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CORRELATION OF PHOTONS IN CASCADE AND THE COHERENCE TIME OF THE 6^3P_1 STATE OF MERCURY*

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We have observed directly the time correlation between successively emitted photons in the 7^3S_1-6^3P_1-6^1S_0 cascade of atomic mercury, and have deduced from it direct measurements of both the natural lifetime of the 6^3P_1 state and the dependence of the mean correlation time (coherence time) on the density of the mercury vapor surrounding the primary radiation region. The density-dependent coherence time constitutes a direct observation of the imprisonment of resonance radiation and provides an experimental confirmation of the theory describing this phenomenon.

Observations of optical photons in cascade have been reported by several authors.[1,2] We describe here the application of delayed coincidence counting techniques to an atomic system, leading to the detailed observation of the time correlation between the successively emitted photons in the 7^3S_1-6^3P_1-6^1S_0 cascade of atomic mercury. Examination of this correlation over a wide range of pressures has yielded a direct and unambiguous measurement of both the natural lifetime of the 6^3P_1 state (the "zero-pressure" mean correlation time) and the dependence of the mean correlation time (coherence time) on the density of mercury vapor surrounding the primary radiation region. The density-dependent coherence time constitutes a direct observation of the imprisonment of resonance radiation, which is responsible for the observed narrowing of resonance lines.

The experimental apparatus, along with the cascade under investigation, is shown schematically in Fig. 1. The desired excitation was produced by bombarding mercury vapor[4] with a well collimated dc beam of 365-eV electrons. Both the mercury vapor and the electron gun were enclosed in a sealed quartz envelope, in which the density of mercury vapor was controlled by controlling the temperature of the lower portion of the source tube. Although the ionization potential of atomic mercury is only 10.38 V, it was necessary to operate the electron gun with the final focusing electrode and the plate at the high voltage mentioned above in order to satisfactorily draw the electrons emitted from the cathode into the radiation tube, some 4 in. away. A cathode-ray gun of this type, rather than a simple small-gap cathode-plate arrangement, was employed in order to keep the filament sufficiently far from the radiation region as to render undetectable its blackbody radiation. The mercury radiation itself was emitted from a pencil beam of about 1 mm in diameter, and was collected by cylindrically symmetric conical mirrors with vacuum-evaporated aluminum faces. The photons of interest were selected by 4358- and 2537-Å interference filters with 100-Å bandwidths and peak transmissions of about 70 and 20 %, respectively. These photons were then converted by high-speed, high-gain photomultipliers

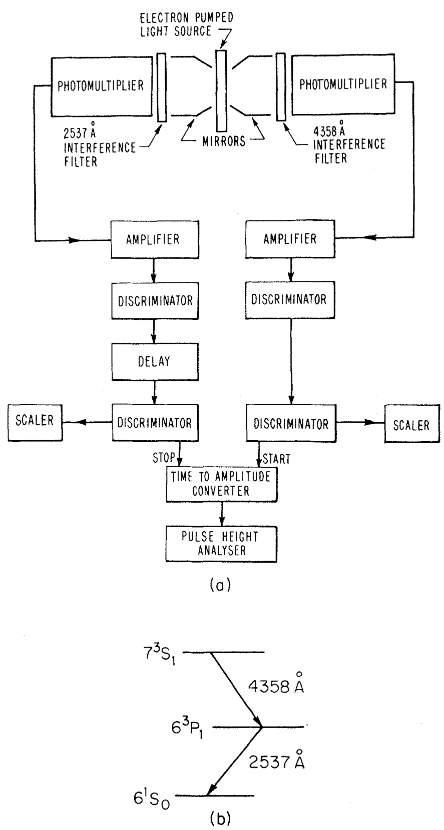

FIG. 1. (a) Block diagram of the apparatus. (b) Cascade under investigation.

to pulses of about 5 nsec full width at half-maximum. After further amplification the pulses were sent to fast discriminators and then to a time-to-amplitude converter. The output pulses of the converter were sent to and stored in a 400-channel pulse-height analyzer.

The time-to-amplitude converter is turned on by a pulse due to a 4358-Å photon and shut off by one due to a 2537-Å photon. Since such an accepted start-stop pulse pair gives rise to an output pulse whose height is directly proportional to the time separation between the input pulses, the resulting distribution of output pulse heights from the converter yields directly the time correlation between the successively emitted photons of the cascade under investigation.

Experiments were conducted at five different pressures, ranging from 2.35×10^{-5} mm to 1.8×10^{-3} mm of mercury. At each of these pressures the voltage on the accelerating electrode of the electron gun was adjusted to insure the best possible beam definition. Furthermore, it was found that the orientation of the electron beam could be kept approximately constant from run to run. It is estimated that the effective thickness of the absorption layer of mercury vapor, that is, the average distance that a detected 2537-Å photon, emitted from the pencil beam, had to travel before leaving the source tube (without being absorbed), was about 3.5 mm. The filament and regulating grid voltages were adjusted so that the "4358" and "2537" single-channel counting rates (C_1 and C_2, respectively) were in the neighborhood of 1000 counts/sec. These single-channel rates were monitored at regular intervals throughout each run by scalers, as indicated in Fig. 1. The over-all single-channel efficiencies, ϵ_1 and ϵ_2, were about 8.7×10^{-4} and 1.3×10^{-4}, respectively.[5]

The decision to run at the above singles rates represents a compromise, necessitated by the desire to satisfy simultaneously a number of requirements, which are not all completely compatible. Perhaps the prime requirement is that the contribution to the singles rates from the light source should be large compared with the dark rates of the photomultiplier tubes themselves. Although statistical considerations alone would also suggest the desirability of running at high counting rates, the desire to minimize systematic errors as well (specifically, the error introduced by dead-time losses in the "stop" input of the time-to-amplitude converter) puts an upper limit on singles rates, especially C_2.

Such requirements originally necessitated cooling of the photomultiplier tubes to about $-150°$C, and the early runs were carried out under these conditions. We were later able to obtain tubes whose dark rates at room temperature were no higher than those previously obtained with the photomultipliers cooled. Hence the remainder of the experiments were carried out with the phototubes at room temperature. In both cases the dark rates, n_1 and n_2, were about 100 counts/sec.

The time-to-amplitude converter and pulse-height analyzer were adjusted so that the resolution of the combination was 5 nsec per channel. An external delay of 550 nsec was inserted in the "2537" channel (see Fig. 1) in order to allow the noise background, due to accidental coincidences, to be determined separately and precisely.

Statistical analysis of the data was employed to determine the noise background, to subtract it from the spectrum in the region where counts from both true and accidental coincidences were stored, and to fit the remaining signal spectrum with an exponential. The "least-squares best fits" to the data were very good, with P, the χ^2 probability, ranging from 0.431 to 0.852.[6]

The curves so obtained, which represent the photon time correlation functions at the respective pressures, are presented in Fig. 2(a). The results of a particular run, at a pressure of 5.3×10^{-4} mm of Hg, are presented in Fig. 2(b).

At 2.35×10^{-5} mm of mercury, the lowest pressure employed, the minimum mean free path for absorption of a 2537-Å photon is nearly 30 times the thickness of the absorption layer ($L = 3.5$ mm). Thus, the coherence time obtained for this pressure is equal to the natural lifetime of the 6^3P_1 state to well within the statistical error of the measurement. The value $(1.14\pm0.14)\times10^{-7}$ sec obtained here agrees with those found by other methods.[7]

As was mentioned earlier, the pressure (or density) dependence of the coherence time is due to the imprisonment of resonance radiation. The problem of radiation imprisonment or "trapping" has been treated extensively in the literature.[8-11] If τ_0 represents the true mean life (zero-pressure coherence time) of the 6^3P_1 state, and τ_c the coherence time at some pressure P, then theoretically it is predicted that[12]

$$\tau_c = \tau_0/(1-x),$$

where according to D'yakonov and Perel[11]

$$x = \frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}\exp(-\omega^2)[1-\exp(-k_0Le^{-\omega^2})]d\omega,$$

and according to Barrat[10]

$$x = 1-\exp[-(\pi/6)^{1/2}k_0L].$$

In both cases[13]

$$k_0 = \frac{1}{5}\frac{2}{\Delta\nu_D}\left(\frac{\ln 2}{\pi}\right)^{1/2}\frac{\lambda_0^2}{8\pi}\frac{g_2}{g_1}\frac{N}{\tau_0}\quad (P=NkT).$$

The quantity x represents the fractional absorption of 2537-Å radiation, or equivalently, the probability for absorption of a 2537-Å photon by a layer of mercury vapor of density N and thickness L.

For the case where the widths of the individual emission and absorption lines are the same and equal to the Doppler width $\Delta\nu_D$ (as is assumed here), k_0 represents the inverse absorp-

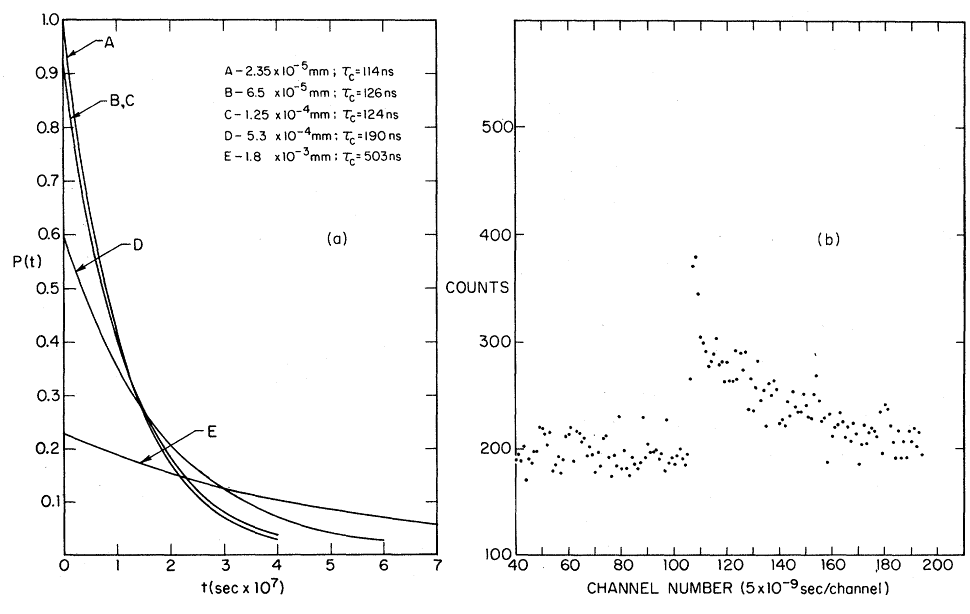

FIG. 2. (a) Experimentally determined time correlation between 4358- and 2537-Å cascade photons for a variety of pressures. Correlation function expressed in normalized, dimensionless form, $P(t) = (\tau_0/\tau_c)\exp(-t/\tau_c)$. For further details on experimental conditions and results consult Table I. (b) Results of a particular run, before analysis. The spectrum below channel 105 is due to accidental coincidences only and serves to determine the noise background. The spectrum above channel 110 contains counts due to both true and accidental coincidences. Here pressure $=5.3\times10^{-4}$ mm; scale $=5\times10^{-9}$ sec/channel; counting time $=20.8$ h. Total number of signal (true) coincidences $\simeq3600$. Analysis of above signal spectrum yields $\tau_c = (1.90\pm0.15)\times10^{-7}$ sec.

Table I. Experimental parameters, results, and predictions of the theory.

Pressure (mm Hg)	Density (atoms/cm^3)	Optical thickness k_0L	Counting time (h)	Average true coincidence rate (counts/h)	$10^7\tau_c$ Expt. (sec)	τ_c/τ_0 Expt.	τ_c/τ_0 Theor. (Perel)	τ_c/τ_0 Theor. (Barrat)
2.35×10^{-5}	7.55×10^{11}	0.033	24	172	(1.14 ± 0.14)[a]	1.00 ± 0.17	1.02	1.02
6.5×10^{-5}	2.08×10^{12}	0.092	41	79	1.26 ± 0.11	1.11 ± 0.17	1.07	1.07
1.25×10^{-4}	4.01×10^{12}	0.176	21	164	1.24 ± 0.11	1.09 ± 0.17	1.13	1.14
5.3×10^{-4}	1.70×10^{13}	0.749	20.8	172	1.90 ± 0.15	1.67 ± 0.24	1.66	1.72
1.8×10^{-3}	5.77×10^{13}	2.54	42.5	123	5.03 ± 0.67	4.41 ± 0.08	4.34	6.30

[a]This value of τ_c is taken to be the experimental value of τ_0.

tion length at the center of the line. In terms more appropriate to the present discussion, $1/k_0$ can be thought of as the minimum mean free path for absorption of a 2537-Å photon. Both of the above quantities, x and k_0, are discussed at length in Ref. 8.

Finally we compare the experimentally obtained coherence times with those predicted by the two theories. The comparison, along with other pertinent information, is presented in Table I. From Table I it can be seen that for $k_0L \ll 1$, agreement between both theories and between the theories and experiment is quite good. As there is relatively little imprisonment for these small optical thicknesses, however, such agreement does not really constitute a conclusive test of the theories. It is seen that the coherence times predicted by the two theories differ ever more markedly as k_0L approaches and exceeds unity. At the largest value of k_0L listed above (2.54) agreement between experiment and D'yakonov and Perel's theory is excellent, while that between experiment and the theory of Barrat is rather poor.

Since Barrat's treatment assumes that all atoms in the vapor have the same velocity, or equivalently that $\Delta\nu_D = 0$, such disagreement is not surprising. Such a treatment does not satisfactorily describe the actual physical situation, and it leads to a calculation of the imprisonment (or absorption) based on consideration of the minimum photon mean free path only. Therefore, it is to be expected that such a treatment would predict a more pronounced imprisonment for a given density, or a greater coherence time, than was actually observed.

The treatment of D'yakonov and Perel, which takes into account the shape of the emission and absorption lines, more closely approximates the actual physical situation. Agreement between the coherence times predicted by this theory and those obtained experimentally is seen to be excellent over the range of densities studied.

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[5]By efficiency here is meant the over-all efficiency, i.e., the probability that an emitted photon of the proper wavelength will give rise to a pulse which triggers the discriminator.

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[12]Our notation here differs from that employed by D'yakonov and Perel. Their τ_0, τ are our τ_c, τ_0, respectively.

[13]The factor of $\frac{1}{5}$ in front of the familiar expression for k_0 (see Ref. 8, p. 100) arises as a result of taking account of the existence of five separate absorption lines, due to the hyperfine structure of mercury and the presence of an isotopic mixture.