Nuclear Resonance Scattering of Synchrotron Radiation by ^{40}K

M. Seto,[1,2] S. Kitao,[1] Y. Kobayashi,[1] R. Haruki,[1] T. Mitsui,[2] Y. Yoda,[3] X. W. Zhang,[4] and Yu. Maeda[5]

[1]*Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan*

[2]*Japan Atomic Energy Research Institute, 1-1-1 Kouto, Mikazuki-cho, Sayo-gun, Hyogo 679-5148, Japan*

[3]*Japan Synchrotron Radiation Research Institute, 1-1-1 Kouto, Mikazuki-cho, Sayo-gun, Hyogo 679-5198, Japan*

[4]*Institute of Materials Structure Science, High Energy Accelerator Research Organization, 1-1 Oho, Tsukuba-shi, Ibaraki 305-0801, Japan*

[5]*Kansai Gaidai University, Hirakata, Osaka, 573-1001, Japan*

(Received 26 August 1999)

We have observed the excitation of the 29.83 keV nuclear level of the radioactive ^{40}K nuclide, which level is not populated by any radioactive source, by synchrotron radiation for the first time. The absolute energy of the level of 29.834 $\pm$ 0.011 keV and the half-life of 4.13 $\pm$ 0.12 ns were obtained from our measurements. Our success in observing the nuclear excitation of radioactive ^{40}K in a KCl powder sample shows the possibility for study of the local vibrational states and the electronic states of potassium in various areas of research.

PACS numbers: 76.80.+y, 27.40.+z, 78.70.Ck

Potassium is one of the most important elements in material and biological sciences, and the extensive and detailed investigation on the local vibrational and electronic states of the element will lead new developments of the areas of the researches. Mössbauer spectroscopy is an effective and well-established method for those researches, and the potassium isotope ^{40}K is the nuclide with the lightest mass for which a Mössbauer resonance has been observed [1,2]. This isotope itself is radioactive with a half-life $t_{1/2}$ of 1.277×10^9 yr and has a natural abundance of 0.0117% [3]. However, the 29.83 keV first excited state is not populated by any radioactive parent nuclide. The observation of the Mössbauer effect, therefore, is impossible by using ordinary radioactive sources. Inbeam methods for nuclear reactions [(d, p) reaction [1] and (n, γ) reaction [2]] on the predominant potassium isotope ^{39}K were adopted for the Mössbauer measurement. In these cases, the damage of the source, which is frequently used as a sample and nuclear reaction occurs on, is serious. In addition to the required 29.83 keV γ rays, other γ rays are strongly emitted from the source and reduce the relative absorption of Mössbauer effects. Though potassium is a very important element for all kinds of material including biological substances, these difficulties have prevented the extensive study on potassium using nuclear resonance methods with some exceptions.

For the hyperfine-interaction studies, the pulsed, highly bright, well-collimated, tunable and polarized synchrotron radiation is very distinct and advantageous as compared to a conventional radioactive source; the regular short pulse of synchrotron radiation offers a high-resolution spectroscopy in time domain that is free of troublesome mechanical motions in ordinary Mössbauer spectroscopy. After pioneering experiments [4,5], the nuclear resonant Bragg scattering of synchrotron radiation was observed in 1984 [6]. Adding to the coherent nuclear resonant scattering, the time evolution of the incoherent nuclear scattering was observed [7]. Furthermore, the observation of the single-nucleus quantum beats in the incoherent scattering has shown a possibility of the incoherent one for hyperfine spectroscopy [8]. On the other hand, one of the most outstanding and important aspects of synchrotron radiation is that the measurement of a direct phonon energy spectrum of a specific element is possible. After the first observation of the phonon energy spectrum of iron using synchrotron radiation in 1994 [9], various kinds of measurements on the local vibrational states of condensed matter have been performed with synchrotron radiation. Furthermore, the use of synchrotron radiation allows the observation of the spectrum of the diffusive motion of ions in liquids at an arbitrary temperature [10]. Though the energy tunability of synchrotron radiation has a possibility to do nuclear resonance excitation experiments of various nuclides, the resonance excitation with synchrotron radiation has been observed only for few isotopes: ^{57}Fe, ^{169}Tm [11], ^{119}Sn [12], ^{83}Kr [13], ^{181}Ta [14], ^{151}Eu [15], and ^{161}Dy [16]. Therefore, the increase of the nuclide that can be excited with synchrotron radiation is highly important and the most important target we believe is the ^{40}K nuclide, which has no appropriate Mössbauer source. The first proposal for the use of the synchrotron radiation as a source for Mössbauer measurements was done in 1974 [17], and the advantage of the synchrotron radiation for ^{40}K was pointed out in the paper. As far as we know, however, the resonance excitation of ^{40}K using synchrotron radiation has not been accomplished yet. We have succeeded in observing the nuclear resonance scattering by ^{40}K for the first time using high-pure and high-brilliant synchrotron radiation of SPring-8 and report the results in this Letter. Our results will lead to a new possibility of the vibrational and hyperfine spectroscopic studies of potassium.

The experiments were performed at the JAERI beam line (BL11XU) of the SPring-8. The storage ring was

 0031-9007/00/84(3)/566(4)$15.00

operated in a 116-bunch mode, giving a bunch distance of 41.3 ns. The electron beam current of the storage ring was 70–40 mA at 8 GeV. The 3rd harmonic of the undulator radiation was reduced to ~2 eV bandwidth at an energy of 29.83 keV with a double crystal Si(3 3 3) monochromator. The energy of the Si(3 3 3) monochromator was calibrated by measuring the Sn K absorption edge (29.2001 keV [18]) for the Sn powder absorber. The incident beam was limited to the size of 1×1 mm^2 with slits, and an Al plate of 2 mm thickness was placed in front of the slits to reduce the first harmonic radiation (9.94 keV) passing through the monochromator. The intensity ratio of the measured harmonics (1st:3rd:4th:5th) of the incident radiation was 2:100:6:2. The average intensity measured by a calibrated Si-PIN photodiode detector was about 10^{11} photons/s for 29.83 keV. The sample was KCl powder purchased from Oak Ridge National Laboratories whose isotope ratio (^{39}K:^{40}K:^{41}K) was 90.42:4.03:5.55. It was measured as a tablet of 4 mm diameter and 3 mm thickness, held in a polyethylene sample holder at room temperature (298 K).

We adopted an incoherent nuclear resonant scattering geometry as shown in Fig. 1. If the recoilless fraction of a sample is large enough, the coherent nuclear resonant scattering of synchrotron radiation is advantageous to the hyperfine spectroscopy [19]. On the other hand, when the recoilless fraction is small enough, most of the scattered radiation is incoherent and is emitted into 4π sr. In the case of the incoherent nuclear resonant scattering, the photons having the transition energy between the excited state and the ground state are directly emitted and the fluorescent x rays following the internal conversion process of the decay are emitted also. For the directly emitted photons, the solid angle of a detector should be so small to obtain the quantum beat pattern with a high visibility, which reflects hyperfine fields [8]. On the contrary, the fluorescent x rays following the internal conversion process lose the information on hyperfine interactions. These facts seem to imply that the incoherent scattering is not easy for the hyperfine spectroscopy as compared to the coherent scattering. The incoherent scattering is nevertheless important and not so

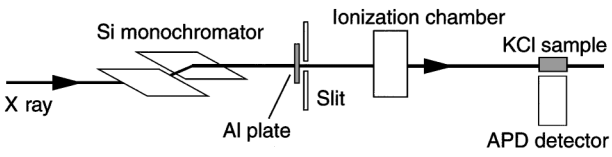

FIG. 1. Schematic drawing of the experimental setup for the incoherent nuclear resonant scattering of synchrotron radiation. With a double-crystal Si(3 3 3) monochromator, incident radiation was reduced to ~2 eV bandwidth at an energy of 29.83 keV and was limited with slits. The first harmonic radiation was reduced with an Al plate of 2 mm thickness. The photon flux on the sample was calibrated with the prompt electronic scattering and the current of an ionization chamber. The powder KCl sample was placed on the Be window of a Si-avalanche photodiode detector.

inconvenient for a sample with a low recoilless fraction and for a nuclide with a low internal conversion coefficient, because directly emitted incoherent photons are dominant in this case. For ^{40}K nuclear resonance scattering, these conditions are valid and 29.83 keV photons are incoherently scattered following the deexcitation because the internal conversion coefficient is low (0.29 [20]) and the recoilless fraction is small for most of the compounds at room temperature [2,21]. Therefore, the incoherent nuclear resonant scattering geometry was suited for our measurements. A Si avalanche photodiode (APD, Hamamatsu Photonics) detector [22] was used, the diameter of which was 3 mm. The efficiency for 29.83 keV photons is about 1%. The sample was placed on the Be window of the APD detector. The sample irradiated with incident pulses of synchrotron radiation emits isotropic radiation incoherently after a delay, and this delayed emission is counted with the detector in this setup. The photon flux on the sample was calibrated with the prompt electronic scattering and the current of an ionization chamber.

For searching the nuclear resonance excitation of ^{40}K, the energy scan of the incident x-ray radiation was carried out by changing the angle of the Si monochromator. As ^{40}K is a radioactive nuclide decaying into ^{40}Ca(89.28%, β decay) and into ^{40}Ar(10.72%, electron-capture decay), the measured background contained the radioactive emission from ^{40}K. The signal due to the resonance excitation has a time structure depending on the incident pulse timing, whereas the radioactive background is constant and is independent of the timing. No signal due to the resonance excitation but only constant radioactive emission is observed a long time after each prompt pulse. Therefore, by limiting the measuring time window to an adequate duration in which the signal due to the resonance excitation can be observed, a good signal-to-noise ratio can be obtained. We have searched delayed counts in the time interval 5–15 ns after the prompt incident pulse. The result of the energy scan is shown in Fig. 2, in which delayed counts normalized by the incident prompt counts are shown. The nuclear resonance scattering by ^{40}K is clearly observed. In the time window, the average count rate of the resonance peak was 20 counts/s, and the average background was as weak as 2 counts/s. For the confirmation of no contribution due to x-ray fluorescence following the internal conversion, between the sample and the APD detector we placed an Al foil of 0.3 mm thickness which cuts 3.3 keV $K\alpha$ x ray, and a conceivable change was not observed in the delayed signal. Therefore, the contribution due to x-ray fluorescence following the internal conversion process can be negligible in the spectrum. In this measurement, the absolute energy of the first excited state of ^{40}K was found as 29.834 ± 0.011 keV. This value agrees with the previous reported value of 29.8299 ± 0.0006 keV [3].

To measure the correct time dependence of nuclear resonance scattering, the window for the time distribution of

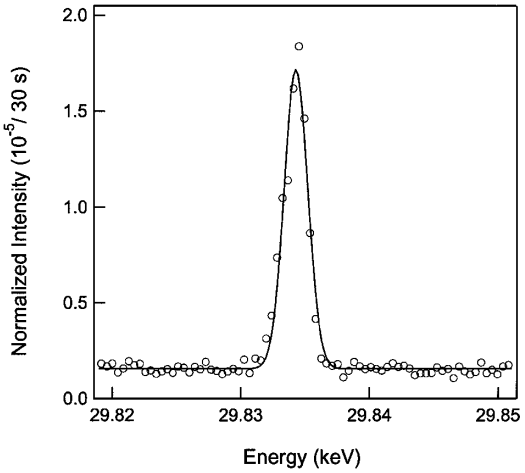

FIG. 2. Energy spectrum of delayed emission from ^{40}K in KCl. Open circles are the delayed counts normalized by the counts of the prompt scattering in the time interval 5–15 ns after prompt electronic scattering. The solid line is a Gaussian fit to the normalized delayed counts, and the peak position was determined to be 29.834 ± 0.011 keV.

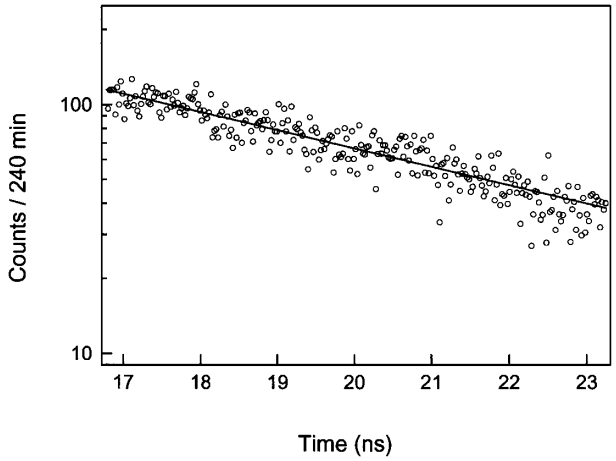

FIG. 3. Time distribution of the incoherent nuclear resonance scattering of synchrotron radiation by ^{40}K in KCl. The measured spectrum (open circles) is fitted by an exponential function, and the half-life of the first excited state of ^{40}K was evaluated to be 4.13 ± 0.12 ns.

incoherent nuclear resonant scattering was set from 17 to 23 ns avoiding the influence of the strong prompt x-ray pulses on the APD detector and the electronic timing system. The time distribution of the incoherent nuclear resonant scattering from ^{40}K in KCl is shown in Fig. 3, and no quantum beat is observed reflecting the cubic-symmetrical electronic states of K atoms in KCl crystal. This is consistent with the result of the Mössbauer measurement at low temperature [2], which showed a single Lorentzian absorption. It should be noted that using synchrotron radiation the measurement at an arbitrary temperature is possible. In the case of ^{40}K nuclear resonance in KCl at room temperature, a contribution due to the secondary x-ray scattering following the coherent nuclear resonance forward scattering is expected to be extremely small because of the small recoilless fraction [2,21]. Therefore, a coherent scattering contribution, the decay profile of which is not exponential, can be ignored in the measured time distribution. Consequently, the assumption that the time distribution is simple exponential is reasonable, and the observed spectrum can be fitted with a simple exponential time distribution. In this case, the correct decay time of the excited state can be obtained directly without complex parameters required for the time distribution of the coherent forward scattering. From this spectrum, the half-life of the first excited state is obtained as 4.13 ± 0.12 ns and this value agrees with the previously reported value of 4.24 ± 0.09 ns [3] within the experimental error.

In conclusion, we have observed the nuclear resonant excitation of the first excited state of ^{40}K and measured the time evolution of the decay for the first time, and we confirmed the energy and the lifetime of the excited state. Although the ^{40}K is a radioactive nuclide, we could obtain the resonance excitation spectrum with a good signal-to-noise ratio (S/N ≈ 10). The signal-to-noise ratio can be further improved by narrowing the time window with a small reduction of the signal. Moreover, the detecting efficiency for 29.83 keV photons can be improved by using the stacked Si-APD detector keeping the time resolution [23]. One of the most important features of the nuclear resonance spectroscopy with synchrotron radiation is the possibility to excite nuclides having no appropriate parent nuclides. Potassium-40 is just that kind of nuclide, and our experimental results show the feasibility of the nuclear resonance spectroscopy of ^{40}K and the effectiveness of the synchrotron radiation. Though quadrupole and magnetic hyperfine interactions were not observed in potassium compounds already measured [21], potassium clusters incorporated into zeolite LTA, which has supercages arrayed in a simple cubic structure and was found to show ferromagnetism below about 4 K [24], may be promising for the measurement. Furthermore, the use of synchrotron radiation allows a direct measurement of the phonon energy spectrum in the solid or the spectrum of diffusive motion of potassium atoms in liquids. This method has a prominent feature that the local vibrational state of the excited nucleus can be obtained. The use of the high-resolution monochromator with a ~5–10 meV bandwidth makes the observation of phonon energy spectra of ^{40}K possible. Furthermore, the reduction of the number of incident photons by using the high-resolution monochromator permits the measurement of delayed photons just after the prompt incident pulses. Therefore, the increase of the signal-to-noise ratio can be possible without the loss of signals. We are now in preparation for the measurement of phonon energy spectrum of potassium. A high-resolution monochromator in the relatively high-energy region is proposed [25]. We should emphasize that we could measure the nuclear resonant scattering of synchrotron radiation by the radioactive ^{40}K nuclide in spite of the

constant radioactive background, and this is the step for the further study using radioactive nuclides. For example, ^{129}I, ^{133}Ba, and ^{238}U are the radioactive nuclides by which the nuclear resonant inelastic scattering is expected to be observed [26]. Our observation of the nuclear resonance excitation of ^{40}K shows that the electronic and vibrational studies on potassium can be possible. Therefore, the promising future of experimental studies on potassium in material and life sciences is opened.

We thank the Experimental Facilities Development Group of the Japan Atomic Energy Research Institute for the development of the BL11XU beam line of SPring-8, the Accelerator Group of SPring-8 for the effort of obtaining pure bunches, and all of the staff of SPring-8 for their support. We also thank Professor Kishimoto of KEK for his support on the APD detector. We are grateful to Professor Kikuta of JASRI and Dr. Harami of JAERI for their helpful suggestions and encouragement. This research was partially supported by the Ministry of Education, Science, Sports and Culture, Grant-in-Aid for Encouragement of Young Scientists, No. 10740205, 1998.

[1] S. L. Ruby and R. E. Holland, Phys. Rev. Lett. **14**, 591 (1965).
[2] D. W. Hafemeister and E. Brooks Shera, Phys. Rev. Lett. **14**, 593 (1965).
[3] P. M. Endt, Nucl. Phys. **A521**, 1 (1990) (the properties of ^{40}K nuclide are compiled and evaluated by the author with special emphasis on nuclear spectroscopy).
[4] R. L. Cohen, G. L. Miller, and K. W. West, Phys. Rev. Lett. **41**, 381 (1978).
[5] A. I. Chechin, N. V. Andronova, M. V. Zelepukhin, A. N. Artem'ev, and E. P. Stepanov, Pis'ma Zh. Eksp. Teor. Fiz. **37**, 531 (1983) [JETP Lett. **37**, 633 (1983)].
[6] E. Gerdau, R. Rüffer, H. Winkler, W. Tolksdorf, C. P. Klages, and J. P. Hannon, Phys. Rev. Lett. **54**, 835 (1985).
[7] U. Bergmann, J. B. Hastings, and D. P. Siddons, Phys. Rev. B **49**, 1513 (1994).
[8] A. Q. R. Baron, A. I. Chumakov, R. Rüffer, H. Grünsteudel, H. F. Grünsteudel, and O. Leupold, Europhys. Lett. **34**, 331 (1996).
[9] M. Seto, Y. Yoda, S. Kikuta, X. W. Zhang, and M. Ando, Phys. Rev. Lett. **74**, 3828 (1995).
[10] X. W. Zhang, Y. Yoda, M. Seto, Yu. Maeda, M. Ando, and S. Kikuta, Jpn. J. Appl. Phys. **34**, L330 (1995).
[11] W. Sturhahn, E. Gerdau, R. Hollatz, R. Rüffer, H. D. Rüter, and W. Tolksdorf, Europhys. Lett. **14**, 821 (1991).
[12] E. E. Alp, T. M. Mooney, T. Toellner, W. Sturhahn, E. Witthoff, R. Röhlsberger, and E. Gerdau, Phys. Rev. Lett. **70**, 3351 (1993).
[13] D. E. Johnson, D. P. Siddons, J. Z. Larese, and J. B. Hastings, Phys. Rev. B **51**, 7909 (1995).
[14] A. I. Chumakov, A. Q. R. Baron, J. Arthur, S. L. Ruby, G. S. Brown, G. V. Smirnov, U. van Bürck, and G. Wortmann, Phys. Rev. Lett. **75**, 549 (1995).
[15] O. Leupold, J. Pollmann, E. Gerdau, H. D. Rüter, G. Faigel, M. Tegze, G. Bortel, R. Rüffer, A. I. Chumakov, and A. Q. R. Baron, Europhys. Lett. **35**, 671 (1996).
[16] I. Koyama, Y. Yoda, X. W. Zhang, M. Ando, and S. Kikuta, Jpn. J. Appl. Phys. Pt. 1 **35**, 6297 (1996).
[17] S. L. Ruby, J. Phys. (Paris), Colloq. **35**, C6–209 (1974).
[18] J. A. Bearden and A. F. Burr, Rev. Mod. Phys. **39**, 125 (1967).
[19] J. B. Hastings, D. P. Siddons, U. van Bürck, R. Hollatz, and U. Bergmann, Phys. Rev. Lett. **66**, 770 (1991).
[20] I. M. Band, M. B. Trzhaskovskaya, and M. A. Listengarten, At. Data Nucl. Data Tables **18**, 433 (1976) (the internal conversion coefficient for ^{40}K is obtained by interpolating the reported values in the reference).
[21] P. F. Tseng, S. L. Ruby, and D. H. Vincent, Phys. Rev. **172**, 249 (1968).
[22] S. Kishimoto, Nucl. Instrum. Methods Phys. Res., Sect. A **309**, 603 (1991).
[23] S. Kishimoto, N. Ishizawa, and T. P. Vaalsta, Rev. Sci. Instrum. **69**, 384 (1998).
[24] Y. Nozue, T. Kodaira, and T. Goto, Phys. Rev. Lett. **68**, 3789 (1992); Y. Nozue, T. Kodaira, S. Ohwashi, T. Goto, and O. Terasaki, Phys. Rev. B **48**, 12 253 (1993).
[25] Yu. V. Shvyd'ko, E. Gerdau, J. Jäschke, O. Leupold, M. Lucht, and H. D. Rüter, Phys. Rev. B **57**, 4968 (1998).
[26] N. N. Greenwood and T. C. Gibb, *Mössbauer Spectroscopy* (Chapman & Hall, London, 1971).