Nuclear Scattering of Synchrotron Radiation by ^{181}Ta

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We report the first observation of excitation of the 6.2 keV nuclear level of ^{181}Ta by synchrotron radiation. The absolute energy of the level was determined to be 6214 ± 2 eV. The time distribution of nuclear forward scattering of synchrotron radiation by a 7.5 μm Ta metal foil was measured, and a decay time of 530 ± 80 ns was observed. Analysis of the time distribution reveals an inhomogeneous broadening of the nuclear resonance in this sample 6 ± 2 natural linewidths.

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The recent rapid development of hyperfine nuclear resonance spectroscopy using synchrotron radiation [1] indicates that this method will become a standard technique in the near future. The broadband, pulsed, well-collimated, and highly polarized synchrotron radiation is very different in character from that of a conventional radioactive source, and offers distinct advantages for hyperfine spectroscopy experiments. However, only a few isotopes have been used in synchrotron-based experiments to date: ^{57}Fe [1], ^{169}Tm [2], ^{119}Sn [3,4], and ^{83}Kr [5,6].

An attractive target is the 6.2 keV nuclear $E1$ ($\frac{9}{2} \rightarrow \frac{7}{2}$) transition in ^{181}Ta. The long lifetime of this nuclear level ($\tau_0 = 8.73$ μs [7]) and the complementary narrow energy width ($\Gamma_0 = 7.5 \times 10^{-11}$ eV, corresponding to a Doppler shift of 3.6 μm/s velocity), together with a very large nuclear magnetic moment ($\mu = +5.2$), make this resonance a very sensitive probe of hyperfine interactions. In the field of Mössbauer spectroscopy this resonance gives more than an order of magnitude better energy resolution than the 14.4 keV resonance of ^{57}Fe ($\tau_0 =$ 141 ns). This has been shown in many experimental studies, including high-resolution measurements of the ^{181}Ta isomer shift in metallic systems and compounds [8], studies of hydrogen diffusion in Ta metal [9], temperature-induced changes in the electronic structure of ferroelectric $LiTaO_3$ [10], and observation of the double NMR Mössbauer resonance [11].

However, the high sensitivity causes severe technical difficulties in the preparation of good radioactive sources for the ^{181}Ta Mössbauer analysis. The source lines are considerably broadened by crystal imperfections or by interstitial impurities such as oxygen or hydrogen [12,13]. These difficulties are reflected in the fact that the narrowest experimental linewidth observed so far in ^{181}Ta Mössbauer spectroscopy is about $15\Gamma_0$ [13]. Another disadvantage of the radioactive sources is the low flux of γ radiation due to the high internal conversion coefficient ($\alpha = 70.5$ [14,15]) and the large photoelectric absorption. In addition, the resonant 6.2 keV γ radiation is strongly shadowed by the intense $L_{\alpha,\beta}$ fluorescence x radiation of Ta.

By contrast, the synchrotron radiation source provides almost perfect experimental conditions. The short pulse of synchrotron radiation allows high-resolution time-domain spectroscopy. The timing experiment also eliminates the problems arising from mechanical vibrations [16] and geometrical broadening. The method is practically free of any background radiation in the time window excluding the synchrotron radiation pulses. Within certain limits, even the broadening of the *absorber* resonance lines does not complicate the hyperfine spectroscopy, because it leads to the decay acceleration, but not to smearing out the temporal quantum beats arising from hyperfine interactions [1].

For these reasons it is of high interest to explore the feasibility of nuclear resonance spectroscopy of ^{181}Ta with synchrotron radiation as the source. In this paper we report the precise determination of the location of the ^{181}Ta resonance, the determination of the absolute energy of the 6.2 keV nuclear transition, and the measurement of the time distribution of nuclear forward scattering.

The experiment was performed at the 27-period wiggler beamline 6-2 at the Stanford Synchrotron Radiation Laboratory (SSRL). The storage ring was run in timing mode with four electron bunches in the ring, providing pulsed radiation with a 195 ns period. During a 12-h period a special single bunch mode with 780 ns between pulses was used for the studies of the time distribution of the ^{181}Ta nuclear decay.

A synchrotron radiation beam of ~3.6 eV bandwidth at an energy of 6.2 keV was prepared by a cooled Si(111) monochromator. Further reduction of the bandwidth down to ~83 meV was achieved by a Si(333)

symmetric-reflection channel-cut monochromator (θ_B = 72.6°). After passing through the ^{181}Ta sample the x rays were counted by an EG&G avalanche photodiode (APD) timing detector [17]. The sample and high-resolution optics were contained in a helium-filled box to minimize flight-path absorption of the 6.2 keV radiation.

A nuclear forward scattering [18] geometry was used, in which radiation is transmitted straight through the sample into the detector. The resonant sample is excited by short pulses of synchrotron radiation, and emits radiation coherently into the forward direction after a delay determined by the lifetime of the nuclear excited state. The prompt flux on the detector, due to nonresonant transmission of the synchrotron pulses, was about 1.5×10^7 photons/s (as measured by a calibrated ion chamber). The APD signal was gated off during the prompt pulse so that only delayed events were counted.

The resonant ^{181}Ta target was a 99.996% pure Ta foil of 15 mm diameter and 3.8 μm thickness. This sample had been previously studied by conventional Mössbauer spectroscopy [13] and revealed an extremely narrow experimental linewidth (measured as a convolution of the source and the absorber lines) of about 57 ± 1 μm/s [(15.8 ± 0.3)Γ_0]. For the present experiment the sample was tilted at an angle of about 30°, so its effective thickness along the beam path was about 7.5 μm.

A major technical challenge for the experiment arose from the large uncertainty in the value of the ^{181}Ta resonance energy. The error range derived from the Mössbauer literature was about ±20 eV, around a central value 6238 eV [14]. Recently, a somewhat more precise value for the resonance energy, 6224 ± 5 eV, was found from a preliminary analysis of high-resolution measurements of conversion electron spectra of Ta compounds [19].

In the search for the nuclear resonance the total delayed count rate, in the time interval 40–90 ns after the prompt flash, was measured as a function of the incident x-ray beam energy. Figure 1(a) shows the peak of the delayed events with a maximum count rate of 0.23 counts/s above a background of 0.013 counts/s. The observed width of the peak, 83 ± 12 meV (FWHM), is in good agreement with the calculated value (87 meV) of the energy bandwidth of the Si(333) monochromator.

For the determination of the ^{181}Ta resonance energy we used the Nd L_{III} absorption edge. The Si(111) monochromator was calibrated by measuring the edge position (6209.5 eV [20]) for the NdF_3 powder absorber. In this measurement the *absolute* energy of the ^{181}Ta resonance was found as 6214 ± 2 eV (the error is dominated by the uncertainty in the published data on Nd L_{III} edge energy). It was possible to make much more precise *relative* measurements of the resonance energy with respect to the pronounced absorption maximum (white line) of the NdF_3 absorber. For this purpose the absorption fine structure of the Nd L_{III} edge was measured by scanning the Si(333) monochromator while simultaneously adjusting the Si(111) monochromator so as to keep the incident intensity constant [Fig. 1(b)]. By this means, the location of the ^{181}Ta resonance was determined to be 1130 ± 80 meV above the maximum procedure for finding the resonance in further studies.

The time distribution of the nuclear forward scattering from the Ta foil was measured during a special 12-h period of single bunch operation of the synchrotron light source. The time distribution is shown in Fig. 2. The experiment data may be approximately described by an exponent with $(1/e)$ decay time of 530 ± 80 ns.

The experimental data were fitted using the dynamical theory of resonant nuclear scattering [21]. For an $E1$ nuclear transition with some inhomogeneous broadening, the response function $R(E)$—the energy-dependent complex amplitude of the wave field transmitted through the absorber—takes the form [22,23]

$$R(E) = \exp -\frac{(iT\Gamma_0/4)(1 + 2i\xi)}{E - E_0 + i\Gamma/2}, \qquad (1)$$

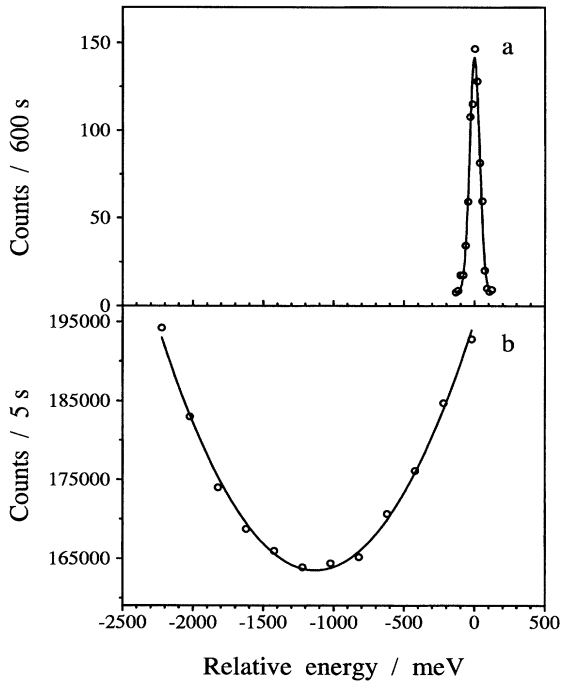

FIG. 1. Determination of the ^{181}Ta resonance energy relative to the absorption fine structure of the Nd L_{III} edge. (a) Delayed count rate of nuclear forward scattering of synchrotron radiation by a 7.5 μm Ta foil (in the time interval 40–90 ns after the prompt pulse). The solid line is a Gaussian fit to the experimental data with a FWHM of 83 ± 12 meV. (b) Intensity of radiation transmitted through the NdF_3 sample in the vicinity of the maximum absorption point. Experimental data in the displayed region are approximated by a parabola (solid line). Both sets of data are plotted as functions of energy relative to the ^{181}Ta resonance peak center.

where $T = (\sigma_0 f_{LM} n z) = 44$ is the effective resonant thickness of the sample, $\sigma_0 = 1.1 \times 10^{-18}$ cm^2 is the resonance cross section, $f_{LM} = 0.96$ is the Lamb-Mössbauer factor, $n = 0.55 \times 10^{23}$ cm^{-3} is the density of resonant nuclei, z is the thickness of the sample, $(E - E_0)$ is the deviation from the resonance energy, Γ is the inhomogeneously broadened width of the resonance, and $\xi = -0.16$ [24] is a parameter which accounts for an interference between electronic scattering and nuclear scattering for the $E1$ transition of ^{181}Ta [22,23]. The fit used a Fourier transform of the response function. The overlap of the decays resulting from consecutive prompt pulses was taken into account. The best fit (solid line in Fig. 2) was obtained with the width of the nuclear resonance $\Gamma = (6 \pm 2)\Gamma_0$, corresponding to 22 ± 7 μm/s

The ability to determine directly the width of the nuclear resonance in a sample is one of the advantages of synchrotron nuclear spectroscopy. In standard Mössbauer spectroscopy the measured linewidth always includes a convolution with the linewidth of the radioactive source. On the contrary, the value obtained here for the inhomogeneous broadening of the nuclear transition, $\Gamma = (6 \pm 2)\Gamma_0$, is a characteristic of the sample, determined only by the chemical environment of the ^{181}Ta nuclei.

Standard Mössbauer absorption experiments using the 6.2 keV nuclear $E1$ transition in ^{181}Ta show a large asymmetry of the resonant line [25], caused by an interference between the nuclear internal conversion and photoelectric absorption processes [22,23]. It has been predicted [26] that an interference between the nuclear and electronic channels should occur during emission and scattering processes as well. This interference was included in Eq. (1). Calculations of the forward scattering time distribution over a larger time range show that the dynamical beats [27] in the time distribution are affected sensitively by this interference. However, the experimentally available time range, much smaller than the expected time of the first dynamical beat minimum for this sample (at about 2.9 μs), did not allow this effect to be studied in the present experiment.

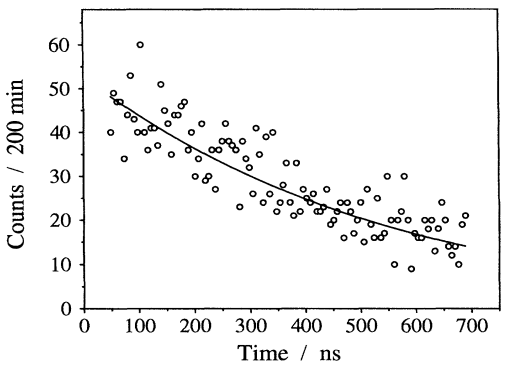

FIG. 2. Time distribution of nuclear forward scattering of synchrotron radiation by the 7.5 μm Ta foil. The experimental data are fitted (solid line) by the dynamical theory of resonant nuclear scattering under the conditions of inhomogeneous broadening of the resonance line. A small background (2.7 counts per channel in the figure) is included in the fit. The width of the nuclear resonance is found to be $\Gamma = (6 \pm 2)\Gamma_0$, corresponding to 22 ± 7 μm/s.

The x-ray beam created by nuclear scattering in the Ta sample (the delayed, coherent forward scattered radiation), had a bandwidth of about 4.5×10^{-10} eV. This qualifies as the most monochromatic beam of synchrotron radiation ever prepared ($\Delta E/E = 7 \times 10^{-14}$). The longitudinal coherence length of such radiation is about 1 km.

This demonstration of the feasibility of hyperfine nuclear resonance ^{181}Ta spectroscopy using synchrotron radiation promises future developments. The general advantages of this method are particularly apparent in the case of ^{181}Ta spectroscopy, where synchrotron radiation provides almost ideal *instrumental resolution* and, therefore, allows direct measurement of the resonance parameters of the sample *alone*. Furthermore, a timing experiment provides a means to distinguish even unresolved lines in some cases [28], thereby avoiding complications due to broadening of the resonance lines of the *absorber*. Thus, the extreme narrowness of the ^{181}Ta resonance can, in principle, be fully exploited for the study of very weak hyperfine interactions. However, the observation of hyperfine splitting of less than 0.1 mm/s (corresponding to quantum beats with period larger than 2 μs) is restricted by the relatively small time window available at the present synchrotron radiation storage rings ($\leq$3 μs). Finally, the pronounced interference between nuclear and electronic scattering ^{181}Ta, observed so far only in adsorption, could stimulate new developments in the dynamical theory of resonant nuclear scattering.

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[1] See e.g., the review; E. Gerdau and U. van Bürck, in *Resonant Anomalous X-Ray Scattering. Theory and Applications,* edited by G. Materlik, C. J. Sparks, and K. Fischer (Elsevier, New York, 1994), p. 589, and references therein.

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