Observation of Nuclear Resonant Scattering Accompanied by Phonon Excitation Using Synchrotron Radiation

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The phonon energy spectrum of a polycrystalline α-Fe foil was observed via nuclear resonant scattering of synchrotron radiation for the first time. The measured spectrum is in good agreement with earlier neutron inelastic scattering data. One of the advantageous features of this method is that excitation of only a specific element is possible. Our results show that this method is applicable to the study of lattice dynamics and open a new field of nuclear resonant scattering spectroscopy.

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Recoilless nuclear resonance absorption of gamma quanta is well known as the Mössbauer effect, which is applied in various fields of science [1]. Before the discovery of the Mössbauer effect, Moon first observed nuclear resonant excitation with recoil using an ultracentrifuge [2]. On the other hand, it has been suggested that the information about phonon spectra can be obtained with a Mössbauer radioisotope source by the nuclear resonance excitation with recoil [3]. However, it is extremely difficult to scan a phonon spectrum with a single line of the Mössbauer source, because the spread of a phonon spectrum (the order of 10 meV) is much wider than even very wide linewidth of the source (the order of 10 μeV). Thus, until today, such experiments could not be achieved. However, by using synchrotron radiation (SR) with bandwidth of the order of meV, it is possible to measure the phonon energy spectrum. The first experiment of nuclear resonant excitation with SR was performed in 1978 by Cohen, Miller, and West [4]. They observed the incoherently emitted electrons in the decay of the 14.4 keV level of the ^{57}Fe nucleus. Subsequently, several experimental studies of the coherent nuclear resonant scattering have been done using SR [5–9]. Hastings *et al.* reported nuclear resonant scattering of SR in the presence of electronic charge scattering with a monochromator which provides an energy resolution of 5 meV [7]. Bergmann, Hastings, and Siddons observed incoherent nuclear resonant scattering of SR from an ^{57}Fe foil [10]. Now we have successfully measured the phonon energy spectrum with SR. In this Letter, we report the first observation of the phonon energy spectrum of a polycrystalline α-Fe foil. Furthermore, we discuss the interpretation of the result and the advantageous feature of this method.

The experiments were performed at the NE3 undulator beam line of the accumulation ring (AR) of TRISTAN at the National Laboratory for High Energy Physics (KEK). During our measurements, the AR has been operated in a single-bunch mode giving 1.2 μs pulse intervals at 20–40 mA and 6.5 GeV. The experimental setup is shown in Fig. 1. The polycrystalline α-Fe foil (1.91 mg/cm^2, enriched in ^{57}Fe to 95.45%) was folded into eight; this folded foil was used as a specimen, and the illuminated area of the foil was about 0.3×10 mm^2. High brilliance radiation was provided by the in-vacuum-type undulator [11]. Two monochromators were included in our x-ray optics as shown in Fig. 1. First, a double-crystal Si(111) premonochromator was used to handle the high heat load of the undulator radiation. Second, a 4-bounce precision monochromator consisting of an asymmetrical Si(4 2 2) channel-cut crystal and a symmetrical Si(12 2 2) channel-cut crystal produced an incident beam with a 6.7 meV bandwidth at the 14.413 keV nuclear resonance in ^{57}Fe; the peak energy of the beam was tuned within the relative accuracy of 0.6 meV. The intensity of the incident beam was monitored with an ionization chamber and a beam flux monitor. It should be noted that the nuclear resonant scattering is divided into nuclear-elastic events and into nuclear-inelastic (spin-flip) events as summarized in Ref. [12]. The energy difference of the two processes is too small (typically $<\mu$eV for ^{57}Fe) to distinguish with our monochromator. However, the detection of such nuclear-inelastic events is not our purpose, and we will concentrate on the inelastic events due to recoil. The nuclear resonant scattering was observed with an avalanche photodiode (APD) detector [13], which detected not only directly emitted 14.413 keV photons but also 6.4 keV Fe-$K\alpha$ photons which follow internal conversion processes. The detecting area of the APD detector is 3 mm in diameter. All the measurements

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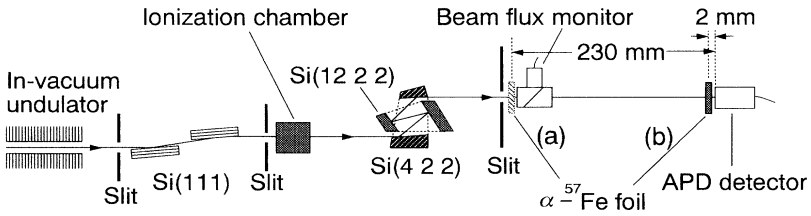

FIG. 1. Schematic drawing of the experimental setup used. The 4-bounce monochromator was used to produce a probe beam with a 6.7 meV bandwidth. The polycrystalline α-^{57}Fe foil was used as a specimen, which was placed at the position (a) 230 or (b) 2 mm. The nuclear resonant scattering was observed with an avalanche photodiode detector.

were performed at 299 K. The specimen, of which the surface was kept perpendicular to the incident beam, was placed between the APD detector and the precision monochromator. The distance between the specimen and the detecting plane of the detector was (a) 230 or (b) 2 mm. When the foil is placed at (a), about 1×10^{-5} of the isotropic radiation from the illuminated area of the foil is incident on the detection area of the detector; this setup is similar to that of the earlier experiments [8,9] in which only coherent forward scattering was observed. On the other hand, about 5×10^{-2} of the isotropic radiation is incident on the detection area when the foil is placed at (b); thus both coherent scattering and incoherent scattering are expected to be observed. We can discriminate between scattering due to the nuclear resonant excitation and nonresonant electronic scattering by using the relatively long lifetime of the nuclear excited state and the pulsed nature of the SR [14]. Therefore, we have measured the energy spectra of the nuclear resonant excitation as a function of the incident x-ray energy by counting the number of emitted photons over the time interval between 202 and 382 ns after each synchrotron pulse. The reason for choosing the time interval is to avoid partially filled ($\leq 10^{-5}$) electron bunches; they were, owing to lack of purity in the single-bunch structure, around 20, 140, and 180 ns after the main electron bunch. The incident photon energy E was controlled by the precision monochromator.

Measured energy spectra are shown as closed circles in Fig. 2. The energy of the first excited state of the ^{57}Fe nucleus is denoted as E_0 in Fig. 2. In the case of position (a), only the recoilless peak is observed. On the contrary, in the case of position (b), nuclear resonant inelastic scattering (both sides of the recoilless peak) is clearly observed. In our experimental setup, the observed ratio of the recoilless scattering intensity to the total intensity depends on the distance between the detector and the sample as observed in Fig. 2; thus, the ratio does not equal the recoilless fraction. This is because coherent forward scattering of which the intensity is almost independent of the distance is contained in the recoilless part, whereas such forward scattering is not contained in the inelastic part. Furthermore, the ratio depends on the time interval of the integrating of delayed counts, because the time response of the recoilless scattering is different from that of the inelastic scattering. They are shown in Fig. 3 with (a) the probe beam energy set on recoilless resonance and (b) the energy tuned to +25 meV from recoilless resonance. In the recoilless resonance (a), the decay time is sped up, as compared with that of an isolated ^{57}Fe nucleus in an excited state with a mean lifetime of 141 ns. The speedup is due to the collective nature of the coherent interaction of the radiation with the system of nuclei [15–17]. On the other hand, in the inelastic scattering (b), the time dependence is almost the exponential characteristic of an isolated ^{57}Fe nucleus. In spectrum (b), the peaks observed around 20, 140, and 180 ns are partially filled

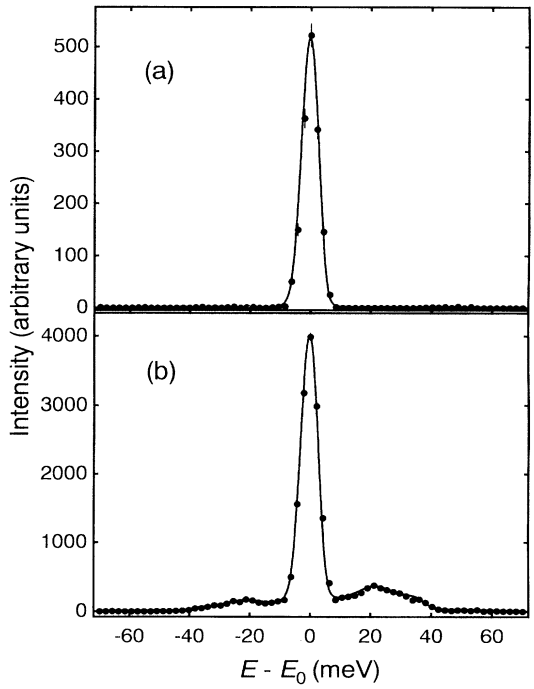

FIG. 2. Energy spectra of nuclear resonant scattering from a polycrystalline α-^{57}Fe foil. The distance of the iron foil from the detecting plane of the APD detector was (a) 230 or (b) 2 mm. The incident photon energy and the energy of the first excited state of the ^{57}Fe nucleus are denoted as E and E_0, respectively. The solid line in (a) is the resolution function of the monochromator of which the intensity was fitted to the measured data. The solid line in (b) is the sum of the recoilless part and the inelastic part calculated from the result of the neutron experiment [19].

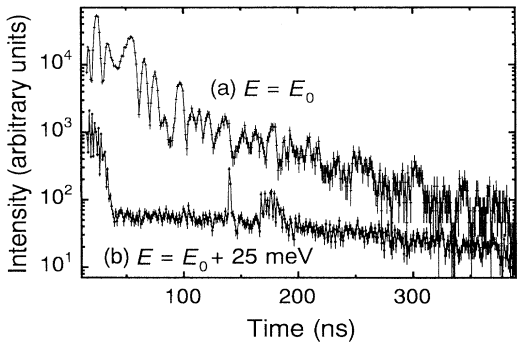

FIG. 3. Line graphs of time responses of nuclear resonant scattering from a polycrystalline α-^{57}Fe foil with (a) the incident photon energy set on recoilless resonance ($E = E_0$) and (b) the photon energy tuned to +25 meV from recoilless resonance ($E = E_0 + 25$ meV). For a comparison of the time responses, the intensity scale of the time spectrum (a) was corrected so that the data collection time of spectrum (a) corresponds to that of spectrum (b).

($\leq 10^{-5}$) electron bunches each spaced 2 ns apart, owing to lack of purity in the single-bunch structure.

It should be confirmed that the observed inelastic scattering really reflects the phonon energy distribution. When the normalized distribution $f(z)$ of the energy levels of the phonons is given, the excitation cross section σ_r with recoil per nucleus can be deduced. Since α-Fe metal has a body-centered-cubic structure, we concentrate on cubic Bravais lattices for simplicity. Following the treatment of Singwi and Sjölander [18], σ_r of a nucleus of mass M and first excited state energy E_0, for an x ray of energy E at absolute temperature T, is given by

$$\sigma_r = \frac{\pi\sigma_0\Gamma\exp(-2W)}{2}\left[\sum_{n=1}^{\infty}\frac{(2W)^n}{n!}\,g_n(E - E_0, T)\right], \tag{1}$$

where

$$g_1(E,T) = \frac{f(|E|)}{2EF(T)}\left[\coth\left(\frac{E}{2k_BT}\right) - 1\right], \tag{2}$$

$$g_n(E,T) = \int_{-\infty}^{\infty} g_1(E - E', T)\,g_{n-1}(E', T)\,dE', \tag{3}$$

$$F(T) = \int_0^{\infty}\frac{f(z)}{z}\coth\left(\frac{z}{2k_BT}\right)dz\,, \tag{4}$$

$$2W = \frac{E^2}{2Mc^2}\,F(T)\,. \tag{5}$$

Γ is the natural width of the excited state of the nucleus and σ_0 is the maximum cross section at recoilless resonance; c is the speed of light, k_B is the Boltzmann constant, and e^{-2W} corresponds to the usual Debye-Waller factor. In Eq. (1), the first term ($n = 1$) in the sum corresponds to a one-phonon process, and the others to multiphonon processes. Using Eq. (1), the inelastic cross section σ_r ($n \leq 12$) was calculated from the $f(z)$ of α-Fe (Ref. [19]) deduced from the dispersion curves obtained from the neutron inelastic scattering experiment; since the calculated Debye-Waller exponent $2W$ is $\sim$0.2, then the multiphonon terms are small compared with the one-phonon term, and σ_r converges sufficiently with a maximum $n = 12$. Then, the recoilless part and the calculated inelastic part each of which was convoluted with the resolution function of the monochromator were added by adjusting only their absolute intensities to fit the sum to the measured data. The calculated spectrum is shown in Fig. 4 [or Fig. 2(b)] as a solid line. Our spectrum is in good agreement with that obtained from the neutron experiment. The contribution from the multiphonon terms ($2 \leq n \leq 12$) shown as a dashed line in Fig. 4 fits the shoulder observed in the high-energy region ($\geq$40 meV). These results have clearly shown that the observed inelastic scattering is due to phonon scattering.

We have successfully obtained the convincing results of nuclear resonant scattering combined with phonon excitation using SR. Now, adding to knowledge of the electromagnetic environment of the nucleus, vibrational information can be obtained. If the incident photon energy is tuned to excite a target nucleus that constitutes a foreign atom in a host lattice, the vibrational character of the local surroundings of the nucleus is obtained, instead of the host lattice vibration. Therefore, this method will be important for the study of impurity or doped atoms. Furthermore, this method is effective for investigating small samples and is useful for the cases where single-crystal samples are not available. It should be noted that this method has the potential for measuring high-energy phonons, the measurements of which are difficult for neutron scattering. Thus, our method is complementary to neutron inelastic scattering. Furthermore, the results clearly indicate that it is possible to excite any nucleus of which the excitation energy is covered with SR, even

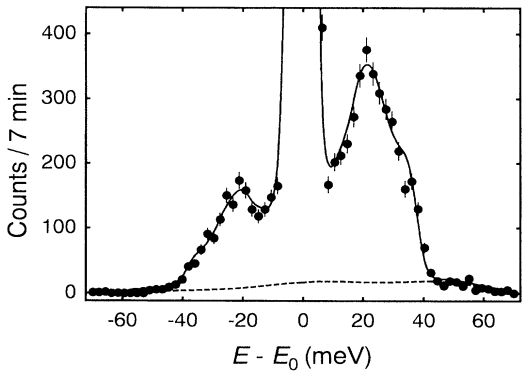

FIG. 4. Energy spectrum of nuclear resonant scattering from a polycrystalline α-^{57}Fe foil. The distance of the iron foil from the detecting plane of the APD detector was 2 mm. This is the same as in Fig. 2(b), except that the intensity scale is magnified and the sum of calculated multiphonon terms ($2 \leq n \leq 12$) convoluted with the resolution function is shown as a dashed line. The accumulation time was converted to the value when the current of the AR was 26 mA. See also the caption of Fig. 2.

though the recoilless fraction is extremely small or zero. Therefore, this method is applicable not only to solid matter but also to liquid or gas; we measured nuclear resonant excitation of ^{57}Fe ions in a hydrochloric acid solution by using this method [20].

If we adopt a detector having a larger detection area, considerable reduction in the measuring time is possible. In fact, we are now making further improvements in the detection system and the monochromators. Furthermore, in new SR sources like the European Synchrotron Radiation Facility (ESRF), Advanced Photon Source (APS), or SPring-8, higher intensity and energy are expected. For these machines this method will not be restricted to nuclides having low-energy excited states, and the energy resolution can be reduced to the order of μeV using a nuclear resonant monochromator. This method represents an important development in nuclear resonant scattering spectroscopy that can be applied in a number of research fields.

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[1] R. L. Mössbauer, Z. Phys. **151**, 124 (1958).
[2] P. B. Moon, Proc. Phys. Soc. London **63**, 1189 (1950).
[3] W. M. Visscher, Ann. Phys. (N.Y.) **9**, 194 (1960).
[4] R. L. Cohen, G. L. Miller, and K. W. West, Phys. Rev. Lett. **41**, 381 (1978).
[5] E. Gerdau, R. Rüffer, H. Winkler, W. Tolksdorf, C. P. Klages, and J. P. Hannon, Phys. Rev. Lett. **54**, 835 (1985).
[6] R. Rüffer, E. Gerdau, R. Hollatz, and J. P. Hannon, Phys. Rev. Lett. **58**, 2359 (1987).
[7] J. B. Hastings, D. P. Siddons, G. Faigel, L. E. Berman, P. E. Haustein, and J. R. Grover, Phys. Rev. Lett. **63**, 2252 (1989).
[8] J. B. Hastings, D. P. Siddons, U. van Bürck, R. Hollatz, and U. Bergmann, Phys. Rev. Lett. **66**, 770 (1991).
[9] S. Kikuta, Y. Yoda, Y. Hasegawa, K. Izumi, T. Ishikawa, X. W. Zhang, S. Kishimoto, H. Sugiyama, T. Matsushita, M. Ando, C. K. Suzuki, M. Seto, H. Ohno, and H. Takei, Hyperfine Interact. **71**, 1491 (1992).
[10] U. Bergmann, J. B. Hastings, and D. P. Siddons, Phys. Rev. B **49**, 1513 (1994).
[11] S. Yamamoto, X. W. Zhang, H. Kitamura, T. Shioya, T. Mochizuki, H. Sugiyama, M. Ando, Y. Yoda, S. Kikuta, and H. Takei, J. Appl. Phys. **74**, 500 (1993).
[12] D. C. Champeney, Rep. Prog. Phys. **42**, 1017 (1979).
[13] S. Kishimoto, Nucl. Instrum. Methods Phys. Res., Sect. A **309**, 603 (1991).
[14] S. L. Ruby, J. Phys. (Paris), Colloq. **35**, C6-209 (1974).
[15] F. J. Lynch, R. E. Holland, and M. Hamermesh, Phys. Rev. **120**, 513 (1960).
[16] G. T. Trammell and J. P. Hannon, Phys. Rev. B **18**, 165 (1978).
[17] G. V. Smirnov, Yu. V. Shvyd'ko, and É. Realo, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 33 (1984) [JETP Lett. **39**, 41 (1984)].
[18] K. S. Singwi and A. Sjölander, Phys. Rev. **120**, 1093 (1960).
[19] V. J. Minkiewicz, G. Shirane, and R. Nathans, Phys. Rev. **162**, 528 (1967).
[20] X. W. Zhang, Y. Yoda, M. Seto, Yu. Maeda, M. Ando, and S. Kikuta, Jpn. J. Appl. Phys. (to be published).