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LETTER TO THE EDITOR

Nuclear parameters of the 140 keV Mössbauer level in ^{99}Tc from Mössbauer spectroscopy

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Abstract. Mössbauer resonance spectra of the 140 keV transition in ^{99}Tc were measured in a variety of source and absorber combinations over the temperature range from 1·6 to 65 K. The following parameters were obtained from these spectra: $\Delta\langle r^2\rangle \simeq 6\times10^{-3}$ fm^2, $T_{1/2}(140) = 237\pm14$ ps and $g(140) = 1{\cdot}03\pm0{\cdot}25$.

Only a few Mössbauer resonance measurements have been reported thus far on the 140 keV, $9/2^+ \to 7/2^+$ γ ray transition in ^{99}Tc. The reasons are the large recoil energy of 0·1 eV which is the highest of all established resonances leading to a recoilless fraction of the order of 10^{-3}, and the need to handle radioactive absorbers. Utilizing a very strong source and fast counting electronics the small resonance effects of 10^{-1} to 10^{-2}% could still be measured with good accuracy within counting periods of the order of days. The short half-life of the 140 keV state in combination with the high spins of both the ground and the excited state reduce considerably the usefulness of this resonance for the measurement of hyperfine interactions. Resolution is always a problem and in nearly all cases the resonance pattern can be approximated well by a single lorentzian (Steiner *et al* 1969).

Since chemical compounds of technetium in most cases gave unmeasurably small resonance effects due to rather low Debye temperatures we could only determine isomer shifts of Tc when it was an impurity in various 3d, 4d and 5d host metals. In all cases, source measurements were performed; that is, dilute alloys of Mo in transition metals were prepared either before or after the neutron irradiation. Spectra were recorded for a Tc metal absorber (2 g cm^{-2}) using a standard electromechanical velocity spectrometer (figure 1). The shifts observed never exceed 5% of the experimental linewidth of about 17 mm s^{-1}. In figure 2 the measured isomer shifts are compared with similar data (Potzel *et al* 1971, Potzel 1971) for the 89 keV resonance of ^{99}Ru. Making use of the systematic trends of isomer shifts of transition metal impurities in various 3d, 4d and 5d hosts, (Wagner *et al* 1973, Kaindl and Salomon 1973) we find $\Delta\langle r^2\rangle(^{99}\text{Tc})/\Delta\langle r^2\rangle(^{99}\text{Ru}) = 0{\cdot}30\pm0{\cdot}15$. Using

$$\Delta\langle r^2\rangle(^{99}\text{Ru}) \simeq 20\times10^{-3}\ \text{fm}^2$$

(Kalvius and Shenoy 1973, Potzel *et al* 1971) one obtains $\Delta\langle r^2\rangle \simeq 6\times10^{-3}$ fm^2.

One single chemical compound (TcO_2) gave large resonance effects. We observed an isomer shift of $0{\cdot}32 \pm 0{\cdot}10$ mm s^{-1} between a (Tc)MoO_3 source and a TcO_2 absorber. If one assumes that the Tc configuration in these two systems is Tc(VI):$4d^1$ and Tc(IV):$4d^3$, respectively, the difference in the charge density at the nucleus is

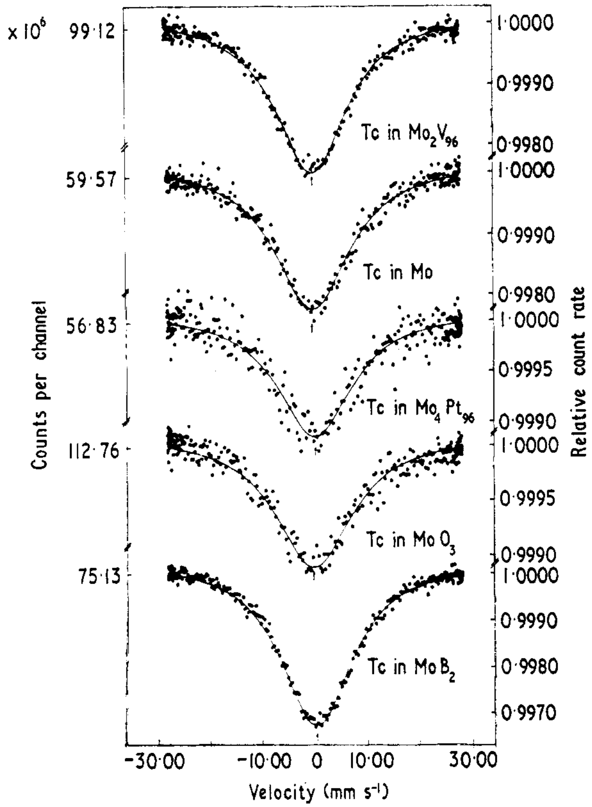

Figure 1. Resonance absorption spectra of the 140·5 keV γ ray in ^{99}Tc at 4·2 K for various source materials. All spectra are measured with a Tc metal absorber.

found to be $19{\cdot}7\ a_0^{-3}$ by using a Dirac–Fock–Slater program. This yields

$$\Delta\langle r^2\rangle = 5{\cdot}5 \times 10^{-3}\ \mathrm{fm}^2$$

in agreement with the above evaluation. There are numerous theoretical discussions of the structure of the low-lying levels in ^{99}Tc, but an estimate for $\Delta\langle r^2\rangle$ is nowhere given. The small value obtained here is in agreement with the interpretation of the 140 keV level as a rotational state (McDonald *et al* 1971).

Table 1. The half-life of the 140 keV level in ^{99}Tc

$T_{1/2}$(ps)	Reference
192 ± 10	Steiner *et al* 1969
160 ± 20	McDonald *et al* 1971
$191 \leq T_{1/2} \leq 271$	Bond *et al* 1972
237 ± 14	Present work

The lifetime of the 140 keV level has been determined by various techniques. The results as listed in table 1 show a considerable scatter. The value of this work was obtained by measurements of the resonance linewidth using a Mo metal source and TcO_2 absorbers of different thicknesses. Extrapolating the linewidth with the usual

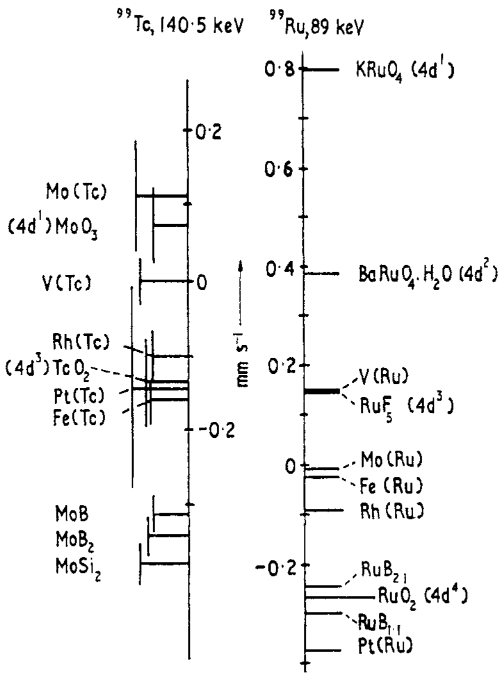

Figure 2. Systematics of the isomer shift for the 140 keV transition in ^{99}Tc and for the 89 keV transition in ^{99}Ru as referred to absorbers. The zero points of the scales are Tc metal and Ru metal respectively.

formalism (Margulies and Ehrmann 1961) to zero absorber thickness, we deduce $T_{1/2} = (237 \pm 14)$ ps. This value is somewhat larger than the one obtained by Steiner *et al* (1969) from similar measurements using Tc metal absorbers. In both cases, the environment of the Tc nucleus is not cubic and line broadenings could be present. Both values should therefore be considered as a lower limit. The errors in table 1 refer only to the statistical uncertainties.

To investigate magnetic hyperfine splittings, the Mössbauer spectrum of a source of 2 at % Mo in a Fe matrix was measured at 4·2 K against our standard Tc metal absorber. The resonance line was broadened to $18 \cdot 2 \pm 0 \cdot 5$ mm s^{-1} FWHM but showed no structure. Using a hyperfine magnetic field of -298 kOe at the Tc nucleus in Fe as obtained from NMR ON measurements at low temperatures (Fox *et al* 1972), a pure magnetic hyperfine pattern was fitted to the resonance pattern leaving the ratio $g(140)/g(0)$ as a free parameter. A least-squares fitting routine employing the full transmission integral was used. This is a necessity in cases of poorly resolved complex hyperfine spectra (Gerdau *et al* 1972). Taking $g(0) = 1 \cdot 257$, we obtain

$$g(140) = 1 \cdot 03 \pm 0 \cdot 25$$

in agreement with earlier PAC data (Zawislak and Rogers 1968, Inia *et al* 1969).

In a few cases the variation of the total intensity of the Mössbauer pattern with temperature was also recorded between 1·6 K and 65 K in order to obtain values for the recoil-free fraction. A γ ray transition with high recoil energy is expected to be the best tool for an investigation of lattice dynamics. Deviations from simple Debye behaviour were found for Tc in Mo. The data can be explained by a change in Debye temperature from 365 K at 4·2 K to 380 K at 65 K. A rather high Debye temperature of $\theta_D = 415 \pm 15$ K was estimated for TcO_2 which proves to be an excellent single-line absorber material.

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