Synchrotron Mössbauer source

G. V. Smirnov
Russian Research Center ''Kurchatov Institute,'' 123182 Moscow, Russia

U. van Bürck
Physik-Department E15, Technische Universität München, D-85748 Garching, Germany

A. I. Chumakov, A. Q. R. Baron, and R. Rüffer
European Synchrotron Radiation Facility, BP 220, F-38043 Grenoble, France
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Mössbauer radiation was generated by synchrotron x rays for use in Mössbauer absorption and scattering spectroscopy. Like a conventional Mössbauer source, the synchrotron Mössbauer source (SMS) emits single-line radiation of about natural linewidth, but in addition the emitted radiation is fully recoilless, highly directed, and of pure linear polarization. First Mössbauer transmission spectra were measured with common resonance absorbers including stainless steel, iron, and Invar. Applications of the SMS are discussed. [S0163-1829(97)02209-1]

I. INTRODUCTION

The idea to use synchrotron radiation (SR) for studying nuclear resonances was introduced in Ref. 1. Originally it was intended to use SR in order to perform a measurement of nuclear resonance absorption or scattering in the energy domain, i.e., a Mössbauer measurement. However, a different method was developed first. The pulsed structure of photons emitted by the electrons circulating in a storage ring provides perfect conditions for time-resolved measurements of nuclear resonance scattering.[2,3] Indeed such measurements provide a direct and efficient way to study nuclear γ resonance[4,5] and its hyperfine structure.[6–8] In the last years this method has developed rapidly (for recent reviews see, e.g., Refs. 9 and 10).

Yet the time domain technique will probably not replace the traditional Mössbauer spectroscopy: the two methods are complementary, having their own features and advantages. It remains therefore a fascinating idea to filter the resonant radiation components from SR for carrying out Mössbauer measurements also at a SR station. The special features of SR (high degree of polarization, extremely small divergence, small beam cross section) will allow new developments in traditional Mössbauer spectroscopy. Moreover, it can be beneficial to combine the two complementary approaches at one and the same experimental station.

In the following we describe a method to extract single-line Mössbauer radiation out of broadband SR. A very strong suppression of the overwhelming nonresonant part of the SR must be provided by such a method, while the nuclear resonant part of the radiation should be preserved. A promising way to do this is to exploit an electronically forbidden but nuclear allowed Bragg reflection, i.e., a so-called pure nuclear reflection.[11,2]

Pure nuclear reflections rely on hyperfine interaction, and therefore they consist in general of several Mössbauer lines. However, a particular case of pure nuclear reflection was found in Ref. 12, which is well matched to the idea of extraction of a single-line nuclear component of SR. Pure nuclear reflectivity within an energy band of about the natural width of the nuclear level was obtained when an $^{57}FeBO_3$ (iron borate) single crystal was heated in an external magnetic field to the Néel temperature, T_N.

In the immediate vicinity of T_N, the magnetic hyperfine interaction nearly vanishes in the crystal while the quadrupole interaction stays constant. Since the magnetic hyperfine field and the electric-field gradient stay at a right angle in iron borate, the nuclear excited substrates are strongly mixed by spin projection. Due to destructive interference in this complex transition regime the low-energy line of the quadrupole doublet disappears in the nuclear Bragg reflection spectrum while the other line stays and forms a nearly single line of almost natural linewidth.[12,13] These results were confirmed by the time dependences of nuclear resonant Bragg reflection of SR from iron borate in approaching T_N (Ref. 14) and in the immediate vicinity of this temperature.[15]

Thus an iron borate single crystal set for a pure nuclear reflection could serve as a single-line source of Mössbauer radiation generated by SR. This synchrotron Mössbauer source (SMS) would emit linearly polarized, highly directed, recoilless γ radiation into a Bragg direction. Mössbauer measurements can be carried out in steady-state mode by moving an absorber or scatterer under study relative to the SMS and by recording the transmitted or scattered intensity in the traditional way in function of the Doppler shift.

With the advent of the third-generation SR sources, which have an extreme spectral density in the range of many Mössbauer transitions, it became possible to realize these ideas. Therefore the aim of the present work was to generate single-line Mössbauer radiation from SR and to test and demonstrate the properties of this new SMS.

II. EXPERIMENTAL SETUP

The experiment was performed at the Nuclear Resonance Beamline[16] of the European Synchrotron Radiation Facility

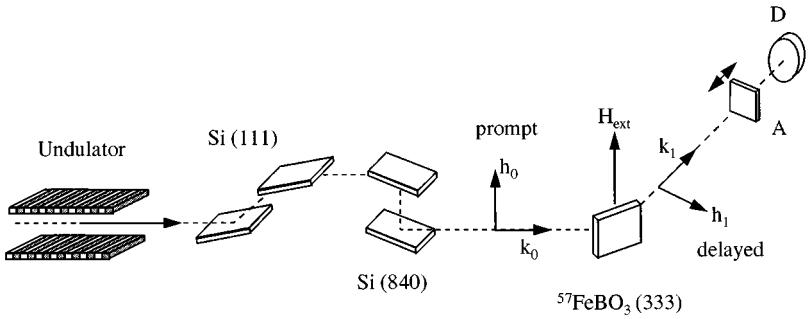

FIG. 1. Scheme of the experimental setup for the generation of synchrotron single-line Mössbauer radiation. Monochromatization and polarization were provided by a double-crystal Si(111) monochromator and a channel-cut Si(840) polarizer, respectively. The pure nuclear Bragg reflection was performed by the reflection (333) of a $^{57}FeBO_3$ single crystal in an oven (see text). The absorber under study (*A*), driven by a Mössbauer transducer, and the APD detector (*D*) are mounted along the diffracted beam.

(ESRF). The ESRF storage ring was operated in the hybrid mode where 1/3 of the ring is filled with 330 equally placed bunches and an additional single bunch is centered in the 2/3 gap. In the multibunch section the spacing between the adjacent bunches is about 3 ns while the time interval between the single bunch and the multibunch section is 910 ns.[16] Typical integral current was 130 mA with 5 mA for the single bunch.

The layout of the experimental arrangement is displayed in Fig. 1. The undulator of the beamline was tuned to provide 14.413-keV radiation in the fundamental. A Si (111) double-crystal monochromator reduced the bandwidth to 2.8 eV at the nuclear resonance energy. An asymmetric Si (840) Bragg reflection was used as a polarizer and for further monochromatization. This reflection has for 14.4-keV radiation a scattering angle of $2\Theta_B = 90.2°$. The π-polarized component of the radiation was reduced by this reflection from a level about 1% (SR is naturally well polarized in the plane of the orbit) to less than 10^{-4}%. The (840) reflection also reduced the bandwidth to ~400 meV. Thus after the (840) reflection, one has an x-ray beam centered at the nuclear resonance energy, of pure linear polarization, with divergence of about 10×35 μrad^2 (vertical×horizontal) and sizes of about 0.5×2.0 mm^2. The flux in this beam was about 300 cps per natural linewidth Γ_0 ($\Gamma_0 = 4.7 \times 10^{-9}$ neV) at 130 mA electron current.

This polarized multipulsed x-ray beam was incident on a single-crystal platelet of iron borate set for the Bragg reflection (333) in a horizontal scattering geometry [the planes (nnn) are parallel to the crystal surface]. The crystal was mounted in an oven where it could be heated up to the Néel temperature (75.35 °C), and an external magnetic field, $H_{\rm ext}$, of about 10 mT was applied along the crystal to magnetize it parallel to the vertical axis. (A simple oven was designed for the Bragg reflection geometry having a platelet homogeneous heater and adjustable magnetic poles as construction elements. The incident and reflected beams pass through Be windows. The temperature is controlled by a Pt-resistor sensor.) The hyperfine magnetic fields are normal to $H_{\rm ext}$ and lie in the crystal plane. The nuclei in the target excited by the short SR radiation pulses respond with a delay which is characterized by the excited-state lifetime. The energy bandwidth of the reemitted radiation was given primarily by the width of the nuclear resonance in the crystal under the conditions of dynamical diffraction.

The crucial question of the present experiment was whether a single reflection of the iron borate crystal would be sufficient to suppress the nonresonant SR to such an extent that Mössbauer experiments can be performed *without any time gating*.

III. MEASUREMENTS, RESULTS, AND DISCUSSION

The energy dependence of the reflectivity of the iron borate crystal was measured beforehand at a Mössbauer diffractometer in the Russian Research Center ''Kurchatov Institute.'' In Fig. 2 the Mössbauer (333) reflection spectrum of the crystal at T_N and in a weak magnetic field is displayed (spectrum B) together with a Mössbauer absorption spectrum taken for calibration using a thin standard absorber of sodium nitroprusside (spectrum A). The Mössbauer source linewidth was determined from the fit of the spectrum (*A*): $\Gamma_s = 2.0\ \Gamma_0$. Using this width the spectrum (B) was fitted with a Lorentzian line of width $1.3\ \Gamma_0$. This value characterizes the energy width of the nuclear reflection from the crystal in case of a noncollimated incident beam. Another important characteristic provided by this measurement is the position of the resonance line. Figure 2 shows that it coincides very well with one of the resonance lines in sodium nitroprusside.

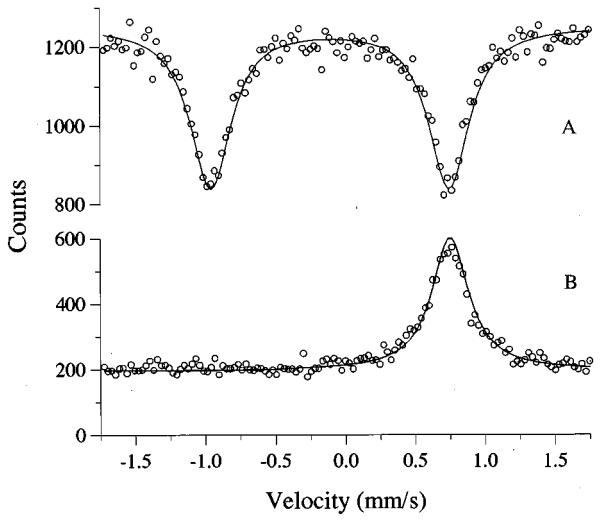

FIG. 2. (*A*) Mössbauer absorption spectrum of resonant γ-radiation from a ^{57}Co (Cr) source of a thin standard absorber of sodium nitroprusside. The solid line is a fit with the transmission integral. (*B*) Mössbauer diffraction spectrum of the same radiation off a $^{57}FeBO_3$ crystal at the Néel temperature [(333) Bragg reflection]. A magnetic field of 10 mT was applied along the crystal surface normal to the scattering plane. The solid line is a fit with a Lorentzian line.

In the adjustment of the iron borate crystal at the SR beam line it was important to get the highest suppression of the electronic scattering from the crystal. Although the (333) reflection in iron borate is electronically forbidden, there are so-called Umweg reflections, where the combination of several allowed reflections provides scattering into the direction of a forbidden reflection. They appear at certain azimuthal angular positions while turning the crystal platelet around the reciprocal-lattice vector of the reflection. The spectrum of these reflections was measured carefully. One of the intense Umweg reflected beams was used to define the optical axis downstream of the iron borate crystal. The Mössbauer driving unit with the absorber and the detector were adjusted on this axis. For the actual experiment an azimuthal angular position was selected as far as possible from all Umweg reflections.

Using the single SR pulse opposite to the 1/3 fill we examined the time dependence of the (333) reflection from the $^{57}FeBO_3$ single crystal. A fast operating avalanche photodiode detector was employed.[17] No time gating was applied. The time dependence showed a weak pulse of prompt radiation and an extended distribution of delayed radiation related to electronic and nuclear scattering, respectively. At room temperature the intensity of the prompt part was only few percent of the delayed intensity. This contribution appeared to be dependent on the crystal temperature and dropped down to approximately 1% of the total reflected intensity while heating the crystal up to T_N. Without discussing this behavior further, we only mention that the Umweg reflection pattern was strongly dependent on temperature and this could be a reason for the intensity change of the prompt radiation. Another reason might be a weak, variable contribution of magnetic x-ray scattering. At the Néel temperature, finally, the electronic reflectivity of the (333) reflection of iron borate reached the extremely low value of about 10^{-10}.

For the Mössbauer-type measurements, the full current stored in the ring was used. Several temperature points near the Néel transition were tested for choosing the working regime of the SMS. When the Néel temperature is approached, the initial four-line reflection spectrum collapses into a single pseudoline,[12–15] but at the same time the nuclear reflectivity decreases. In a compromise between intensity and linewidth, that temperature was chosen where the resonance linewidth was not decreasing any more. The linewidth was measured with a 1-μm stainless-steel (SS) absorber enriched in ^{57}Fe to 95%. The reflected intensity at this temperature was about $\approx$100 cps with a 1% contribution due to electronic scattering. The quantities are given for an integral electron current of 130 mA stored in the ring. It is worth noting that a similar radiation flux into a solid angle of 10×35 μrad^2 would require a radioactive Mössbauer source of about 1000 Ci of ^{57}Co.

To illustrate the properties of the SMS we performed measurements using common nuclear resonant absorbers: SS, iron, and an Invar alloy (all absorbers were enriched in ^{57}Fe up to 95%). Single-line spectra with a resonance effect of $\varepsilon=70$ and 86% $[\varepsilon=100\%(I_\infty-I_r)/I_\infty]$ were observed for SS foils of 1 and 10 μm thicknesses, respectively, yielding a Lamb-Mössbauer factor of the SMS equal unity.

In order to demonstrate the high degree of polarization of the SMS, Mössbauer spectra of a thin iron foil were measured (see Fig. 3). Spectrum (*A*) was taken with the foil nonmagnetized. The characteristic pattern of six lines was developed in this case. Spectra (*B*) and (*C*) were measured when the foil was magnetized in an external magnetic field of 60 mT at $H_{\rm ext}$ perpendicular and parallel to the magnetic polarization vector of the radiation, respectively. Within the statistics, only the four or two resonances related to the nuclear $\Delta m=\pm1$ and $\Delta m=0$ transitions, respectively, were observed. This result is a clear evidence of the polarization state of the source radiation: the radiation is of pure linear polarization. A comparably high degree of polarization for radiation from a Mössbauer source has been obtained only very recently.[18]

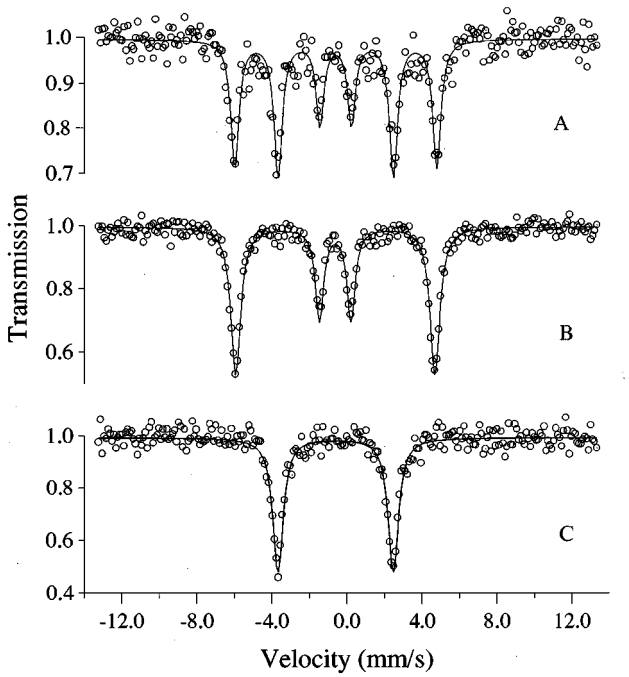

FIG. 3. Mössbauer transmission spectra of an ^{57}Fe foil of 1.3 μm thickness measured with radiation from the SMS. (*A*) foil nonmagnetized; (*B*) foil magnetized: $H_{\rm ext}\perp h_1$ and $H_{\rm ext}\perp k_1$; and (*C*) foil magnetized: $H_{\rm ext}\| h_1$. The solid lines are fits using the transmission integral with 100% linearly polarized and fully recoilless radiation.

The advantage of the high polarization was demonstrated by measuring a Mössbauer spectrum of an Invar alloy ($Fe_{65}Ni_{35}$). There is a complicated inhomogeneous distribution of the magnetic hyperfine field strengths in such alloys.[19] By using polarized radiation, one is able to select either two or four lines of the six line spectrum and thus to simplify essentially the observation of the field distribution. In Fig. 4 the Mössbauer spectrum of a thin Invar foil magnetized along the magnetic polarization vector of the γ radiation is shown (spectrum *B*) and compared with the analogous spectrum of the thin iron foil (spectrum *A*). An asymmetric distribution of the hyperfine magnetic field strengths is directly seen from the data. Note that with an unpolarized source, spectra of similar simplicity can only be obtained indirectly.[20]

Additional source parameters were obtained from the fit with the transmission integral in case of the Mössbauer spectrum of the magnetized iron; see Fig. 4, curve *A*. The fit yields a source recoilless factor of unity (1% of nonresonant

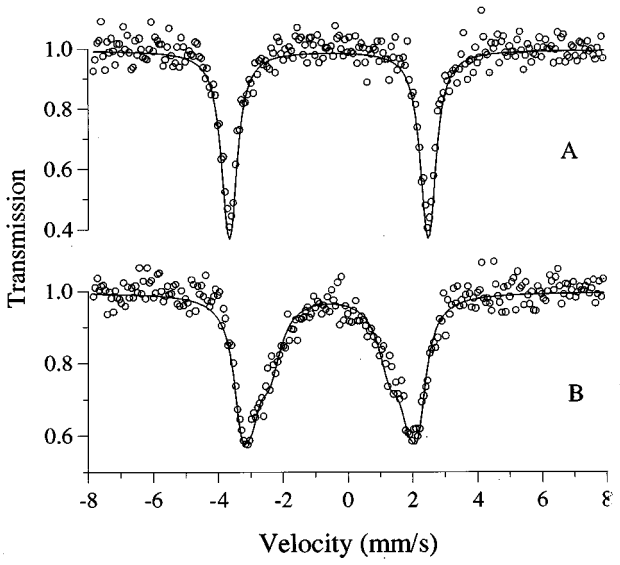

FIG. 4. Mössbauer transmission spectra of thin foils of iron metal and Invar alloy measured with radiation from the SMS. (*A*) iron foil of 1.3 μm thickness, magnetized: $H_{\rm ext}\|h_1$; (*B*) Invar foil of 1 μm thickness, magnetized: $H_{\rm ext}\|h_1$. The solid lines are fits using the transmission integral. The field strength distribution is simulated by three Lorentzians.

background was taken into account) and a source linewidth of 2.9 Γ_0. Thus the Mössbauer radiation filtered from SR had a linewidth almost twice broader than expected from the previous measurement of the reflection using Mössbauer radiation, Fig. 2, curve *B*. The reason for this result is the collective nature of the nuclear excitation in our source.[4,21] The resonance width in coherent nuclear resonance scattering is determined by the number of nuclei contributing in phase to the Bragg reflection, and thus by the deviation from the Bragg reflection condition.[22,23] In the present case the divergence of the incident SR beam was comparable to the intrinsic angular width of the Bragg reflection. Therefore the conditions for a proper phasing of a large nuclear ensemble and hence for a strong broadening of the resonance were fulfilled. Note that a narrower source linewidth could be obtained by increasing the incident beam divergence or by shifting the direction of incidence slightly out of the exact Bragg position.

IV. CONCLUSION

A source of coherent Mössbauer radiation from ^{57}Fe nuclei has been developed. It emits single-line radiation, which is highly directed, fully polarized and fully recoilless. The radiation is generated by the pure nuclear diffraction of SR from a single crystal of iron borate at Néel temperature. The single-line character of the source is due to the collapse of the nuclear Bragg reflection spectrum at Néel temperature in the presence of a small magnetic field. High directionality and polarization are defined by the undulator and the polarizer, respectively. Recoil-less emission is provided by the pure nuclear Bragg scattering, where an extremely high suppression of the electronic scattering by a factor of about 10^{-10} was achieved. In the working regime the energy bandwidth of the resonant radiation was about 15 neV, the beam intensity 100 s^{-1}, the beam divergence 10×35 μrad^2, and the beam cross section 0.5×2.0 mm^2 (vertical$\times$horizontal). Without changing the experimental optics, one could gain about a factor of 2 in intensity by using a more efficient detector. Also, if the subsequent experiment could tolerate about 1%–2% contamination of the other linear polarization, another factor of 2 should be gained from the replacement of the Si(840) polarizing crystal by a standard nested monochromator.[16]

One aspect of greatest practical importance is that the proposed method allows one to use a SR storage ring for Mössbauer experiments in any fill mode. In this respect, the only alternative is a method employing crossed polarizers for the suppression of the prompt intensity.[24,25] However, this method will always yield a multiline source radiation spectrum, and the suppression of the prompt radiation is less.

There is still a problem with a possible Doppler shift of the SMS. For many Mössbauer experiments it is more convenient or even required to move the source rather than the absorber or scatterer. One possibility would be to move an especially light oven containing the borate crystal in the way presented in Ref. 26. Another possibility is to move an intermediate nonresonant Bragg reflector.[27,28]

The combination of the special properties of the new SMS opens wide perspectives for its application both in transmission and diffraction Mössbauer experiments. The high degree of polarization can be employed for studies of all kinds of static distributions of magnetic moments in materials (see the Invar example), e.g., also of magnetic domain structures in crystals, of moments in metal glasses or in magnetic tapes, etc. It also gives the possibility to excite selectively a part of the nuclear target or a subsystem characterized by a particular hyperfine environment and to study the hyperfine interaction in this part. The ideal resonance purity of the source radiation (100% recoilfree) allows one to measure directly Lamb-Mössbauer factors. The directionality of the source radiation provides the possibility to investigate anisotropic diffusion. Another group of experiments could benefit from the very small beam cross section, which allows the use of small samples as required, e.g., for high-pressure studies or the investigation of small parts of the sample in case of sample or temperature inhomogeneities.

Special applications are opened in the field of nuclear resonant dynamical diffraction of γ radiation, where the extreme brilliance of the new source is of greatest importance. A review of the phenomena in this field can be found for instance in Ref. 29. For diffraction studies it is of interest that the new SMS has an extremely low divergence in both dimensions perpendicular to the beam, thus allowing for instance the study of *n*-beam multiple resonant diffraction.[30]

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