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UTILIZATION OF THE SPECIFIC CHARACTERISTICS OF THE SYNCHROTRON RADIATION IN EXPERIMENTS OF THE ^{57m}Fe MÖSSBAUER LEVEL EXCITATION

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Some properties of the synchrotron radiation (SR) – such as its excessive spectral brightness and the inherent amplitude modulation – give rise to a hope on the feasibility of the direct excitation of low-lying (Mossbauer) nuclear states and on performance of a series of time-resolving experiments. The main difficulty in performing these experiments lies in the selection of the small rate of resonant quanta in the gross SR beam flux. For instance, I_{res}/I_{tot} for the intrinsic width of the ^{57m}Fe Mossbauer level is of the order of 10^{-14}.

In the present paper an experimental arrangement is proposed and investigated which provides

- preliminary monochromatization of SR by Ge monocrystals,
- suppression of the coherent background using a purely nuclear reflection at hematite single crystal α-^{57}Fe$_2$O$_3$,
- suppression of the coherent and non-coherent background resulting from both SR beam high-extent polarization and the use of reflection angles on hematite close to $2\theta = 90°$,
- reduction of the non-coherent background by means of analyzing crystal upstream the detector.

Electronic units applied provided selection of the energy spectrum range needed, measurement of the Mossbauer spectrum and time distribution.

Preliminary results of the measurements are presented.

The properties of the synchrotron radiation (SR) give rise to a hope on feasibility of the direct excitation of low-lying (Mossbauer) nuclear levels and hence on performance of a series of rather interesting experiments on investigation of the time dependance of the resonant diffraction. To perform such an experiment without SR one has to design ultra-rapid choppers* and superintense sources. For instance the SR beam brightness on existing electron storage rings in quantum energy range of 10–30 keV in the bandwidth equal to that of a Mossbauer level amounts to $\approx 10^{11}$ photons/cm^2 sec sr[1]), while under the same conditions brightness of the most powerfull source (≈ 0.1 Ci/mm^2) is 2×10^9 photons/cm^2 sec sr. To carry out a time-resolving experiment with a conventional source chopping of the quantum flux is needed thus reducing the mean brightness by a factor of ≈ 100. In contrast to that the time distribution of the SR intensity in storage rings (e.g. 1 nsec pulses spaced by 0.1–1 μsec intervals) fortunately suits these experiments** ^{57}Fe seems to be an optimum Mossbauer isotope with respect to a sum of characteristics for first experiments.

1. The experimental arrangement; estimates relevant

The work was carried out on the VEPP-3 storage ring SR beam line (electron energy up to 2.2 GeV, bending radius is 6.15 m, current up to 100 mA). Total flux of quanta of all energies per milliradian of horizontal angle interval is $\approx 10^{16}$ photons/sec mrad. Flux of quanta with the energy of 14.4 keV in the energy range equal to the intrinsic width of ^{57m}Fe level ($\Delta E = 4.5 \times 10^{-9}$ eV) amounts to 10^2 photons/sec mrad. Thus the content of resonant quanta in the gross SR beam is about 10^{-14} therefore measurements in the direct beam are obviously impossible.

In the present paper an experimental configuration is proposed shown in fig. 1a. To reduce the quantum flux incident on the crystal which contains Mossbauer nuclei a preliminary monochromatization of the SR beam is envisaged. A monochromator consists of two perfect Ge crystals positioned so as to achieve maximum dispersion. The double crystal monochromator of this type is necessary for stable selection of required wavelength from the SR beam and besides for reduction of the diffuse background. The monochromatization is effected in the vertical plane.

Downstream the monochromator the quanta strike a hematite single crystal positioned for purely nuclear Bragg reflection so as to reduce considerably the coherent non-resonant background pro-

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* Possible realization of such choppers (e.g. a vibrating quartz plate or a resonant absorber) are now under development by the authors.

** Other possible applications of SR in Mossbauer experiments and nuclear spectroscopy are discussed in[1,2]) duced by electronic diffraction. A forbiddenness factor (i.e the intensity ratio of non-resonant γ-rays reflection on electrons and nuclei in the forbidden reflection (777) to that in the allowed (666) reflection) is $K_f \lesssim 10^{-5}$, as determined from the curve of purely nuclear reflection[3]).

A goniometer bearing the hematite crystal is positioned in the plane parallel to the direction of the γ-ray beam reflected the monochromator This together with making use of the high extent of SR beam polarization (e.g at $E = 2.2$ GeV and $\lambda = 0.86$ Å the average polarization $p = 90\%$ thus the intensity ratio of π to σ components $I_\pi/I_\sigma = 5\times10^{-2}$) and choosing the angle of purely nuclear Bragg reflection close to 90° ($2\theta_B^{777} = 82°$ and the reflected γ-ray beam travels along the electric field vector in the incident wave) provide further reduction of both the flux of the coherent nonresonant quanta and non-coherent diffuse background

The higher-order harmonics are suppressed by a combination of the planes (III) Ge and (777) α^{57} Fe_2O_3 $\lambda_0 = 0.86$ Å ($E = 14.4$ keV) past the monochromator ($\theta_B = 7°30'$) is used for the experiment on the search for the purely nuclear reflection at λ_0, the reflection due to the electronic diffraction on the hematite is forbidden, $\lambda_0/2$ is eliminated by (222) Ge plane array, $\lambda_0/3$ can pass the monochromator but is not reflected on the hematite as cancelled by (21 21 21) plane array, $\lambda_0/4$ penetrates the monochromator and can be reflected on the hematite. By estimates its contribution to the intensity on the detector is small ($I_{\lambda_0/4}/I_{\lambda_0} = 3\times10^{-5}$) and is readily eliminated by means of energy amplitude analysis

A scintillator counter (NaI with the photomultiplier FEU-85) was used as a detector

A Mossbauer absorber placed upstream the monochromator was made of polycrystal hematite (15 mg/cm^2 of ^{57}Fe). Its thickness provides a total absorption of the resonant quanta at $V_{res} = 0$

Let us make an estimate for background conditions and a value of effect, i e. a change in the counting rate for two velocities of the absorber $V_{res} = 0$ and $V_{res} = \infty$ (actually $V > 5$ cm/sec) in the experiment configuration above described.

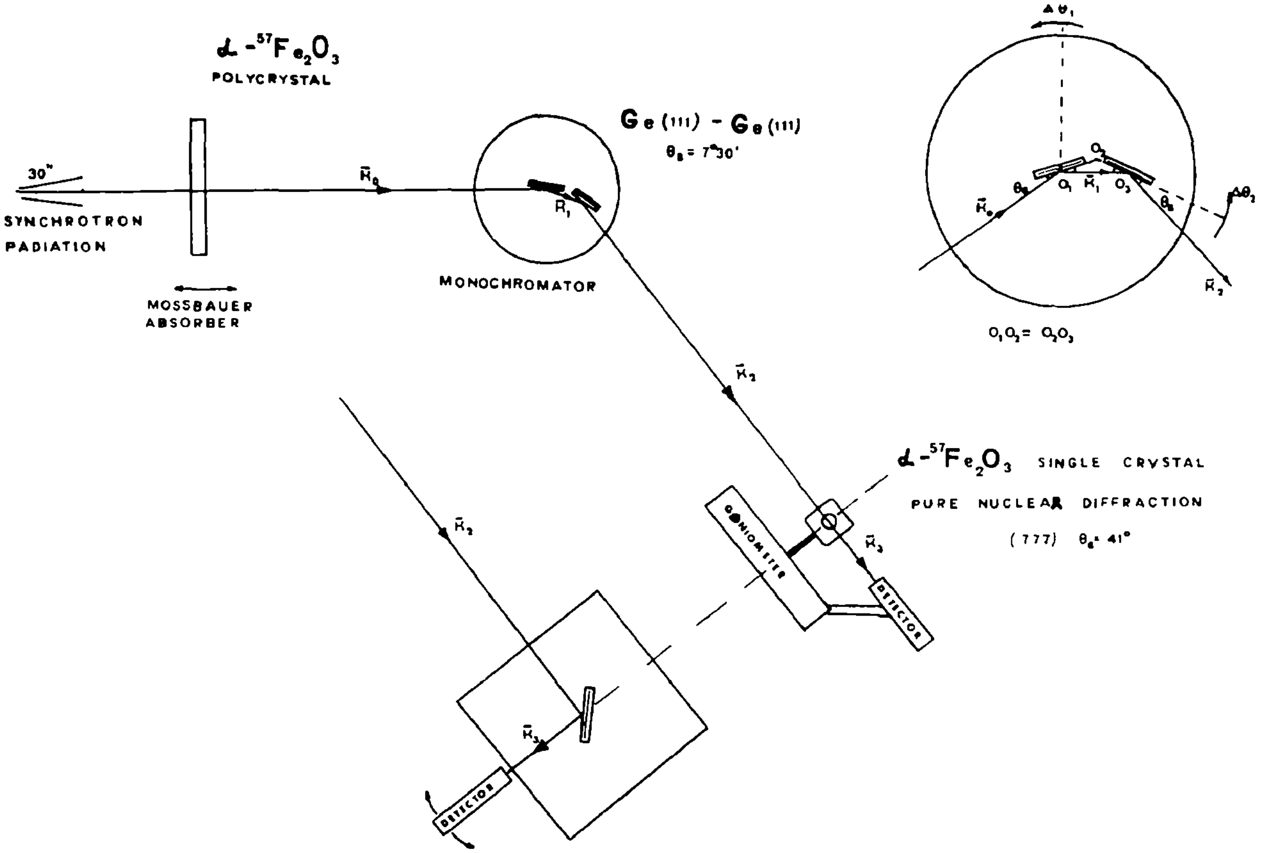

Fig 1 a) Layout of the experiment on ^{57m}Fe Mossbauer level excitation by the synchrotron radiation, b) The monochromator schematic view

The background conditions are primarily determined by the coherent electronic (but not nuclear resonance) scattering in the hematite monocrystal

$$I^s_{e\,coh} = \frac{\dot{N}^s(\lambda)\left(\frac{\Delta\lambda}{\lambda}\right)_m K^2_{Ge} K_{\alpha Fe_2O_3} \Delta\varphi K_f K_s}{Z^s q}, \tag{1}$$

where $I^s_{e\,coh}$ is the intensity on the detector resulting from the coherent background; S is the polarization index of the SR beam, $\dot{N}^s(\lambda)$ is the SR spectral intensity on the detector for a given polarization, $(\Delta\lambda/\lambda)_m = 2\times10^{-4}$ is the transmission bandwidth of the double crystal monochromator; $K_{Ge} = 0.6$ and $K_{\alpha Fe_2O_3} = 3\times10^{-2}$ are the X-ray reflectances within the rocking curves on Ge(III) and hematite $\alpha\,^{57}Fe_2O_3$ (666) monocrystals respectively (measured on a double crystal X-ray spectrometer with Mo K_α radiation), $K_f \lesssim 10^{-5}$ is the forbiddeness factor of X-ray reflection on the hematite single crystal, $\Delta\varphi = 2\times10^{-4}$ is the hematite mosaic structure factor which determines the usable SR beam horizontal angular range, K_s is the ratio of the polarization factors of the hematite monocrystal (666) to (777) reflections; Z^s is a factor accounting for the convolution of the SR intensity distribution over λ and the vertical angle θ with the rocking curves of the Ge monochromator crystals, $q = 2$ is the non-resonant attenuation in the Mossbauer absorber.

The intensity of the resonant quanta may be written as follows

$$I^s_{res} = \frac{\dot{N}^s(\lambda)\,\frac{\Delta E}{E}\,(^{57m}Fe)\,K^2_{Ge} K_{\alpha Fe_2O_3}\cdot L}{Z^s q} \tag{2}$$

here is addition to eq. (1)· I^s_{res} stands for the intensity on the detector caused by the nuclear resonant diffraction; $\Delta E/E\,(^{57m}Fe) = 3.2\times10^{-13}$ is the intrinsic width of the ^{57}Fe Mössbauer level at 14.4 keV; $L = 20$ is a factor accounting for the hyperfine splitting and for the resonant line broadening in the nuclear diffraction on the $\alpha\,^{57}Fe_2O_3$ crystal.

The estimates of $I^s_{e\,coh}$ and I^s_{res} are summarized in the table in which one can see that the effect over background ratio $\varepsilon = I_{res}/I_{e\,coh}$ (at the counting rate of ≈ 10 photons/sec) amounts to $\approx 7\%$. It is essential that the estimation was effected for a continuous measurement. The detector gating accounting of the radically different de-excitation time for quanta scattered on electrons and on nuclear may greatly improve the conditions of the experiment.

2. Monochromatization of the synchrotron radiation

The monchromator schematic view is shown in fig 1b. The 1st crystal was placed in the center of a disk rotated around SR beam stepwise by 3″. The rotation axis of the 2nd crystal was placed on the same disk The 2nd crystal was mounted so that its rotation axis, its center and the axis of the disk should stay in an isosceles triangle vertices It is important that this configuration persisted while the angle between the two crystals was varied The 2nd crystal could be rotated with respect to the 1st one by 5″ steps The monochromator design provided a total scattered SR shielding which was very important for the background considerations The adjustments of the monochromator were provided by shifts with respect to the SR beam line

Use was made of Ge monocrystals (30 mm in

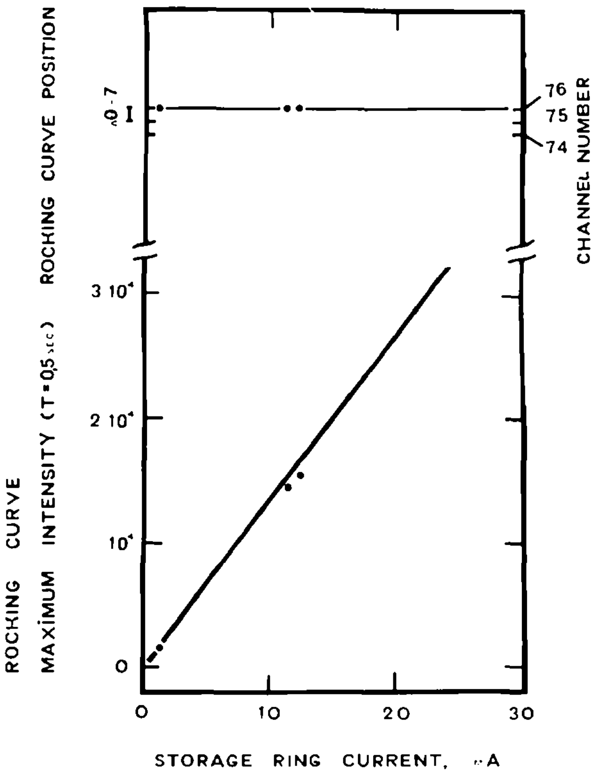

Fig 2 The SR beam thermal effect in the monochromator Above the dependence of the reflection angle for radiation outgoing the monochromator on the 3rd Ge crystal position Below the reflection intensity vs electron current in the storage ring

diameter, $\approx 300\,\mu$m thick), the rocking curve width was $\approx 15''$ (Mo K_α, $SiO_2[20\bar{2}2]$, without dispersion correction) with the reflectance of ≈ 0.6 within the rocking curve The crystals were fixed on the flat metal surface of the arm support with an oil drop

The fine adjustment of the monochromator at the needed γ energy was made with the radiation of the Mössbauer source ^{57}Co (0 2 Ci) After adjustment the monochromator was mounted on the SR beam line The transmission bandwidth of $\approx 30''$ was observed when rotating the monochromator assembly with respect to the SR beam

As the power in the direct SR beam was as high as ≈ 2 W/mm^2 the testing of the monochromator stability on the SR beam was needed To this end the beam outgoing the monochromator was analysed by a 3rd Ge crystal put in the place of the hematite crystal on the goniometer The beam went in collimated (two 0 1 mm slits spaced 1 5 m apart) The 3rd crystal rotation was driven by a synchronous motor The detector output was fed to a multichannel analyzer switched on the "scaler" operation mode Figs 2a and 2b show the slope of the rocking curve and its intensity respectively vs the electron current in the storage ring The persistence of the slope position is an evidence that there are no shift in the monochromator transmission band (the errors are evaluated from spread in the slope positions as seen in repetitive rocking curve scans at fixed current) The linear growth of the intensity indicates the persistence of the Ge crystals reflectance

Thus for the regime under which the storage ring was operated there was no thermal effect This seems to be due to the Mossbauer absorber in the SR beam acting as a thermal filter it reduces the heat flux incident the 1st monochromator crystal by an order of magnitude while only 2 times attenuating 14 4 keV radiation Besides the crystal supports are designed for an effective heat removal off the thin monochromator crystals

Utilization of greater electron currents or energies in a storage ring or of monochromators with a more narrow rocking curves may require either

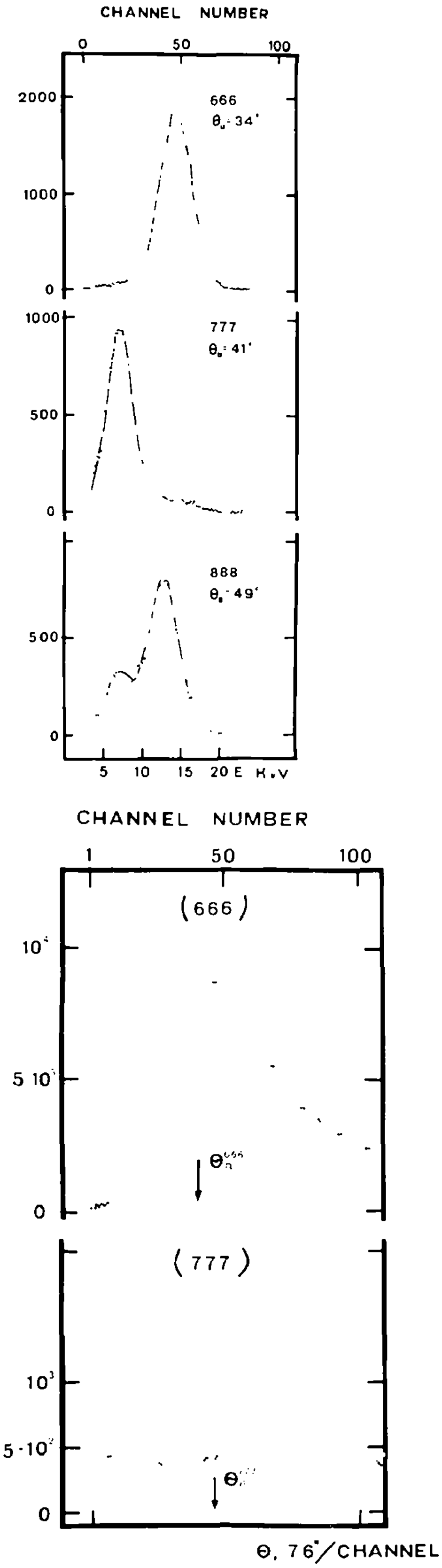

Fig 3 Reflection of the monochromatized SR beam at the hematite (see fig 1a) NaI crystal (0 1 mm thick, 17 mm in diameter) as a detector a The hematite rocking curves for the allowed (666) reflection and in the vicinity of the forbidden (777) reflection b) The energy (amplitude) spectra measured in the allowed (666) and (888) reflections and in the vicinity of the forbidden (777) one

an effective forced cooling or more elaborate filters with total external reflection (thus cutting-off the short wave radiation and at the same time suppressing the higher-order harmonics) as well as absorbers with properly chosen K-edge positions.

3. Reflection of the monochromatized synchrotron radiation at the hematite monocrystal

A hematite α-Fe_2O_3 (85% of ^{57}Fe) single crystal was used with dimensions of 7×4 mm^2, mosaic structure angle of $\approx 40''$ and X-ray reflectance of 3×10^{-2} within the rocking curve. Fig. 3 (right) shows the hematite rocking curves in the monochromatized SR beam for the allowed (666) reflection and in the vicinity of the forbidden (777) one (see the experiment schematic in fig. 1a). The angular width of the peak of 3′ matches to the horizontal divergence of the SR beam striking the hematite. Two humps on top were caused by the presence on the reflecting spot of two large grains in the hematite monocrystal. Similar curves were observed for some other allowed reflections. No peak could be seen in the vicinity of the forbidden (777) reflection.

To determine the energy spectrum of the radiation the amplitude spectra of (666), (888) reflections and of the forbidden (777) reflection vicinity were measured (see fig. 3a). The spectrum of the strong (666) reflection consisted of the single 14.4 keV peak due to the diffracted radiation at $\lambda_0 = 0.86$ Å. In the amplitude spectrum of the (888) reflection the coherent reflection was considerably suppressed (because the incident SR beam was polarized and $2\theta_B$ (888) $\simeq 98°$ which was close to 90°) therefore the hematite iron characteristic fluorescence was displayed at $E = 6.3$ keV (fig. 3b). The intensity of this isotropic radiation in the (777) reflection vicinity ($2\theta_B = 82°$) was much greater than that of the diffuse background at 14.4 keV.

Thus the background was suppressed so well in the chosen configuration that the relatively feeble fluorescence became dominant. The further background reduction (besides gating) is possible via the scintillator size diminution (as the collimated beam area of 6 mm^2 is much less than that of the utilized NaI crystal 17 mm in diameter) or via installation of low-Z filters and an analyzing crystal upstream the detector or by an amplitude discrimination. Moreover the analyzing crystal reduces the elastic diffuse background by the ratio of its acceptance angle to the solid angle of the hematite as seen from the detector. The latter measurement is also performed in our experiment.

The estimated intensities and the measured quantities corresponding to the storage ring operation parameters are summarized in table 1. One can see that the disagreement in intensity is not greater than by an order of magnitude for both allowed and forbidden reflections. Such agreement should be thought of as a satisfactory one because the estimates for intensity do not seem to be too precise.

4. Time measurements

A block diagram of electronics is presented in fig. 4. It comprises a reference pulse former, a γ-detection channel, a time to amplitude convertor and an amplitude analyzer. The electronics is realized on the basis of units described in refs. 6 and 7.

Time measurements were carried out in the configuration shown in fig. 1a. The feeble (444) reflection $\theta_B = 22°$ of hematite was used with the structure factor $F_{444} = 0.06$ related to one iron atom. In addition to the configuration of fig. 1a an analyzing crystal of pyrolitic graphite was installed downstream the hematite. We believe these conditions to simultate most closely a low counting rate and a low diffuse scattering background expected in the experiment. The instantaneous dif-

TABLE 1

The comparison of calculated and measured data for storage ring parameters being used ($E_e = 2.0$ GeV, $I_e = 10$ mA)

S	λ^s		I^s_{coh} (666) (photons/sec)		I^s_{coh} (777) (photons/sec)		I^s_{res} (777) (photons/sec)
	nonres	res	calc	exp	calc	exp	calc
σ	6	3	4.3×10^5	6.6×10^4	1.2	0.3	2×10^{-2}
π	15	7	1.4×10^4		1.8		2×10^{-4}

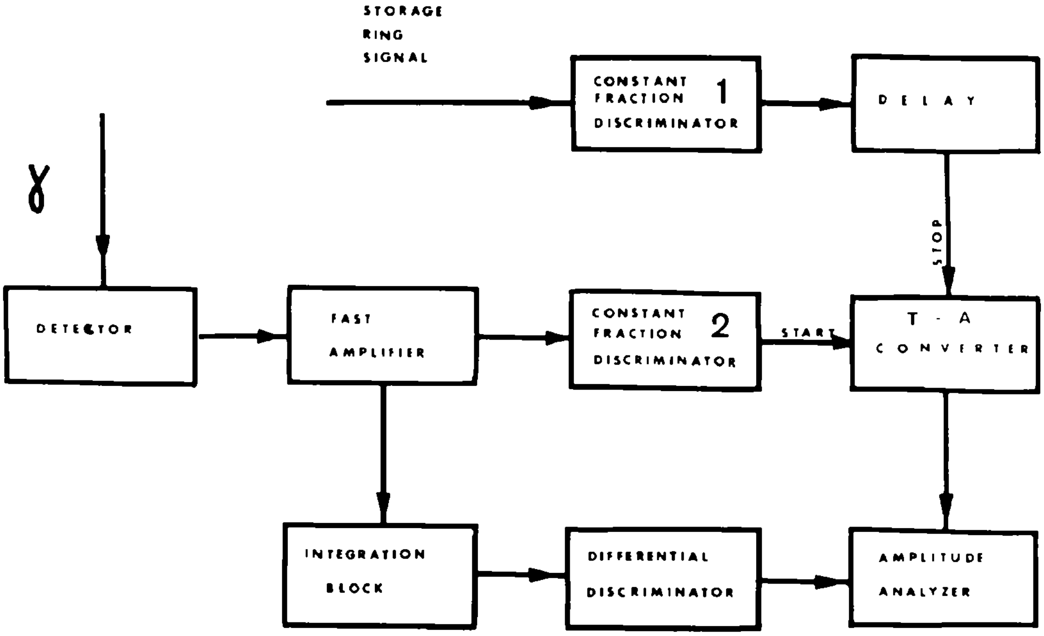

Fig 4 The block diagram of electronics employed

fraction peak and background of random coincidences of SR pulses are shown in fig 5 The peak full width at half maximum is 9 nsec. The peak to background ratio amounts to $\approx 1\ 7\times 10^3$ To subtract the electronics noise contribution a test was made at the same conditions but the scintillator was removed There was no noise (1 count per 10^3 sec) The resolution and peak-to-background ratio now achieved make it possible to measure intensities 10^3 times smaller than that of the instantaneous scattering in 25 nsec past SR flash

5. Discussion and further development

In the experiment configuration presented above the major difficulty in Mössbauer experiments with the synchrotron radiation appears to have been overcome The conditions are found at which the background resulting from the instantaneous electronic diffraction of the huge SR quantum flux is suppressed. The results obtained make it possible to excite experimentally the ^{57m}Fe Mossbauer level

The VEPP-3 storage ring operation parameters that were available ($E = 2$ GeV, $I = 30$ mA) were not high enough for these experiments to be performed within the scheduled time quota. The work is to be continued in 1978 The further improvement to enhance the resonant quanta flux

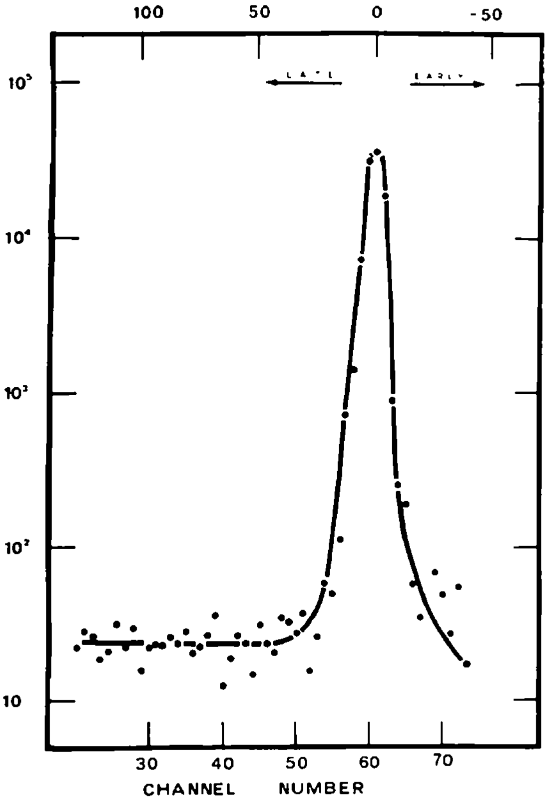

Fig 5 The time distribution of scattered quanta on the NaI detector The width of the peak is due to time resolution of the detector and electronics and equals to 9 nsec

will result from mounting of the apparatus on the SR beam line of the VEPP-4 damping magnet[8]). At a given value of current VEPP-4 at 4.5–5 GeV will yield 150 times more resonant quanta than VEPP-3 at $E = 2.2$ GeV. Later on the experiments may be continued on the SR beam line of a superconductive wiggler to be installed on VEPP-3, there the resonant quanta flux is to be 100 times higher than on the existing beam line.

It is worth mentioning that by means of the "nuclear Bragg monochromatization", of which the virtues and the problems in realization are discussed in[1]), one can separate from the SR beam a γ-ray beam with the following properties:

- provisions to perform a Mossbauer experiment on whatever nucleus;
- availability of an intense quantum flux in a narrow solid angle,
- inherent amplitude modulation of radiation with characteristic time of 0.1–1.0 nsec;
- the radiation is naturally polarized.

This quantum beam is to become a perfect Mössbauer sources for various fields where the Mössbauer effect is widely applied (e.g. Mössbauer spectroscopy, pure and applied studies on Mossbauer diffraction, investigations of hyperfine coupling in solids, etc.)

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