Europhys. Lett., 22 (4), pp. 305-310 (1993)

Nuclear Bragg Diffraction of Synchrotron Radiation in the Presence of Acoustic Vibrations.

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(received 11 December 1992; accepted 4 March 1993)

PACS. 76.80 - Mössbauer effect; other gamma-ray spectroscopy.
PACS. 63.20 - Phonons and vibrations in crystal lattices.
PACS. 78.70 - Other interactions of matter with particles and radiation.

Abstract. – The time evolution of nuclear Bragg diffraction of 14.4 keV synchrotron radiation in a 57 FeBO₃ crystal was studied when the crystal was excited to magnetoelastic vibrations by a r.f. field of 2.93 MHz. The vibrations did not modify the time evolution of the diffraction. This result is due to the fact that after the excitation by the SR pulse the nuclear vibrations cause only a phase modulation of the re-emitted radiation. Energy spectra, by contrast, are extremely sensitive to vibrations. Thus the present case is an instructive example of complementarity between energy and time domain Mössbauer spectroscopy. Time domain spectroscopy, in particular, will allow to reveal phenomena, which are blurred in the energy spectra by vibrations.

Introduction. – Mössbauer spectra are sensitive to nuclear motions. Harmonic vibrations of frequency f generate satellite lines in the spectra [1], which are spaced apart by multiples of $2\pi f\hbar$. The natural linewidth Γ_0 of the nuclear resonance and the wavelength λ_0 of the γ -radiation determine the scale of sensitivity of the Mössbauer spectra to vibrations. Vibrations with frequency $f_0 = \Gamma_0/2\pi\hbar$ and with amplitude $u_0 = \lambda_0$ effectively disturb Mössbauer spectra. For ⁵⁷Fe these values are $f_0 = 1.125$ MHz and $u_0 = 0.861$ Å. The creation of sidebands (or of line broadening due to unresolved sidebands) by nuclear vibration is a well-known fact in Mössbauer emission and absorption spectroscopy (for a review see, e.g., [2,3]). Recently also Mössbauer diffraction spectra were studied under the influence of magnetoelastic vibrations excited by an external r.f. field [4]. It was found that the vibrations produced a strong broadening of the lines of the diffraction spectrum to such an extent that the spectrum formed a region of high reflectivity about $(100 \div 200)\Gamma_0$ wide.

One often associates a broadening of the resonance line in an ensemble of resonators with an acceleration of the coherent response of this ensemble. Such a correspondence was observed, *e.g.*, in the nuclear resonant Bragg scattering of Mössbauer radiation [5,6] and of synchrotron radiation (SR) [7] in single crystals. The goal of the present paper was to clarify



Fig. 1. – Scheme of the experimental set-up. The SR pulse from DORIS III was monochromatized by a double Si-reflection and Bragg-reflected by a 57 FeBO₃ single crystal. A constant magnetic field H_{c1} and a magnetic r.f. field $H_{r.f.}$ were applied to the crystal. The 57 FeBO₃ resonance absorber and the SBA analyser, shown in the dashed box, were installed only in the control experiments.

whether the strong broadening of the Mössbauer diffraction spectra observed in [4] will also be connected with a speed-up in the time evolution of the nuclear resonant diffraction. For this purpose the effect of nuclear vibrations on the time evolution of nuclear resonant diffraction was studied.

Experiments and results. – The experiment was performed at HASYLAB on the high-resolution spectrometer [8]. The experimental set-up is shown in fig. 1. Radiation of 14.4 keV was filtered from the SR beam by a double Si(111) reflection resulting in a beam divergence of 20 µrad. The 14.4 keV radiation pulse excited the ⁵⁷Fe nuclei of a ⁵⁷FeBO₃ crystal set for the pure nuclear Bragg reflection (333) at a Bragg angle of $\Theta_{\rm B} = 15.4^{\circ}$. A perfect single-crystal platelet of ⁵⁷FeBO₃ of size (6 × 10) mm² and of thickness 55 µm with (111) surface [6] was employed. A constant magnetic field $H_{\rm cl} = 5$ Oe was applied perpendicular to the scattering plane in order to provide a definite magnetization of the crystal. Nuclear vibrations were excited by an external magnetic r.f. field $H_{\rm r.f.}$ of frequency f = 2.93 MHz and of variable amplitude ((0 ÷ 16) Oe), which was applied in the (111) plane perpendicular to the constant field. Plate modes of acoustic vibrations with wavelength $\Lambda \ge 1$ mm were excited this way [9]. The vibrations were polarized predominantly in the (111) plane of the crystal parallel to $H_{\rm r.f.}$ [4]. Therefore the nuclear response in low-order (n.n.n.) Bragg reflections should be influenced most effectively by these vibrations.

Figure 2 shows the measured time evolution of nuclear diffraction in the presence of acoustic vibrations excited by r.f. fields of different amplitudes. The time spectra are obviously insensitive to the nuclear vibrations. It was possible to fit all three spectra using the theory for a non-vibrating crystal [10]. A slight modification of the quantum beat period, which can be attributed to heating of the crystal [11] by a few degrees due to the r.f. field, was taken into account in the fit by a decrease of the hyperfine field (0.5% in fig. 2b) and 2.1% in fig. 2c)). The fit of the measured spectra proves the complete insensitivity of the time spectra to the excited nuclear vibrations.

In order to demonstrate that nuclear vibrations were actually excited, additional measurements were performed. Firstly, a resonance absorber was installed behind the diffracting ⁵⁷FeBO₃ crystal. It was another ⁵⁷FeBO₃ crystal platelet of 15 µm thickness, which was magnetized by a constant magnetic field H_{c2} perpendicular to the scattering plane. Figure 3*a*) shows the time spectrum obtained when the diffracting crystal was at rest. Comparison with fig. 2*a*) shows that the resonance absorber causes pronounced modifications of the time spectrum. Figure 3*b*) shows the spectrum, when a r.f. field of $H_{r.f.} = 6.8$ Oe was applied to the



Fig. 2. – Time spectra of the pure nuclear Bragg reflection ${}^{57}\text{FeBO}_3(333)$, measured at different amplitudes of the 2.93 MHz r.f. field $H_{r.f.}$: 0 (a)), 6.8 Oe (b)) and 15.8 Oe (c)). The solid lines are the theoretical simulations assuming the crystal at rest in all three cases.

diffracting crystal. The spectrum now resembles much more the one of fig. 2a). Obviously the majority of the reflected radiation passes through the resonance absorber. This is possible only if the spectrum of the reflected radiation is strongly broadened. Both time spectra of fig. 3 were fitted using the theory for nuclear resonant diffraction in the presence of acoustic vibrations [12]. The parameters used were the same as in the case of the spectra of fig. 2. For the fit of the spectrum of fig. 3b) a Rayleigh distribution of the vibration amplitudes was assumed [3].

In the second control experiment the energy distribution of the reflected radiation was measured by conventional Mössbauer analysis using a standard black absorber (SBA) [13]. When recording the energy spectra the large prompt background was eliminated by a 10 ns gate. The energy spectra measured at different values of the r.f. field amplitude are shown in fig. 4. Increasing the r.f. amplitude causes a strong broadening of the resonance lines. The theoretical spectra used for the fitting of the energy spectra of fig. 4 were computed using the theory [12]. Both control experiments prove that nuclear vibrations were indeed excited in the crystal by the applied r.f. field.

Interpretation and discussion. – In the following a short explanation of the observed insensitivity of the diffraction time spectra is presented. The full theoretical treatment will be given elsewhere [12].

Consider the resonant scattering of SR by an individual nucleus which has a transition energy E_0 . According to the classical treatment a vector-potential component $A(\mathbf{r}, t)$ of the



Fig. 3. – Time spectra of the radiation reflected by the $^{57}{\rm FeBO}_3$ crystal and transmitted through the $^{57}{\rm FeBO}_3$ resonance absorber, measured at different r.f. field amplitudes $H_{\rm r.f.}$: 0 (a)) and 6.8 Oe (b)). The solid lines are the theoretical simulations on the basis of the theory [10,12]. The most probable value a_R of the projection of the vibration amplitude on the beam direction was (0.8 \pm 0.1) Å (obtained from the fitting).

radiation emitted by a nucleus is given by

$$A(\mathbf{r}, t) = \frac{1}{cr} \exp\left[ik\rho(t)\right] j(\mathbf{k}, t) \,. \tag{1}$$

Here $\rho(t) = |\mathbf{r} - \mathbf{u}(t)| \cong \mathbf{r} - \mathbf{k}\mathbf{u}(t)/k$ is the distance between the points of observation and emission, $\mathbf{u}(t)$ is the displacement of the nucleus from the origin, and $\mathbf{k} = (E_0/c\hbar)(\mathbf{r}/r)$ is the wave vector of the emitted radiation; $j(\mathbf{k}, t)$ is the correspondent component of the current density vector of the nucleus.

In the present case of nuclear resonant excitation by a SR pulse, the excitation time is extremely short both with respect to the period of the nuclear vibrations $2\pi/\Omega$ (about 300 ns) and to the nuclear lifetime $\tau_0 = \hbar/\Gamma_0$ ($\tau_0 = 141.1$ ns for ⁵⁷Fe). Therefore the excitation can be considered as instantaneous and the nuclear current density is as usual

$$j(\mathbf{k}, t) \propto \exp\left[-\frac{i}{\hbar}\left(E_0 - i\frac{\Gamma_0}{2}\right)t\right]\Theta(t).$$
 (2)

Inserting eq. (2) into eq. (1), one obtains

$$A(\mathbf{r}, t) \propto \frac{1}{cr} \exp\left[-\frac{i}{\hbar} \left(E_0 - i\frac{\Gamma_0}{2}\right)t - i\mathbf{k}\mathbf{u}(t)\right] \Theta(t).$$
(3)

This case is analogous to the situation where a Mössbauer source is exposed to ultrasound vibrations (see, *e.g.*, [14]). The radiation emitted by a vibrated nucleus is phase modulated, due to the changed distance between the detector and the emitter. The phase, however, is lost in the process of detection, and the time spectra $I(\mathbf{r}, t) \propto |A(\mathbf{r}, t)|^2$ do not reveal the vibration:

$$I(t) \propto \exp\left[-\frac{\Gamma_0 t}{\hbar}\right] \Theta(t).$$
 (4)



Fig. 4. – Energy spectra of the reflected radiation obtained by means of the SBA analyser measured at different r.f. field amplitudes $H_{\rm r.f.}$: 0 (a)), 6.8 Oe (b)) and 15.8 Oe (c)). The prompt background was eliminated by a 10 ns gate. Note that time gating leads to modifications seen in the wings [15]. The solid line are fits computed on the basis of the theory [12]. The most probable values a_R of the projection of the vibration amplitudes on the beam direction were (0.7 ± 0.1) Å in b) and (1.4 ± 0.1) Å in c) (obtained from the fitting).

The corresponding energy spectrum I(E) of the emitted radiation is obtained via the Fourier transformation of $A(\mathbf{r}, t)$ into $A(\mathbf{r}, E)$. In the special case of harmonic vibrations $u(t) = a \cos(\Omega t + \phi)$ the transformation leads to the expression (see, e.g., [14])

$$I(E) \propto \sum_{n = 0; \pm 1; \dots} \frac{J_n^2(ka)}{(E - E_0 - n\hbar\Omega)^2 + (\Gamma_0/2)^2} \,.$$
(5)

This example shows that by contrast to the time spectra the effect of vibrations is clearly seen in the energy spectra by the appearance of satellite lines at energies $E_0 + n\hbar\Omega$.

In the experiment the collective response of many nuclei was measured. The acoustic vibrations excited in the experiment had wavelengths in the order of 1 mm [9]. In this case the Bragg-reflected intensity can be considered as the incoherent sum over the contributions from different crystal domains, in each of which all nuclei vibrate in unison [4, 12]. This model was successfully employed to fit the energy spectra of resonant diffraction in the presence of acoustic vibrations [4]. It should therefore also work for the time spectra. Then the wave field emerging from a vibrated crystal domain is that of the domain at rest, times a common phase modulation factor due to vibration in unison. The loss of this phase factor in the process of detection explains the observed vibration insensitivity of the time spectra.

The picture should change completely in the case of non-unison vibrations of nuclei in volumes

comparable in size with the diffracting volumes (*e.g.*, coherence volume [16] or Fresnel zones). In this case those nuclei, which contribute to the diffractional signal, have different vibration phases. Then the time response will become sensitive to the acoustic disturbances.

Conclusion. – Time spectra of nuclear Bragg diffraction of synchrotron radiation by a perfect ⁵⁷FeBO₃ single crystal were studied in the presence of acoustic vibrations excited by a r.f. field. The experiment revealed that the time evolution of the resonant re-emission by the nuclei was not affected by the vibrations. This result was explained by the fact that in the present conditions the radiation emitted by a vibrated crystal was only phase modulated.

The insensitivity of the time spectra with respect to nuclear vibrations in unison is in strong contrast to the well-known vibration sensitivity of the energy spectra. The present experiment is an instructive example of the fact that energy domain and time domain Mössbauer spectroscopy can yield complementary information.

The observed insensitivity could be exploited in such cases, where an effect of interest is blurred in the energy spectrum by sidebands or line broadening due to nuclear vibrations. Time domain spectroscopy could provide in this case the possibility to distinguish between vibrational and other origins of such disturbances. Special examples would be the radiofrequency perturbation of excited nuclear levels, *e.g.*, excited-state NMR, and studies of nuclear γ -resonance in long-lived isomers.

This work has been funded by the Bundesministerium für Forschung und Technologie under contracts No. 05 405 AYB7, KA2TUM/6 and KfK/IB-6.4 and by the RSC Kurchatov Institute.

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