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¹⁰T. J. Schmutge and C. D. Jeffries multiply the electronic relaxation through forbidden transitions by a factor N_e/N_n which renders it always negligible; they also multiply W by N_e/N_n in the equation for the evolution of P_n . This is perfectly correct when there is a fast spin diffusion, but then w must be multiplied by the same factor, which disappears from the stationary

state equation.

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ISOMER-SHIFT MEASUREMENT IN W^{182}

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We report here on an unambiguous Mössbauer measurement of the isomeric shift of the $2^+ - 0^+$, 100-keV transition in W^{182} , observed between metallic tungsten and WCl_6 . The radius of the 2^+ rotational state is thus different from that of the 0^+ ground state. Preliminary results for the isomer shifts in W^{184} and W^{186} are reported at the end of this paper. It is impossible with our present limited understanding of the electronic wave functions in W and WCl_6 to determine a reliable value for the difference between the ground- and excited-state radii, ΔR . An estimate of $|\Psi_S^2(0)_W - \Psi_S^2(0)_{WCl_6}|$ is presented below, from which we conclude that the mean square radius of the 2^+ state is greater than that of the ground state and that the value of $\Delta\langle R^2 \rangle / R^2 \approx 1.3 \times 10^{-4}$. The uncertainty in this value is too great to distinguish between the value expected for a simple rotational stretching and that calculated by Udagawa and Sheline¹ in which they take account of the Coriolis antipairing effect. We present our results and analysis in the spirit of stimulating further investigations of isomer shifts between rotational states in even-even nuclei as well as encouraging additional work on electronic wave functions in the tungsten compounds.

It is well known that the rotational spectra of deformed even-even nuclei are characterized to a good approximation by a simple band structure of the form $E = \hbar^2 I(I+1)/2\mathcal{I}$. More exactly, there are deviations from this ideal rotational spectrum which become increasing-

ly important for high-spin members of the band. It was originally suggested² that most of the deviations could be accounted for by a higher order term proportional to $I^2(I+1)^2$ arising from a rotational-vibrational interaction. This centrifugal stretching would also produce an increase in nuclear deformation proportional to $I(I+1)$. However, attempts to account for the deviations from an $I(I+1)$ energy spectrum by including the contributions of beta and gamma bands have met with varying success. More recently, it has been suggested,³ within the framework of a pairing model applied to deformed nuclei, that the Coriolis force should reduce the effective pairing interactions and thus increase the moments of inertia of the higher angular-momentum states. This antipairing effect would not be expected to produce a corresponding change in the deformation. Thus the knowledge of the change in deformation, together with the deviations from the $I(I+1)$ energy rule, may elucidate the roles of the various interactions.

The change in nuclear deformation may be deduced from a measurement of the Mössbauer isomer shift. The importance of such measurements has stimulated searches for a positive isomer-shift effect between rotational states with, however, little success. Fink and Kienle⁴ have recently reported the observation of a small isomer shift between the 0^+ and first 2^+ states in both Gd^{156} and Gd^{158} , measured between the metal and the trivalent ion. Their

result, a shift of 0.03 ± 0.01 of the experimental linewidths, is subject to considerable uncertainty since their spectra are broad (about seven times natural width) and show unresolved hyperfine structure. The existence of small centroid shifts is markedly dependent on a detailed understanding of the hyperfine interaction. Moreover, it is possible to observe centroid shifts which are not isomeric if the source is polarized or if the crystal is anisotropic.

The Mössbauer absorption spectra of the 100-keV transition were observed at 4.2°K in tungsten metal and tungsten-hexachloride absorbers using a Ta^{182} source in tantalum metal. The results are shown in Fig. 1(a) and 1(b). Both absorbers show unsplit absorption lines whose widths are equal, within experimental error, to the natural linewidth $2\Gamma = 0.20$ cm/sec. The centroid of the WCl_6 resonance is clearly seen to be shifted towards negative velocities (i.e., source moving away from absorber). The resonant energy in tungsten metal is thus greater than in WCl_6 . The magnitude

of the shift is 0.25 ± 0.05 mm/sec, corresponding to an energy change of about 8×10^{-8} eV. The experiment has been repeated several times with the same results.

The usual expression used for the isomeric shift⁵ is

$$\Delta E = (2\pi Z e^2 / 5) S'(Z) R^2 \times \{ \Psi_s^2(0)_I - \Psi_s^2(0)_{II} \} \{ \Delta \langle R^2 \rangle / R^2 \} \quad (1)$$

where the first expression in brackets is the change in *s* electronic wave function at the nucleus between absorber I and II, and the second is the fractional change in mean square radius between excited and lower states. $S'(Z)$ is a relativistic correction. There is little doubt that the value of the *s* wave function at the nucleus in W metal is greater than it is in WCl_6 ; the free-atom configuration of tungsten is $(5d)^4(6s)^2$; thus $\Delta \Psi_s^2(0)$ is positive. From the sign of the energy shift we conclude that ΔR is positive; that is, the 2^+ state has a greater mean-square radius than the ground state.

An accurate estimate of the magnitude of $\Delta \Psi_s^2(0)$ requires at least a knowledge of (1) the degree of covalent bonding as well as its effects on the *s* wave function density for WCl_6 ; (2) the shielding effect of the *5d* and *6s* electrons on the other *6s* electrons; (3) the shielding effect of the *5d* on the *5s* electrons (which can result in a reversal of the sign of the isomer shift⁵); and (4) the number of localized *6s* electrons in W metal. None of this information is really known. A detailed examination of isomer shift versus compound structure might result in firm values, but this information is not now available. The following estimate is based on much-used simple recipes.⁵ We assume that the covalent bonding can be obtained from electronegativity and bond-length arguments.⁶ We would then conclude that WCl_6 is about 50% ionic. We further assume that the 50% covalent bonding does not contribute to the *s* electron density nor does it materially modify the shielding estimates. We also assume, following Brix and Kopferman,⁷ that each *5d* and (other) *6s* electron gives a 20% shielding effect on the *6s* electrons. The shielding effect of the *5d* on the *5s* electrons is ignored. Finally we assume that both *6s* electrons in W metal are effective. With these assumptions and taking the values of $S'(Z) = 5.5$ and $\Psi_{6s}^2 = 0.71 \times 10^{26}$ from Ref. 5, we obtain $\Delta \Psi_s^2(0) = 0.2 \times 10^{26}$

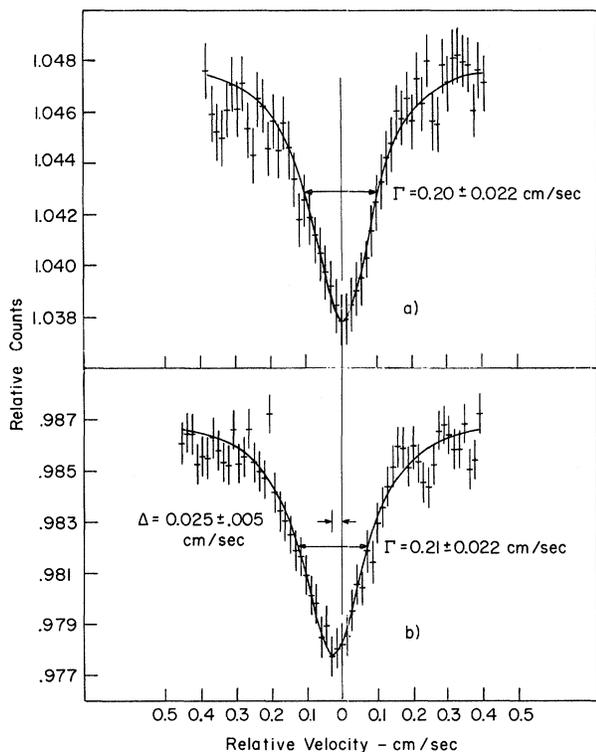


FIG. 1. Mössbauer spectra of the 100-keV, $2^+ \rightarrow 0^+$ transition in W^{182} taken at 4.2°K. (a) Natural tungsten absorber, 12 mg/cm². (b) Natural WCl_6 absorber, 115 mg/cm².

and $\Delta\langle R^2\rangle/R^2 = 1.3 \times 10^{-4}$.

If we make the usual assumptions that the nucleus is axially symmetric and incompressible then, following Kopferman,⁸ we can replace $\Delta\langle R^2\rangle/R^2$ by $2\alpha\Delta\alpha$, where α is the parameter describing the surface deformation and given by $R(\theta) = R[1 + \alpha P_2(\cos\theta)]$; α is related to the more commonly used deformation parameter β by the approximate relation $\alpha \approx 2\beta/3$, so that $\Delta\beta/\beta = (9/4)\alpha\Delta\alpha/\beta^2$. Taking β for W^{182} as 0.24,⁹ we obtain

$$(\Delta\beta/\beta)_{\text{I.S.}} = 2.5 \times 10^{-3}, \quad (2)$$

where the subscript "I.S." means as determined from the isomer shift.

This crude value is approximately that expected from the known deviations of the W^{182} rotational level from the $I(I+1)$ rule assuming only a rotational stretching mechanism. The model we use for centrifugal stretching is essentially that used by Diamond, Stephens, and Swiatecki¹⁰ and by Davidov and Chaban¹¹ (and analogous to that used for molecules). That is, we assume harmonic forces, $\mathcal{J} = 3B\beta^2$, and only coupling with the β vibrational band. Then

$$E = \frac{1}{2}C(\beta - \beta_0)^2 + (\hbar^2/6B\beta^2)I(I+1), \quad (3)$$

where β_0 is the ground-state deformation. To find the rate of change of the equilibrium deformation, we set the differential with respect to β equal to zero and obtain

$$\Delta\beta/\beta = (\hbar^2/3BC\beta^4)I(I+1). \quad (4)$$

Eliminating C from Eqs. (3) and (4), we obtain the energy as a function of the change in equilibrium deformation,

$$E = \frac{\hbar^2}{2} \frac{I(I+1)}{\mathcal{J}} \left[1 - \frac{\Delta\beta}{\beta} \right]. \quad (5)$$

From the precisely measured energies of the 2^+ and 4^+ state, 100.09 and 329.36 keV, respectively,¹² we obtain for the 2^+ state, $(\Delta\beta/\beta)_{\text{energy}} = 4 \times 10^{-3}$, somewhat larger than the value estimated from the isomer-shift data.

In the accompanying Letter, Udagawa and Sheline argue that the approximation $\mathcal{J} \propto \beta^2$ is a poor one for W^{182} and that the deviation from the $I(I+1)$ energy rule arises principally from the Coriolis antipairing effect. Their "microscopically" calculated value of $\Delta\beta/\beta = 2.0 \times 10^{-3}$ is in somewhat better agreement with $(\Delta\beta/\beta)_{\text{I.S.}}$. The approximations necessary to obtain this

latter value are too crude to allow a decision between the two basically different interpretations.

A more meaningful experiment at this time is the measurement of the variation of $\Delta\beta/\beta$ on going from W^{182} to W^{184} and W^{186} and such measurements are in progress. The energies of the transitions (111 and 122 keV for W^{184} and W^{186} , respectively) are high, and the Mössbauer resonant absorptions are small ($\sim 0.25\%$). The following are preliminary results¹³: Using a Re^{184} -in- W source, which yielded broadened lines, the observed isomer shift between WCl_6 and W metal is 0.17 ± 0.46 mm/sec. Using a Re^{186} source, lines of approximately natural width are obtained for W^{186} in W metal and WCl_6 ; the observed isomer shift is -0.10 ± 0.22 mm/sec. Calculations based on Eq. (5), as well as those of Ref. 1, show that the isomer shift in W^{186} should be approximately twice that in W^{182} , i.e., ~ 0.5 mm/sec. Our preliminary result implies an even smaller centrifugal stretching contribution in W^{186} relative to W^{182} than that calculated in Ref. 1.

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CENTRIFUGAL-STRETCHING AND CORIOLIS-FORCE ANTIPAIRING EFFECTS IN $W^{182}\dagger$

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In the preceding paper,¹ Cohen *et al.* have reported observing unambiguously the Mössbauer isomer shift of the $2^+ - 0^+$, 100-keV transition in W^{182} . Unfortunately the difference of the s -electron density at the W nucleus between the emitter and the absorber is not well known. Therefore, it is somewhat difficult to extract from the measured isomer shift with high accuracy the information about the nuclear factor, the change in mean radii, $\delta\langle R^2 \rangle / R^2$, between the isomeric pair. However, Cohen *et al.*¹ have estimated the change in the s -electron density and deduced the $\delta\langle R^2 \rangle / R^2$ value. The result obtained is

$$\delta\langle R^2 \rangle / R^2 \approx 1.3 \times 10^{-4}. \quad (1)$$

Assuming the centrifugal stretching due to the rotation to be the origin of this change in the nuclear radii, one can extract $\Delta\beta$, the change in the deformation between the 2^+ and 0^+ states¹:

$$\Delta\beta = 6.1 \times 10^{-4}. \quad (2)$$

Following the argument given by Moszkowski² and Diamond *et al.*,³ one can also relate this $\Delta\beta$ to B_β , representing the contribution from the centrifugal stretching to the nonadiabatic parameter B obtained from fitting the ground state rotational spectrum to an expansion of the form

$$E_I = (\hbar^2/2\mathcal{I}_0)I(I+1) + BI^2(I+1)^2 + CI^3(I+1)^3 + \dots \quad (3)$$

The relation between $\Delta\beta$ and B_β obtained in this way is

$$B_\beta = -\frac{\hbar^2}{24\mathcal{I}_0^2} \frac{\partial \mathcal{I}}{\partial \beta} \Delta\beta. \quad (4)$$

Thus, if the derivative $\partial \mathcal{I} / \partial \beta$ were known one could obtain experimental information about the B_β value. This provides more detailed

information about the nonadiabatic effects of the nuclear rotation that cause deviations of the spectrum from the simple $I(I+1)$ rule.

The aim of our paper is, first, to show that the observed values of $\Delta\beta$ and B_β (obtained from the observed $\Delta\beta$ value with the use of a calculated value of $\partial \mathcal{I} / \partial \beta$) can be successfully reproduced from a realistic calculation based on the microscopic nuclear model,⁴ and second, to show that the rest of the experimental value of B , i.e., $B - B_\beta$, can be well explained in terms of the Coriolis-force antipairing (CAP) effect.⁵

For these purposes, following a method recently proposed by the present authors,⁶ calculations of the centrifugal stretching and the CAP effects were made on the lowest order nonadiabatic parameter A and the values of $\Delta\beta$ and $\partial \mathcal{I} / \partial \beta$. The parameter A is given by

$$A = -2\mathcal{I}_0^4 B / \hbar^8. \quad (5)$$

The method proposed in Ref. 6 is based on the cranking and the pairing-plus-quadrupole-force models. The detail has been described in Ref. 6 and will not be repeated.

However, two remarks about the choice of the parametric values used in the present calculations are in order: (i) The deformation parameter⁷ ϵ has been determined from the empirical intrinsic electric quadrupole moments, Q_e^{exp} , by using the relation

$$Q_e^{\text{exp}} = Q_e^{\text{theo}} = 2 \sum_i \langle p \rangle q_{ii}^{(0)} \rho_i^{(0)} \quad (6)$$

where $q_{ii}^{(0)}$ is the quadrupole moment of the single proton orbit i and $\rho_i^{(0)}$ is the occupation probability of nucleons in that orbit.⁶ (ii) The single-particle energies used in the present calculations are essentially the same as those