

(see reference 8) indicate that inclusion of the $V_v(\mathbf{h})$ can have little effect on the relative positions of the energy levels calculated for lithium using the Seitz potential.⁹

⁹ The Seitz potential does not satisfy the condition $(\nabla V)_{r_s=0}$, and this may require a significant correction.

All these calculations have been performed for $r_s=3.21$, and a lattice constant $a=6.518$.

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Low-Temperature Influence on the Technetium-99m Lifetime*

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The rate of decay of a nuclear isomer has been demonstrated to be influenced by its electronic environment. This effect has been utilized to detect environmental changes of Tc^{99m} in its metallic lattice at low temperatures. The effect of low temperature on the decay constant of Tc^{99m} in technetium metal was measured. Measurements were made at 77°K and 4.2°K. Since the metal is superconducting at 4.2°K, a measurement was made at 4.2°K in a magnetic field sufficient to destroy superconductivity. Results of the experiments indicate a negligible change in the decay constant for the metal at 77°K, whereas a noticeable change in the decay constant was observed for the superconducting metal at 4.2°K:

$$\lambda(4.2^\circ\text{K superconducting}) - \lambda(293^\circ\text{K}) = (6.4 \pm 0.4) \times 10^{-4} \lambda(293^\circ\text{K}).$$

Measurements on the low-temperature normal-state metal indicate a gross removal of this influence:

$$\lambda(4.2^\circ\text{K normal}) - \lambda(293^\circ\text{K}) = (1.3 \pm 0.4) \times 10^{-4} \lambda(293^\circ\text{K}).$$

THE decay of a radioactive nucleus is generally quite unaffected by the physical surroundings of the nucleus. However, in two examples, the decay constant has been shown to be affected slightly by chemical surroundings, i.e., in the decay by K capture of Be^7 and in the internal conversion² of Tc^{99m} . This change in decay constant is attributed to a change in the electron density near the nucleus.

Tc^{99m} is ideally suited to this type of experiment because of its extremely small isomeric transition energy of 2 kev. This low transition energy insures interaction with electrons only beyond the second shell where they may be more easily affected by external means. Since technetium has been found to be a superconductor,³ it was decided that an investigation of the decay constant at low temperatures might be of interest.

I. THE EXPERIMENT

The method employed for the measurement of minute differences of decay constant was the differential method of Rutherford, which has been used extensively in in-

vestigations of this nature.^{1,2,4,5} Two sources whose decay constants are very nearly equal are compared by placing the sources in identical ionization chambers and measuring the difference in ion current as a function of time. The difference in their decay constants may then be found by plotting the difference current times the inverse decay factor, $\exp(\lambda t)$, against time. The slope of the straight line which best fits these data is equal to the difference in decay constant times the initial strength of one source. This calculation and the standard deviations have been discussed in complete detail in the literature.⁶

The ionization chambers used in the present work were constructed of brass tubing $7\frac{1}{2}$ in. in diameter and 15 in. long with $\frac{1}{2}$ -in. stainless steel tubes soldered in the ends to permit essentially 4π counting geometry. The chambers were filled to a pressure of 200 psi with pure aged argon. A vacuum tube electrometer was used to measure ion currents.

Metallic technetium sources were prepared by using the method outlined by Bainbridge *et al.*⁷ Tc^{99m} was chemically separated from its parent Mo^{99} , ground-state technetium was added, and an oxide of technetium was electrodeposited from this solution onto a copper-plated

* This work is described in greater detail in a thesis submitted by Don H. Byers in partial fulfillment of the requirements for the degree of Doctor of Philosophy to the University of Kansas, 1958.

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¹ Kraushaar, Wilson, and Bainbridge, *Phys. Rev.* **90**, 610 (1953).

² Bainbridge, Goldhaber, and Wilson, *Phys. Rev.* **90**, 430 (1953).

³ J. G. Daunt and J. W. Cobble, *Phys. Rev.* **92**, 507 (1953).

⁴ Leininger, Segrè, and Wiegand, *Phys. Rev.* **81**, 280 (1951); **76**, 897 (1949).

⁵ Bouchez, Daudel, Daudel, Muxart, and Rogozinski, *J. phys. radium* **10**, 201 (1949).

⁶ See reference 2, Sec. II.

⁷ See reference 2, p. 434.

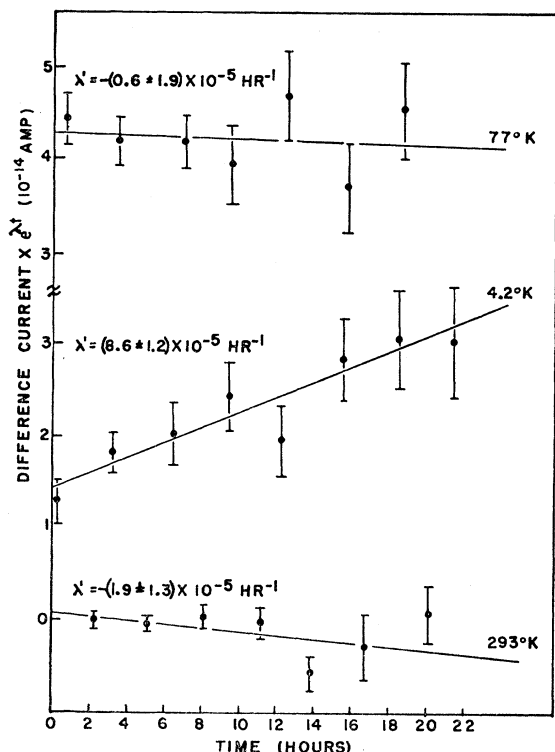


FIG. 1. Lifetime alteration at various temperatures. The increase in decay constant over the room-temperature comparison source is denoted λ' .

Nichrome cathode. The cathode deposit was reduced to the pure metal by heating the cathode wire in an hydrogen atmosphere. A thin film of aluminum was evaporated over the active layer in order to prevent reoxidation in the atmosphere. Both radio-isotopes were obtained from Oak Ridge National Laboratory. The reduced and aluminized active cathode wires were cut into equal lengths and sealed in source holders constructed from $\frac{1}{16}$ -in. diameter, 0.004-in. wall stainless steel tubing. The sources were carefully balanced at the outset of the experiment by tinning the source holders of the stronger sources with solder, and then filing away the solder until a balance was reached.

Four identical sources were prepared for each experiment. One source was selected as the comparison source and was kept at room temperature. The remaining three were kept in various low-temperature baths between measurements. An error is introduced in the measured value of the difference in decay constant due to the fact that the source must be warmed to room temperature during the measurements of difference current. Providing the difference current measurements are performed at equal intervals of time and each measurement has equal duration, the error may be corrected by multiplying the observed difference in decay constant by the ratio of the duration of the experiment to the accumulated time at low temperature.

Three sources were compared to determine the effect of temperature. One of these was left at room temperature along with the comparison source. This provided an over-all check of the methods, the source preparation techniques and the measuring equipment. A second test source was kept in a liquid nitrogen bath (approximately 77°K), and the third was left in liquid helium at 4.2°K. The results of this experiment are shown in Fig. 1. There is no difference in lifetime between the first two test sources and the comparison source, indicating first, satisfactory apparatus and techniques and second, that the electron distribution in technetium metal at 77°K is not altered enough to be detected. The third test source did, however, display a definite increase in decay constant.

In order to demonstrate that metallic technetium was present on the source wires in sufficient quantities to behave as the bulk material, a rough measurement of the critical field at 4.2°K was undertaken. The measurement was made by observing the shift in frequency of an oscillator due to the exclusion of flux by superconducting wires placed along the axis of its tank coil.

Measurements of Daunt and Cobble⁸ on 102.7 mg of Tc indicate that $\partial H_c/\partial T$ at the critical temperature is 300 to 400 gauss/degree. Assuming that the usual parabolic relation between critical field and temperature applied at 4.2°K, the critical field should equal 1440 to 1920 gauss. Our measurement on 4 wires (about 100 micrograms of technetium) yields a value of 1800 ± 200 gauss. Thus it may be concluded that the thin films used do not differ greatly in their superconducting properties from the bulk material. As a check on the apparatus, a similarly prepared lead specimen was found to have a critical field of 600 ± 50 gauss at that temperature. The accepted value for this measurement is 552 gauss.⁸

The experiment was repeated in liquid helium at 4.2°K, with two sources, one in zero field, the other in a

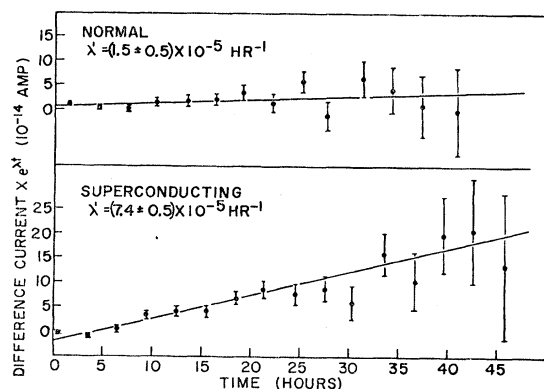


FIG. 2. Lifetime alteration at 4.2°K. Both sources were held at 4.2°K. The normal state was attained in a magnetic field of 5300 gauss. The increase in decay constant over the room-temperature comparison source is denoted λ' .

⁸ J. G. Daunt and J. W. Mendelsohn, Proc. Phys. Soc. (London) A160, 127 (1937).

field of 5300 gauss. Since the high field is well above the critical field, the low-temperature effect could be effectively separated from any possible superconductive effect. Results of this experiment (Fig. 2) confirmed the previous value of the altered decay constant in superconducting technetium. The low-temperature normal-state source displayed only a fraction of this change.

II. RESULTS

In order to calculate the difference in decay constant of two sources, one must know the decay constant of one source and the initial strength of the other. In practice the decay constant and initial strength of the same source is determined from the plot of log intensity *vs* time for that source. All data were assigned standard deviations on the basis of counting statistics coupled with the error introduced through the uncertainty in difference current measurements. The current resolution of the equipment for a typical measurement is 7×10^{-16} amp. The initial intensity of the comparison source in the final experiment was 7.66×10^{-11} amp. Standard deviations in the result were calculated according to the method outlined by Birge.⁹ Straight lines were fitted to the data by the method of least squares. The calculation of the coefficients of the straight lines, as well as the summations necessary for the standard deviation calculations, were carried out on an IBM 650 computer.

Table I lists all experimental results. The room temperature check source (see Fig. 1) displayed no altered lifetime within experimental accuracy, indicating the proper functioning of the apparatus and the purity of the sources with respect to active contaminants. This is further indicated by the ability to repeat the results in liquid helium using sources prepared separately from different shipments of the activity. The plot of log (intensity of one source) *vs* time for the last experiment contained 30 points spread over a period of twelve half-lives (72 hours). These data displayed no perceptible deviation from a straight line. This fact demonstrates the constancy of chamber sensitivity, freedom of the chambers from saturation effects and purity of the source with respect to active contaminants of half-life other than six hours. An active contaminant of the strength of 10 parts per million should have been quite noticeable. The value of λ appearing in Table I was found from these data.

TABLE I. Experimental results. The following table lists the change, λ' , in the decay constant for metallic Tc^{99m} under various conditions of temperature and magnetic field. A positive value for λ' indicates a decreased lifetime over that of the room-temperature field-free metal.

Source	$\lambda' \times 10^5 \text{ hr}^{-1}$	$(\lambda'/\lambda) \times 100$
293°K (check source)	-1.9 ± 1.3	not significant
77°K	-0.6 ± 1.9	not significant
4.2°K superconducting (temp. experiment)	8.6 ± 1.2	0.075 ± 0.010
4.2°K superconducting (field experiment)	7.4 ± 0.5	0.064 ± 0.004
4.2°K normal (field experiment, 5300 gauss)	1.5 ± 0.4	0.013 ± 0.004
Value of decay constant	$0.1156 \pm 0.0002 \text{ hr}^{-1}$	

Results of these experiments indicate that the electron distribution is not sufficiently altered at 77°K for detection by the methods of this experiment, whereas a definite alteration is experienced at 4.2°K in the superconducting material. The comparatively small value of increased decay constant in the normal metal at 4.2°K demonstrates that the effect is attributable to superconductivity.

In comparison with lifetime alterations found in technetium metal at extremely high pressures,¹⁰ one may predict that the volume contraction due to low temperature should not produce a measurably altered lifetime. This fact is substantiated by the results at 77°K and by the removal of the effect by magnetic field. There has been reported¹¹ a volume change at the superconducting transition. The dimensional change, however, amounts to only 1 part in 10^7 and will not account for the effect found by us. We are led to the conclusion, therefore, that the increased electron density in the vicinity of the nucleus in the superconducting metal is brought about by a redistribution of the electrons rather than a density change in the material.

One or both of two influencing factors could be responsible for a decreased lifetime in the superconductor. Certain conduction electrons may be permitted a closer approach to the nucleus in the superconducting material, or altered motion of the superconducting electrons may alter the shielding effects sufficiently to permit a slight contraction of the bound electron shells. The methods of this experiment do not permit any distinction between these possibilities.

ACKNOWLEDGMENT

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⁹ R. T. Birge, Phys. Rev. **40**, 225, 226 (1932).

¹⁰ K. T. Bainbridge (private communication).

¹¹ J. L. Olsen and H. Roher, Helv. Phys. Acta **30**, 49 (1957).