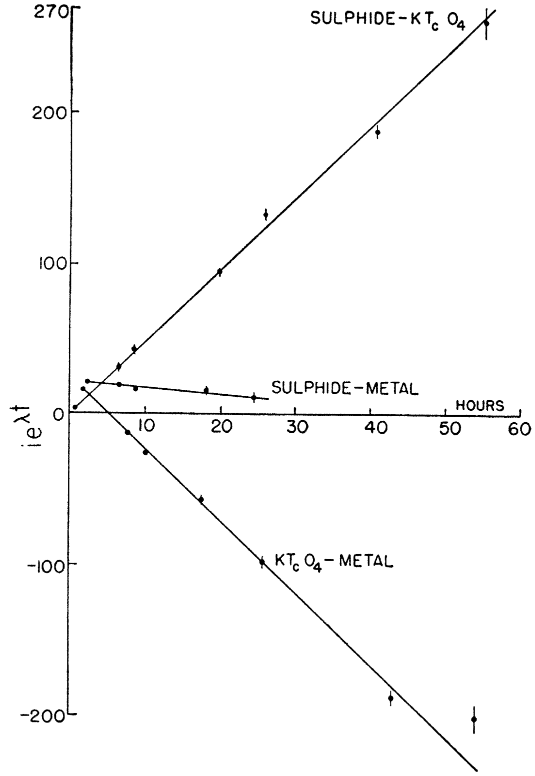

FIG. 1. $ie^{\lambda t} = i_0 - \Delta\lambda I_0 t$ (divisions) plotted against t. The statistical errors of each point are shown. The lines indicate the least-squares fit to the points. $I_0 = 15030$ divisions for sources sulfide-$KTcO_4$, 12350 for sulfide-metal, 13270 for $KTcO_4$-metal. The initial time points, $t=0$, for each curve are 0.75, 2.45, and 1.83 hours, respectively.

influence can best be tested by using an isomer in different chemical states which has a highly converted low energy isomeric transition.

We have carried out a series of experiments with Tc^{99m} (6 hours), an isomer with a 2-kev highly converted isomeric transition followed promptly by a 140-kev partially converted gamma-ray[1,2] which can be conveniently measured. We find that the decay constant of the 2-kev isomeric transition does assume different values which are dependent on the chemical state of the element.

When long-lived Tc^{99} is used as a carrier, λ for Tc^{99m} in $KTcO_4$, as the dry salt, is $1+0.0030\pm0.00010$ times greater than λ for Tc^{99m} electrolytically deposited on copper.[3] In another experiment the same value, within the expressed error, was found for $KTcO_4$ in basic aqueous solution.

The experimental data for the dry salt $KTcO_4$ and the sulfide[3] (Tc_2S_7) compared with electrolytically deposited Tc^{99m} are shown in Fig. 1. λ for Tc^{99m} as the sulfide is $1+0.00031\pm0.00012$ times greater than λ for Tc^{99m} electrolytically deposited on copper. The radiations from the long-lived Tc^{99} carrier, ~25 micrograms for an individual source, have a negligible effect on these measurements.

The measurements were made by the balance method introduced by Rutherford[4] in which two identical ionization chambers housing the sources under comparison are connected to collect ions of opposite sign. Thus the difference current produced by two closely matched sources is small and can be measured with a precision[4-7] approaching that defined by the statistics of the number of separate ionization events which have occurred during an observation.[6,8] The difference current is $i = i_0e^{-\lambda t} - I_0\Delta\lambda te^{-\lambda t}$ at any time t, where i_0 is the initial unbalance current, I_0 is the initial intensity of the stronger source whose transition constant is λ, and $\lambda - \Delta\lambda$ is the transition constant of the other source.

The observed effect is discussed theoretically by Slater in an accompanying letter.

We are indebted to Mr. M. McKeown for checking the purity of the sources with a NaI scintillation spectrometer, to Dr. G. Friedlander for advice on some chemical aspects of this work, to the Brookhaven Reactor Group for irradiations, to Miss Jean Snover for help in the computations, and to the Isotopes Division of the AEC for the long-lived Tc carrier material. The ionization chambers were constructed under ONR contract with Harvard University.

* Research carried out under contract with the AEC.

† On leave from Harvard University, Cambridge, Massachusetts.

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The Effect of Chemical Combination on the Internal Conversion in Tc*

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BAINBRIDGE, Goldhaber, and Wilson[1] have shown experimentally that the probability of a certain internal conversion in Tc^{99m} depends on the state of chemical combination. In this conversion, the nucleus loses about 2 kev of energy, undergoing an electric octopole transition of $\Delta l = 3$. This energy is only enough to remove an M or N electron from the Tc atom, and since these electrons lie toward the outside of the atom, it was thought that their wave functions might be appreciably affected by chemical combination, with consequent appreciable effect on the conversion probability. In the reference just cited, it was found in fact that there is such an effect, of the order of three-tenths of a percent, and that furthermore, $KTcO_4$ has a somewhat greater conversion probability than Tc metal. It is the purpose of this note to show that this is not unreasonable theoretically.

In the first place, a very rough calculation has been made of the relative contributions of the various electronic shells to the internal conversion probability. The matrix component which has to be computed is the integral of the product of the wave function of the bound electron in the Tc atom or crystal, the wave function of an electron in the continuum with an energy 2 kev greater than the bound electron, and a quantity representing the potential of the nuclear octopole. Since the electromagnetic wavelength of a gamma-ray of 2 kev is about 6A, and the distances concerned in the integral are a rather small fraction of an angstrom, we are justified in representing this potential electrostatically, so that it is given by a spherical harmonic of $l=3$, times $1/r^4$. The integral then involves a product of three spherical harmonics, one from each wave function and one from the potential. Such integrals are zero unless certain relations exist between the l values, resulting in selection principles, which in the present case result in possible transitions from the bound s states to an f state in the continuum, from a bound p state to a d state in the continuum, and from a bound d state to a p state in the continuum. The functions of angles are easily computed, and the matrix component, whose square is proportional to the transition probability, reduces to an integral over r, of the product of the radial wave functions of bound and continuum states, times $1/r^4$, times the volume elements r^2dr.

To compute these radial integrals, a very rough numerical integration for both the bound and continuum wave functions has been made, using as a potential the Fermi-Thomas potential for Tc, corrected for exchange. The integrand in each case proves to start for small r's proportionally to r (the singularity in the

TABLE I. Contribution of electron shells to transition probabilities.

Electron	Transition probability	Ionization potential, ev
$3p$	0.60	430
$3d$	0.27	260
$4p$	0.10	45
$4d$	0.03	7
	Sum 1.00	

quantity $1/r^4$ being canceled by positive powers arising from the wave functions), to rise to a maximum, and to become negligibly small for values of r greater than about 0.2A, so that the effect depends on the behavior of the wave function for small values of r. The required integrals have been computed numerically. For transitions from the bound s wave functions to the continuum, the probabilities prove to be negligibly small, for a rather trivial reason: the integrand is partly positive, partly negative in the range of integration on account of the innermost node of the s functions, and the contributions almost cancel. For the transitions from the p and d electrons, in the free atom, the relative contributions to the internal conversion, as computed, are given in Table I. These values are to be regarded as very rough approximations, on account of the very crude numerical integration which has so far been used to approximate the wave functions. In Table I we give also very rough approximations to the ionization potentials of the various shells, obtained by interpolation between known x-ray term values.

In the metal or in $KTcO_4$, as contrasted to the free atom, these probabilities will be affected only as the charge density of the various electronic shells is altered at distances less than 0.2A, which alone contribute to the transition probabilities. A rough estimate indicates that the $3p$ and $3d$ electrons will not be affected to a significant extent. The $4p$ shell, which certainly does not contribute to binding, will be somewhat squeezed, and very rough estimates indicate that it might increase its charge density in the required region by a few percent of its total density, giving an increase of probability of a few tenths of a percent over all. The change in the $4d$ shell is less easy to estimate. These electrons doubtless take part in the binding, both in the metal and the $KTcO_4$. In the metal, they are certainly somewhat squeezed as compared with the free atom. In the $KTcO_4$, an ionic picture, regarding the $(TcO_4)^-$ ion as being made up of $Tc^{+7}(O^=)_4$, would lead to a large removal of $4d$ electrons from the neighborhood of the Tc, piling them up around the oxygens. No one, however, supposes that this really happens, and the writer suspects that the actual density of valence electrons near the Tc atom is not very different in $KTcO_4$ from what it is in the free atom or the metal. In other words, it is uncertain whether the contribution 0.03 of the $4d$ electrons in the free atom would be increased or decreased in the crystals. These considerations, in other words, lead to the order of magnitude of a fraction of a percent for the total change, as has been observed, probably arising mostly from the $4p$ wave functions.

As to why the probability of internal conversion is greater in $KTcO_4$ than in the metal, the most straightforward explanation seems to be that the Tc atom is squeezed more in the $KTcO_4$. The Tc—Tc distance in the metal is about 2.7A.[2] The distance has not been measured in $KTcO_4$, but an interpolation between similar tetrahedral ions indicates a probable distance from Tc to O of about 1.7A. Discussion of the sizes of the atoms shows that this indicates a much greater squeezing in the $KTcO_4$, on account of the close distance of approach of the O atoms. This effect would probably be felt on the $4p$ electrons as well as on the $4d$'s; the effect would also be strong on the $5s$'s, but would not appear in the internal conversion probability on account of the small contribution of these electrons.

Of course, it is to be realized that the wave functions of the valence electrons surrounding the Tc, in the ion $(TcO_4)^-$, will be quite different from what they are in the isolated atom, and work is under way to investigate the actual form of wave functions in ions of this sort. It will not be possible to make real calculations of the effect of chemical combination on internal conversion probability until such wave functions have been studied.

I am much indebted to Dr. Bainbridge and Dr. Goldhaber for pointing out to me the interesting relations of this problem to the electronic wave functions in molecules and solids. Similar experiments may well be able to throw considerable light on the electronic distributions in compounds of various sorts.

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Proton-Proton Scattering at 240 Mev by a Magnetic Deflection Method*

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PROTON-PROTON scattering at 240 Mev has been observed for eight angles ranging from 171.3° to 108.1° center of mass. The cross section has the previously observed isotropic behavior down to about 15° (referring to the more commonly used supplementary scattering angle for the conjugate proton) and then increases sharply by a factor of 3.5 at 8.7°. This increase at the small c.m. angle, caused in part by Coulomb effects, has the same general trend observed by Chamberlain, Segrè, and Wiegand[1] at 11.3° for 345-Mev protons. The average value of the cross section (excluding the value at 8.7°) is 4.66 ± 0.39 mb/sterad., based on a $C^{12}(p, pn)C^{11}$ cross section of 49 ± 3 mb. The error in the average value includes the statistical error, the 6 percent error in the C^{11} cross section, a 4 percent error in the beta-counter calibration, and other estimated errors.

The apparatus has been described briefly before.[2] The detectors were photographic plates placed in the tank of the Rochester cyclotron near the lower pole tip. The protons scattered by protons in a hydrocarbon $(CH_2)_n$ target arrive at a given position on a plate in a nearly collimated group which corresponds to a well-defined angle of proton-proton scattering, while the protons scattered by the carbon nucleons arrive uncollimated. The angles at which the protons entered the plates were measured with a protractor eyepiece attached to a microscope. Entrance angle distributions were obtained for both hydrocarbon and carbon targets of suitable thicknesses, the incident beam being monitored by the C^{11} activity induced in each target. The number of protons scattered by protons was determined by subtracting the carbon target distribution from the hydrocarbon target distribution. As a result of energy loss in the thinnest carbon targets that could be used, thin polystyrene $(CH)_n$ targets were used to check the distribution of protons scattered by the carbon nucleons in the polyethylene $(CH_2)_n$ targets for the laboratory angles 83.1° and 85.4°.

The center-of-mass cross sections obtained are presented in Table I and Fig. 1. The indicated errors are the statistical errors in the number of tracks in the proton-proton peak and carbon background.

TABLE I. Experimental differential scattering cross section.

$\pi-\theta_{\mathrm{c.m.}}$	$d\sigma/d\Omega$, mb/sterad.
71.9°	4.33 ± 0.22
45.2°	4.81 ± 0.25
36.6°	4.90 ± 0.28
28.3°	4.43 ± 0.21
27.2°	4.38 ± 0.38
18.6°	4.59 ± 0.31
13.0°	5.16 ± 0.39
8.7°	15.8 ± 1.6