MÖSSBAUER HYPERFINE SPECTRA OF Ta^{181} IN Ta AND IN W METALS*

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The possibility of using the 6.8-μsec half-life,[1] 6.2-keV[2] γ ray of Ta^{181} for Mössbauer effect studies has been recognized for some time; however, only one case of any success has been reported.[3,4] The appeal of this resonance lies in the fact that the natural linewidth Γ_N relative to the energy E of the γ ray is $\Gamma_N/E = 1.1 \times 10^{-14}$, or about 30 times narrower than that found for Fe^{57}. As well, a recoil-free fraction f approaching one is expected even at room temperature. Mössbauer-effect studies using Ta^{181} might shed some new light on the character of such a deformed nucleus and on the nature of chemical bonding in Ta compounds.

A Kr-filled proportional counter was used for (somewhat) improved detection of the 6.2-keV γ rays in the presence of the nearby Ta x rays.[2] An energy of 6.15 ± 0.06 keV was assigned to the gamma ray based on an "on-off" experiment in which pulse-height spectra of the radiation from a source of W^{181} (Ta^{181} parent) in Ta through a 5-mg/cm^2 Ta^{181} absorber were taken at Doppler velocities of ± 7.2 and 0 mm/sec. These two spectra were identical except for a peak at 6.16 keV where a 2% change showed that resonance absorption had taken place. Muir[2] has recently remeasured the energy to be 6.2 ± 0.1 keV using a coincidence technique.

The Mössbauer spectrum of our best Ta source and Ta absorber is presented in Fig. 1(a). The central peak has a linewidth of about 1.0 mm/sec, which is to be contrasted with the ideal linewidth $2\Gamma_N = 0.007$ mm/sec. The source of this broadening is no doubt due to quadrupole effects despite the "nominally" cubic environment. A velocity sensitivity to quadrupole effects some 700 times greater than that found for Fe^{57} is expected due to the large quadrupole moment of Ta, a large Sternheimer antishielding factor, and the lower γ-ray energy. Furthermore, Ta metal is extremely difficult to free of such interstitial impurities as O_2, N_2, and C, which likely produce the observed field gradients.

The resonance strength for Ta corrected for x rays by a background factor of 2.5 is 0.05 for the strongest dip. For such broad lines the area under the resonance curve is a better measure of f. For our case, provided there is no self-absorption, one calculates an area $= (\pi/2) f_s f_a \sigma_0 n \Gamma_N = 37\Gamma_N$, which is applicable when the resonance absorption is very small. The f subscripts refer to source and absorber, σ_0 is the resonant cross section, and n is the thickness of the absorber in atoms/cm^2. The observed area is $31\Gamma_N$ with the above background correction. This is indicative that few, if any, hyperfine contributions lie outside the velocity scan used and that f is indeed very large. Quantitative comparisons are not possible unless the background correction factor is determined more precisely.

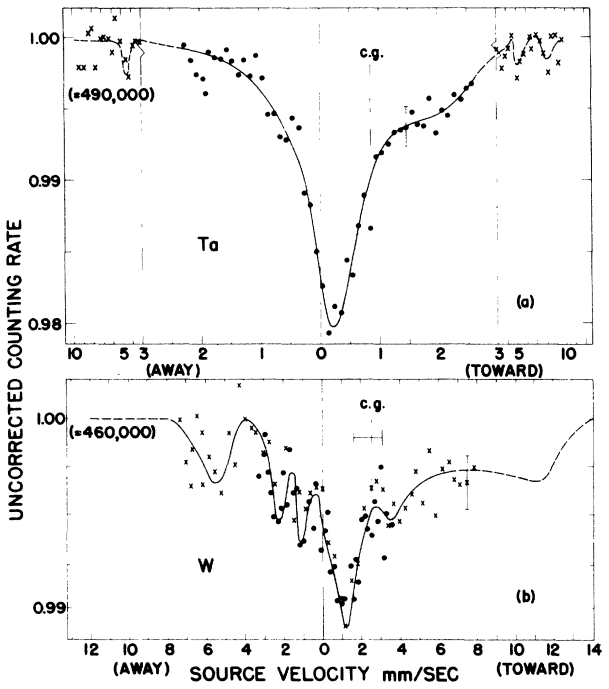

FIG. 1. (a) Mössbauer velocity spectrum of Ta^{181} in Ta versus treated Ta absorber. High drive data taken above 3 mm/sec have been normalized to same base line. (b) Similar data for Ta^{181} in W source. Results shown are a composite of three different velocity scans, data for two of which are shown. The center of gravity, c.g., of the spectrum as drawn is shown. Limits of error account for uncertainties in drawing the curves for high velocities and for an additional small peak farther out suggested by our crude quadrupole assignment.

The Mössbauer spectrum for a Ta^{181}-in-W source and the same Ta absorber is given in Fig. 1(b). Although the lines are broader than found for Ta-Ta, a partial resolution of a hyperfine structure is evident. The area under the curve is $43\Gamma_N$ after applying a background correction factor estimated to be 2, compared to a computed area of $38\Gamma_N$. If we assume the absorber hyperfine spectrum is narrow enough to be considered as a single line relative to the W source, the observed spectrum may be readily compared with the family of computed spectra[3] arising from quadrupole splitting for the known transitions from the excited to ground state assuming various values of the excited-state quadrupole moment Q_e. Similarities exist, but no single choice of field gradient and Q_e gave quantitative agreement.

In addition to the data presented in Fig. 1, resonance data have been obtained for several other Ta and W sources and Ta absorbers; generally, these earlier attempts yielded broader lines, weaker resonance effects, and slightly different chemical shifts[5] (center of gravity of spectrum), which could be crudely correlated with the analyzed O_2 content which varied from 500 to 2500 at. ppm. The strongest, narrowest line to date was obtained using a W single-crystal source and the Ta absorber used above. The slightly skewed single line had an uncorrected resonance strength of 4.5% and a linewidth of 0.26 mm/sec, and was located at 0.9 mm/sec (toward). Preliminary evidence for other weak lines at Doppler velocities below ±5 mm/sec prevent an accurate determination of the chemical shift. The activity in this weak source was deeply annealed, giving rise to a background correction factor greater than 4, and furthermore its low strength prevented a ready determination of the hyperfine spectra. Preliminary results from a Pd source with Ta absorber suggest a weak resonance, while none was seen with a Hf^{181} in HfC source and TaC absorber. No effect (greater than 0.2%) was observed for the W source and a TaC absorber at Doppler velocities less than ±0.5 mm/sec, whereas Cohen, Marinov, and Budnick[3] reported a 0.6 ± 0.15% effect at zero velocity. Their analysis implies the same chemical shift for Ta in W and in TaC.

Sources were prepared by evaporating about 1 mCi of activity in solution in HCl-HF onto the host, reducing in H_2 at 800°C, and diffusion annealing in high vacuum (about 10^{-6} Torr) at an elevated temperature. W sources, contained in a Ta crucible, were heated for 90 minutes at about 2250°C. The single-crystal source was treated at 2500°C. Ta sources were treated at 2000°C for 30 minutes. Absorbers, 0.0003 in. thick, were treated at 2300-2400°C for 1-2 hours at $(1\text{-}2)\times10^{-6}$ Torr and then etched in HNO_3-HF to final thickness.

The deviations of the center of gravity of the spectra from zero represent the chemical shifts and are indicated in Fig. 1. The chemical shift shown for Ta^{181} in "identical" chemical environments is consistent with a sizable change in nuclear radius ΔR associated with the 6.2-keV transition,[6] as well as a large sensitivity of a high-Z atom with low γ-ray energy to changes in electron configuration. We note that Fe, when dissolved in a host with more outer d and s electrons, suffers a decrease in $\psi^2(0)$, due possibly to an increase in the $3d$ electronic wave functions and an increased shielding of the s electrons. $\psi^2(0)$ is the electronic wave function at the nucleus. Apparently the same is true of Ta dissolved in W (taking $\Delta R^2/R^2 > 0$), where we observe a decreased energy relative to Ta.

The observation[7] of nuclear magnetic resonance in Ta foils shows that sources and absorbers having narrower lines should be possible, allowing accurate measurements of excited-state moments. From the chemical shifts for Ta in two or more known valence states [where the changes in $\psi^2(0)$ may be accurately assigned], one will be able to determine $\Delta R^2/R^2$. Knowing $\Delta R^2/R^2$, one can determine $\psi^2(0)$ for other compounds such as TaC, whose chemical bonding is much debated.

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EVIDENCE FOR THE PHOTOPRODUCTION OF $Y=0$ STATES WITH MASSES GREATER THAN 1900 MeV*

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This Letter reports evidence for the existence of $Y=0$ states with masses greater than 1900 MeV obtained from studies of the associated photoproduction of K^+ mesons with neutral $Y=0$ hyperons.[1] The experiments were carried out at the Cambridge electron accelerator. The reactions studied were[1]

$$\gamma + p \rightarrow K^+ + Y^*, \qquad (1)$$

in which the production angle and momentum of the K^+ were measured. Identification of the various $Y=0$ states is based on kinematics and presumes that the reactions proceed primarily through the two-body final states denoted by Eq. (1).[2] This method has been used successfully in the past at photon energies to 1.2 BeV.[3] Its applicability to the higher energies of this experiment is discussed below.

The K^+ mesons were photoproduced on a liquid hydrogen target by an external bremsstrahlung beam from the accelerator and detected by a momentum-velocity analysis system consisting of bending magnets, quadrupole lenses, scintillation counters, and two differential gas Cherenkov counters in series. Measurements are reported for a central momentum of 2.63 BeV/c at two laboratory angles: 5°, with maximum bremsstrahlung energies ranging from 4.0 to 6.0 BeV; and 10°, with maximum bremsstrahlung energies from 3.0 to 6.0 BeV. Complete Cherenkov pressure curves were taken at several of these points to ascertain the contribution of the pion tail to the measured K^+ rates. In no case did this pion background exceed 10%. A typical pressure curve is shown in Fig. 1. Experimental details, as well as a complete analysis of all the data, will appear in a later publication. The present Letter is concerned chiefly with the probable existence of the new, high-mass states.

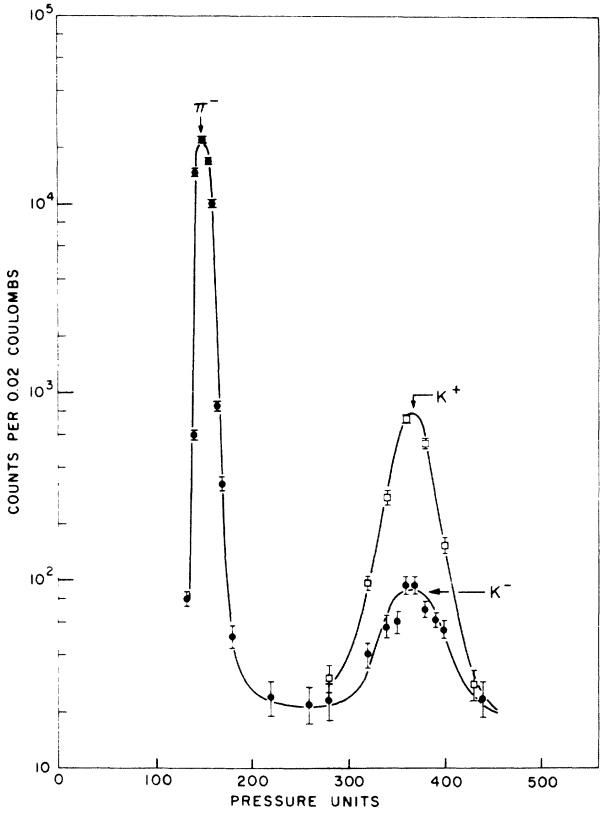

FIG. 1. A representative Cherenkov-counter pressure curve for pions and kaons. Two Cherenkov counters placed along the line of flight of the particles were connected electronically in a coincidence arrangement. The momentum of the particles was 2.63 BeV/c. The difference in widths for the pions and kaons is due to the momentum spread of the particles which was 10%.

Reaction (1) is the simplest Y^* photoproduc-