\,(1 + \cos^2\theta')\,\sin^2\phi]$$
$$\times[\cos^2\theta + \cos^2\theta'$$
$$+ \sin^2\theta\,\sin^2\theta'\,\sin^2\phi]^{-1}. \qquad (4)$$

Let us remember that the degree of polarization is expressed in terms of the Stokes parameters in the following way: $P = (\sum_{i=1}^{3} \xi_i^2)^{1/2}$. The polarization matrix, defined by (4) is real. This means that in general the scattered radiation is partially linearly polarized, with the orientation of the plane of polarization of the scattered radiation depending on the orientation of the photon wave vectors relative to the antiferromagnetic axis before and after the scattering.

Expressions (3) and (4) were derived within the kinematical approximation of diffraction theory. To allow for multiple scattering of the Mössbauer radiation in the crystal, we must use a modification of those expressions.[6,12] Here let us remark too, that in the crystallographic diffraction peaks, where Rayleigh and nuclear scattering are present, the interference of these two types of scattering shows up in the energy dependence of the intensity and polarization of the scattered Mössbauer radiation and is very important and informative (Fig. 2). However, since the character of this interference depends on the details of not only the magnetic but also the crystallographic structure, the generalized formulas of the type (3) and (4) do not describe it.

The interesting physics of Mössbauer diffraction and the information it conveys in the study of crystals has been a stimulus in the development of theoretical and experimental investigations in this field. Such investigations have resulted in the discovery of many effects that are interesting both from a basic and an applied point of view and many proposals have been made for the practical application of this diffraction. Among the first of these proposals is that Mössbauer diffraction can be used to determine the phase of the x-ray structure factor,[13] which, in the case of complicated

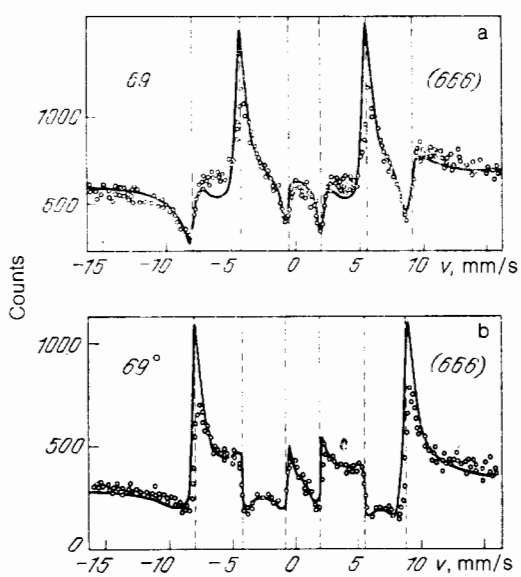

FIG. 2. Interference of nuclear and Rayleigh scattering in a crystallographic diffraction peak and its dependence on the nuclear structure factor F_τ^N (Ref. 51). The hematite (666) reflection. The antiferromagnetic axis is (a) perpendicular to the scattering plane, (b) in the scattering plane.

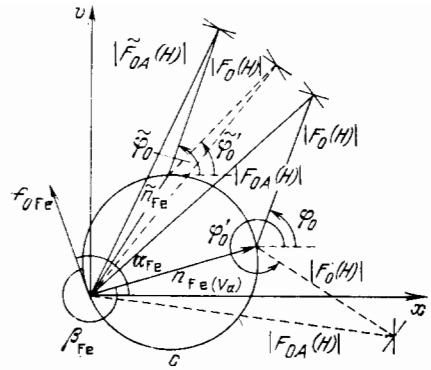

FIG. 3. Principle of determining the phase φ_0 of the x-ray structure factor $F_0(H)$. $F_{0A}(H)$, $\{\tilde{F}_{0A}(H)\}$, are the total nuclear and Rayleigh structure factors, which depend on the gamma ray energy, n_{Fe}, ($\tilde{n}_{Fe}$) is the nuclear resonance scattering amplitude.

structures, is an extremely difficult problem in x-ray structural analysis. The underlying principle here is the possibility of varying in a controlled way the phase of the nuclear component F_τ^N of the structure factor in (1) by means of a small change in the gamma-ray energy for which the Rayleigh factor F_τ^R remains unchanged. This procedure makes it possible to recover the phase of the Rayleigh amplitude F_τ^R by measuring at several energies the intensity of the Mössbauer reflection, which is proportional to $|F_\tau^R + F_\tau^N|^2$ (Fig. 3). Another attractive possibility for the application of Mössbauer diffraction is associated with its possible use in the direct (diffraction) determination of the magnetic structure of crystals as well as the structure of crystal field gradients by a method similar to magnetic neutron diffraction and based on the observation of nuclear diffraction peaks.[7,57]

Still another unique potential for practical application is afforded by the possibility of energy analysis of the diffracted Mössbauer radiation with a resolution of the order 10^{-8} eV, which makes it possible to distinguish with a correspondingly uniquely high energy resolution Mössbauer radiation scattered elastically and inelastically by the crystal.[14,15] Mössbauer diffraction experiments of this sort do not require the nuclei of Mössbauer nuclei to be present in the sample (in this case the Mössbauer photons interact with the crystal exactly as x-rays, since the amplitude F_τ^N in (1) vanishes under these conditions) and for the energy analysis of the scattered radiation ordinary techniques using a Mössbauer absorber are employed (Fig. 4). Such measurements permit the study of low-frequency dynamics and are particu-

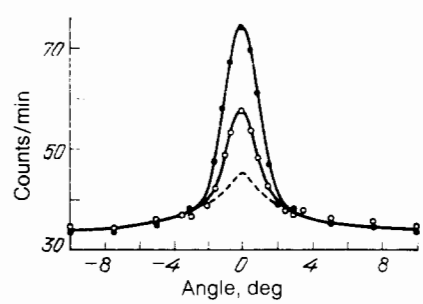

FIG. 4. Angular dependence of Rayleigh scattering of 14.4 keV Mössbauer radiation from a silicon single crystal, near the (444) reflection.[52] The upper curve is the total scattering intensity; the middle one shows the results of measurements with a resonance absorber in front of the scattered radiation detector; the dashed line, obtained by subtracting the first two, is the intensity of inelastic scattering of gamma rays.

larly useful for investigating phase transitions and biological objects.

3. MULTIPLE SCATTERING EFFECTS

For the time being in the discussion we have not assumed a high degree of perfection for the crystalline samples in which the Mössbauer diffraction takes place. In highly perfect crystals interesting physical effects occur in Mössbauer diffraction, and are associated with multiple scattering of the radiation in the crystal. Foremost among these effects is the Kagan-Afanas'ev effect,[16] which involves the suppression of nuclear absorption of the Mössbauer gamma rays in a perfect crystal under conditions of nuclear diffraction of the photons. This effect is the analog of the Bormann effect, which is well known in x-ray diffraction, but the former exhibits substantial differences from the latter because of the nuclear scattering mechanism in the case of Mössbauer radiation. In particular, in the Kagan-Afanas'ev effect the absorption can be totally suppressed, whereas in the case of the Bormann effect the suppression cannot in principle be total. This difference arises because the parameter

$$\Delta = F_{11}F_{22} - F_{12}F_{21}, \tag{5}$$

whose vanishing means that the absorption can be totally suppressed, can in fact vanish in the case of Mössbauer radiation, while in the case of x rays it cannot [in (5) F_{ip} is the structure factor for scattering from the direction i to the direction p; it describes both the scattering without change in direction of the wave vector ($i=p$) as well as with a change in wave vector from $\mathbf{k}$ to $\mathbf{k}'$ or the reverse]. The reason that (5) cannot vanish in the case of x-ray diffraction is that the structure factors include the Debye-Waller factor $\exp(-|\mathbf{k}-\mathbf{k}'|^2\langle r^2\rangle/3)$, (where $\langle r^2\rangle$ is the mean square amplitude of thermal vibrations of the atom in the crystal), which for forward scattering is rigorously equal to unity but for scattering through some finite angle is always different from unity, and hence prevents (5) from vanishing. In the case of nuclear resonance scattering, on the other hand, the Debye-Waller factor is replaced by the product of two Lamb-Mössbauer factors $\exp(-k^2\langle r^2\rangle/3)$ and $\exp(-k'^2\langle r^2\rangle/3)$, and as a result each of the terms in (5) contains the same product of Lamb-Mössbauer factors and hence the factor Δ is not prevented from vanishing. The physical reason for this difference is that the process of nuclear resonance scattering is slow compared to the oscillations of an atom in the crystal, and consists of the absorption and subsequent emission of a photon by a nucleus, while the scattering of an x-ray photon is fast and it occurs in one step. Consequently, the resonance photon "sees" the resonance nuclei in their equilibrium positions, and the thermal motion of the atoms does not disrupt the effect of suppression of the nuclear absorption. On the other hand, when the x-ray photons are scattered they see an instantaneous picture of the positions of the atoms in the crystal, which, because of the thermal motion deviates from an ideal periodic lattice and therefore does not permit complete suppression of the electronic absorption. A review of the experimental work on observations of the suppression effect can be found in Ref. 17.

Another interesting effect which shows up in Mössbauer diffraction in a perfect crystal is the formation of a collective nuclear excitation in a crystal (a "nuclear exciton"). Kagan, Afanas'ev, and Kohn[18] have shown that the time characteristics of the decay of this kind of exciton and the angular characteristics of the radiation emitted in this decay are radically different from the characteristics of an isolated excited nucleus. In particular, when the Bragg condition is fulfilled exactly the decay of a nuclear exciton that is formed in the diffraction process differs from the exponential decay of an isolated nucleus, $\exp(-t/\tau_N)$, where τ_N is the lifetime of the excited nuclear level of an isolated nucleus, and goes faster, according to the relation

$$I(t) \sim \left(\frac{t}{\tau_N}\right)^2 e^{-t/\tau_N}. \tag{6}$$

Experimental investigations of this effect have been reported in Refs. 1, 19, 55, and 56 (Fig. 5).

4. THE PROBLEM OF THE MÖSSBAUER SOURCE

Many of the fields of Mössbauer diffraction discussed in the preceding sections have been developed by investigators in the Soviet Union using standard sources of Mössbauer radiation. For example, the first observations of nuclear diffraction peaks (magnetic and quadrupolar) were made by the group of Sklyarevskiĭ[20,21] at the I. V. Kurchatov Institute of Atomic Energy, Moscow, and the group of Kuz'-min[22] at Moscow State University. The Kagan-Afanas'ev effect was also observed for the first time at the Institute of Atomic Energy by the groups of Voĭtovetskiĭ[23] and Sklyarevskiĭ.[24] The first unambiguous determination of the magnetic structure of a crystal by Mössbauer diffraction was accomplished in the work of Labushkin's group[25] (Fig. 6). Investigations of Rayleigh scattering of Mössbauer radiation, a field in which the pioneers were O'Connor[26] and Albanese,[14] have also been quite actively developed[27] particularly in the group of Gol'danskiĭ, who have applied it to biological studies.[28] In the group of Smirnov at the Institute of Atomic Energy in-

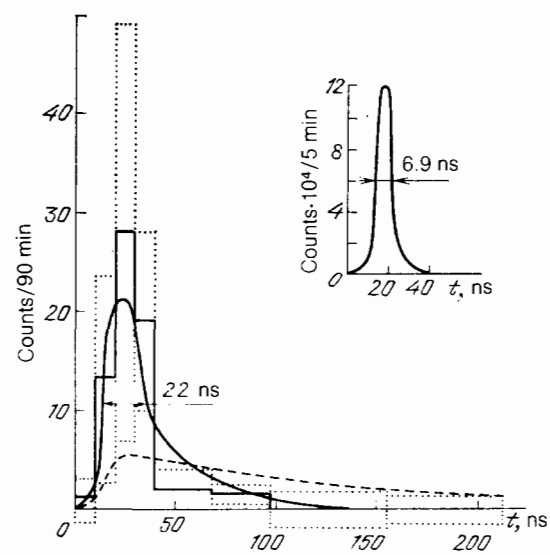

FIG. 5. Time distribution of gamma quanta obtained by Mössbauer filtration of synchrotron radiation in a hematite crystal.[1] The solid curve is the calculation for a nonexponential decay.[18] The dashed line corresponds to the calculation for the decay of an isolated Fe^{57} nucleus. The points on the histogram show the statistical error of the measurements. The inset shows the exciting synchrotron radiation pulse, measured using the nonresonance scattering of the radiation.

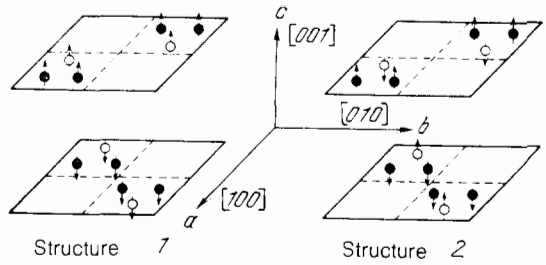

FIG. 6. Unambiguous determination of the magnetic structure of Fe_3BO_6, obtained by magnetic Mössbauer diffraction,[25] which shows that the magnetic structure of Fe_3BO_6 is described by structure 1, while structure 2 is completely inconsistent with the results of the measurements (see Fig. 10). The dark circles and open circles are the iron ions in 8d sites and the 4c sites, respectively. The arrows show the direction of the magnetic moments of the iron ions.

vestigations have also been carried out on the acceleration of the decay of excited nuclear Mössbauer levels in a crystal under diffraction conditions, using a traditional Mössbauer source[19] (the observation of this effect with the use of synchrotron radiation has been noted above[1]).

Most of the works cited are unique physical investigations, but broad extensions of these studies are hindered by technical problems, mainly connected with the low activity and brightness of existing ordinary Mössbauer radiation sources based on radioactive decay. Therefore, quite recently, along with attempts to increase the activity of ordinary Mössbauer sources, suggestions have been put forward for the creation of sources for the directional emission of gamma rays which, although of rather low total activity, could provide high brightness in a selected direction. In particular, investigators have discussed the possibility of extracting a narrowly directional, highly monochromatized beam of gamma rays from the continuous spectrum of a powerful x-ray source by diffracting the radiation into nuclear diffraction peaks[6,29] or by coherent Coulomb excitation of Mössbauer transitions by charged particles in crystals.[30,31,57] Time has shown, however, that practicable separation of Mössbauer gamma rays has come about only with the advent of synchrotron radiation in the x-ray region.

Another possibility of separating out a beam of photons monochromatized to the Mössbauer level is associated with the phenomenon of total external reflection of an x-ray beam from a material containing a Mössbauer isotope[32,33] and was noted directly in connection with the use of synchrotron radiation for this purpose. At grazing angles of incidence onto a sample of these materials there exists a small range of angles φ near zero grazing angle, $(1-n_e)^{1/2}<\varphi<(1-n_r)^{1/2}$, (where n_r and n_e are, respectively, the indices of refraction of the medium for resonance Mössbauer radiation and x-rays of nearly the same energy as the Mössbauer transition) in which the only photons that undergo total external reflection are those that have undergone nuclear resonance interaction with the sample, and as a result, the reflected radiation is highly monochromatized. Outside of this range, at smaller grazing angles, photons which interact only with electrons also undergo total internal reflection, and in this case monochromatization by total external reflection does not occur. In order to get rid of this parasitic reflection, the authors of Refs. 32 and 33 have suggested depositing on top of the layer containing the Mössbauer nuclei a film of a material with dielectric properties which will allow for the quenching of the nonresonant part of the radiation. The details of their calculations show that using a few (up to four) reflections from such layers under conditions of total external reflection also makes possible the separation a beam of gamma rays monochromatized to the level of Mössbauer radiation from the continuous synchrotron radiation spectrum.

5. MÖSSBAUER FILTRATION OF SYNCHROTRON RADIATION

For all the conceptual simplicity of separating from synchrotron radiation a line with an energy width the order of the width of a Mössbauer level, the realization of this idea is an extremely complex experimental problem. The complexity stems from the fact that we are dealing with the separation of a line of the order of 10^{-8} eV wide out of the continuous synchrotron radiation spectrum. If one realizes that only a line of the order of 1–10 eV wide can be separated out of the synchrotron radiation continuum comparatively easily with the use of ordinary diffracting grating monochromators, then it becomes clear that further monochromtization utilizing the Mössbauer effect must reduce the width of the line by another 8–9 orders of magnitude, and here the background radiation problem becomes very acute.

The solution of this problem is supplied by another important property of synchrotron radiation—its pulsed nature. The typical duration of a burst of synchrotron radiation is of the order 1–0.1 ns, with an interval of the order of 1 μs between pulses. This time structure of synchrotron radiation makes it possible to use time delay techniques to detect the monochromatized radiation that is essentially free of background radiation that has not undergone resonance scattering from Mössbauer nuclei. The principle of this separation lies in the slowness of the Mössbauer scattering, the time of which is of the order of the lifetime of the Mössbauer level (for Fe^{57} this is 10^{-7} s), which is by far longer than the time of interaction of synchrotron radiation with the sample by any other channel.

Let us turn now to a more detailed exposition of the results on filtration of synchrotron radiation[1,3] using by way of example the most recent work.[2] The work was performed on the DORIS storage ring at the HASYLAB in Hamburg. A diagram of the experiment is shown in Fig. 7. The synchrotron radiation beam from the storage ring was first monochromatized by two germanium single crystals and then (after passing through a collimating slit and a krypton cell, the latter used for calibrating the energy of the radiation), it was directed onto the nuclear monochromator. The intensity of the beam incident on the nuclear monochromator was $(3–6)\cdot 10^{10}$ photons per second. For the further monochromatization, the investigators used the principle, described above, involving separating out the Mössbauer line by scattering the beam into a purely nuclear (in this case, magnetic) diffraction peak. For the nuclear Bragg monochromator they used two single crystal films of yttrium iron garnet (YIG) enriched to 88% in Fe^{57}. The films were arranged in the proper position for consecutive twofold purely nuclear scattering of the peak corresponding to the strong (200) nuclear reflection.

The radiation monochromatized in this way was detected with a germanium semiconductor detector (with an ener-

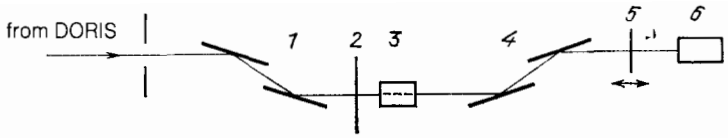

FIG. 7. Experimental arrangement for observing filtration of synchrotron radiation.[2] 1) monochromator, 2) slit, 3) krypton cell, 4) nuclear Bragg monochromator, 5) resonance absorber, 6) germanium detector.

gy resolution around 1 keV) connected with the storage ring pulse circuit by a delayed coincidence circuit. The spectrum of delayed coincidences of the germanium detector is shown in Fig. 8. For the lower curve the energy distribution of the beam incident on the nuclear monochromator did not include the energy of the Mössbauer line, at 14.4 keV, while for the upper curve, the energy of the beam was centered on that line. A conspicuous difference between the two curves immediately strikes the eye. This difference is accounted for by the presence, in the first case (the lower curve), of only the fast component of the scattering, due to diffuse scattering of the radiation by the electrons of the crystal. In the second case (the upper curve), on the other hand, besides the fast-scattering peak that is also present in the first case, there is a delayed component that is due to nuclear resonance scattering via the Mössbauer level. The authors do not discuss the details of the time delay of the radiation that has undergone nuclear scattering, but they note that it differs from the corresponding delay in the noncoherent decay of an isolated excited Mössbauer nucleus. In their work they were also able for the first time to record a Mössbauer spectrum of the synchrotron radiation beam monochromatized in this way, using for this purpose a Mössbauer absorber placed in front of the germanium detector (Fig. 9). This spectrum reproduced the characteristic features of the well known Mössbauer transmission spectrum of YIG, and was in accord with the theoretical calculations of Mössbauer diffraction in YIG.

The results of that investigation show that the end result of the Mössbauer filtration of synchrotron radiation is a beam of Mössbauer photons with an angular divergence less than 70 mrad and an intensity of one photon per second. The authors also note that under the conditions of their experiment they believe it is realistic to anticipate increasing the intensity of the monochromatized beam by about 20 times, to say nothing of the enormous possibilities of increasing the intensity of a beam monochromatized by Mössbauer filtration of synchrotron radiation at existing and planned specialized synchrotron radiation sources, specifically by the use of undulators and wigglers.

The results of the very latest investigations bear out the optimism of the authors of Ref. 2. For instance, in the experiments on Mössbauer filtration of synchrotron radiation reported in Ref. 54, temporal beats caused by the hyperfine splitting of the Mössbauer line, have been observed in the intensity of the filtered beam, and in investigations where synchrotron radiation[55] or a standard source[56] were used, a significant increase in the decay rate of Mössbauer levels was observed under conditions of nuclear diffraction.

6. FUTURE MÖSSBAUER EXPERIMENTS WITH SYNCHROTRON RADIATION

The results reported in Ref. 2 and the optimistic prognosis for the further increase in the intensity of beams obtained by Mössbauer filtration of synchrotron radiation make it important to examine the imminent new possibilities opened up by this new method of obtaining highly monochromatized beams of gamma rays. First of all, of course, one should remark on the broad prospects for expanding the number of isotopes of various chemical elements for which it is possible to do Mössbauer experiments. The issue here is that although many isotopes have low-lying excited nuclear levels which in principle permit observation of the Mössbauer effect, sources of radiation suitable for the experiments have been developed for only a small number of them. The impediment is frequently the absence of suitable radioactive decay chains that have sufficiently long lifetimes for the experiments and which have as the penultimate link the required nucleus in a low-lying excited state. The method of Mössbauer filtration of synchrotron radiation reduces the source problem because to achieve the filtration it is sufficient to have the required isotope with the level for the Mössbauer studies only in the ground state. For instance, Ruby[34] (see also Ref. 35), in proposing the idea of Mössbauer filtration of synchrotron radiation, immediately suggested creating by this method a source for carrying out Mössbauer experiments on the isotope K.[40] Of course, this is only one example, and the number of nuclei that are candidates for entering the list of Mössbauer isotopes by this means is quite comparable to the number of isotopes in which the Möss-

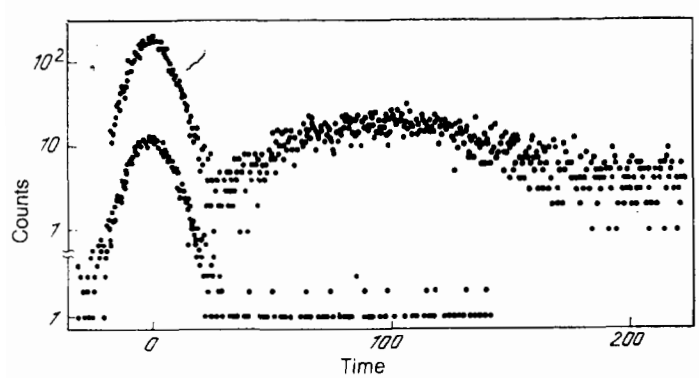

FIG. 8. Time dependence of scattering intensity of synchrotron radiation in resonance (upper curve) and out of resonance (lower curve). The peak around 100 ns does not correspond to the time characteristics of noncoherent decay of the Mössbauer level.[2] Time measured in ns.

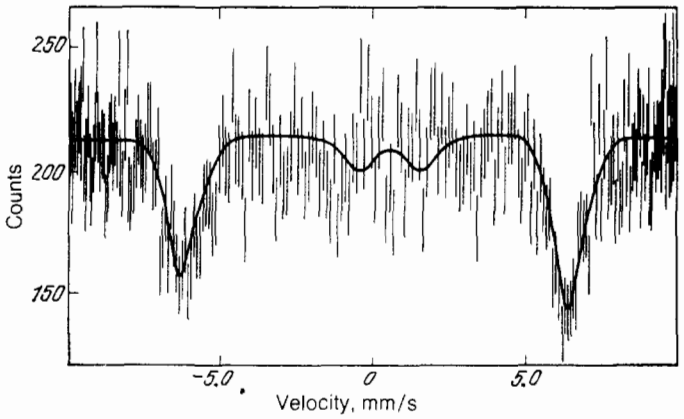

FIG. 9. Mössbauer spectrum of the radiation after scattering of synchrotron radiation by two YIG crystals. Solid curve: theoretical calculation of the spectrum.[2]

bauer effect has been observed to the present time.

Another promising direction for study is easily discerned in the analysis of a series of Mössbauer diffraction experiments which are unique and are frequently achieved at the very limits of what is possible. In this connection should be mentioned the work of the group of G. V. Smirnov, of the Institute of Atomic Energy, on the acceleration of the decay of a nuclear level upon its coherent excitation under diffraction conditions, as well as the work of that group, carried out in cooperation with West German investigators headed by R. Mössbauer,[17] in studying the Kagan-Afanas'ev effect with the use of conventional sources. In these experiments it was possible with a uniquely fast Mössbauer radiation modulator, to measure the acceleration of the nuclear decay of a nucleus by coherent excitation. Nonetheless, as a discussion of these results and the results obtained with synchrotron radiation[1] shows, there exists here a problem of distinguishing the noncoherent sources of line broadening, which cause an apparent acceleration of the nuclear decay. The corresponding experiments could of course be carried out more effectively with synchrotron radiation. The results of the investigation of the Kagan-Afanas'ev effect (suppression of the inelastic channels of nuclear reactions) are also directly related to Mössbauer filtration of synchrotron radiation, since these results reveal the conditions that are optimum for the realization of this effect, and they show that the appropriate experiments will be developed with the use of synchrotron radiation.

From an applied point of view broad and important prospects are being opened up in connection with studying the structure of magnetic fields and crystal field gradients in crystals. In this direction, in addition to the exploitation of these possibilities for determining magnetic and crystal structure afforded by the observation of purely nuclear reflections, one should bear in mind also the possibilities stemming from the very weak dependence of Rayleigh nonnuclear scattering of synchrotron radiation on the orientation of the atomic magnetic moment and on the asymmetry of the spatial distribution of the atomic electron density in the crystal, and the scattering anisotropy associated with it.[36,37] However, before turning to a discussion of the nonnuclear scattering mechanism, let us note another recently discovered feature of coherent nuclear scattering which proves useful in magnetic and other structural investigations by the Mössbauer diffraction. This has to do with the large amount of information implicit in the interference of the components of the hyperfine nuclear splitting of the levels of nuclei located at crystallographically nonequivalent sites in the unit cell. In the scattering this interference is manifested in a very strong dependence of the hyperfine splitting spectrum on the details of the structure, in particular on the magnetic structure.[38] This dependence made it possible for Kovalenko and coworkers[25] to remove all ambiguity in the magnetic structure of Fe_3BO_6 (Fig. 10).

Moving now to another possibility, not connected with nuclear scattering, for using synchrotron radiation for a direct, i.e., diffraction, method of determining the magnetic structure of crystals, let us note at once that this possibility was mentioned quite some time ago in connection with x-ray studies[36] and moreover, was realized in experiments which used x-ray tubes as the radiation source.[39,40] Now, with the advent of synchrotron radiation sources, the situation has changed qualitatively. Actually, the spin-dependent part of

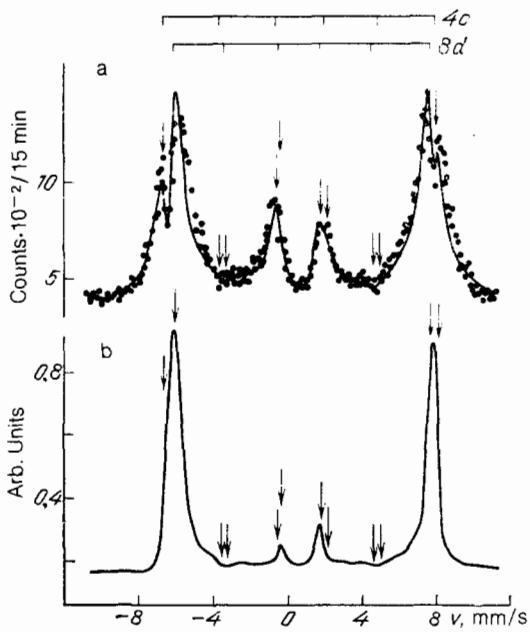

FIG. 10. Experimental and theoretical spectra of the (700) reflection from a Fe_3BO_6 crystal,[25] demonstrating the interference of nuclear scattering from the iron nuclei occupying crystallographically nonequivalent sites, and permitting a unique determination of the magnetic structure of Fe_3BO_6 (see Fig. 6). The results of the calculations for structures 1 and 2 (see the curves in Fig. 6) are given by the solid curves of Figs. 2a and 2b, respectively. The resonance positions for the iron nuclei in the 4c- and 8d-sites are shown by the arrows.

the synchrotron radiation (x-ray) scattering amplitude is 3–4 orders of magnitude less than the isotropic component and as a result the observation of magnetic scattering entails such great difficulties that the above-mentioned observation of magnetic diffraction peaks in the scattering of radiation from x-ray tubes can be considered no more than a demonstration of the effect. The incomparably greater intensity of synchrotron radiation relative to the radiation from x-ray tubes provides a bright outlook for studying the magnetic structure of crystals with the use of synchrotron radiation[41] by a completely practicable method, with the prospect that these studies will prove to be an important complement to the chief method of determining directly the magnetic structure of crystals by neutron diffraction.[37,53] In discussing investigations of the magnetic structure (as well as the structure of crystal field gradients) it should be kept in mind that the advantages of synchrotron radiation are its continuous spectrum and its high degree of polarization. The latter is important, since the anisotropy of Rayleigh scattering shows up most strongly near the absorption lines of the crystal,[42,43] and to obtain the maximum effect of magnetic scattering or scattering associated with the asymmetry of the electron density distribution in the crystal, it is favorable to have a frequency-tunable radiation source. It is also important that the magnetic scattering effect is strongest for polarized radiation (with a large component, as a rule, of circular polarization), while the radiation scattered into magnetic diffraction peaks is also characterized by a polarization that is different for the polarization in the crystallographic diffraction peaks. All these points, as can be seen, underscore the advantages and the potentialities for the use of synchrotron radiation in investigations of the magnetic structure of crystals.

The potential exists for using synchrotron radiation (through the technique of Mössbauer filtration) in the study of surface properties,[44,45] and in particular using it to observe inelastic channels of the decay of Mössbauer nuclear levels. These possibilities have been demonstrated in the work reported in Ref. 46, in which electron internal conversion was observed under conditions of Mössbauer diffraction, and it was shown that the method is highly sensitive to the perfection of the surface layers.

Let us note also the possibility of using synchrotron radiation for carrying out new Mössbauer diffraction experiments which would be very problematic if standard sources of Mössbauer radiation were used. For instance, the coherent synchrotron-radiation-induced excitation of a cascade of gamma transitions in a single crystal has been studied theoretically.[47] It was found that, in comparison to the case of coherent excitation of the first nuclear level, which has been the main topic of discussion in this paper, the coherent excitation of a cascade of low-lying nuclear gamma transitions exhibits interesting features, especially in the angular characteristics of the coherent emission of the gamma rays of the cascade. In Ref. 47 the case of an arbitrary number of gamma transitions was treated, but since the cases of practical interest involve only two or three transitions in the cascade, we shall discuss the results using by way of example a cascade of two Mössbauer gamma transitions. Let us note here that observation of the coherent excitation of a cascade of two Mössbauer gamma transitions would evidently be simplest to carry out experimentally using the same iron isotope, Fe^{57} in which the coherent excitation of the first Mössbauer level was observed,[1,4,5] since in this isotope there is another low-lying level at 136.4 eV with which the Mössbauer effect has also been observed. It is also important that this level decays with overwhelming probability through the first Mössbauer level with the emission of a 122 keV gamma ray.

Thus, as applied to the isotope Fe^{57} the coherent excitation of the cascade means the following. A 136.4 keV beam of synchrotron radiation excites in the crystal the second Mössbauer level, which decays via a cascade of gamma rays of energy 122 and 14.4 keV and returns the nucleus and the crystal as a whole to its initial state. This sequence satisfies the conditions for a coherent process (the amplitudes are summed, and not the cross sections of the process in the individual nuclei of the crystal). In connection with the coherence of the process under discussion, the spatial periodicity of the crystal lattice has a significant effect on the process, in particular on the angular distribution of the gamma rays, which turns out to differ from the distribution described by the well known gamma-ray angular correlations in a cascade, that are observed in noncoherent nuclear decay.[48]

Analysis of the process shows that the crystal lattice introduces an additional correlation into only the last gamma ray of the cascade (in the case of Fe^{57} this is the 14.4 keV gamma ray). In the kinematical approximation—that is neglecting scattering of the cascade gamma rays in the crystal—the angular distribution of the radiation emitted coherently in the second transition of the cascade is given by the correlation $W(\mathbf{k}_s,\mathbf{k}_2)$ between the direction of the incident synchrotron radiation beam (of wave vector $\mathbf{k}_s$) and the direction of the emission of the photon (of wave vector $\mathbf{k}_2$) of the second step of the cascade:

$$W(\mathbf{k}_s,\ \mathbf{k}_2) = f'\delta(\mathbf{k}_p - \mathbf{k}_2 - \boldsymbol{\tau}) \times \int \sigma(E_s)\, I(E_s) W(\mathbf{k}_s,\ \mathbf{k}_1,\ \mathbf{k}_2)\, dE_s\, d\Omega_{k_1}, \quad (7)$$

where $\sigma(E_s)$ is the cross section for excitation of the second level and depends on the energy E_s of the synchrotron radiation, f' is the Lamb-Mössbauer factor for the second step of the cascade, $I(E_s)$ is the intensity of the synchrotron radiation, $\mathbf{k}_1$ is the wave vector of the gamma ray in the first step of the cascade, $\mathbf{k}_p = (\mathbf{k}_s/|\mathbf{k}_s|)(\omega_2/c)$, ω_2 is the frequency of the photon of the second step of the cascade, c is the velocity of light, τ is a reciprocal lattice vector of the crystal, $W(\mathbf{k}_s;\mathbf{k}_1,\mathbf{k}_2)$ is the well known angular correlation function in an noncoherent cascade of gamma transitions induced by a synchrotron radiation photon, and the integration in (7) is carried out over the directions of emission of the first photon of the cascade.

Ultimately, the connection between the direction of the beam and the direction of emission of the radiation in the final step of the cascade is given by a relation which is completely analogous to the Bragg condition in ordinary diffraction (i.e., when the energies of the initial beam and the scattered beam are identical) and which requires that the argument of the delta function in (7) vanish. There is an important difference, however, and that is that in this case the analog to the Bragg condition does not contain the wave vector of the photon with the energy of the second Mössbauer level, $\mathbf{k}_s$, whose modulus differs substantially from that of the wave vector $\mathbf{k}_2$ of the final photon and from the magnitude of $\mathbf{k}_p$. Therefore the geometrical Bragg condi-

tions for coherent excitation of the cascade are the same as for diffraction through the first excited level. It should also be noted that the use of synchrotron radiation for studying coherent nuclear decays of the type examined above makes it possible to investigate the role played by phonon processes in the decays, i.e., to study nuclear transitions accompanied by the emission or absorption of phonons of the crystal lattice. The continuous synchrotron radiation spectrum allows the excitation of these processes with an integrated probability $1-f'$, whereas the probability of excitation of phonon process with the radiation from standard Mössbauer sources is vanishingly small because of the narrowness of the Mössbauer line.

Let us draw attention to yet another possible area of application of Mössbauer filtration of synchrotron radiation—creating a standard source of electromagnetic radiation in the x-ray wavelength region. Here an important advantage of the Mössbauer filtration method is that the standard does not decay with time, in contrast to ordinary radioactive Mössbauer radiation sources. A Mössbauer radiation source based on the filtration of synchrotron radiation could serve as both an energy standard and a length standard in the corresponding ranges, since the Mössbauer radiation monochromaticity, which is unique in this energy range, makes it possible to reproduce both the energy and the wavelength of the radiation with a relative error of the order of 10^{-12} (for Fe^{57}), and in principle with an even smaller error with other isotopes. Here attention should be drawn to two circumstances. On the one hand, such high precision in the determination of energy and length is for the time being greater than is needed for any practical purposes, and on the other hand, as experiments have shown[1,19] the energy width of the line obtained by Mössbauer filtration of synchrotron radiation can exceed rather substantially the natural linewidth of the Mössbauer line. Therefore the necessity may arise of obtaining conditions of Mössbauer filtration of synchrotron radiation under which this line broadening does not occur.

A completely unexpected way of narrowing the Mössbauer line obtained by the filtration of synchrotron radiation has been demonstrated by experiments on Mössbauer diffraction using a standard source, in which interference was observed in nuclear resonance scattering from nuclei occupying crystallographically nonequivalent sites in the unit cell,[38,49] and experiments in which the phase transition from a magnetically ordered state to a paramagnetic state was studied.[50] In particular, in Ref. 50, besides a number of features of the phase transition from the antiferromagnetic to the paramagnetic state, in the same study of the antiferromagnetic substance $FeBO_3$, in the immediate vicinity of the Néel temperature, a magnetic (nuclear) diffraction peak was observed in which the energy width of the unsplit scattered line was close to the natural width of the Mössbauer level. The authors of Ref. 50 propose the use of the phenomenon that they observed for obtaining a line with a width close to the natural linewidth in experiments on Mössbauer filtration of synchrotron radiation.

7. CONCLUSIONS

In conclusion it should be stated that the first experiments on Mössbauer filtration of synchrotron radiation and the immediately succeeding work have already shown that this process has opened up a new chapter in the study of coherent gamma and x-ray optics and offers interesting possibilities both for physical investigations and for the application of this phenomenon in solid state physics and in the nuclear physics of solids. There is no doubt that the present progress observed in the improvement of synchrotron radiation sources and the scale with which these sources are being used will serve as a basis for new and interesting experiments and investigations that are being opened up in this new field.

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