Precision measurement of the lifetime of the $7\,^3S_1$ state of atomic mercury*

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We report the first observation of the atomic cascade $9\,^1P_1$-$7\,^3S_1$-$6\,^3P_1$ in mercury. The exponential distribution of time delays between detection of the $9\,^1P_1$-$7\,^3S_1$ (5676-Å) photon and the $7\,^3S_1$-$6\,^3P_1$ (4358-Å) photon was measured, and a value of 8.2(0.2) nsec was obtained for the mean lifetime of the $7\,^3S_1$ level.

INTRODUCTION

The cascade coincidence method has distinct advantages over other means for determining lifetimes of excited atomic levels.[1,2] It is applicable to very highly excited states, unlike the Hanle-effect method,[3] in which one is restricted to states which can be reached from the ground state with resonance radiation or by the rather difficult technique of stepwise excitation.[3,4] In contrast to beam-foil spectroscopy[5-10] and pulsed-electron-beam excitation,[11,12] it is completely free from the effects of unwanted cascading. In this paper we will describe a precision lifetime measurement in mercury and give criteria for the optimization of such experiments.

APPARATUS

Figure 1 shows a schematic diagram of the apparatus. A light source was prepared by evacuating (to ~ 10^{-8} torr) a Pyrex envelope containing a commercial cathode-ray-tube electron gun, sealing the lamp off, and then transferring a small quantity of triply distilled Hg through a breakoff seal connecting the lamp to a previously prepared capsule of Hg. In the region where observations were made the Pyrex envelope had an inner diameter of 12.5mm. The gun provided an easily controlled and steady beam of low-energy electrons. It was typically run at a final accelerating potential of 100 V and gave about 1 μA in the interaction region. The electron beam had a diameter of approximately 1mm.

As shown in Fig. 1, photomultipliers were set up on opposite sides of the lamp, with narrow-band dielectric interference filters to select the desired wavelengths, and collimating lenses to collect photons in cones of about 40° half-angle. Figure 2 shows the relevant energy levels of atomic mercury. An Amperex 56DVP photomultiplier with a bialkali cathode was used to detect the 4358-Å photons, and a trialkali 56TVP was used for the 5676-Å photons. The latter was thermoelectrically cooled to −25°C to reduce dark counts. With gains adjusted to ~ 10^9 these tubes required no further amplifiers, and the anode pulses were fed directly to Chronetics 101 fixed −100-mV threshold discriminators. The discriminator outputs went to the start and stop inputs of an EG&G TH200A/N time-to-amplitude converter (TAC). The TAC output pulses were measured and stored by a multichannel pulse-height analyzer (PHA). In the early runs this was a RIDL 34-12B; in the later ones a Northern Scientific NS-600 was substituted.

Measurements were made at room temperature where the mercury pressure in the lamp was roughly 1.4×10^{-3} torr. In order to rule out the effect of radiation trapping on the lifetime measurement, it is necessary to estimate the equilibrium density of atoms in the terminating $6\,^3P_0$ level. A recent calculation[12] indicates that the $6\,^3P_0$ direct electron excitation cross section has fallen to 10^{-18} cm^2 at an incident energy of 40 eV. The most likely indirect process is electron excitation to the $6\,^3D_1$ level followed by spontaneous emission, or to the $6\,^3P_2$ level followed by cascade decay via the $7\,^3S_1$ level. The available data[13] indicate that these cross sections are less than 10^{-18} cm^2 at 50 eV. Taking 10^{-17} cm^2 as an estimate for all processes, we find that about 10^{10} metastables/sec are produced in the source region. On the average each one remains in this region for 3×10^{-5} sec so the equilibrium density of $6\,^3P_0$ atoms is on the order of 4×10^5 per cm^3. This is far too small to produce a noticeable degree of radiation trapping.[14]

The ideal PHA spectrum consists of an exponential distribution whose decay constant is the inverse of the intermediate state lifetime, plus a flat background due to events in which the two photons are from different atoms and thus are not time-correlated. In order to measure this background, we inserted an extra length of cable in the stop channel to shift the zero-time-delay channel to the middle of the PHA range. To the left of this channel the spectrum contains only the flat background,

and this can then be used to subtract the background in the region to the right of the prompt channel.

OPTIMIZATION

Although it may at first glance appear desirable to maximize the signal-to-background ratio, this does not yield the optimum rate of information collection. The latter is obtained when the fractional error in the signal is smallest, for a given run time T. Let N_A be the rate at which atoms emit the 5676-Å photons in the first stage of the cascade, and N_B the rate of emission of 4358-Å photons. If the over-all detection efficiencies are η_A and η_B, then the start and stop single rates will be

$$C_A = \eta_A N_A, \quad C_B = \eta_B N_B, \tag{1}$$

respectively. In the region between zero time delay and a time delay equal to one $7\,^3S_1$ lifetime, τ, there will be

$$B = C_A C_B \tau T \tag{2}$$

random coincidences and

$$S = 0.63\beta\eta_B C_A T \approx \beta\eta_B C_A T \tag{3}$$

true coincidences. Here β is the branching ratio for the $7\,^3S_1$-$6\,^3P_1$ decay relative to all decays from that level. In practice we can only measure the sum of these two and then do a background subtraction. The latter makes only a small contribution to the error, however, so that the scatter in the net signal is approximately equal to the square root of the raw signal before background subtraction. It is then an easy matter to show that the "noise-to-signal" ratio is given by

$$(S/N)^{-1} = (S/N)_\infty^{-1}(1 + \beta/N_B\tau)^{1/2} \tag{4}$$

in which

$$(S/N)_\infty^{-1} = (\tau/\eta_A\eta_B\alpha\beta)^{1/2}T^{-1/2} \tag{5}$$

is the value which $(S/N)^{-1}$ approaches asymptotically as the lamp intensity becomes infinite ($N_B \to \infty$). We have introduced α, the rate at which atoms enter the $7\,^3S_1$ level via the $9\,^1P_1 \to 7\,^3S_1$ transition divided by the total rate at which this level is populated, and have used the fact that in the steady state

$$N_A/\alpha = N_B/\beta. \tag{6}$$

Hence we see that the rate of information collection increases monotonically with source intensity, but as a practical matter there is little improvement beyond the point at which

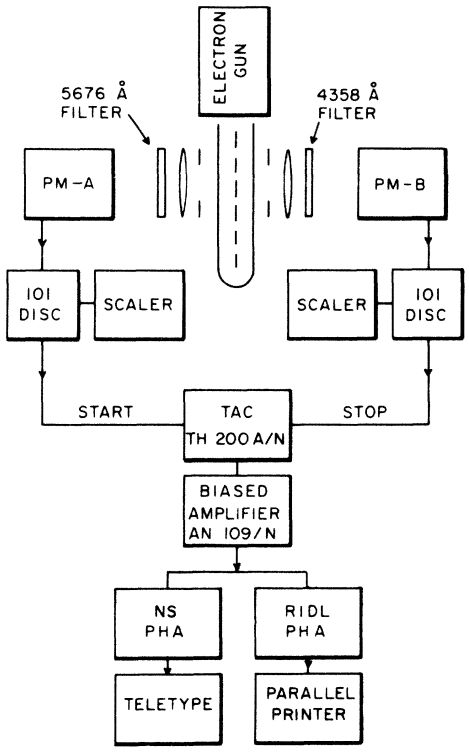

FIG. 1. Schematic diagram of the apparatus.

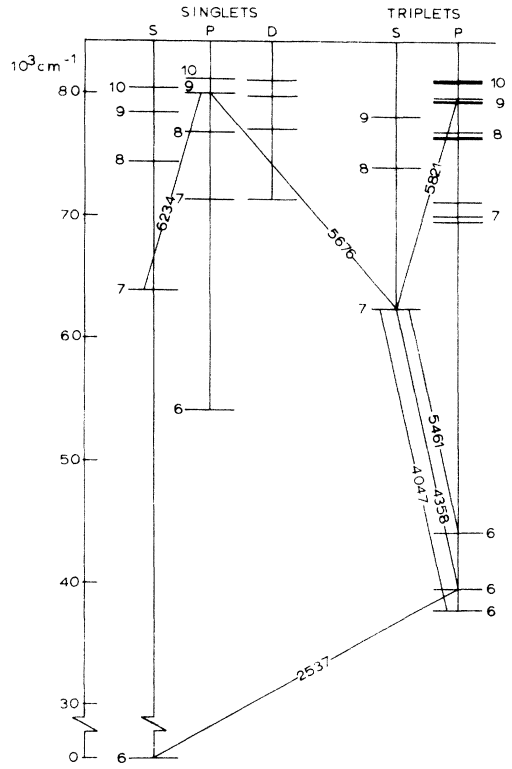

FIG. 2. Relevant energy levels of atomic mercury and transition observed in this experiment.

$$N_B \gtrsim \beta/\tau. \tag{7}$$

From Eqs. (2) and (3) we have

$$S/B = \beta/N_B\tau; \tag{8}$$

thus Eq. (7) is equivalent to

$$S/B \lesssim 1. \tag{9}$$

For this experiment S/B was approximately 0.6 for the RIDL data and 0.3 for the NS data. It can also be seen from Eq. (5) that it is advantageous to make τ as small as possible and α, β, and η's as large as possible. In particular, one wishes to avoid populating the $7\,^3S_1$ level by direct electron excitation or by cascading via levels other than the $9\,^1P_1$ (the worst offender in this case is the $7\,^3P_2$). The optical-excitation cross-section for the $7\,^3S_1 - 6\,^3P_1$ transition peaks near threshold and falls rapidly at higher incident-electron energies, whereas the $9\,^1P_1 - 7\,^3S_1$ cross section is nearly flat out to quite high energies,[13] so our choice of 100-eV electrons reduces this problem greatly.

RESULTS

The complete data are shown in Fig. 3. The differential nonlinearity was approximately ± 1.5% for the RIDL over the range of interest, while for the Northern Scientific PHA it was only about ± 0.5%. These figures were determined from

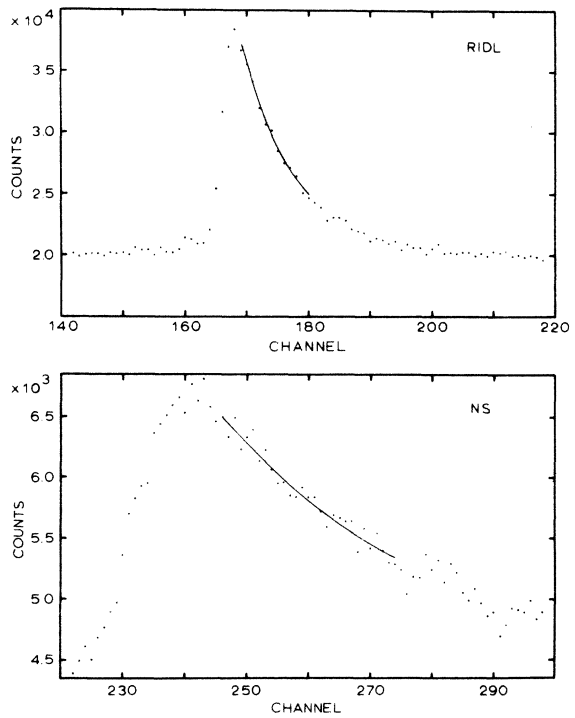

FIG. 3. TAC-PHA spectra for runs with both the RIDL analyzer and the NS analyzer.

TABLE I. Summary of the measured lifetimes for the $7\,^3S_1$ state of atomic mercury.

Author	Ref.	Method	$\tau(7\,^3S_1)$ (nsec)
Brannen *et al.*	17	Cascade	11.2(0.2)
Agárbiceanu *et al.*	18	Double resonance	11.16(0.6)
Barrat *et al.*	19	Hanle effect	8.1(0.2)
Pardies	20	Cascade	9.7(2.0)
Present work		Cascade	8.2(0.2)

separate linearity runs in which an extra time delay was added to the stop channel to move the signal completely out of the region of interest. These spectra were fitted to quadratic curves which were then used to compute the relative channel widths. It was not possible to accumulate sufficient statistics to do a channel-by-channel linearity analysis, and it was in fact the small-scale variations of linearity that ultimately limited the experimental accuracy. We were, however, able to avoid two well-known sources of spectrum distortion. By choosing a sufficiently low stop channel singles rate, one can avoid the background droop due to the start/stop nature of the TAC. Since the TAC is halted by the first stop pulse that comes along following a valid start, the apparent rate of coincidences in which the time delay equals t is given by the rate at which these occur, multiplied by the probability that no stop pulses come along between 0 and t. The latter factor is $e^{-C_B t}$ from the Poisson distribution. It can be made arbitrarily close to 1 throughout the entire time range by choosing a low enough C_B. The second type of distortion has been discussed by Lowe *et al.*,[15] and involves a mutual interaction of the correlated and uncorrelated events. It is negligible in our case because η_B is quite small.

The absolute time calibration was carried out by the method described by Baker *et al.*[16] The average channel width in Fig. 3 is 922.4 psec for the RIDL and 219.8 psec for the NS.

The data were fit to exponential curves by an iterative nonlinear least-squares computer analysis program. The final result was

$$\tau(7\,^3S_1) = 8.2(0.2) \text{ nsec}. \tag{8}$$

Table I summarizes the lifetime obtained for this state by other observers.[17-20] The agreement with the most recent experiments is excellent. The discrepancy with Agárbiceanu *et al.* may be due to their use of a high-pressure nitrogen buffer gas. We cannot suggest a reason for the disagreement with Brannen *et al.*

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