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Effect of Compression on the Decay Rate of Tc^{99m} Metal*

R. A. PORTER, *Theoretical Division, The Lawrence Radiation Laboratory, University of California, Livermore, California*

AND

W. G. McMILLAN, *Department of Chemistry, University of California, Los Angeles and the RAND Corporation, Santa Monica, California*

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Motivated by Bainbridge's measurement, we have attempted a theoretical calculation of the change in lifetime of the internal conversion of Tc^{99m} in compressed Tc metal. We have employed the Thomas-Fermi statistical potential, corrected for the self-potential of the electron in question, to obtain the initial and final state electronic wave functions for two volumes: the normal uncompressed state, and ten percent compression. Because the energy available is so low ($\sim$2 kev), the only contributions to the internal conversion coefficient come from the M and higher shells, and mainly from the $3p$ and $3d$ levels. The principal contribution to the *change* in the internal conversion coefficient comes, however, from the valence electrons, particularly the $4p$, $4d$ and $5s$ levels. In order to relate compression to pressure, we have estimated the compressibility of technetium metal to be 0.27 megabar^{-1}. From this compressibility and the assumption that the internal conversion coefficient is linear in pressure, we calculate for the experimental pressure of 0.1 megabar a fractional decrease in lifetime of (2 to 4)$\times$10^{-4}, the quoted variation residing in the uncertainty of the structure of the $4p$ band. This result agrees with Bainbridge's measurement, (2.3$\pm$0.5)$\times$10^{-4}, within the accuracy of our calculation.

I. INTRODUCTION

THE decay rate of the 2-kev electric octupole isomeric transition of Tc^{99m} is known to be measurably dependent upon the external environment. Differences in the lifetime have been observed for different chemical compounds,[1] for the metal under high static pressure,[2] and for the superconducting state.[3] Of these environmental effects, that due to compression of the metal is most amenable to a theoretical treatment. In the present work we attempt a calculation of this effect on the basis of a simple metallic model. Although technetium is far from being a simple metal it appears possible to incorporate the most essential features without excessive complication. The use of a consistent treatment of initial and final wave functions for different atomic volumes makes the fractional change in lifetime, to be compared with experiment, consider-ably more reliable than any single element of the calculation.

II. THEORY OF THE DECAY RATE OF Tc^{99m}

Because of the very low transition energy and the large change in angular momentum involved, the decay of Tc^{99m} occurs almost entirely by internal conversion. The decay rate is therefore proportional to the internal conversion coefficient α, which is given by the relation[4]

$$\alpha = (2\pi^3 W)\mathfrak{S} |(f|\mathfrak{H}|i)|^2, \qquad (1)$$

where W is the transition energy, $\mathfrak{H}$ the interaction Hamiltonian, i and f are the initial and final electronic wave functions, and $\mathfrak{S}$ indicates summation over all directions of emission and all substates of the initial (bound) and final (continuum) states. The present calculation will be nonrelativistic, since only valence and near-valence electrons are involved. We shall treat the metallic problem as spherically symmetric, and thus ignore effects of the lattice structure except for the periodicity of the Bloch waves. The interaction Hamiltonian is therefore

$$\mathfrak{H} = ie\mathbf{B}_L{}^M \cdot \nabla + (ie/2)\nabla \cdot \mathbf{B}_L{}^M + e\Phi_L{}^M, \qquad (2)$$

* Based on a thesis submitted by R. A. Porter in partial fulfillment of the requirements for the degree of Doctor of Philosophy, Department of Chemistry, University of California, Los Angeles.

[1] Bainbridge, Goldhaber, and Wilson, Phys. Rev. **84**, 1260 (1951); **90**, 430 (1953).

[2] K. T. Bainbridge (private communication). We are much indebted to Professor Bainbridge for permission to quote this result prior to publication. Previous mention of his work has also been made by Chem. Eng. News **30**, 654 (1952) and by R. Daudel, J. phys. radium **13**, 562 (1952).

[3] D. H. Byers and R. Stump, Phys. Rev. **112**, 77 (1958).

[4] In this section we employ the notation and units of M. E. Rose, *Multipole Fields* (John Wiley and Sons, New York, 1955).

where $\mathbf{B}_L{}^M$ and $\Phi_L{}^M$ are the vector and scalar potentials for an outgoing electromagnetic wave of multipole order L. Since we are dealing with an electric-type transition, it is convenient to use the conventional gauge, for which

$$\mathbf{B}_L{}^M = [(2L+1)(L+1)]^{\frac{1}{2}} h_{L-1}(kr) \mathbf{T}_{L,L-1}{}^M(\theta,\phi), \quad (3)$$

$$\Phi_L{}^M = i[L/(L+1)]^{\frac{1}{2}} h_L(kr) Y_L{}^M(\theta,\phi), \quad (4)$$

where h_L is a spherical Hankel function, $Y_L{}^M$ a spherical harmonic and $\mathbf{T}_{L,L-1}{}^M$ an irreducible tensor on the unit sphere.

Because virtually all the internal conversion occurs in the near zone of the radiation field, only the electrostatic term $\Phi_L{}^M$ contributes significantly to α. (A rough calculation showed that the terms in $\mathbf{B}_L{}^M$ contribute about 0.1% to the total decay rate.) In addition, the spherical Bessel function part of h_L is negligible compared to the spherical Neumann function part n_L in the region of interest near the nucleus. Thus, on dropping the small terms just mentioned and performing the angular integration which results when Eqs. (2) and (4) are combined with (1), we obtain

$$\alpha(W,L) = 4\pi^2 W e^2 \sum_{f,i} \left| \left(\frac{L(2L+1)(2l_i+1)}{4\pi(L+1)(2l_f+1)} \right)^{\frac{1}{2}} \right.$$
$$\left. \times C(Ll_i l_f; 00) \int R_{lf} n_L R_{li} r^2 dr \right|^2, \quad (5)$$

where R_{li} and R_{lf} are the radial wave functions of the initial and final states of angular momentum l_i and l_f, respectively, and $C(Ll_i l_f; 00)$ is a Clebsch-Gordon coefficient.

III. INITIAL STATE WAVE FUNCTIONS

The present calculation of wave functions for Tc metal makes use of the spherical Thomas-Fermi (TF) potential as applied to metals.[5] We have attempted to remove the electron self-interaction present in this model by investigating two types of corrections. In the first, the potential is taken to be

$$V(r) = V_{TF}(r), \quad V_{TF} \geq e/r,$$
$$V(r) = e/r, \quad \text{otherwise}, \quad (6)$$

where V_{TF} is the TF potential. This method, which approximates the proper behavior near the atomic cell boundary, $r = r_s$, will be referred to as the cutoff method. It was employed by Latter[6] in his TF calculations for free atoms. In the second method an electron wave function is calculated for a given level in the unmodified TF potential. This wave function is then used to calculate the electron self-potential, which is in turn subtracted from the TF potential. Finally this corrected

[5] P. Gombas, *Die Statistiche Theorie Des Atoms* (Springer-Verlag, Vienna, 1949).

[6] R. Latter, J. Chem. Phys. **24**, 280 (1956).

TABLE I. Energy term values (in Rydbergs) for technetium.

Level i	Boundary condition ii	Normal volume ($\eta=1$) TF iii	Cutoff iv	SPC v	Experiment vi	($\eta=1.1$) SPC vii
2s	$\psi=0$	−200	−200	−212	−225	
2p	$\psi=0$	−183	−183	−197	−206, −197	
3s	$\psi=0$	−32.6	−32.6	−36.8	−40.1	
3p	$\psi=0$	−26.3	−26.3	−30.5	−33.0, −31.5	−30.4
3d	$\psi=0$	−15.1	−15.1	−19.6	−19.0, −18.8	−19.4
4s	$\psi=0$	−2.72	−2.74	−4.31	−5.29	
4p	$\psi'=0$	−1.21		−2.50	−3.09	−2.42
	$\psi=0$	−1.00	−1.04	−2.47	−3.09	−2.35
4d	$\psi'=0$	+0.69		−0.19		−0.16
	$\psi=0$	+1.69	+1.59	+0.44		+0.58
5s	$\psi'=0$	+0.51		−0.30		−0.23
	$\psi=0$	+3.34		+2.32		+2.70

potential is used to obtain a new wave function for the given level. This method will be referred to as the self-potential correction (SPC) method.

For core electrons (M shell and below) the initial wave functions ψ_i are computed by requiring that $\psi_i(r_s) = 0$, where r_s is the radius of a sphere of volume equal to that of the atomic cell. For the near-valence $4p$ band and the valence ($4d$ and $5s$) bands, wave functions are computed for the top $[\psi_i(r_s) = 0]$ and bottom $[\psi_i'(r_s) = 0]$ of the band. The values of matrix elements for intermediate positions in the band are obtained by an interpolation scheme which will be discussed later (Sec. V).

The energy term values computed with the TF potential and its two modifications at the normal volume ($r_s = 2.84$ Bohr radii) are given in the third to fifth columns of Table I. No direct experimental values of these quantities for Tc exist; however, interpolated values from neighboring elements have been given by Siegbahn.[7] These are listed in the sixth column of Table I. The two values given for some of the low-lying levels reflect relativistic splittings, which are ignored here. It can be seen that the cutoff method is only a slight improvement on the TF potential, while the SPC method is a substantial improvement. This latter method was therefore made the basis for subsequent calculations.

The wave functions for higher pressures can be most easily found by repeating the calculation for smaller values of r_s and relating the compression $\eta = V_0/V$ to the pressure by means of a separate pressure-volume relation. Unfortunately, no experimental pressure-volume data exist for technetium metal, but the compressibility can be estimated satisfactorily enough for the present purpose. This estimate will be discussed in Sec. VI. Only one compression in addition to the normal volume was investigated, this being $\eta = 1.1$. The energy term values from the SPC method for this compression are given in the last column of Table I. From this table it can be seen that only the M and higher shells can contribute to internal conversion, since for lower levels the electron binding energy is

[7] K. Siegbahn, *Beta- and Gamma-Ray Spectroscopy* (Interscience Publishers, New York, 1955).

greater than that available from the nuclear transition (2 kev = 147 Rydbergs).

The band structure of the $4d$ and $5s$ bands was approximated by the second-order perturbation method of Silverman.[8] In this method the energy E_k of a valence electron characterized by a wave number vector $\mathbf{k}$ is represented by

$$E_{\mathbf{k}} = E_0 + k^2 E_2 + O(k^4). \qquad (7)$$

For the $5s$ band[9]

$$E_2(5s) = \tfrac{1}{3} r_s{}^3 u_0{}^2(r_s) [r_s f_p{}'(r_s)/f_p(r_s)], \qquad (8)$$

where u_0 is the radial wave function for $k = 0$ and f_p is a radial p-function which satisfies the radial part of the equation

$$(\nabla^2 + V - E_0) f_p = 0. \qquad (9)$$

This result was first obtained by Bardeen.[10] By a method exactly analogous to that of Silverman for s bands, we have derived for the $4d$ band the expression

$$E_2(4d) = r_s{}^3 u_0{}^2(r_s) [27 r_s f_f{}'(r_s)/f_f(r_s)$$
$$+ 28 r_s f_p{}'(r_s)/f_p(r_s)]/105, \qquad (10)$$

where f_f is a radial f-function satisfying (9).

Assuming that all seven valence electrons above the $4p$ level lie in the $4d$ and $5s$ bands, the numbers N_{4d} and N_{5s} of electrons in these bands can be calculated from the following conditions: (i) the number of electrons in the bands must add to seven, and (ii) the energies (chemical potentials) of the highest filled level in each (partially filled) band must be the same. Since the density of states $\rho(k)$ is assumed to have the free-electron form [a consequence of neglecting terms of order higher than k^2 in Eq. (7)], the first condition leads to the relation

$$7 = N_{4d} + N_{5s} = (4 r_s{}^3/9\pi)(5 K_{4d}{}^3 + K_{5s}{}^3), \qquad (11)$$

where the K's are the maximum occupied values of k in the bands. The second condition leads to the relation

$$E_0(4d) + E_2(4d) K_{4d}{}^2 = E_0(5s) + E_2(5s) K_{5s}{}^2. \qquad (12)$$

Equations (11) and (12) can be solved simultaneously for the K's, from which the N's can be obtained. For each band the values of E_2, N, and K, along with the values of E_0 from Table I, are listed in Table II for $\eta = 1.0$ and $\eta = 1.1$. It will be noticed that as expected, electrons pass from the $5s$ band to the $4d$ band on compression.

IV. FINAL STATE WAVE FUNCTIONS

In calculating the wave function of the ejected electron in its final state, we have assumed that (a) inside an atomic cell, the potential is the same as for

TABLE II. Valence band parameters for technetium metal (in atomic units).

	4d band		5s band	
	$\eta = 1.0$	$\eta = 1.1$	$\eta = 1.0$	$\eta = 1.1$
E_0	−0.19	−0.16	−0.30	−0.23
E_2	1.98	2.13	1.74	1.53
K	0.69	0.72	0.81	0.80
N	5.26	5.47	1.74	1.53

the initial state; and (b) outside the cell, the radial part of the wave function eventually has the form of a spherical wave,

$$R_{lf} = A \sin(kr)/r.$$

The kinetic energy is determined by the difference between the binding energy of the electron in its initial state and the energy available from the nuclear transition, 2 kev. The values of l_f are determined by the usual vector model selection rules. Our procedure was to calculate the wave function in a region which included, besides the atomic cell, a small region outside the cell (extending in our calculations to $2.25 r_s$) in which the potential was set equal to e/r. The purpose of this outer region was to enable the wave function to settle down to a constant amplitude A by which it could be characterized outside the cell. In all cases the amplitude of the last two extrema in $r R_{lf}$ agreed to better than one part in 10^4. The portion of the wave function inside the atomic cell is used to calculate the matrix element; in the final calculation, the square of the matrix element is then multiplied by $(2m/E)^{\frac{1}{2}}/A^2 \pi \hbar$, where E is the energy of the final state, in order to normalize the final state wave function to unit energy range.

V. RESULTS FOR THE CONVERSION COEFFICIENT

It was shown previously that only the M shell and above contribute to internal conversion in Tc^{99m}. A rough calculation was performed using the unmodified TF potential to determine the relative importance of the different levels and of different values of l_f. It was found that the $3s$ and $4s$ levels contribute less than 1% to the absolute value of α, and an even smaller percentage to its *change* on compression. Values of l_f above the lowest for a given initial state were also found to give insignificant contributions to $\Delta \alpha / \alpha$. (It was at first thought that the $3d$, $l_f = 3$ matrix element might be important, and it was carried in the more refined calculation; however, its contribution to $\Delta \alpha / \alpha$ turned out to be negligible.) The reason for these results has been indicated by the work of Slater,[11] who calculated the relative contributions of the different electron levels of the free Tc atom to the decay rate of Tc^{99m}. He found, as we also have found, that almost the entire contribution to the matrix element occurs in the region $0 \leq r/r_s \leq 0.1$. The reason that the s levels contribute so little is that the s wave functions have a node in the

[8] R. A. Silverman, Phys. Rev. **85**, 227 (1952).

[9] The wave function u_0 in Eq. (8) is normalized such that $\int_0^{r_s} u_0{}^2 r^2 dr = 1$.

[10] J. Bardeen, J. Chem. Phys. **6**, 367 (1938).

[11] J. C. Slater, Phys. Rev. **84**, 1261 (1951).

TABLE III. Contributions of various types of wave functions to the conversion coefficient (SPC).

Initial state	Boundary condition	l_f	$\eta=1.0$		$\eta=1.1$	
			N_i	$10^{-8}(N_i\alpha_{fi})$	N_i	$10^{-8}(N_i\alpha_{fi})$
$3p$	$\psi=0$	2	6	5.4486	6	5.4492
$3d$	$\psi=0$	1	10	7.9140	10	7.9165
	$\psi=0$	3	10	0.0610	10	0.0610
$4p^a$	$\psi'=0$	2	4.28	0.4911	4.28	0.4735
	$\psi=0$	2	1.72	0.2360	1.72	0.2376
$4p^b$	$\psi'=0$	2	2.4	0.2754	3.6	0.2655
	$\psi=0$	2	3.6	0.4940	2.4	0.4973
$4d$	$\psi'=0$	1	3.20	0.0860	3.27	0.0892
	$\psi=0$	1	2.06	0.1704	2.19	0.1899
$5s$	$\psi'=0$	3	0.79	0.0004	0.76	0.0005
	$\psi=0$	3	0.95	0.0008	0.77	0.0015

[a] Using Eq. (17).
[b] Using Eq. (18).

middle of this region, causing a partial cancellation. The p and d functions do not have a node in this region. (The contribution of the $5s$ band to α is also very small, but it makes a larger contribution to $\Delta\alpha/\alpha$ since the effect of compression is greatest for this band.) Similarly the higher values of l_f do not contribute significantly because the indicial behavior of R_{l_f} is like r^{l_f}; thus for high l_f the important part of the wave function near the nucleus is smaller.

Before proceeding to a description of our more accurate calculation, we must describe our method of estimating the matrix elements for values of k between $k=0$ ($\psi_i'=0$ boundary condition) and $k=k_{max}$ ($\psi_i=0$ boundary condition) in the $4p$ and valence bands. We express the initial state wave function as a Bloch wave,

$$\psi_i=u_k(r)\exp(i\mathbf{k}\cdot\mathbf{r}).$$

Expanding both u_k and $\exp(i\mathbf{k}\cdot\mathbf{r})$ in powers of k and keeping only terms up to k^2 gives

$$\psi_i=u_0+k[u_1+iru_0\cos(\mathbf{k}\cdot\mathbf{r})]+k^2[u_2+iru_1\cos(\mathbf{k}\cdot\mathbf{r})$$
$$-(r^2/2)u_0\cos^2(\mathbf{k}\cdot\mathbf{r})]. \quad (13)$$

But $u_1=i\cos(\mathbf{k}\cdot\mathbf{r})[ru_0+\text{terms in } f_{l_i\pm1}$ such as to make[8] $u_1(r_s)=0]$. Thus the term linear in k makes no contribution to the matrix element due to the vanishing
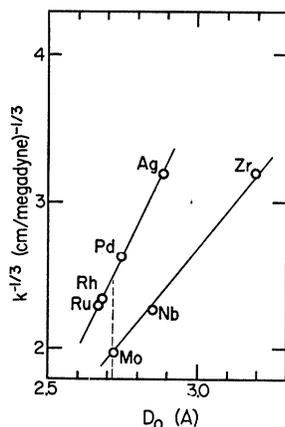
FIG. 1. Metallic bond force constant k as a function of nearest-neighbor bond length D_0 for some elements of the second transition period, after Pauling and Waser.[13] The dashed line represents the nearest-neighbor distance in technetium.

of the angular integral. Since ψ_f is nearly independent of k, the contribution $\alpha_{fi}\sim|\int\psi_i^+\mathfrak{H}\psi_jd\tau|^2$ of a given initial and final state can therefore be approximated reasonably by a form parabolic in k:

$$\alpha_{fi}(k)=\alpha_{fi}(0)+(k^2/k_{max}^2)[\alpha_{fi}(k_{max})-\alpha_{fi}(0)]. \quad (14)$$

Higher terms in k should have little effect since the higher terms in (13) are proportional to higher powers of r.

The total contribution to the conversion coefficient averaged over a partially filled valence band is found from

$$\alpha_{fi}=\int_0^K\alpha_{fi}(k)\rho(k)dk\bigg/\int_0^K\rho(k)dk. \quad (15)$$

For both the $4d$ and $5s$ bands, we have made the approximation that $\rho(k)\sim k^2$, so that, using Eq. (14)

$$\alpha_{fi}=[1-(3K^2/5k_{max}^2)]\alpha_{fi}(0)$$
$$+(3K^2/5k_{max}^2)\alpha_{fi}(k_{max}). \quad (16)$$

K is obtained from Table II, and k_{max} from the free-electron relation $k_{max}=(9\pi/2)^{\frac{1}{3}}/r_s$.

The filled $4p$ band presents a difficult problem, since the values of the matrix element for the $\psi=0$ and $\psi'=0$ boundary conditions differ much more than might naively be expected from the narrowness of the band. We have not attempted a detailed calculation of the structure of this band, but have instead employed two approximations to the density of states $\rho(k)$ which we feel represent limits to the true band structure. The first is a symmetrical band shape $\rho(k)\sim k^2(K-k)^2$, for which

$$\alpha_{fi}=(5/7)\alpha_{fi}(0)+(2/7)\alpha_{fi}(k_{max}). \quad (17)$$

The second is a parabolic shape, $\rho(k)\sim k^2$, cut off at $k=K$, for which

$$\alpha_{fi}=\tfrac{2}{5}\alpha_{fi}(0)+\tfrac{3}{5}\alpha_{fi}(k_{max}). \quad (18)$$

This uncertainty in the $4p$ band calculation is one of the major sources of error in our calculation of $\Delta\alpha/\alpha$.

In our more refined calculation of α and $\Delta\alpha/\alpha$, we employed the SPC potential described previously. The grid used was fine enough to guarantee an accuracy in the amplitude of ψ_f of 1 part in 10^4. The matrix elements found to be unimportant in the earlier crude calculation were neglected. The results[12] are summarized in Table III. The electron number weighting N_i for a given initial wave function and boundary condition is obtained by using Eq. (16) together with Table II for the $4d$ and $5s$ bands, and Eq. (17) or (18) for the $4p$ band. With the notable exception of the $4p$, $\psi'=0$

[12] After the calculations were completed, it was discovered that a value of 137 Rydberg=1.82 kev had been used by mistake for the transition energy instead of the intended value 147 Rydberg =2.0 kev. We have since found that recent measurements by M. S. Freedman et al., Phys. Rev. 108, 836 (1957), give the value of 2.15 ± 0.03 kev=159 ± 2 Rydberg. We do not believe that the essential conclusions regarding the comparison of our results with the experimental results of Bainbridge are affected by this error.

TABLE IV. Sensitivity of internal conversion matrix elements to the potential.

Initial state	Boundary condition	l_f	Uncorrected TF potential $10^{-7}\alpha_{fi}$			Self-potential correction $10^{-7}\alpha_{fi}$		
			$\eta=1.0$	$\eta=1.1$	$10^5(\Delta\alpha_{fi}/\alpha)$[a]	$\eta=1.0$	$\eta=1.1$	$10^4(\Delta\alpha_{fi}/\alpha)$[a]
$3p$	$\psi=0$	2	8.5110	8.5100	−0.17	9.0810	9.0820	+0.7
$3d$	$\psi=0$	1	7.3097	7.3081	−1.1	7.9140	7.9165	+1.8
$4p$	$\psi'=0$	2	0.0985	0.0948	−2.7	0.1147	0.1106	−2.9

[a] $\Delta\alpha_{fi}\equiv\alpha_{fi}(\eta=1.1)-\alpha_{fi}(\eta=1.0)$, α taken as 1.44×10^9 for both cases.

case, all matrix elements increase upon compression. We cannot say whether the strikingly anomalous behavior of this matrix element reflects physical reality or is simply a mathematical quirk.

The total conversion coefficient is obtained by summing the contributions given in Table III. Using (17) for the $4p$ band, we obtain

$$\alpha=1.4408\times10^9, \quad \eta=1.0,$$
$$\alpha=1.4419\times10^9, \quad \eta=1.1.$$

Using (18) for the $4p$ band, we obtain

$$\alpha=1.4451\times10^9, \quad \eta=1.0,$$
$$\alpha=1.4471\times10^9, \quad \eta=1.1.$$

The uncertainty in the $4p$ band calculation thus introduces an uncertainty of about a factor two in $\Delta\alpha$.

To test the sensitivity of our calculation to the potential, a few matrix elements were also computed in the uncorrected TF potential with better numerical accuracy than in the crude TF calculation mentioned before. The values of α_{fi} and $\Delta\alpha_{fi}/\alpha$ are listed in Table IV for both the SPC and TF potentials. It is seen that the absolute values of α_{fi} deviate by $\sim5\%$. The values of $\Delta\alpha_{fi}/\alpha$ differ even as to sign for the $3p$ and $3d$ matrix elements, but surprisingly the large negative value of $\Delta\alpha_{fi}/\alpha$ for the $4p$, $\psi'=0$ matrix element is fairly accurately reproduced. The self-potential correction is therefore seen to be a most essential feature of the calculation.

VI. COMPRESSIBILITY OF Tc METAL

In order to relate the results quoted in the last section to the pressure experiments of Bainbridge, we must make an estimate of the compressibility of technetium metal. A rough estimate can be obtained

TABLE V. Zero-pressure compressibilities (megabars^{-1}) and crystal structures of elements neighboring technetium.

41 Nb	42 Mo	43 Tc	44 Ru	45 Rh
0.581 bcc	0.367 bcc	hcp	0.349 hcp	0.368 fcc

73 Ta	74 W	75 Re	76 Os	77 Ir
0.488 bcc	0.324 bcc	0.269 hcp	hcp	0.273 fcc

from the zero-pressure compressibilities of neighboring elements, which are listed in Table V.[13,14] The analogous elements from the row below technetium are also listed, since they have similar atomic volumes and crystal structures. Because the elements in these groups are fairly incompressible, there is probably no great error in assuming that the compressibility of technetium is constant up to $\eta=1.1$. From Table V one can infer for technetium a compressibility $\beta\sim0.3$ megabar^{-1}.

A semiempirical estimate of the compressibility can also be made by utilizing the regularity, observed by Waser and Pauling,[13] between the nearest-neighbor bond length D_0 and the inverse cube root of the metallic bond force constant k, defined by

$$k=9V_0/nD_0^2\beta,$$

where V_0 is the normal volume of the unit cell containing n equivalent nearest-neighbor bonds. This regularity is indicated in Figs. 1 and 2, which correspond, respectively, to the two periods listed in Table V. In Fig. 2 it will be seen that rhenium, the analog of technetium, unfortunately lies between the two linear sequences. Assuming a similar behavior for technetium, we estimate a value $k^{-\frac{1}{3}}\sim2.1$, corresponding to $\beta\sim0.27$ megabar^{-1}.

The crude result just obtained can be used to orient the results of the last section with respect to Bainbridge's pressure experiment. It indicates that a compression of $\eta=1.1$ corresponds to a pressure $p\sim0.34$
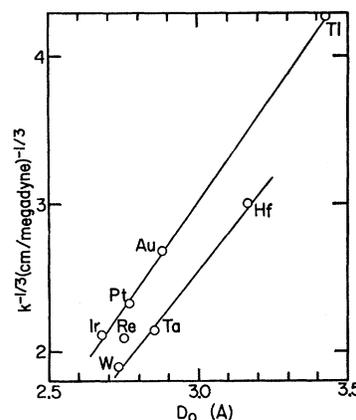
FIG. 2. Metallic bond force constant k as a function of nearest-neighbor bond length D_0 for some elements of the third transition period, after Pauling and Waser.[13] The rhenium point is calculated from the data of Bridgman.[14]

[13] Except for rhenium, the compressibilities in Table V are taken from the compilation of J. Waser and L. Pauling, J. Chem. Phys. 18, 747 (1950).

[14] The rhenium datum is taken from P. W. Bridgman, Proc. Am. Acad. Arts Sci. 84, 111 (1955).

TABLE VI. Relative contributions of electron states to the decay rate of Tc^{99m}.

| | % of Total decay rate | |
Electron state	Slater	This work
3p	0.60	0.38
3d	0.27	0.53
4p	0.10	0.05
4d	0.03	0.01

megabar. Thus if we assume that the internal conversion coefficient is linear in compression (or pressure) and use Eq. (17) for the 4p band, we find

$$[\alpha(p=0.1 \text{ megabar})-\alpha(p=0)]/\alpha=2.3\times10^{-4},$$

while if Eq. (18) is used for the 4p band, we obtain

$$[\alpha(p=0.1 \text{ megabar})-\alpha(p=0)]/\alpha=4.0\times10^{-4}.$$

These theoretical results agree with the experimental value of Bainbridge,[2]

$$[\alpha(p=0.1 \text{ megabar})-\alpha(p=0)]/\alpha=(2.3\pm0.5)\times10^{-4},$$

within a factor of two. The coincidental agreement of experiment with the first of our theoretical numbers is not to be taken as evidence that Eq. (17) is necessarily a better representation of the 4p band structure, since other errors in the calculation may be of the same order as the difference between the two theoretical results quoted.

VII. DISCUSSION

The absolute value of α for technetium is of interest in its own right, since it is the highest ever calculated. Using the experimental half-life $t_{\frac{1}{2}}$ of 6.0 hours, the rate of gamma emission N_γ is given by

$$N_\gamma=\frac{\ln2}{t_{\frac{1}{2}}(1+\alpha)}=7.0\times10^{-6} \text{ yr}^{-1}.$$

This can be compared with the independent estimate of N_γ based on some nuclear model. For example, on a simplified model in which transitions are ascribed to a single nonrelativistic proton moving in a uniform central velocity-independent field,[15] one obtains, using a transition energy of 1.82 kev,[12]

$$N_\gamma=8.8\times10^{-6} \text{ yr}^{-1}.$$

The agreement between these two numbers for N_γ is respectable considering the many approximations involved.

The relative contributions of the different electron levels have been computed by Slater[11] for the free atom. His results are compared with ours in Table VI. The divergence between the two sets of results seems large even when account is taken of the difference in physical state (free atom vs metal) and the fact that Slater used $1/r^4$ for the radial dependence of the electromagnetic field rather than the spherical Hankel function used here. A measurement of the spectrum has been made by Freedman et al.,[16] but they obtain the relative contributions $M_1:M_{2+3}:M_{4+5}:N::3:3:1:0.9$, in violent disagreement with both Slater's and our calculations.

VIII. ACKNOWLEDGMENTS

[15] S. A. Moszkowski, in Beta- and Gamma-Ray Spectroscopy, edited by K. Siegbahn (Interscience Publishers, New York, 1955).
[16] M. S. Freedman et al., see reference 12.