Effect of atomic charge state on nuclear lifetimes: ^{197}Au

M. Ulrickson, R. Hensler, D. Gordon, and N. Benczer-Koller
Department of Physics, Rutgers University, New Brunswick, New Jersey 08903*

and

H. DeWaard†
Bell Laboratories, Murray Hill, New Jersey 07974
(Received 21 September 1973)

The mean life of the 77.3-keV level in ^{197}Au has been measured simultaneously in neutral gold atoms and in atoms with a mean charge of $\bar{q} = +9.5 \pm 1.5$. Mean lives of $\tau^0 = 2.76 \pm 0.03$ nsec and $\tau^{+9.5} = 2.84 \pm 0.12$ nsec were obtained. These results indicate that even the removal of 10 electrons from this atom does not affect the electronic structure of the inner shells, and therefore has little effect on the internal-conversion coefficients and lifetime of this nuclear state. These results are in good agreement with both relativistic and nonrelativistic calculations.

[RADIOACTIVITY ^{197}Au; $E_x = 77.3$ keV, measured $T_{1/2}$ in neutral and ionized atoms, ^{197}Au(^{16}O, ^{16}O′), $E = 45$ MeV, ^{16}O-γ delayed coin; calculated ICC.]

I. INTRODUCTION

The influence of the chemical environment on the rate of disintegration of nuclei decaying by either electron capture or internal conversion has been under investigation for many years.[1] Recently such studies have been extended to other than purely chemical interactions affecting electronic densities, such as high pressure,[2] superconductivity,[3] large electric fields in anisotropic crystals,[4] and, for impurities in metals, interactions with valence and conduction bands of the host.[5] In all these cases the direct chemical or physical effects on the electronic density near the nucleus are relatively small, and therefore only small variations of the nuclear disintegration rates are expected. Observed changes in the disintegration constant λ are in the range $10^{-4} \leq \Delta\lambda/\lambda \leq 5 \times 10^{-2}$.

Bombardment with energetic heavy ions can excite the nucleus of an atom, while also leaving that atom in a highly ionized state. In this case, the electronic configuration of the atom is drastically changed so that rather large modifications of the nuclear decay rate might result through alteration of the internal-conversion coefficient of the nuclear transition.

Recently, in a Mössbauer effect experiment, Potzel and Perlow[6] observed a 20% narrowing in the resonance width of the 77-keV γ ray from the decay of the first excited state of ^{197}Au nuclei in the particular compounds AuCN and $KAu(CN)_2$, while the resonance for metallic ^{197}Au exhibited the linewidth corresponding to the measured lifetime. If this effect were indeed due to a change of the nuclear lifetime caused by a change in the internal-conversion rate (the internal-conversion coefficient for this transition is $\alpha = 4.2$), some strong chemical modification of the electronic structure must take place in these compounds as compared with the metal. An even larger change of lifetime might be expected for a ^{197}Au nucleus in a highly ionized atom. After the present investigation was started, a new measurement was carried out by Pfeiffer *et al.*[7] These authors measured the linewidth as a function of absorber thickness with greater statistical precision than Potzel and Perlow, and they conclude from an extrapolation to zero absorber thickness that no unusual line narrowing exists. However, the study of nuclei in highly ionized atoms is interesting *per se*, insofar as the nucleus can be viewed as a probe of the electronic structure of the ionized atom, and we report here the results of such an investigation.

II. EXPERIMENTAL PROCEDURE

Levels of ^{197}Au were populated by Coulomb excitation with 45-MeV ^{16}O ions. The target consisted of 200-μg/cm^2 ^{197}Au evaporated onto both sides of a 750-μg/cm^2 carbon foil. Those ^{16}O ions backscattered at a mean angle of 170° were detected in an annular surface-barrier detector, while γ rays

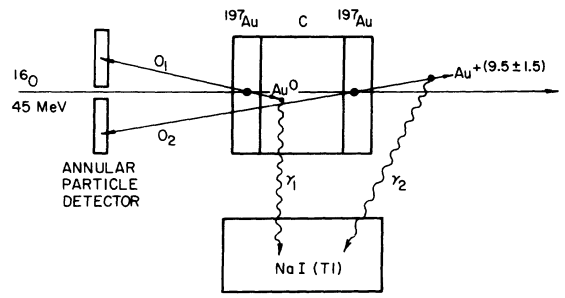

FIG. 1. The target and detector geometry utilized for the present measurement. The subscripts 1 and 2 refer to backscattered ^{16}O ions and γ rays from Au nuclei in the upstream and downstream target layers, respectively.

in coincidence with the backscattered ions were detected in a 3.8-cm-diam by 2.5-cm-thick NaI (Tl) scintillation detector placed 4 cm from the target at 90° with respect to the beam (Fig. 1).

Standard fast-slow time-to-amplitude conversion coincidence circuits were used. The time spectrum was calibrated with an air trombone. The decay of the 77-keV level yields the delayed coincidence spectrum, while the decays of the 279- and 269-keV states provide a prompt time distribution with full width at half maximum (FWHM) = 2.3 nsec. Figure 2 shows a typical γ-ray energy spectrum as well as the γ-ray windows selecting the prompt and delayed contributions. Energy windows were also set on the spectrum of ^{16}O ions backscattered from both Au target layers (Fig. 3). The ^{197}Au atoms from the upstream layer of gold recoil into the

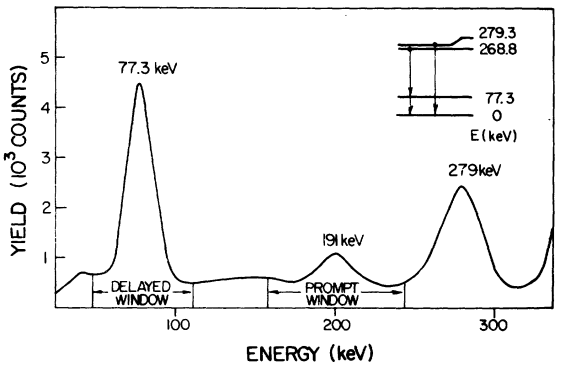

FIG. 2. A typical γ-ray energy spectrum obtained with the NaI(Tl) detector in coincidence with ^{16}O ions backscattered into the annular detector from the upstream Au target layer. A similar spectrum was obtained for γ rays in coincidence with ^{16}O ions from the downstream layer. The energy windows used for the slow-coincidence requirement and for dividing the time spectrum into prompt and delayed contributions are illustrated.

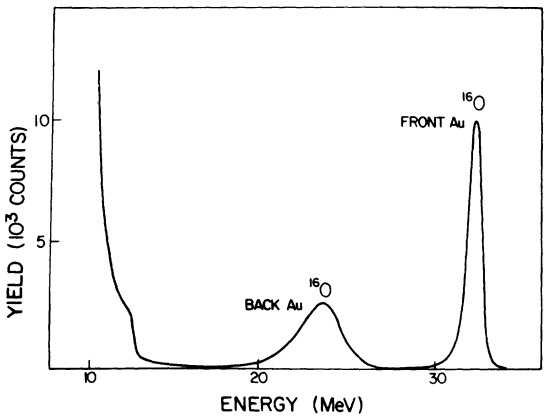

FIG. 3. The spectrum of ^{16}O particles backscattered from the target into the annular detector. The peaks are due to ^{16}O ions scattered from the downstream (back) and upstream (front) Au target layers, respectively.

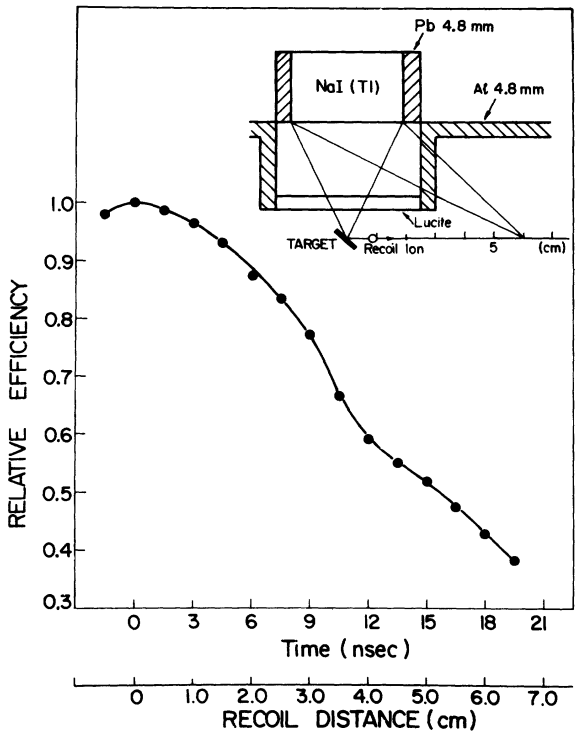

FIG. 4. The relative efficiency of the NaI(Tl) detector as a function of distance downstream from the target position, measured with a ^{241}Am source. This curve reflects the change in solid angle subtended by the detector as well as the differential γ-ray absorption by the target chamber and shielding. The target-detector geometry is illustrated in the inset. The time scale is established from the distance scale through use of the Au ion recoil velocity $v = 0.33$ cm/nsec. The statistical uncertainties are within the size of the data points.

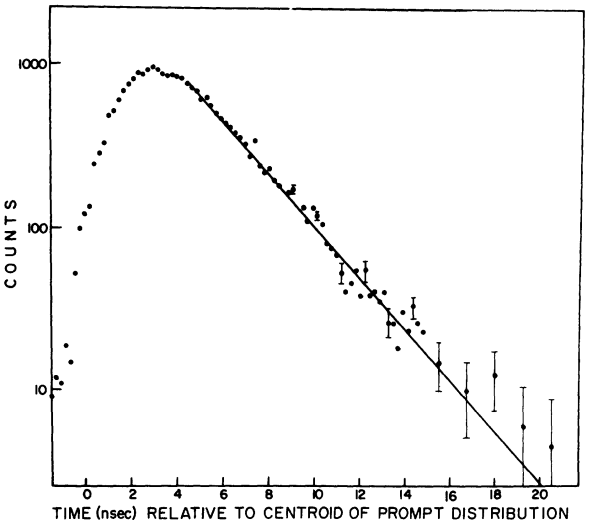

FIG. 5. The delayed coincidence time spectrum obtained from the decay of neutral ^{197}Au nuclei at rest in the carbon layer. These data have been corrected for the time-dependent and -independent backgrounds as described in the text. This spectrum was obtained utilizing γ rays observed in the "delayed" window indicated in Fig. 2 in coincidence with ^{16}O ions backscattered from the front Au surface and lying in the peak indicated in Fig. 3. The solid line is a least-squares fit to an exponential function.

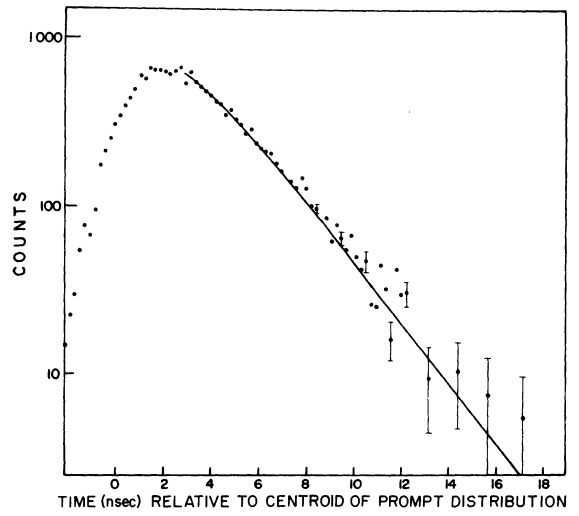

FIG. 6. The delayed coincidence time spectrum obtained from the decay of ^{197}Au with charge $\bar{q} = 9.5 \pm 1.5$ recoiling into vacuum. These data have been corrected for time-dependent and -independent backgrounds as described in the text. This spectrum was obtained utilizing γ rays observed in the "delayed" window indicated in Fig. 2 in coincidence with ^{16}O ions backscattered from the back Au surface and lying in the peak indicated in Fig. 3. The solid line is a least-squares fit of the data to the function in Eq. (2).

carbon and stop before their nuclei decay, and thus are neutral when the γ ray is emitted. However, the 12.0-MeV ^{197}Au atoms from the downstream layer of gold recoil into vacuum and have a mean equilibrium charge of $\bar{q} = 9.5 \pm 1.5$ when they decay. This mean charge was determined from the semiempirical estimate of Nikolaev and Dmitriev.[8] Since the mean velocity of the gold ions recoiling into vacuum is 0.33 cm/nsec, they move on the average an appreciable distance down the beam line before they decay. In order to correct for this effect, the efficiency of the γ-ray detector was measured as a function of recoil distance with a ^{241}Am source which was moved along the beam direction (Fig. 4).

There were two sources of background in the delayed coincidence time spectra; a constant background independent of time, and a tail that corresponded to a long-lived impurity. A few tenths of one percent of ^{191}Ir and ^{193}Ir known to be present in the Au foil could account for both the magnitude and the shape of the tail. After the constant background was subtracted, the long-lived portion of the spectrum was fitted to an exponential function with decay constant 6.5 nsec, corresponding to the average mean life of the impurity states [^{191}Ir* (82.3 keV, 5.6 nsec) and ^{193}Ir* (73.0 keV, 8.9 nsec)] weighted by their isotopic abundance and relative excitation probability.

III. RESULTS

Figure 5 shows the corrected delayed coincidence time spectrum from neutral gold atoms at rest in the carbon at the time of nuclear decay. The solid line is the least-squares straight-line fit to these data. The mean life obtained from this fit is $\tau = 2.76 \pm 0.03$ nsec. These data were also analyzed using a technique presented previously[9] whereby the delayed distribution is generated from the observed prompt distribution and is given by

$$D(t) = \frac{1}{\tau} \int_0^t e^{-(t-t')/\tau} P(t')\, dt' . \qquad (1)$$

The best fit obtained by this technique gave $\tau = 2.77 \pm 0.04$ nsec, in excellent agreement with the straight-line method and with the recent electronic measurement of Lynch[10] who obtained $\tau = 2.78 \pm 0.04$ nsec.

Figure 6 shows the delayed coincidence spectrum from ionized gold atoms recoiling into vacuum. Because of the change in solid angle and efficiency of the γ-ray detector due to decay of nuclei distributed along the beam axis, no straight-line fit can be made to these data. A delayed distribution was therefore generated which includes an effi-

ciency correction factor

$$D(t)=\frac{1}{\tau}\,\frac{\int_0^t e^{-(t-t')/\tau}P(t')\epsilon(t-t')\,dt'}{\int_0^\infty \epsilon(t'')\,dt''}, \qquad (2)$$

where $\epsilon(t)$ is the efficiency shown in Fig. 4. The mean life obtained from the best fit to the data is $\tau = 2.84 \pm 0.12$ nsec.

From the present data, a relative change $(\tau^0 - \tau^{+\bar{q}})/\tau^0 = (-0.029 \pm 0.045)$ is found between the lifetimes of the 77-keV state in neutral ^{197}Au atoms and in ^{197}Au ions of charge $+\bar{q}$. An analogous experiment was carried out by Young, Brenn, and Sprouse[11] who measured the Coulomb excitation probability of the 77.3-keV state using ^{16}O ions and determined the lifetime of the state by γ-γ coincidence techniques. They report a conversion coefficient $\alpha = 2.6 \pm 0.4$ and a ratio $\tau_{Au}/\tau_{vac} = 1.03 \pm 0.06$, or $(\tau_{Au} - \tau_{vac})/\tau_{Au} = +0.03 \pm 0.06$ for the lifetimes of the state as measured with the atom in a metal environment or in vacuum.

IV. DISCUSSION

Measured[12] and calculated[13] (Table I) internal-conversion coefficients for the 77.3-keV transition in the neutral Au atom are in good agreement. We have calculated the conversion coefficients for gold nuclei in ions with a mean charge $\bar{q} = +10$ by the procedure outlined below. The wave functions corresponding to the [Xe] $(4f)^{14}$ $(5d)^1$ electron configurations were calculated from the nonrelativistic Hartree-Fock-Slater program of Herman and Skillman.[14] It is assumed that the gold ion reaches this configuration in a time short compared with the lifetime of the 77-keV state.

The s-shell conversion coefficients for the ionized atom α_s^{+10} were obtained by normalizing the calculated conversion coefficients[13] α_s^0 for the neutral atom by the ratio of calculated s-electron densities at the nucleus:

$$\alpha_s^{+10}/\alpha_s^0 = |\psi_s(0)|^2_{+10}/|\psi_s(0)|^2_0 .$$

This procedure was justified by Band, Sliv, and Trzhaskovskaya,[15] who showed that conversion of all s electrons takes place in a region well inside the radius of the K shell, where the shape of the wave function is not affected by changes in the charge distribution in the outer shells. Thus, the squares of the radial integrals that appear in the calculations of conversion coefficients are directly proportional to the s-electron densities at the nucleus. Due to the (10%) $E2$ admixture in the 77-keV transition, conversion in the p shells is comparable in magnitude to that in the s shells. Band, Sliv, and Trzhaskovskaya[15] also showed that $E2$ p conversion is not restricted to the inner region of the atom but occurs within a region defined by the radius of the L shell. In this case, we have compared the radial p wave functions of the $\bar{q} = +10$ ion and the neutral atom for all radii. It appears that the shapes of the radial $p(+10)$ and $p(0)$ wave functions are almost indistinguishable up to a distance well beyond the radius of the L shell. Hence the conversion coefficients were obtained by normalizing α^0 by the average ratio of wave functions in the region where conversion takes place. The results are displayed in Table I.

TABLE I. Calculations of subshell internal-conversion coefficients for neutral (Ref. 13) and ionized (this work) gold atoms. The uncertainty in the relative conversion coefficients is discussed in the text. $\alpha^0 = 4.188$; $\alpha^{+10} = 4.195$; $(\alpha^0 - \alpha^{+10})/\alpha^0 = -0.0017 \pm 0.0030$.

Subshell		Au^0	Au^{+10}
$2s$	L_I	1.819	1.819
$2p$	L_{II}	0.791	0.791
	L_{III}	0.569	0.569
$3s$	M_I	0.415	0.415
$3p$	M_{II}	0.199	0.199
	M_{III}	0.154	0.154
$4s$	N_I	0.113	0.113
$4p$	N_{II}	0.053	0.053
	N_{III}	0.038	0.038
$5s, p$	O	0.035	0.044
$6s$	P	0.002	0.000

The conversion of s and p electrons in the $n = 2$ to 4 shells together constitutes 99% of the total conversion for the neutral atom, the remaining 1% being contributed by the O and P shells. This small fraction of the conversion was estimated for the $\bar{q} = +10$ gold ion in the same way as outlined for the other shells, even though the approach used for these shells might not be strictly valid here. However, even an error of 30% in the estimates for s and p conversion for these outer shells will only lead to a change of total conversion coefficient of 0.3%. This uncertainty is included in the calculated total relative change of conversion coefficient: $(\alpha^0 - \alpha^{+10})/\alpha^0 = -0.0017 \pm 0.0030$. The corresponding relative change in the mean life is $[(\tau^0 - \tau^{+10})/\tau^0]_{calc} = 0.0014 \pm 0.0024$.

Thus, it turns out that, even though 10 electrons are removed from this atom, the conversion of the remaining electrons increases to such an extent as to leave the total conversion coefficient almost constant. Complete relativistic calculations carried out by Obenshain *et al.*[16] yield $(\alpha^0 - \alpha^{+10})/\alpha^0 = -0.001$. The small relative change in conversion implies that no effect on the lifetime of the state should have been observed within 0.3%. This is in agreement with the experimental result.

We would like to thank R. Klein for preparing the sandwich target.

*Supported in part by the National Science Foundation.

†Permanent address: Natuurkundig Laboratorium, Groningen, The Netherlands.

[1]E. Segrè, Phys. Rev. 71, 274 (1947); S. DeBenedetti, F. deS. Barros, and G. Hoy, Annu. Rev. Nucl. Sci. 16, 31 (1966).

[2]R. A. Porter and W. G. McMillan, Phys. Rev. 117, 795 (1960).

[3]D. H. Byers and R. Stump, Phys. Rev. 112, 77 (1958); A. Olin and K. T. Bainbridge, Phys. Rev. 179, 450 (1969).

[4]P. Huber, St. Gagneux, and H. Leuenberger, Phys. Lett. 27B, 86 (1968).

[5]H. Nève de Mevergnies, Phys. Rev. Lett. 29, 1188 (1972).

[6]W. Potzel and G. J. Perlow, Phys. Rev. Lett. 29, 910 (1972).

[7]L. Pfeiffer, R. S. Raghavan, C. P. Lichtenwalner, and K. W. West, Phys. Rev. Lett. 30, 635 (1973).

[8]V. S. Nikolaev and I. S. Dmitriev, Phys. Lett. 28A, 277 (1968).

[9]J. W. Tape, N. Benczer-Koller, R. Hensler, and J. R. MacDonald, Phys. Lett. 40B, 635 (1972).

[10]F. J. Lynch, Phys. Rev. C 7, 2160 (1973).

[11]L. E. Young, R. Brenn, and G. D. Sprouse, Bull. Am. Phys. Soc. 18, 655 (1973).

[12]I. J. van Heerden, D. Reitmann, and H. Schneider, Nuovo Cimento 11, 167 (1959).

[13]R. S. Hager and E. C. Seltzer, Nucl. Data A4, 1 (1968); O. Dragoun, H. C. Pauli, and F. Schmutzler, *ibid.* A6, 235 (1969).

[14]F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice Hall, Englewood Cliffs, New Jersey, 1963).

[15]I. M. Band, L. A. Sliv, and M. B. Trzhaskovskaya, Nucl. Phys. A156, 170 (1970).

[16]F. E. Obenshain, private communication; C. C. Lu, T. A. Carlson, F. B. Malik, T. C. Tucker, and C. W. Nestor, Jr., Atomic Data 3, 1 (1971).