Measurement of the lifetime of the $^3P_1$ metastable state of Mg by dye-laser excitation*

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We have measured the lifetime of the $^3P_1$ metastable state of Mg and obtained a value of 2.2 ± 0.2 msec. The level was excited with a dye laser tuned to the intercombination line ($3s\,3p\ ^3P_1 \leftrightarrow 3s^2\ ^1S_0$). In addition to the $^3P_1$ state, the $^3P_2$ and $^3P_0$ levels were populated by mixing collisions owing to the presence of an inert buffer gas. The populations of all three metastable levels were monitored as a function of time by observing the absorption of Mg resonance radiation ($^3P_{0,1,2} \rightarrow {}^3S_1$) out of each level. The mixing time was short compared to the $^3P_1$ lifetime, so that the populations of the three metastable levels were in thermal equilibrium with each other. This produced an apparent lengthening of the $^3P_1$ lifetime, which would introduce a large systematic error if the degree of mixing were not known. Conventionally, one observes the fluorescence from the excited levels to obtain their lifetimes. Because the lifetimes of the $^3P_2$ and $^3P_0$ states are too long, fluorescence from these states cannot be observed, and the observation of fluorescence from the $^3P_1$ state alone provides insufficient information to determine its lifetime.

The transition probabilities for the emission lines from the metastable $3s3p\,^3P_{0,1,2}$ states in Mg are of considerable astrophysical interest. Only the intercombination line ($3s3p\,^3P_1 \rightarrow 3s\,^1S_0$) has been observed in the laboratory, and there is a long-standing discrepancy between theoretical and experimental determinations of the lifetime of the $^3P_1$ state. Several workers[1-5] have calculated the lifetime; most recently Garstang[4] and Warner[5] obtained values of 4.2 and 0.5 msec, respectively. The principal experimental check on these calculations is a measurement by Boldt[6] of the oscillator strength for the intercombination line which yields a value for the $^3P_1$ lifetime of 1.9 ± 0.4 msec. Both Garstang and Warner independently conclude that the experimental value may be in error, and the compilers of the NBS tables of atomic transition probabilities[7] average Boldt's absorption-tube measurement of the oscillator strength with Garstang's theoretical number.

Stimulated by a report[8] that the $^3P_1$ state in Ca could be excited by an intense pulse from a tunable dye laser, we undertook an experiment to attempt to resolve the discrepancy between theoretical and experimental values for the $^3P_1$ lifetime of Mg.

The relevant energy-level structure of Mg is shown in Fig. 1. Large numbers of Mg atoms were excited to the $^3P_1$ level by passing an intense 0.5-μsec pulse of 4571-Å radiation from a dye laser through a tantalum-lined quartz absorption cell containing Mg vapor and an inert buffer gas. The number of atoms in the $^3P_1$ state was monitored as a function of time by observing the absorption by the metastables of 5173-Å Mg resonance radiation.

A block diagram of the apparatus is shown in Fig. 2. Mirror M-1 is specially coated to transmit 90% of the 4571-Å laser line while reflecting 95% of the 5173-Å line, enabling one to pass both beams collinearly through the cell. Mirror M-2 was coated to transmit 90% of the 5173-Å line to the ½-m spectrophotometer while reflecting 95% of the 4571-Å laser line for a second pass through the cell. The flashlamp-pumped dye laser was tuned with a diffraction grating and the 0.1-J pulse had a bandwidth of 0.5 Å. The quartz absorption cell was heated in an oven, and measurements were made over the temperature range 400–600°C. The Mg vapor pressure varied with temperature between 10^{-2} and 1.0 Torr. A buffer gas was used to prevent rapid diffusion to the walls and to keep the cell windows clean. The windows were outside the oven. The cell was lined with tantalum foil to prevent reaction of the hot Mg vapor with the quartz, and it was permanently attached to a vacuum gas-handling system to facilitate rapid changes in buffer gas and buffer-gas pressure and to monitor the background contamination.

Before measurements were made, the cell was thoroughly outgassed in a vacuum of 10^{-6} Torr at a temperature of 850°C. After cooling, the Mg was distilled into the sample. Measurements of background contamination as a function of time with the vacuum-pump valve off showed that it did not exceed 10^{-5} Torr during runs in which lifetimes were measured.

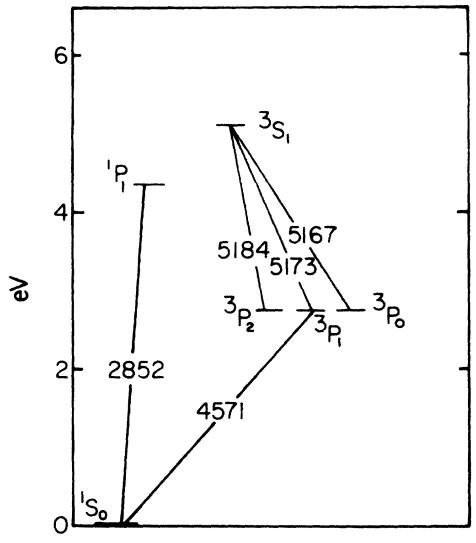

FIG. 1. Relevant energy-level structure of Mg.

The measurements were made in the following manner. The cell was filled with a high pressure of buffer gas and heated to the operating temperature. The laser was tuned until 5173-Å absorption signals were observed on the oscilloscope. The decay of the absorption signal was photographed, and the time constant of the decay was measured from this photograph. A typical photograph is shown in Fig. 3. The buffer gas was pumped out by stages to obtain lifetime measurements as a function of buffer-gas pressure.

We observed no pressure dependence of the lifetime for either He or Ne buffer gases. Most measurements were made with Ne in the pressure range 50–400 Torr.

Because of the small energy separation between the $^3P_2$, $^3P_1$, and $^3P_0$ metastable levels, these states were mixed by collisions with the buffer gas in a time short compared to the 0.5-μsec laser pulse, and absorption could be observed out of all three levels by tuning the spectrometer to the 5184-, 5173-, and 5167-Å lines, respectively. The lifetimes of $^3P_2$ and $^3P_0$ states are about six orders of magnitude longer than the lifetime of the $^3P_1$ state,[4] but the decay of all three absorption signals was characterized by the same time constant of 6.4 ± 0.6 msec. The error represents the rms scatter in the decay constants obtained from the 20 photographs of our final run. Because the mixing time was short compared to the lifetime of the $^3P_1$ level, the populations of the three levels were in thermal equilibrium with each other and were so tightly coupled that the ratio of the populations remained constant as they all decayed via the $^3P_1$ level. The populations of the $^3P_2$, $^3P_1$, and $^3P_0$ levels were calculated to be in the ratio of 4.5:2.83:1 at 600°C, which corresponds to the observed ratio of the absorption signal amplitudes. A solution of the rate equations[9] for the populations of the three levels including the effects of mixing and the decay of the $^3P_1$ state shows that the populations of all three levels decay at the same rate with a time constant of 2.94τ, where τ is the lifetime of the $^3P_1$ level. This lengthening of the decay time can be understood by realizing that a metastable atom spends only 34% of its time in the $^3P_1$ level. Therefore, we conclude that the lifetime of the $^3P_1$ state in Mg is 2.2 ± 0.2 msec.

From the above discussion it is clear that observation of the fluorescence (4571 Å) from the $^3P_1$ level would not in itself be sufficient to measure the $^3P_1$ lifetime. One must have knowledge of the degree of mixing to avoid a large systematic error, and this requires some sort of absorption monitoring of the $^3P_2$- and $^3P_0$-level populations.

Care was taken to avoid errors due to nonlinearities in the absorption signal resulting from excessively high metastable populations or self-reversal in the resonance lamp. Measurements were made over a wide range (more than an order of magnitude) of Mg density and there was no evi-

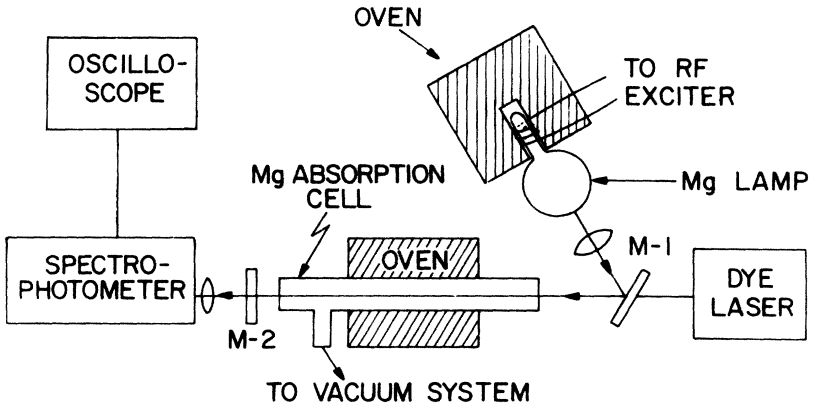

FIG. 2. Block diagram of the apparatus.

FIG. 3. Typical absorption signal. The time scale is 5.0 msec/cm and the time constant of the decay is 2.9τ, where τ is the lifetime of the $^3P_1$ state.

dence of radiation trapping. The most worrisome source of potential error was quenching due to collisions with trace amounts of H_2 and N_2 gas. We measured the quenching in cells filled with these two gases and the results are shown in Fig. 4. From this data we conclude that background contamination levels below 10^{-3} Torr would have no effect on the observed decay times. Our sample preparation precluded this level of contamination. The specifications on the reagent-grade buffer gases which we used indicated a level of contamination well below the critical amount. In any event, contamination in the buffer gas itself would have shown up when we varied the buffer-gas pressure.

Our value of 2.2 ± 0.2 msec for the lifetime of the $^3P_1$ state of Mg is in good agreement with the experimental value of 1.9 ± 0.4 msec obtained by Boldt, but does not agree with the theoretical calculations. The history of the experimental and theoretical investigations of this lifetime is interesting. The first experimental and theoretical

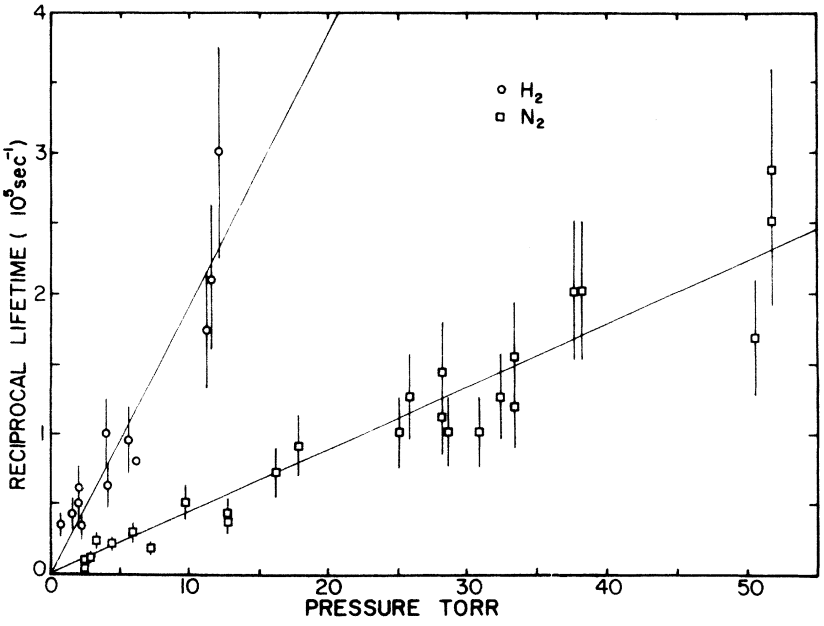

FIG. 4. Reciprocal of the time constant for the decay of the metastable levels due to quenching by H_2 and N_2 is plotted as a function of pressure. Because of the rapid mixing of the metastable states, the population of all the levels decayed with the same time constant, and the quenching cross sections for the individual levels could not be determined.

investigation of the $^3P_1$ lifetime was made by Frayne[1] in 1929. He calculated the lifetime to be 4 msec, which was in rough agreement with his spectral line intensity measurements. Buried in his paper, however, is the statement that a lifetime of 2 msec would be a better fit to his data. Subsequent theoretical calculations have consistently given lifetimes on the order of 4–5 msec, with the exception of those of Warner, while the experiments of Frayne in 1929 and Boldt in 1958, and our present measurement indicate the lifetime to be about 2 msec. These three experiments involve entirely different techniques and each experiment has a different source of systematic error, yet they are in agreement with each other. This indicates to us that the theoretical calculations are in error.

Note added in proof. We wish to thank J. L. Carlsten and G. A. Victor for bringing to our attention the results of recent unpublished work done at the Center for Astrophysics, Harvard College Observatory. G. A. Victor and C. Laughlin have calculated the lifetime of the $^3P_1$ level to be 3.6 msec and C. J. Mitchell, using the hook method, determined the lifetime to be 3.7 msec.

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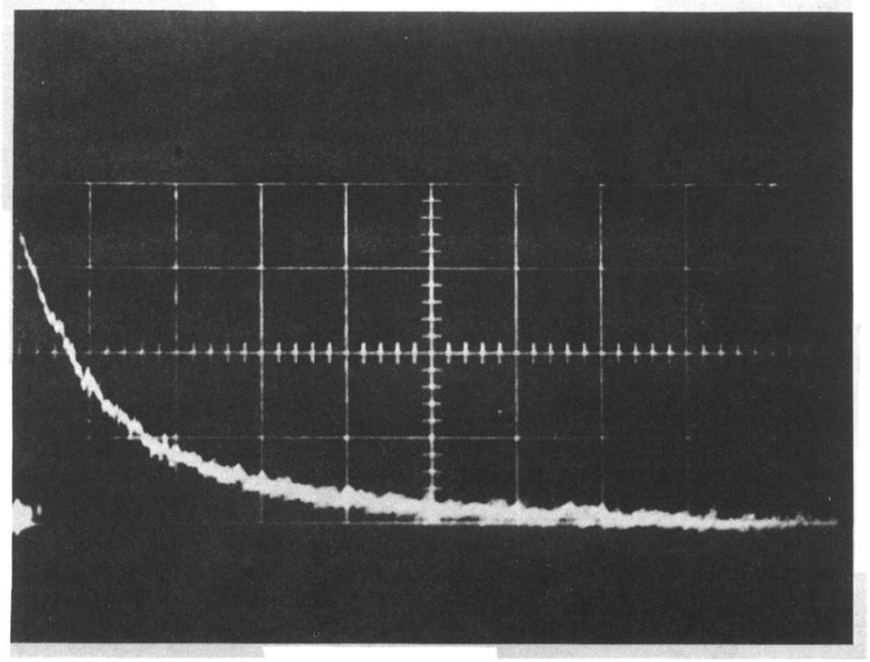

FIG. 3. Typical absorption signal. The time scale is 5.0 msec/cm and the time constant of the decay is 2.9τ, where τ is the lifetime of the $^3P_1$ state.