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QUADRUPOLE SPLITTING OF THE 6.2 keV GAMMA RAYS OF ^{181}Ta IN RHENIUM METAL*

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Abstract:

Recoilless nuclear resonance absorption of the 6.2 keV γ rays of ^{181}Ta was used to study the hyperfine interaction of ^{181}Ta in rhenium metal. From the completely resolved Mössbauer spectra measured at room temperature parallel and perpendicular to the [0001] axis of the single-crystalline ^{181}W(Re) sources, the sign and magnitude of the nuclear quadrupole interaction, $e^2qQ(7/2) = -(2.15\pm0.02)\ 10^{-6}$ eV, the quadrupole moment ratio, $Q(9/2)/Q(7/2) = 1.133\pm0.010$ and the isomer shift of ^{181}Ta(Re) relative to Ta metal, IS = $-(14.00\pm0.10)$ mm/s were derived.

The recent observation of large isomer shifts of the 6.2 keV γ rays of ^{181}Ta between sources of ^{181}W diffused into single crystals of the second and third row transition metals and a Ta metal absorber has demonstrated the high resolution inherent in this gamma resonance, when applied to solid state pro-

blems.[1] The total range of these isomer shifts is by far the largest in terms of the natural linewidth $\Gamma = \hbar/\tau$ ($\sim 10^4$ times Γ) or even in terms of the presently observed minimum experimental linewidth W ($\sim$500 times W)[2] compared to all the gamma resonances ever observed. The sensitivity of the 6.2 keV gamma resonance for magnetic hyperfine studies has already previously been demonstrated by measurements of the magnetic moment of the excited state in externally applied magnetic fields of the order of 1.5 - 3.0 kOe.[3,4] Likewise a very high resolution is expected for the measurement of electric quadrupole interactions (EQI) due to the large electric quadrupole moment of the ground state of ^{181}Ta, $Q(7/2) = +(3.9 \pm 0.4)$ b.[5] Despite these features the 6.2 keV gamma resonance has not been applied to studies of magnetic dipole or electric quadrupole hyperfine interactions in solids up to now. This is at least partly connected with the difficulties in observing the resonance at all, a fact which is a direct consequence of its high sensitivity.

The present paper reports on a measurement of the electric quadrupole splitting of the 6.2 keV γ rays of ^{181}Ta in sources of ^{181}W diffused into single-crystalline rhenium metal. This is the first application of the ^{181}Ta gamma resonance to nuclear quadrupole interaction studies. From the completely resolved split spectra the sign and magnitude of the electric field gradient (EFG) at the ^{181}Ta site in rhenium metal, the ratio of the quadrupole moments of the excited state to that of the ground state and the isomer shift of ^{181}Ta(Re) relative to Ta metal are derived.

The experimental technique employed was similar to the one described previously.[1,4] The ^{181}W activity was produced by neutron activation of 93% enriched ^{180}W metal in a total neutron flux of $2 \cdot 10^{21}$ n/cm^2. From a high-

purity single crystal of rhenium metal ($\rho_{300}/\rho_{4.2}$ = 10500)[6], oriented to within one degree by reflection of coherent light on an etched surface and subsequently by the Laue backscattering method, thin dics (about 1 mm thick) were cut with a spark cutter parallel and perpendicular to the [0001] axis. After electropolishing them in a solution of 6% perchloric acid in methanol, the dissolved ^{181}W activity was dropped onto the discs, dried and reduced in an atmosphere of H_2 at 850°C. Finally the diffusion was done in high-vacuum (at 10^{-7} Torr) for one hour at a temperature of about 2500°C. We used the same Ta metal absorber (4 mg/cm^2 thick) as in our previous work.[1,4] The Mössbauer transmission experiments were performed at room temperature with a sinusoidal electromechanical velocity drive and a 2048 channel analyzer operated in multiscaler mode[7], and the 6.2 keV γ rays were detected with an argon filled proportional counter.

The velocity spectra obtained are presented in Fig. 1. As the 6.2 keV transition has pure E1 character with the spin sequence 9/2→7/2, the hyperfine spectrum resulting from an EQI alone consists of 11 hyperfine components, of which seven are $\Delta m = \pm 1$ transitions, three are $\Delta m = 0$ transitions and one is a mixed transition. In the diagrams of Fig. 2 the dependence of the line-positions on the ratio of the quadrupole moments Q(9/2)/Q(7/2) is presented for an axially symmetric EFG, with the direction of observation perpendicular (a) and parallel (b) to its axis. By observing the emission spectrum parallel and perpendicular to the [0001] axis of the single-crystalline ^{181}W(Re) sources, the assignment of individual transitions to the observed lines could be done uniquely.

Both Mössbauer spectra were simultaneously least-squares fitted with a superposition of dispersion modified Lorentzian lines

$$N(v) = N(\infty)\left[1 - \sum_{i=1,11} \frac{A(i)\,(W/2)^2}{(v-v_o(i))^2 + (W/2)^2}\left(1 + 2\xi\,\frac{v-v_o(i)}{W/2}\right)\right],$$

where the amplitude 2ξ of the dispersion term was set equal to $2\xi = 0.30$[4]. (N = number of counts, W = experimental linewidth (FWHM), v = Doppler velocity, A(i) and $v_o(i)$ = amplitude and position of the ith component, respectively). The z-component of the axially symmetric electric field gradient eq, Q(9/2)/Q(7/2), the isomer shift IS, W/2, and for each of the two spectra $N(\infty)$ and an amplitude factor A_{tot}, describing the total summed effect, were used as free parameters for the 8 parameter fit. Within statistical accuracy the measured spectra are in agreement with the assumption of an axially symmetric EFG, expected from the point symmetry of the hexagonal rhenium lattice.

The results of the least-squares fit analysis are presented in Table 1, with negative signs for IS and $e^2qQ(7/2)$, since the measured interactions occurred in the source. The observed linewidth W = 0.60±0.04 mm/s reflects a considerable line broadening, only part of which is due to geometrical broadening (about 0.1 mm/s for the solid angle $\Omega = 4\pi/500$ used). Because of the source preparation technique the W concentration in the ^{181}W(Re) sources is expected to vary between zero and a maximum value, estimated to be less than 0.5%. This should also give rise to a sizable line broadening of the order of 0.1 mm/s, caused by the concentration dependence of the isomer shift.[8] Because of the line broadening the total resonance effect A_{tot}, summed over all components, is rather small (∿1%) in both spectra.

With the electric quadrupole moment of the $7/2^+$ ground state Q(7/2) = +(3.9±0.4)b[5] values for the electric quadrupole moment of the 6.2 keV state and for the EFG eq at the ^{181}Ta nucleus in rhenium metal can be derived. Using our experimental value

$$Q(9/2)/Q(7/2) = 1.133 \pm 0.010$$

we obtain

$$Q(9/2) = +(4.4 \pm 0.5)b$$

for the quadrupole moment of the $9/2^-$ excited state and

$$eq = -(5.5\pm0.5)\cdot10^{17} \quad V/cm^2$$

for the EFG at ^{181}Ta(Re) at room temperature.

The ground state and the 6.2 keV state of ^{181}Ta have been classified as intrinsic proton states with the Nilsson assignments $7/2^+[404]$ and $9/2^-[514]$, respectively.[9] Assuming the same intrinsic quadrupole moment for both states, the Nilsson model predicts a value of $Q(9/2)/Q(7/2) = 1.17$, which is slightly larger than our experimental value of 1.133 ± 0.010. Neglecting band mixing, we obtain for the ratio of the intrinsic quadrupole moments Q_o a value of $Q_o(9/2)/Q_o(7/2) = 0.97\pm0.01$, showing that the deformation of the $9/2^-[514]$ excited state may be slightly smaller than that of the $7/2^+[404]$ ground state.

Our value for the EFG at ^{181}Ta in rhenium metal may be compared with the results of nuclear specific heat[10] and nuclear acoustic resonance[11] measurements for pure rhenium metal. In agreement with our result the sign of the EQI was reported negative.[10] From the quadrupole coupling constant for ^{185}Re in rhenium metal and the quadrupole moment of the ground state of ^{185}Re ($Q(^{185}Re) = +(2.3\pm0.9)$ b[12]), a value of $eq = -(4.9\pm1.9)\cdot10^{17}$ V/cm^2 can be derived for the EFG in rhenium metal at 4.2 K. This value is in good agreement with our room temperature result for ^{181}Ta(Re), though of less accuracy due to the large error in the value for $Q(^{185}Re)$. The only other 5d element for which the EFG has been measured as an impurity in rhenium metal is iridium. From the quadrupole splitting of the 73 keV γ rays of ^{193}Ir(Re) again a value of $|eq| = 5\cdot10^{17}$ V/cm^2 has been obtained at 4.2 K.[13] Within experimental accuracy the EFG's for Re, Ta and Ir in rhenium metal agree with each other, though the value for ^{181}Ta(Re) has been measured at room temperature. In order to clarify the situation a measurement of the temperature dependence of the EFG of Ta(Re) is highly desirable. This can be

achieved over an unusually large temperature region using the 6.2 keV gamma resonance of ^{181}Ta.

The negative sign of the EFG is rather striking, since the direct contribution of the ionic cores of the hexagonal rhenium lattice, calculated with the help of deWette's tables[14] is positive and only of the order of $q_{latt} = 6 \cdot 10^{14}$ V/cm^2 per unit charge on the rhenium ion cores. This positive direct contribution should be greatly enhanced by the atomic Sternheimer anti-shielding effect.[15] The negative experimental value for the EFG reveals therefore the crucial role played by the conduction electrons, which obviously even change the sign of the direct lattice contribution by their strong shielding effect.[10] Such a strong negative contribution, caused by a repopulation of Bloch states of different symmetry at the Fermi surface under the influence of the perturbing lattice potential, has been studied theoretically by Watson et al.[16] According to their theory, it is linearly related to both q_{latt} and the density of electronic states at the Fermi energy. Therefore this "overshielding" effect should be most important for the transition metals with a high density of d-states at the Fermi level, and they predicted it to be of the order of $-100q_{latt}$ for the case of Re. Recently another strong indication has been found for a dominant contribution of the conduction electrons to the EFG in cadmium metal, measured by TDPAC technique.[17] The EFG found is considerably larger than the direct lattice contribution and shows a temperature dependence opposite to the one expected from the lattice expansion. On the other hand the EFG in hafnium metal was found to be positive,[18] and that for ^{181}Ta(Hf) exhibits only a relatively small temperature dependence of 5% between 4.2 K and 300 K.[19]

The above discussion shows, that presently only very few accurate

experimental data are available for the EFG's (including their signs) in hexagonal 5d transition metals. The superior resolution of the 6.2 keV γ resonance of ^{181}Ta may hopefully provide more such data in the near future.

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FOOTNOTES AND REFERENCES

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Table I. Summary of experimental results obtained by a simultaneous least-squares fit of spectra (b) and (c).

isomer shift IS	-(14.00±0.10)	mm/s
Q(9/2)/Q(7/2)	1.133±0.010	
e^2 q Q(7/2)	-(2.15±0.02) 10^{-6}	eV
linewidth W	0.60±0.04	mm/s
effect A_{tot}	1.30±0.18%	spectrum (b)
	0.94±0.18%	spectrum (c)

FIGURE CAPTIONS

Fig. 1. Velocity spectra of the 6.2 keV γ rays of ^{181}Ta in rhenium metal versus a Ta metal absorber, with direction of observation parallel (c) and perpendicular (b) to the [0001] axis. The solid lines represent the result of a simultaneous least-squares fit of both spectra. Positions and intensities of the individual components are represented in (a) by solid lines (Δm = ±1) and dashed lines (Δm = 0, perpendicular to [0001] axis), respectively.

Fig. 2. Dependence of the line positions expected for the 9/2(E1)7/2 ground state transition in ^{181}Ta on the ratio of the quadrupole moments Q(9/2)/Q(7/2) for an axially symmetric electric field gradient: (a) perpendicular to the axis of the EFG, and (b) parallel to it. The widths of the lines are proportional to the intensities of the components.

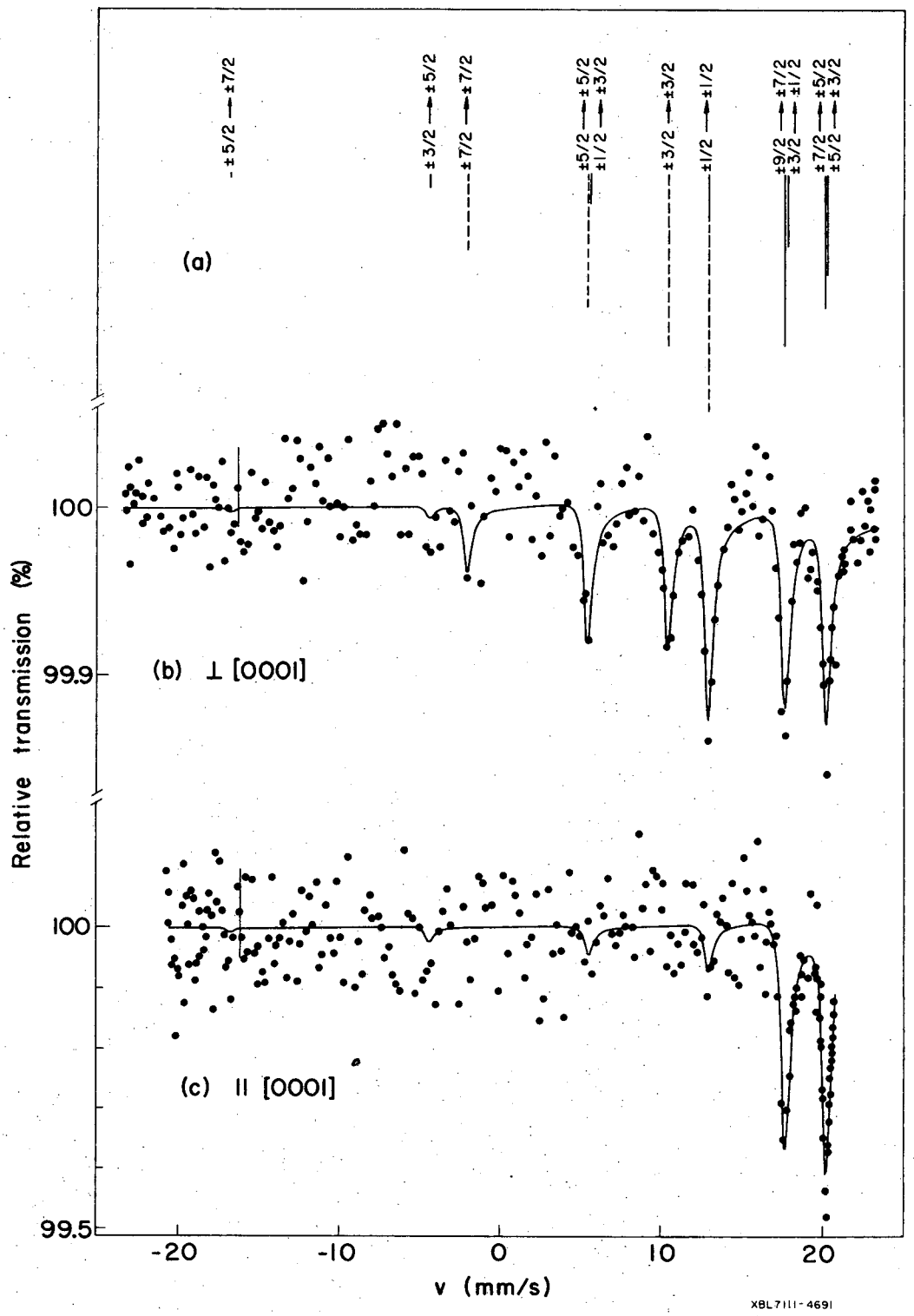
-±5/2 → ±7/2
±3/2 → ±5/2
±7/2 → ±7/2
±5/2 → ±5/2
±1/2 → ±3/2
±3/2 → ±3/2
±1/2 → ±1/2
±9/2 → ±7/2
±3/2 → ±1/2
±7/2 → ±5/2
±5/2 → ±3/2
(a)
(b) ⊥ [0001]
(c) ∥ [0001]
Relative transmission (%)
100
99.9
100
99.5
-20
-10
0
10
20
v (mm/s)
XBL7111-4691

Fig. 1

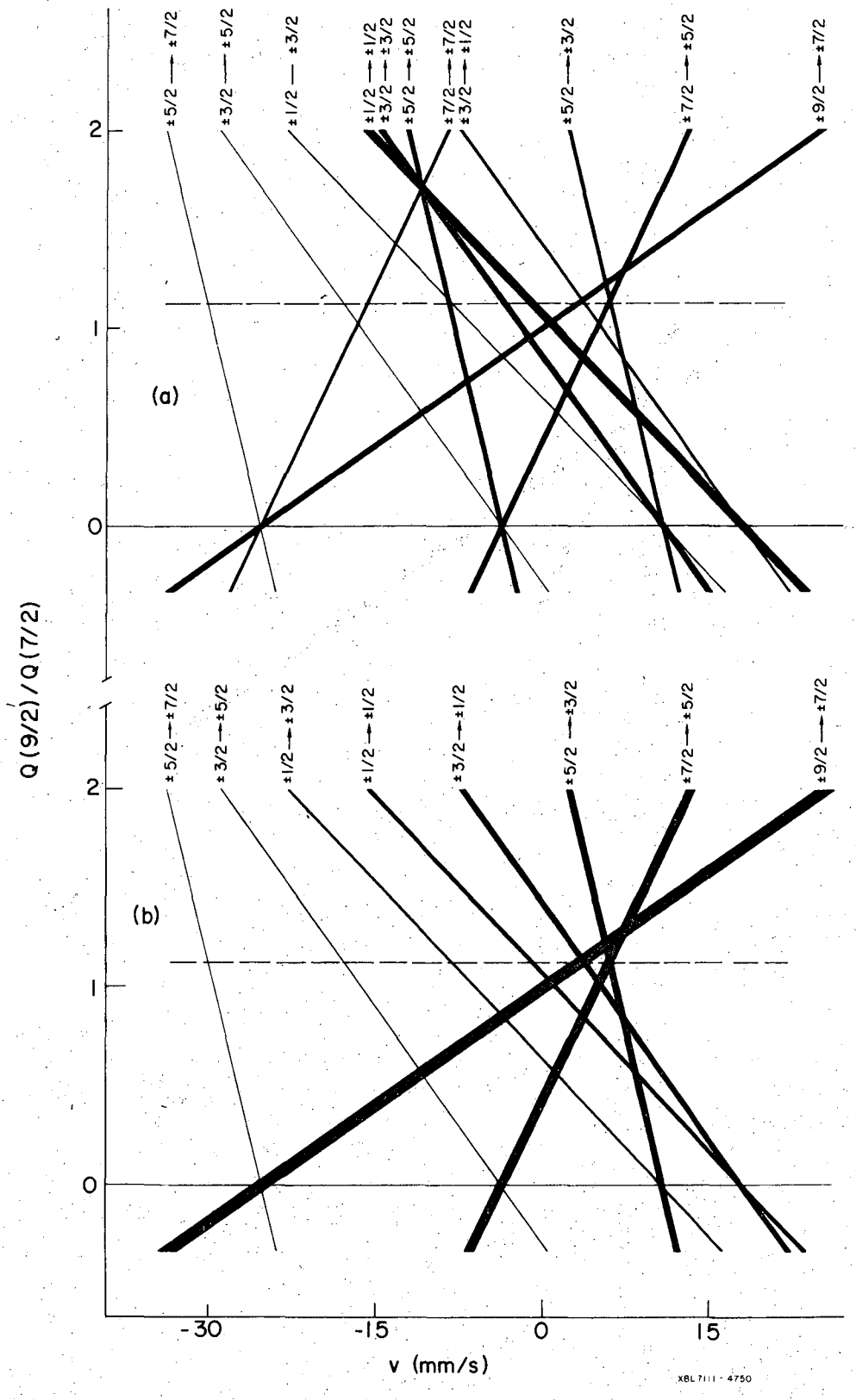
Q(9/2)/Q(7/2)
2
1
0
(a)
±5/2 — ±7/2
±3/2 — ±5/2
±1/2 — ±3/2
±1/2 — ±1/2
±3/2 — ±3/2
±5/2 — ±5/2
±7/2 — ±7/2
±3/2 — ±1/2
±5/2 — ±3/2
±7/2 — ±5/2
±9/2 — ±7/2
(b)
±5/2 — ±7/2
±3/2 — ±5/2
±1/2 — ±3/2
±1/2 — ±1/2
±3/2 — ±1/2
±5/2 — ±3/2
±7/2 — ±5/2
±9/2 — ±7/2
-30
-15
0
15
v (mm/s)
XBL 7111 - 4750

Fig. 2

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