THE MÖSSBAUER EFFECT ON TUNGSTEN ISOTOPES

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We have investigated the recoilless nuclear resonance absorption in the 100.09 keV level of W^{182} and the 99.07 and 46.48 keV levels of W^{183}, as a function of relative velocity of source and absorber. The experimental arrangement permitted us to measure both by the usual method, in which one resolves the line with a scintillation counter, as well as by using a Du Mond type focusing crystal diffraction γ spectrometer with a resolving power of a few tenths of a per cent. The Debye temperatures which were determined from the magnitude of the effect were 280^{+30}_{-10} deg for the source (tantalum metal), and 320^{+70}_{-40} deg for the absorber (tungsten metal). Contrary to the conclusions of de Nercy et al. [4] the observed effect was never less than the theoretical value.

The total widths of the levels were measured. The width of the 100.09 keV level agrees with the data found from Coulomb excitation, [7] and from delayed coincidences, [6] but contradicts the result of an earlier experiment on nuclear resonance absorption in W^{182} [5] The experimental values of the widths (see the table) are in good agreement with theory. [8]

IN studying recoilless nuclear resonance absorption (Mössbauer effect), the resonance line is usually resolved by using a scintillation counter.* In the case of a complex spectrum containing intense lines which are close to the resonance line, the low resolving power of the scintillation counter makes the experiment impossible. In order to get rid of this limitation, to resolve the line we have used a Du Mond type focusing crystal diffraction spectrometer of high resolving power. [2,3]

DESCRIPTION OF APPARATUS

The arrangement of our equipment is shown in Figs. 1 and 2. The source S is on the focal circle. The γ radiation is filtered by the absorber X and falls on the bent crystal C. Diffracted quanta whose wave length satisfies the Bragg condition pass through the multislit collimator K and are recorded by the scintillation counter D. The source 7 and absorber 11 (Fig. 2) are fixed to copper cold pipes whose lower ends are immersed in liquid nitrogen 9 contained in the Dewar 4. Their temperatures are regulated by means of the copper-constantan thermocouples T_1 and T_2. The cold pipe for the absorber is connected to that for the source through the side rods 8 with strap spring couplings.

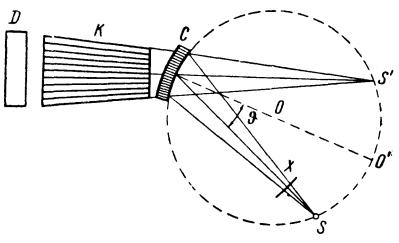

FIG. 1. Diagram of the apparatus.

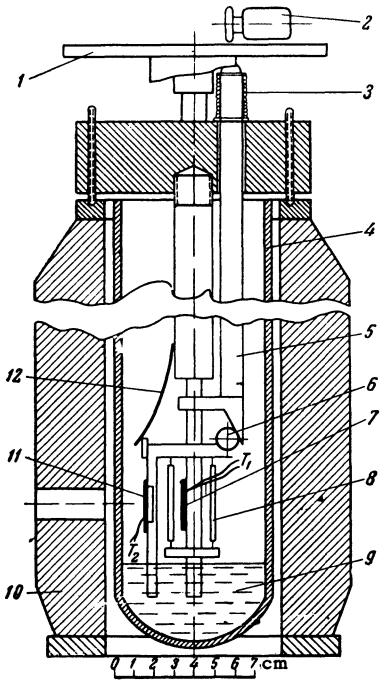

FIG. 2. Construction of cryostat and arrangement for moving the absorber.

*An exception is the work of Mitrofanov and Shpinel', [1] who used a magnetic spectrometer for analysis of the conversion electrons emitted following the resonance absorption.

The system as a whole forms a parallelogram, which produces the displacement of the source relative to the absorber. By means of the spring 12, the upper edge of the parallelogram is pressed through the roller 6 against the flat inclined surface of the beam 5. The beam carries out a sawtooth motion with a velocity which is constant (in absolute value), being pressed by the spring 3 against the special screw profile below the disk 1. The disk is put in motion by the dc motor 2. The required range of velocities is fixed by choosing the distance from the motor roller to the center of the disk. The velocities are also changed by changing the motor voltage. The apparatus permits the selection of any relative velocity of source and absorber within the limits 0.01—3 cm/sec. The placing of source and absorber in the same Dewar and their close coupling is a guarantee that the relative vibrations will be small.

The control experiments consisted in recording the currents in a small coil attached to the absorber and placed in a uniform magnetic field. They showed that the velocity remains constant in absolute value (changing sign twice during a revolution of the disk) to within ± 10%. The additional errors due to the presence of vibrations were smaller than this throughout the whole range of velocities. The apparatus described was fixed rigidly to a lead shield 10, mounted on the source carriage of the diffraction spectrometer.

The pulses from counter D (Fig. 1) are amplified and, after passing through a single-channel differential discriminator, go through a commutator arrangement to the input of one of five counting channels. The counting channels are switched on once a minute simultaneously with the switching on of the voltage driving the motor 2, so that each channel corresponds to a definite velocity of the absorber relative to the source. The channels were not switched when the sign of the velocity changed, i.e., only the absolute value of the velocity was fixed, and not its sign. We determined the relative difference of the numbers of pulses accumulated in the channel corresponding to the particular velocity and in the zero velocity channel, $(n_V - n_0)/n_V$. Data could be taken for four different velocities simultaneously (the fifth channel corresponded to zero velocity). Sometimes, in accumulating pulses corresponding to a selected velocity, data from several channels were taken. The apparatus was designed for continuous round-the-clock operation.

RESULTS OF MEASUREMENTS

For the first investigations we chose W^{182} and W^{183}. After capture of thermal neutrons by the tantalum source, one forms the two active nuclides Ta^{182} and Ta^{183}: $Ta^{181}(n,\gamma)Ta^{182}(n,\gamma)Ta^{183}$. Using such a source with an absorber of natural tungsten, one could expect to observe resonance absorption in the 100.09 keV level of W^{182} and in the 46.48 and 99.07 keV levels of W^{183}.

The Mössbauer effect on the 100 keV level of W^{182} has been observed previously (cf. [4,5]). The source used was metallic tantalum irradiated with thermal neutrons; the absorber was a foil of metallic tungsten. Tantalum and tungsten have cubic lattices and are not ferromagnetic; there should therefore be no Zeeman or Stark effects. One should therefore expect that the level is not split and has the natural width determined by its lifetime. It would seem therefore that the resonance absorption effect should be described accurately by the simplest theory. But according to the data of de Nercy et al. [4] the effect appeared to be 2—3 times smaller than calculated. This result together with similar data obtained by these au-

Level	Total level width Γ, 10^{-7} eV			
	experiment		theory	
	data of other authors	our data	omitting (1/2–3/2) interaction	including (1/2–3/2) interaction[8]
100.09 keV W^{182}	3.6 [6] 3.6 [7] * 7.3 [5]	3.6 ± 0.9 ($T_{1/2} = 1.3 \cdot 10^{-9}$ sec)	4,0	
99.07 keV W^{183}	7.9 [7] **	8 ± 3 ($T_{1/2} = 5.7 \cdot 10^{-10}$ sec)	8.9	7.4
46.48 keV W^{183}		31 ± 3 ($T_{1/2} = 1.5 \cdot 10^{-10}$ sec)	23	27

*Value obtained for $\alpha = 4.0$. Recalculation for the more likely value $\alpha = 5.0$[11] gives 5.1×10^{-7} eV.

**Obtained for $\alpha = 4.0$. Recomputation for $\alpha = 5.0$ gives 11×10^{-7} eV.

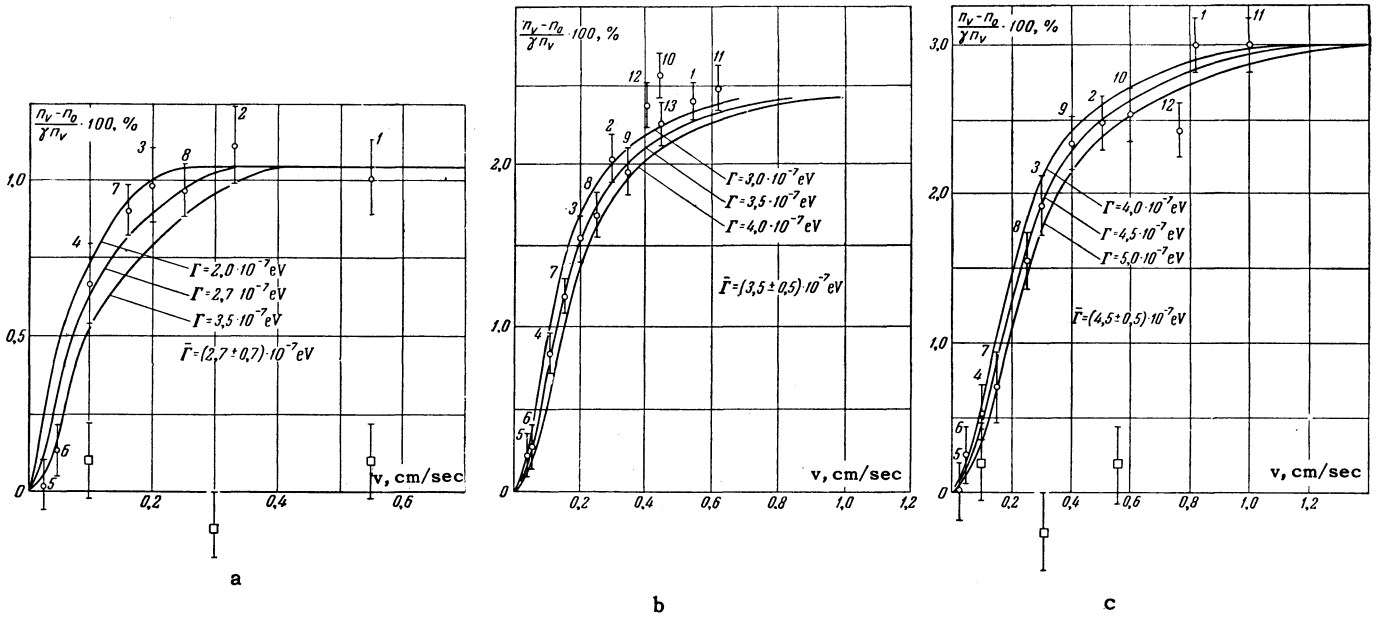

FIG. 3. Effect on the 100.09 keV level of W^{182} (T_s = 111° K, T_a = 101° K) for absorber thicknesses: a) x = 2.4 × 10^{-3} cm; b) x = 7.2 × 10^{-3} cm, c) x = 18.6 × 10^{-3} cm).

thors for various other transitions led them to the conclusion that the present theory in general gives too high values for the probability of recoilless absorption (or emission) for relatively longlived levels ($T_{1/2} \gtrsim 10^{-9}$ sec) and gives too low values for shortlived levels ($T_{1/2} \lesssim 10^{-10}$ sec).

Lee et al [5] studied the dependence of the resonance absorption cross section on the relative velocity of source and absorber. They found a total level width of Γ = 7.3 × 10^{-7} eV, whereas experiments on delayed coincidences [6] and Coulomb excitation [7] gave Γ = 3.6 × 10^{-7} eV (cf. the table). In view of the abovementioned absence of internal magnetic fields and electric field gradients, it is difficult to explain such a broadening, so the authors were inclined to attribute it to experimental errors.

So far as we know, resonance absorption in levels of W^{183} has not been observed previously. The lifetimes of the levels were also not known (the reduced probability for Coulomb excitation B(E2) had been determined [7] for the 99 keV level). Besides, a knowledge of them is of some interest, since the rotational spectrum of W^{183} is not trivially simple, because of the interaction of the K = ½ and K = ³⁄₂ bands. [8] Furthermore, a comparison with experiment of the theoretical predictions of the magnitude of the recoilless resonance absorption for three different levels of the same source-absorber pair seemed to us to be a good test of the accuracy of the theory and a test of the justification for proposed corrections. [4]

The source for our experiments was a piece of tantalum wire 0.2 mm in diameter, 30 mm long, irradiated in the water-water modernized reactor (VVRM) at a flux of ~ 3 × 10^{14} neuts/cm² sec. The absorption in the 100 keV level of W^{182} was investigated after a suitable delay for decay of the Ta^{183} in the irradiated sample ($T_{1/2}$ = 5.2 d). The spectrum was then relatively simple, and the measurements were carried out in the direct beam (source in position S′ in Fig. 1), with the desired line selected by the scintillation counter. The results obtained with tungsten absorber foils of thickness $x_1 = (2.4 \pm 0.2) \times 10^{-3}$, $x_2 = (7.2 \pm 0.6) \times 10^{-3}$ and $x_3 = (18.6 \pm 0.8) \times 10^{-3}$ cm, are shown in Figs. 3a, b, c, respectively. The circles show the points obtained when cooled with nitrogen (T_S = 111 ± 2° K, T_a = 101 ± 2° K), the squares are data for control points corresponding to source and absorber at room temperature. The experimental values are corrected for the fraction γ of the pulses from the line being investigated, in the total number of quanta recorded by the scintillation counter (γ = 48 ± 4, 45 ± 4, 37 ± 5% respectively for x_1, x_2, and x_3).

The relative difference in the counting rates corresponding to saturation ($n_v \to \infty$) and zero velocity (n_0) is given by the well known relation (cf., for example, [10])

$$\frac{n_\infty - n_0}{n_\infty} = f\left[1 - \exp\left(-\frac{f' N\sigma_0 x}{2}\right) J_0\left(\frac{if' N\sigma_0 x}{2}\right)\right];$$

$$\sigma_0 = \frac{\lambda^2}{2\pi}\,\frac{2I'+1}{2I_0+1}\,\frac{1}{1+\alpha}. \qquad (1)$$

Here f is the fraction of quanta emitted without recoil, f′ the fraction of quanta absorbed without re-

coil, $N = (N_A/A)\rho p$ is the number of absorbing nuclei per cc of absorber, p is the fraction of the resonant isotope in the absorber, x is the absorber thickness, J_0 is the Bessel function, λ the wavelength of the resonance radiation, I_0 and I are the spins of the ground state and excited state, and α is the total internal conversion coefficient for transition from the excited state.

The data in Figs. 3a, b, c give three nonlinear equations for determining the two unknowns f and f'. A graphical solution of the equations with $\alpha = 5.0$ [11] and p = 26.4% gives the values $f = 0.040^{+0.020}_{-0.004}$, $f' = 0.083^{+0.060}_{-0.030}$. This result was found using the data for the thinnest and thickest absorbers. The errors represent the differences between this solution and the two other solutions of the same equations.

Using the expressions for f and f'

$$f = \exp\left\{-\frac{6R}{k\Theta}\left[\frac{1}{4} + \Phi\left(\frac{\Theta}{T}\right)\Big/\frac{\Theta}{T}\right]\right\}, \qquad \Phi\left(\frac{\Theta}{T}\right) = \frac{T}{\Theta}\int_0^{\Theta/T}\frac{t}{e^t-1}\,dt, \tag{2}$$

we find $\Theta_{Ta} = 283^{+28}_{-6}$ and $\Theta_W = 320^{+70}_{-40}$ deg. In Eq. (2), R is the recoil energy, k is the Boltzmann constant, Θ is the Debye temperature and T the temperature of the source (or absorber).

Values of the function Φ are tabulated (cf., for example, [9]). Typical values of these quantities, obtained by other methods, [12] are $\Theta_{Ta} = 245°$, $\Theta_W = 310°$. Our values are no smaller than the usually accepted values, and consequently the experimental values of f and f', which give the magnitude of the recoilless resonance absorption in the 100 keV level of W^{182} are in any case not smaller than the theoretical values.

The solid curves in Fig. 3a, b, c were calculated from the relations [10]

$$\frac{n_v - n_0}{n_v} = f\frac{S(v) - S(0)}{S(\infty)\,[1 - fS(v)/S(\infty)]};$$

$$S(v) = \int_{-\infty}^{+\infty}\frac{1}{1+\varepsilon^2}\exp\left\{-\frac{f'N\sigma_0 x}{1+(\varepsilon+\varepsilon_v)^2}\right\}d\varepsilon,$$

$$S(0) = \int_{-\infty}^{+\infty}\frac{1}{1+\varepsilon^2}\exp\left\{-\frac{f'N\sigma_0 x}{1+\varepsilon^2}\right\}d\varepsilon, \qquad S(\infty) = \int_{-\infty}^{+\infty}\frac{1}{1+\varepsilon^2}\,d\varepsilon,$$

$$\varepsilon = \frac{E - E_0}{\Gamma/2}, \qquad \varepsilon_v = \frac{v}{c}\frac{E_0}{\Gamma/2}, \tag{3}$$

where E is the energy, E_0 the resonance energy, Γ the total width of the resonance, v the velocity of the absorber relative to the source, and c the velocity of light.

In Fig. 3a, b, c we indicate the values of Γ, which serves as a parameter, and the average value $\bar{\Gamma}$. The final result (see the table) was obtained by taking the average of all the $\bar{\Gamma}$, where the error is taken to be the deviation of the two extreme values of $\bar{\Gamma}$ from the average value. Our value agrees with the data obtained from delayed coincidence measurements [6] and from Coulomb excitation. [7] The results of Lee et al [5] are apparently incorrect.

In investigating the resonance absorption in the levels of W^{183} at 99 and 46 keV, the lines of interest could not be resolved by the scintillation counter because of the complexity of the spectrum, so the source was placed at position S corresponding to the maximum of reflection of the desired line from the bent crystal. The source was the same tantalum wire as in the experiment on W^{182}, but the experiment was begun soon after the end of the irradiation.

In Fig. 4 we show a portion of the spectrum obtained from the crystal diffraction spectrometer in the region of the 99 keV line of W^{183} and the 100 keV line of W^{182}. The result of the experiment on resonance absorption when we tune to the 99 keV line is shown in Fig. 5. The absorber was a

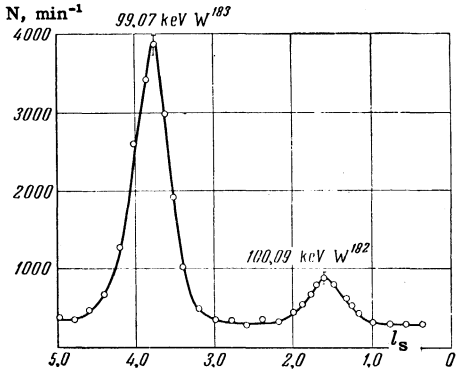

FIG. 4. Portion of spectrum obtained with the crystal diffraction spectrometer in the region around 100 keV

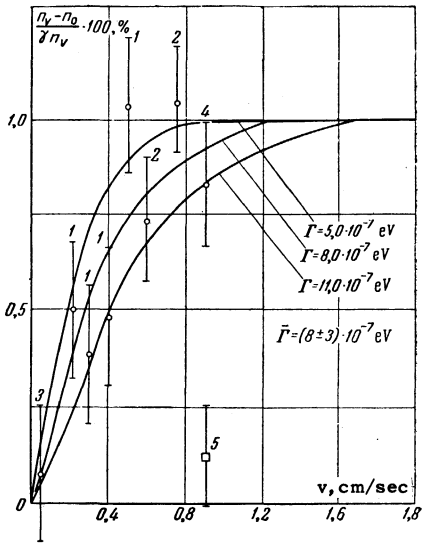

FIG. 5. Effect for the 99.07 keV level in W^{183} (T_S = 111° K, T_a = 101° K). Absorber thickness x = 7.2 × 10^{-3} cm.

7.2×10^{-3} cm thick foil, the same as in the experiment with W^{182}. The experiment was done at "nitrogen temperature" ($T_S = 111°$ K, $T_a = 101°$ K). The square point 5 indicates a control measurement. It was obtained with the diffraction spectrometer tuned to the 107.9 keV line of W^{183}, which corresponds to a transition to the excited level and consequently gives no resonance absorption.[11] Other conditions of the experiment were kept unchanged.

From the 99 keV level, one can have a transition either directly to the ground state or as a cascade through the 46 keV level. We then get an additional factor $(1 + \beta)^{-1}$ in the expression (1), where β is the ratio of the probabilities for cascade and direct transitions. Taking $\alpha = 5.0$, $\beta = 1.34$,[11] $p = 14.4\%$ and using the values $f = 0.40^{+0.020}_{-0.004}$, $f' = 0.083^{+0.060}_{-0.030}$ obtained from the experiment on W^{182}, we find from (1) the value $(n_\infty - n_0)/n_\infty = 0.53\%$, which is almost twice as small as the experimentally observed value (Fig. 5). In order to get agreement between the theoretical (solid curves in Fig. 5) and experimental results, one must use somewhat larger values of f and f', equal respectively to 0.05 and 0.13. This choice is not unique, but the nonuniqueness has little effect on the value of Γ, since the absorber is "thin" ($f'N\sigma_0 x \lesssim 0.46$) and the theoretical curves are close to a Breit-Wigner curve $y = \epsilon_V^2/(4+\epsilon_V^2)$ for any f' in the range $0 \lesssim f' \lesssim 0.13$. The value found for Γ is in good agreement with calculations starting from data on Coulomb excitation (cf. columns 3 and 2 in the table).

In Fig. 6a, b we show the curves obtained when we tune to the 46.48 keV line of W^{183}. We used an absorber thickness $x_1 = (2.4 \pm 0.2) \times 10^{-3}$ cm. The curves in Fig. 6a are for "nitrogen temperatures," those in Fig. 6b were taken with source and absorber at room temperature ($T = 300 \pm 2°$ K). The low energy of the transition and the large mass of the nucleus results in an extremely large value for the effect of recoilless resonance absorption in the 46 keV level of W^{183}. The limiting value $(n_\infty - n_0)/n_\infty$ which, according to computation, could be observed in our experiment, reaches ~ 28% at "nitrogen temperatures," and even with source and absorber at room temperature it is still ~ 7%. Unfortunately our equipment did not permit us to reach velocities greater than ~ 3 cm/sec, while a velocity of ~ 10 cm/sec is required for reaching saturation.

The solid curves were computed from relation (3) with $\alpha = 11.0$.[11] The values of f and f' were determined from (2), using the Debye temperatures

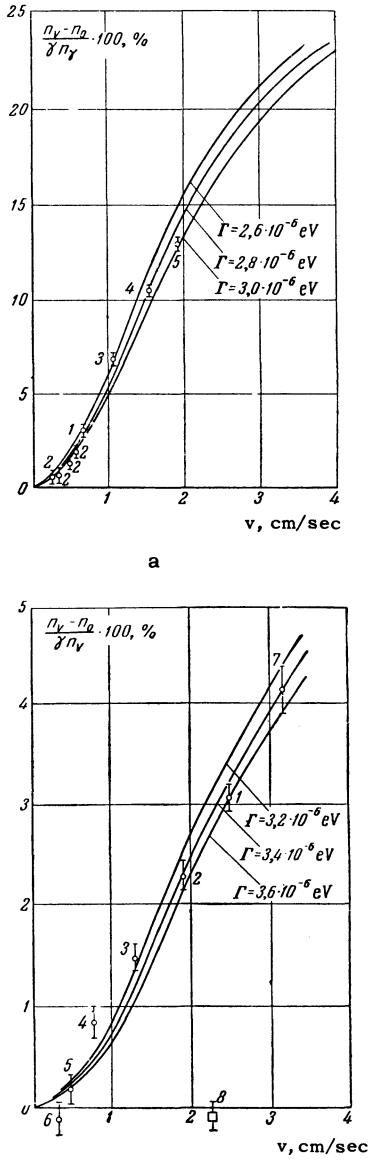

FIG. 6. Effect for the 46.48 keV level in W^{183}. Absorber thickness $x = 2.4 \times 10^{-3}$ cm: a) $T_S = 111°$ K, $T_a = 101°$ K; b) $T_S = T_a = 300°$ K.

$\Theta_{Ta} = 283°$ K and $\Theta_W = 320°$ K found in the experiment on W^{182}. The square point on Fig. 6b is a control point. It was found by tuning to the nearby line at 52.6 keV, corresponding to a transition to an excited level.[11] The average value of the total width Γ of the 46.48 keV level in W^{183}, as found from the data of Fig. 6a and b, is given in the table.

DISCUSSION OF RESULTS

The comparison made between the theoretical and experimental values of the recoilless resonance absorption effect have shown that for the

levels at 100 and 99 keV the observed effect is not lower (and possibly even somewhat higher) than predicted by theory. The value of the effect for the 46 keV level is also in agreement with theory, so far as one can judge from our incomplete curves. Thus our data contradict the results of de Nercy et al,[4] at least that part which refers to the 100 keV transition in W^{182}.

The experimental total line widths can be compared to the theoretical values. For the case of W^{183}, such a comparison was made only for the absolute probabilities of E2 transitions, which are known from experiments on Coulomb excitation. For M1 transitions there are data only on relative intensities.[8] The theoretical values of the total level widths as calculated by Kerman,[8] taking into account the interaction of the $\frac{1}{2}$ and $\frac{3}{2}$ bands in W^{183}, are given in column 5 of the table. In column 4 we give values calculated by us* on the usual theory, which does not include interaction between the $\frac{1}{2}$ and $\frac{3}{2}$ bands.[13] A comparison shows good agreement between theory and experiment.

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*In the computation we used the value $Q_0 = 6.5 \times 10^{-24}$ cm² for the quadrupole moment. To determine B_0 and $(g_K - g_R)^2$, we used data on relative intensities of M1 and E2 transitions from the 99 and 207 keV levels of W^{183}.[11]

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