OBSERVATION OF RECOILLESS EMISSION OF ^{61}Ni GAMMA RAYS FOLLOWING COULOMB EXCITATION*

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We have observed the recoilless emission of gamma rays from ^{61}Ni after the 67.4-keV energy level of this nucleus was populated by Coulomb excitation. The projectiles used for the excitation were 25-MeV oxygen ions from the Oak Ridge National Laboratory tandem Van de Graaff. The total Mössbauer absorption observed by this method for ^{61}Ni agrees well with previous measurements[1] using the radioactive source

$$^{61}\mathrm{Co}\xrightarrow[99\ \mathrm{min}]{\beta^-}{}^{61}\mathrm{Ni}^*.$$

The metallic nickel targets used for the present experiment were irradiated for periods up to 120 hours. These targets were held at a temperature of ~78°K during the entire irradiation period. There were no changes in the Mössbauer spectra observed during these measurements which could be attributed to radiation damage in the target. The maximum absorption, which occurs at zero velocity, remained constant throughout our longest measurement time which was 120 hours.

Previous studies[2] of the Mössbauer effect following Coulomb excitation were done using ^{57}Fe for the target and absorber, and to within experimental accuracy of the measurements there was no effect observed.

The observation of the Mössbauer effect in these experiments can be summarized in the following scheme: (1) The relevant energy level, in this case the 67.4-keV level of ^{61}Ni, is populated by Coulomb excitation. (2) The Coulomb-excited nucleus which recoils during the interaction must dissipate its kinetic energy and come to rest in the crystal lattice in a time short compared to the lifetime of the excited level. (3) The gamma ray must be emitted from the target nucleus without recoil and has to be absorbed recoil free in an absorber which contains ^{61}Ni nuclei. (4) The transmission of these gamma rays through the absorber is measured as a function of the relative velocity between target and absorber.

The criteria for the selection of a nucleus in the present experiment were these: (1) The Mössbauer effect for the nucleus should have been observed previously such that the important parameters of the spectrum are known. (2) The target material should be metallic because we expected that a metallic crystal would return to thermal equilibrium in a time short compared to the lifetime of the excited state of the nucleus after the passage of the projectile through the target. This may not be true for insulators. (3) The Mössbauer effect should be measurable at or above liquid nitrogen temperature. (4) The gamma intensity of the Coulomb-excited Mössbauer level must be high compared with the background contribution[3] at the gamma energy of interest from the following processes: Coulomb excitation populates not only the level of interest but also other low-energy levels of the target nucleus. In addition, the passage of the ions through the target produces a high intensity of characteristic x rays. Bremsstrahlung from the projectile is not negligible in some cases. Of the possible candidates we have chosen ^{61}Ni because this nucleus satisfied all four conditions listed.

Figure 1 shows schematically the arrangement of our apparatus. The $^{16}O^{4+}$ beam enters the chamber along the horizontal axis. The target is in vacuum and is cooled to 78°K by direct contact with liquid nitrogen. The cur-

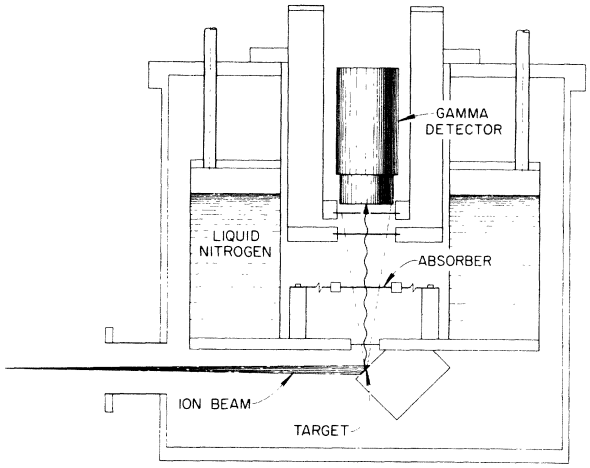

FIG. 1. Target chamber used in this experiment.

rent of the 25-MeV oxygen ions at the target was a maximum of 0.5 μA, and the average was about 0.35 μA during the entire run. The counting rate for this current was 3.5×10^3 counts/min in the 67.4-keV photopeak. The heat input to the target due to the oxygen ions was, on the average, 2 watts.

The target was made by electroplating nickel enriched to 92% ^{61}Ni on a copper plate. The thickness of the deposit was 2.8 mg/cm^2. This thickness gives 87% of the thick-target yield for 25-MeV oxygen ions, but since the target makes an angle of 45° to the incoming beam, the actual yield is 99%. The broadening of the Mössbauer lines due to this thickness is at most 10% of the natural linewidth.

The absorber shown in Fig. 1 was a circular plate of natural nickel of 445 mg/cm^2 thickness. The abundance of ^{61}Ni is 1.25% in natural nickel. The absorber was moved sinusoidally by an electromechanical velocity drive. Both the absorber and velocity drive were held at 78°K during the measurements.

The gamma-ray detector was a Harshaw integral line detector with a 3-in.×3-in. NaI(Tl) crystal. Due to the sinusoidal motion of the velocity drive it was convenient to store the Mössbauer spectrum in one bank of the memory of a multichannel analyzer and a nonresonant background spectrum in a second bank of the memory. The background spectrum is characteristic of the motion of the velocity drive. The Mössbauer spectrum was divided by the background, channel by channel, to produce a normalized velocity spectrum.

The shape of the Mössbauer spectrum of ^{61}Ni in nickel metal is determined by the following parameters: (1) the magnetic moments and spins of the ground- and first-excited states, which are $|\mu_g| = (0.746\pm0.007)\ \mu_N$,[4] $J_g = \frac{3}{2}$, and $|\mu_e| = (0.35\pm0.06)\ \mu_N$, $J_e = \frac{5}{2}$, respectively; (2) the mean lifetime of the first excited state, $\tau = 7.6\times10^{-9}$ seconds; and (3) the effective magnetic field $H_{\text{eff}} = (-70\pm15)$ kG acting on the ^{61}Ni nucleus. It may be seen from a calculation using these parameters that the Mössbauer spectrum consists of 35 closely spaced unresolved lines. This spectrum can be approximated very well by a triangular shape and is in agreement with the measured spectrum.[1]

Two independent runs at the tandem Van de Graaff have been made. Figure 2(a) shows the final result of our second run. To take into account the attenuation of the observed effect due to background counts, we have multiplied the deviations from unity and the errorbars by a constant factor λ = (signal + background)/signal. A typical gamma spectrum is shown in Fig. 2(b). The shape of the measured Mössbauer spectrum agrees well with the calculated spectrum using the nuclear parameters as given above. If the target and absorber in our measurements are in the same chemical state, then the isomer shift is zero. This is, of course, true only if the Coulomb-excited target atoms are on regular lattice sites. The observed isomer shift is $S_I = (0\pm0.5)$ mm/sec. The maximum absorption which occurs at zero velocity as seen in Fig. 2(a) is (3.1 ± 0.3)%. The Mössbauer spectrum obtained in the first run has

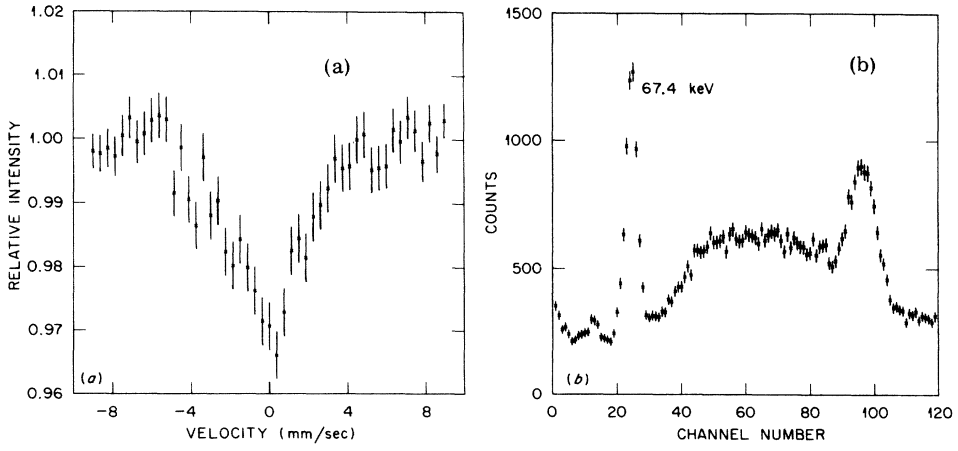

FIG. 2. (a) Mössbauer spectrum of ^{61}Ni following Coulomb excitation. In this spectrum the background has been accounted for. (b) The gamma spectrum from the Coulomb-excited ^{61}Ni nuclei in the energy range below 325 keV.

also the expected triangular shape. The isomer shift is $S_I = (0.0 \pm 0.5)$ mm/sec; the maximum absorption at zero velocity is $(3.7 \pm 1.4)\%$.

From time to time we interrupted the measurement of the absorption spectra in order to measure the maximum absorption at zero velocity, $M(v=0)$, as a function of irradiation time of the target. The individual measurements made during the first run at the tandem have large statistical errors so that only the average is significant, and the result is $\langle M(v=0)\rangle = +(2.6 \pm 0.9)\%$. All $M(v=0)$ values quoted here contain a factor λ. The data obtained for $M(v=0)$ versus irradiation time during the second run are shown in Fig. 3. A least-squares fit to these measurements indicates that there is no observable change in $M(v=0)$ with irradiation time. Any radiation damage in the target seems to reach a saturation value rapidly and to remain constant for the period covered by this measurement.

The average value of $M(v=0)$ is $(3.0 \pm 0.2)\%$. This result is consistent with that obtained in the first run, and in agreement with the maximum absorption of the Mössbauer spectrum, Fig. 2(a). However, it is only $(74 \pm 6)\%$ of the value $(4.05 \pm 0.15)\%$ obtained previously.[1] The total absorption as given by the area of the absorption dip is equal to that observed in reference 1 to within the accuracy of our measurement. A measurement of $M(v=0)$ was also made with an absorber whose thickness was 832-mg/cm^2 natural nickel. The result was $M(v=0) = (3.7 \pm 0.4)\%$. This value should be compared to $(6.1 \pm 0.3)\%$ in reference 1; therefore, $M(v=0)$ is $(61 \pm 7)\%$ of the full effect.

From a comparison of the total absorption measured in this experiment with that obtained in reference 1, we conclude that the average Debye-Waller factor in our target does not differ by more than 10% from that in an undisturbed nickel crystal. This result and the reduced value of $M(v=0)$ leads us to the conclusion that the Coulomb-excited nuclei in their final positions sample a wider distribution of magnetic fields, electronic charge densities, electric field gradients, and binding energies than do nuclei in an undamaged nickel crystal. A model which could lead to an understanding of these results has been worked out by Dederichs et al.[5] They have calculated the probability that an atom of high kinetic energy comes to rest on a lattice site during the slowing down process in a solid. In the cases they considered they obtain the result that at least 30% of these atoms do not find a final position on a lattice site during this process. This result is consistent with our experiment.

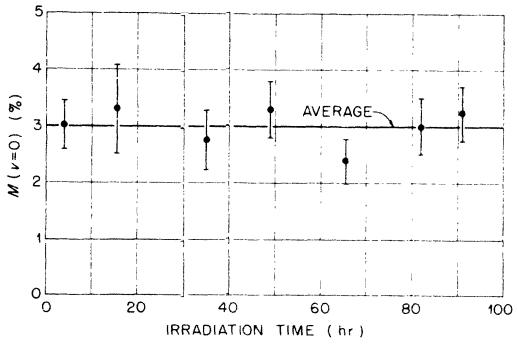

FIG. 3. Mössbauer absorption at zero velocity $M(v=0)$ versus irradiation time of the target. The background has been taken into account for each data point.

This experiment establishes the feasibility of Mössbauer measurements using gamma rays from Coulomb excitation. Since this method is applicable to all stable nuclei with low-lying energy levels, it can be an important tool in both nuclear and solid state physics.[6]

We wish to acknowledge the continuing interest of Horst Wegener throughout the course of this experiment. One of us (D.S.), is grateful to the Deutsche Forschungsgemeinschaft for financial support. We thank Frank DiCarlo for his constant vigilance at the control panel of the tandem.

Note added in proof.—We have learned from Y. K. Lee et al. of John Hopkins University that they have observed the Mössbauer effect with ^{57}Fe using Coulomb excitation.

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MÖSSBAUER EFFECT FROM COULOMB-EXCITED LEVELS IN Fe^{57}†

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We have observed a large Mössbauer effect in Fe^{57} following Coulomb excitation. Our results indicate that despite the large recoil energy of the nucleus following the Coulomb excitation, the Mössbauer fraction is essentially undiminished.

Recently, the presence of recoilless emission of γ rays from nuclei which have been excited by processes which impart large recoil energies to the excited nuclei has been reported by others.[1-4] The observation of a large Mössbauer effect in Fe^{57} is significant for the investigation of these phenomena.

Coulomb excitation was produced by a 1.5-μA beam of 3-MeV alpha particles from the Johns Hopkins University Van de Graaff accelerator. The target and absorber were 1.9 mg/cm^2 of 91% enriched Fe^{57}. The target was thermally coupled to an aluminum backing which was cooled by contact with an alcohol–dry-ice mixture.

The recoilless emission of the 14.4-keV level was observed following Coulomb excitation of the 137-keV level. The 14.4-keV γ ray was detected in coincidence with the preceding 122-keV γ ray (Fig. 1). This was necessary due to the large Mössbauer fraction which allowed self-absorption in the target making it difficult to observe directly the 14.4-keV line in the presence of x rays and bremsstrahlung background.

The detectors were thin NaI(Tl) crystals. The coincidence circuit had a resolving time of 0.5 μsec. The coincidence technique reduced the total background to less than 8%. The rate of 137-keV level excitation to be expected was calculated according to Alder et al.[5] and was in agreement with the rate of 122-keV γ rays observed. On the basis of the 122-keV γ-ray counting rate, one can estimate the number of 14.4-keV γ rays which should be detected in the absence of nuclear resonant self-absorption. However, the observed coincidence counting rate of 3 counts per second was several times less than that expected from this calculation. The existence of self-absorption in the target suggests that perhaps a correction should be made to the $E2$ nuclear matrix element for the 14.4-keV transition as reported by Thomas and Grace.[6]

The width and depth of the absorption dips indicate that the difference in hyperfine splitting between source and absorber is less than 10%. Additional spectra including all the hyperfine lines will be taken to obtain a more accurate determination of the hyperfine splitting of the Coulomb-excited nucleus.

The central absorption dip, uncorrected for background, is 17% of the nonresonant transmission (Fig. 2). Because of the very thick target and uncertainties due to vibrations, it is difficult to obtain a value for f, the Mössbauer fraction for the source. Comparison of the Coulomb-excitation results with those ob-

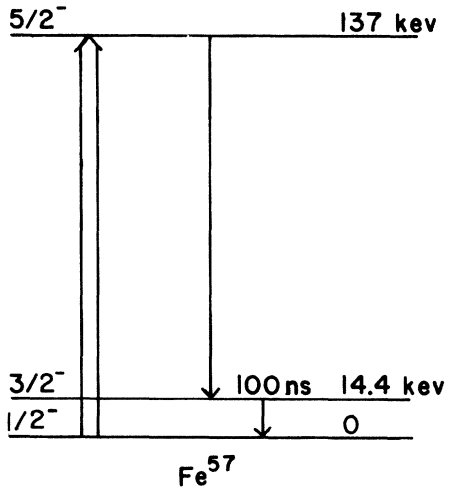

FIG. 1. Energy-level diagram indicating Coulomb excitation of the 137-keV level. The 14.4-keV γ rays were detected in coincidence with the preceding 122-keV γ rays.