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MÖSSBAUER EFFECT IN K^{40} USING AN ACCELERATOR*

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For producing nuclear states for investigation by recoilless-emission techniques, the only methods used heretofore have involved the use of radioactive parents. No radioactive parent exists for K^{40} and therefore this method of populating the first excited state of K^{40} at 29.4 keV[1] cannot be used. However, it can be made by many nuclear reactions such as $K^{40}(p,p')K^{40}$, $K^{39}(n,\gamma)K^{40}$, $A^{40}(p,n)K^{40}$, and $K^{39}(d,p)K^{40}$. Using the last, which was selected mainly to minimize the difficulties of making a suitable target, we have observed recoilless emission of the 29.4-keV radiation from the first excited state of K^{40}.

Since about 25 eV is adequate to eject an atom from its lattice position, and since the above production methods give the excited K^{40} atom a kinetic energy E_k in the range $100\ \text{eV} \leq E_k \leq 2\times10^5$ eV, it is evident that radiation-damage effects will be crucial to the experiment. The usual model for the stopping of such atoms invokes a "heat spike" at the end of the track where initially the local temperature is very high.[2] For example, if 10 keV is shared via multiple elastic collisions over 10^4 atoms, the local temperature might be described as about 10^4 °K. These spikes rapidly relax, and in times of the order of a few times 10^{-11} sec the material recrystallizes. Especially in insulators, and certainly in chemically complicated materials, one does not expect this process to leave the K^{40m} in the same microenvironment each time, and the Mössbauer effect should become correspondingly blurred.

From the point of view of rapid recovery from the radiation-damage effects, potassium metal would be preferred target material. Unfortunately, this very soft material ($\Theta_D \approx 100$°K despite the low mass of the isotope) would not be expected to give rise to an appreciable resonant fraction f. In fact, the usual calculation leads to $f = e^{-8} \approx 0.03\%$ for the maximum possible effect from such a source at 78°K. It was hoped that other alloys or compounds might be found to be more suitable as source materials. For example, typical force constants in the potassium halides lead to Debye temperature of approximately 215°K for the potassium atom; f at 78°K would then be about 20%.

Figure 1 is a schematic diagram of our equipment. Both the target and absorber were cooled with liquid nitrogen. The absorber was 10 mg of KCl, enriched to 12.5% in K^{40}, and distributed over an area of 6.3 mm diameter. The detector was a NaI(Tl) scintillator 0.25 mm thick. In the pulse-height spectrum obtained with 0.2 μA of 3.5-MeV deuterons incident on a thick potassium metal target, the peak from the 29-keV gamma ray was three to four times the background generated by higher energy γ rays and neutrons produced in the target. The

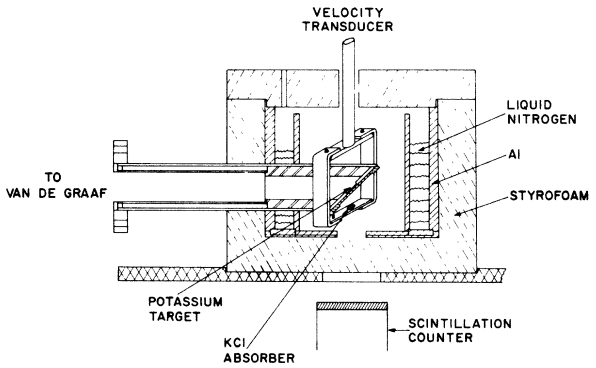

FIG. 1. Schematic diagram of target, absorber, and velocity spectrometer.

counting rate for events in this peak was of the order of 2000 counts/sec. The absorber was moved with a loud-speaker–type electromechanical drive, its velocity was measured with a pick-up coil in a permanent magnet, and the velocity was slaved to the regularly stepped address of the multichannel analyzer through a feedback amplifier.

No resonant absorption greater than 0.3% was observed with thick (~0.2 mm) targets of KBr and KOH. This may have resulted partly from poor cooling under the beam spot because of the insulating properties of these targets.

Resonant absorption was obtained, however, with nominally metallic potassium targets. Figure 2 shows the results of the second "metallic" run as returned from the computer. Here the solid line is a Lorentzian line shape whose parameters of amplitude, width, and position were chosen to minimize the least-mean-square difference from the 400 experimental points. The results for this run are $(1.19 \pm 0.07)\%$, 3.8 ± 0.5 mm/sec, and 0.02 ± 0.12 mm/sec, respectively.

Two further "metallic" runs were made with fresh material for targets, and they gave the same results as the above run within the estimated errors. The first "metallic" run had given an absorption dip with almost twice the amplitude and 40% greater linewidth.

The average of all four runs is -0.02 ± 0.09 for the isomer shift. The linewidths averaged to 4.1 ± 0.35 mm/sec. From the half-life of 3.9 nsec,[3] the minimum experimental linewidth (corresponding to a thin absorber and a source with natural linewidth) is found to be 2.5 mm/sec. The observed broadening to 5/3 times the natural line is fully accounted for by the absorber thicknesses. In fact, these experimental results are consistent with a Debye temperature of 180°K for the potassium in KCl rather than the 215°K anticipated above. In any case the source linewidth must be rather close to the natural width.

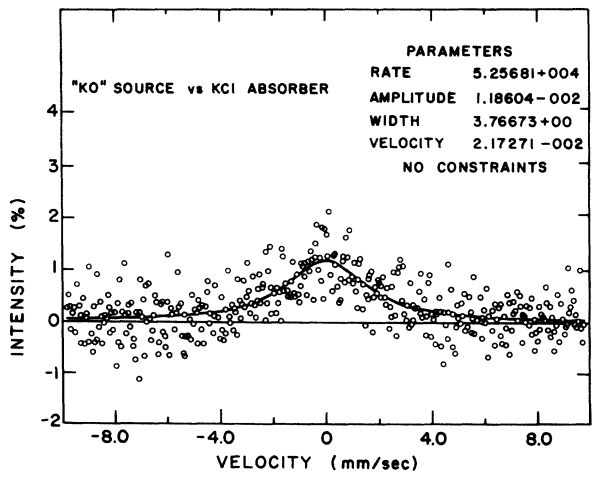

FIG. 2. Resonant absorption spectrum for the 29-keV gamma ray from K^{40}. The curve is the result of a least-squares fit of a Lorentzian line to the data.

The surface of these targets was oxidized because of exposure to air during their preparation. In an effort to test the importance of this oxygen layer, the target for the fourth run was exposed to air for a longer time. However, as indicated above, no change was observed in the spectrum.

We do not know the explanation of our large effect. We speculate that our target is actually some kind of potassium oxide for which the elastic constants are much higher than for potassium metal. This hypothesis is supported by the work of Stone and Pillinger[4] who observed Mössbauer effect in Np^{237} after both α and β decay, where the source Am^{241} or U^{237} was dissolved in neptunium oxide. The radiation-damage problems following α decay are as difficult as those following (d,p) reactions. It is noteworthy that the intensity of Mössbauer absorption following α decay was only about one-fourth that found when the isomeric state was formed from β decay. A second fact which suggests a potassium oxide source was discovered by a short investigation of the background in the scintillation counter. A major fraction of this background has a lifetime very close to that of 66-sec F^{17}. We tentatively assume that this comes from the reaction $O^{16}(d,n)F^{17}$ so that oxygen must be present in the pertinent part of the target.

It is expected that further experimentation will lead to a more satisfactory source. It is anticipated that even without significantly better sources, valuable information on the moments and isomer shift can be found for this unusually well-understood nucleus. It is also expected that this method of exciting low-energy γ rays may be extended to other isotopes; also it should revive interest in the Coulomb-excitation method for producing recoilless radiation.

We wish to extend our thanks to Ronald Amrine, John Bicek, and Richard Davies for their excellent help in adapting our experiment to the Van de Graaff accelerator.

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MÖSSBAUER EFFECT OF THE 29.4-keV NEUTRON CAPTURE GAMMA RAY OF K^{40}†

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Capture of a thermal neutron by K^{39} leads to the formation of an excited level in K^{40} with energy of about 7.8 MeV. After the emission of one or more energetic gamma rays, approximately 30% of the neutron captures result in the population of an excited level at 29.4 keV. We have observed Mössbauer resonance absorption of the 29.4-keV gamma ray resulting from the decay of this latter state. Emission of the energetic photons preceding the formation of the 29.4-keV level leaves the K^{40} nucleus with a distribution of recoil energies up to a maximum of about 800 eV. Energies of this order can be expected to displace the atom from its normal lattice site. However, our measurements indicate that radiation damage from this effect does not substantially diminish the recoilless fraction, f, in insulators (KCl and KF) or in metals (K). It appears, therefore, that thermal neutron capture can be used to excite a variety of new nuclei which have not been accessible for Mössbauer study through radioactive decay.

In this experiment a $\frac{1}{2}$-in.-diameter thermal neutron beam of about 3×10^6 neutrons/sec was extracted from the Omega West reactor. The targets, consisting of natural potassium in the forms K, KCl, and KF, were all about 0.3 cm thick. The resonance was observed at two temperatures, 4 and 78°K, and using two absorber thicknesses, 4.3 and 10.6 mg/cm^2, of K^{40}. Each absorber was $\frac{3}{4}$ in. in diameter and consisted of KCl enriched to 30.3% in K^{40}. The effective neutron cross section of K^{39} for production of the 29.4-keV isomer is about 0.6 b. With the arrangement described, typical counting rates were about 2500 counts/min. Target and absorber were mounted in a cryostat fitted with thin Al windows for passage of the neutron beam and the 29.4-keV gamma ray.

Motion of the absorber was obtained with a velocity sweep system in which the sinusoidal

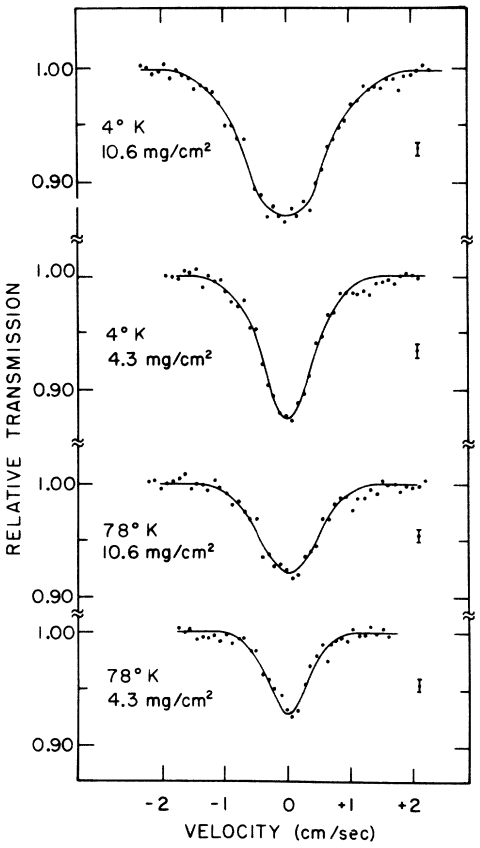

FIG. 1. Relative transmission of 29.4-keV gamma rays from a KF source through a $K^{40}Cl$ absorber as a function of velocity, temperature, and absorber thickness.