

Effect of Imprisonment of Radiation in Sodium Vapor on the Measured Lifetime of the 3^2P States*

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The effect of imprisonment of radiation in sodium vapor on the measured lifetime of the 3^2P states has been investigated in an experiment in which the atoms in the vapor were excited with 10-nsec pulses of resonance radiation and the elapsed times between the exciting and the fluorescent pulses were recorded using a time-to-amplitude converter and kicksorter. At vapor densities below 10^{10} atoms/cm³, where there is no multiple scattering, the measured lifetime reaches a constant value of $(1.63 \pm 0.04) \times 10^{-8}$ sec, equal to the natural lifetime of the 3^2P states. In the density range 10^9 – 10^{13} atoms/cm³, the variation of the effective lifetime with the vapor density is in good agreement with the predictions of Milne's theory of radiation trapping; agreement with Holstein's theory is observed only in the range 10^{12} – 10^{13} atoms/cm³. The results of a subsidiary Hanle experiment show that a multiply scattered photon of sodium resonance fluorescence loses its original polarization and that, at high sodium vapor pressure, coherence is not preserved.

I. INTRODUCTION

WHEN sodium vapor at low pressure is illuminated with a short pulse of sodium resonance radiation, the atoms become excited to the 3^2P states and decay emitting resonance fluorescence. It is usually accepted that the decay process is described by $N_t = N_0 \exp(-t/\tau)$, where N_t and N_0 are the numbers of excited atoms at times t and 0, respectively, and τ , defined as the mean lifetime of the 3^2P states, is a specific property of the sodium atoms. If the vapor density is increased, the resonance radiation becomes imprisoned in the vapor and measurements of decay rates yield effective lifetimes τ' which are significantly longer than τ and which are not only a property of the isolated atoms, but also of the whole vapor. The precise knowledge of τ as well as detailed information about the relationship between τ' and the alkali vapor pressure are of fundamental interest to workers in the fields of atomic physics and atomic collisions, radio-frequency spectroscopy, upper-atmosphere physics, and plasma physics.

Although there have been several successful attempts to measure τ using methods of magnetorotation at the edge of an absorption line,¹ Hanle effect,² and fluorometry,³ there has been no systematic experimental investigation of the direct influence of multiple scattering in alkali vapor on the lifetimes of the resonance states. Theoretical treatments of the effect^{4,5} have been used by various authors to correct experimental results for radiation trapping.⁶

In the present investigation, the lifetime of the 3^2P resonance states in sodium has been studied in the most

direct way. The atoms were excited with 10-nsec pulses of resonance radiation and the time intervals between excitation and decay were determined as a function of the vapor density. Similar techniques of time measurement have been used previously in lifetime determinations for various excited levels in inert gases,^{7,8} but the excitation was carried out with electron beams whose energy spread caused the simultaneous excitation of several atomic levels, thus complicating the separation of the decay processes. At very low vapor densities, where there is no imprisonment of radiation and $\tau' = \tau$, the method yields accurate values of τ and thus of oscillator strengths. The values of τ' may be compared with the predictions of both Milne's⁴ and Holstein's⁵ theories of radiation trapping. The theory of Barrat⁹ does not apply to the imprisonment of sodium resonance radiation, as is evident from the results of a complementary Hanle experiment.

II. EXPERIMENTAL

The arrangement of the apparatus is shown in Fig. 1. The sodium resonance radiation emitted by the lamp was passed through the Kerr cell¹⁰ which was placed between two crossed polaroids and which transmitted 10-nsec light pulses with a repetition rate of 60 pulses/sec. The transmitted light was focused in the fluorescence cell,¹¹ in which the pressure of the sodium vapor was controlled by the temperature of the liquid metal contained in a side-arm. The fluorescent light pulses emerging from the side-window of the cell were detected with a photomultiplier.

The Osram lamp was operated at a current of about 1 A and emitted Na_D lines which were only slightly self-reversed. The exciting radiation was made incident

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¹ G. Stephenson, Proc. Phys. Soc. (London) **A64**, 458 (1951).

² G. V. Markova and M. P. Chaika, Opt. i Spektroskopiya **17**, 319 (1964) [English transl.: Opt. Spectry. (USSR) **17**, 170 (1964)].

³ W. Demtröder, Z. Physik **166**, 42 (1962).

⁴ E. Milne, J. London Math. Soc. **1**, 1 (1926).

⁵ T. Holstein, Phys. Rev. **72**, 1212 (1947).

⁶ See, for instance, R. Seiwert, Ann. Physik **18**, 54 (1956).

⁷ S. Heron, R. W. P. McWhirter, and E. H. Rhoderick, Proc. Roy. Soc. (London) **A234**, 565 (1956).

⁸ W. R. Bennett, *Advances in Quantum Electronics* (Columbia University Press, New York, 1961).

⁹ J. P. Barrat, J. Phys. Radium **20**, 633 (1959).

¹⁰ The Kerr-cell modulator, Model ILM70, was manufactured by Electro-Optical Instruments of Pasadena, California.

¹¹ L. Krause, Appl. Opt. **5**, 1375 (1966).

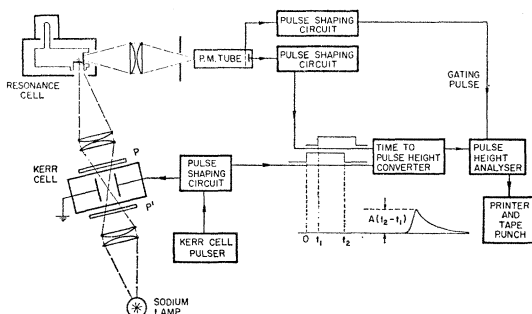


FIG. 1. The apparatus used for lifetime measurements. P and P' are the two crossed polaroids. The amplitude of the ramp pulse produced by the time-to-pulse-height converter is proportional to $(t_2 - t_1)$, the time overlap between the two rectangular pulses.

on the cell at an oblique angle to decrease the amount of stray light scattered from the windows. The cell was mounted in an electrically heated oven which was divided into two parts: one contained the main body of the cell and the other maintained the side-tube containing the liquid sodium at a slightly lower temperature, which determined the density of the fluorescing vapor. Both parts were maintained at temperatures constant within 0.3°C by means of temperature controllers¹² which employed thermistors as sensing elements. The fluorescence cell was connected by a tube of 3-mm bore to the vacuum system which maintained a vacuum of about 10^{-7} Torr throughout the experiments to remove all gases other than the sodium vapor. The fluorescent light was focused by lenses of large aperture on the photocathode of a Philips 56TVP photomultiplier which was housed in a liquid-air cryostat and was operated at 2700 V.

The time intervals between excitation and decay were measured as follows. A small fraction of the 35-kV pulse, which opened the Kerr shutter, was diverted to a pulse-shaping circuit¹³ to shape it into a rectangular pulse of 100-nsec duration and constant height, whose leading edge corresponded to the opening of the shutter. The output pulse from the photomultiplier was similarly shaped and the two pulses were directed into a time-to-pulse-height converter¹³ which converted the time overlap between them into a ramp pulse of height proportional to the time overlap. The ramp pulse was then counted, according to its amplitude, in a channel of a 400-channel kicksorter.¹⁴ The resulting spectrum, built up over many repetitions of the incident light pulse, gave the distribution of the times elapsed between the excitation and decay of atoms in the vapor. The kicksorter was gated by pulses taken from the

penultimate dynode of the photomultiplier in order to eliminate pulses too small to be shaped uniformly. The accumulated count was read out by means of a Teletype printer and tape punch.

The kicksorter was calibrated by placing various delay cables in one input of the time-to-pulse-height converter. These shifted the recorded spectrum by a number of channels corresponding to the known time delay of the particular cable. The resulting time calibration was accurate to better than 2%.

III. RESULTS AND DISCUSSION

A typical pulse-height spectrum, containing a total of about 10^5 counts accumulated during a period of a few hours, is shown in Fig. 2. The peak is due to the prompt spectrum, which has a half-width of 10 nsec. The slight undulations in the exponential part of the curve are due partly to statistical error and partly to the imperfect shape of the sample pulse taken from the Kerr cell. The spectrum also contains some background arising from light leaking through the Kerr cell and from the small dark current in the photomultiplier, which amounted to 21 counts/channel uniformly distributed in time. This background was subtracted before subjecting the results to further analysis, which was carried out with an IBM 1620 computer and yielded the apparent lifetimes τ' . The exponential character of the decay is demonstrated in Fig. 3 by the appearance of the semilogarithmic plot of the experimental spectrum.

It was thus possible to measure a range of apparent lifetimes over a range of sodium vapor pressures, as is shown in Fig. 4. At low vapor densities, τ' became constant and equal to τ . As the vapor pressure increased, τ' rose steeply. The individual values τ' are subject to experimental error of 1% caused by uncertainties in the background count and to error arising from the possibility of "pile-up," in which more than 1 photon per exciting light pulse could be detected by the photomultiplier, but only the first photon would be registered by the time-measuring system. Such pile-up, which

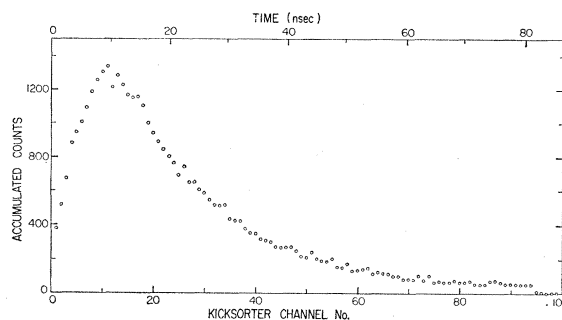


FIG. 2. A pulse-height spectrum showing the exponential decay of 3^2P sodium atoms, registered in 100 channels of a 400-channel kicksorter. The spectrum was taken at a vapor density of 7×10^9 atoms/cm³. The Kerr cell was open during the first 18 nsec.

¹² G. D. Chapman, Ph.D. thesis, University of Windsor, 1966 (unpublished).

¹³ G. Copley, M.Sc. thesis, University of Windsor, 1966 (unpublished).

¹⁴ Only 100 channels of the Victoreen P.I.P. 400 kicksorter were used in any given experiment.

would favor decays occurring just after the exciting pulse, was minimized by keeping the intensity of the exciting light so low that an average of only 1 photon per 10 exciting pulses was observed. A correction was made for this effect, which was negligible for low values of τ' , but which amounted to as much as 20% for values of $\tau' \geq 10\tau$. The effect on τ' of small changes in the depth at which the fluorescence was observed was found to be smaller than the experimental error.

The oscillator strength is twice as large for the $3^2S_{1/2} \leftarrow 3^2P_{3/2}$ transition as for the $3^2S_{1/2} \leftarrow 3^2P_{1/2}$ transition and thus the probability of trapping the 5890 Å radiation is twice as great as it is for the 5896 Å component. Consequently, although the $3^2P_{1/2}$ and $3^2P_{3/2}$ states exhibit lifetimes τ which are indistinguishable from one another at low vapor densities, at higher densities the respective values $\tau'(\frac{1}{2})$ and $\tau'(\frac{3}{2})$ differ and the observed experimental pulse-height spectra each consist of two exponentials corresponding to the two separate decay rates. It was not possible to separate these exponentials, because at low vapor densities the difference in lifetimes is very small and at higher densities only a short portion of the decay was observed. Thus the measured lifetimes τ' lie between $\tau'(\frac{1}{2})$ and $\tau'(\frac{3}{2})$.

The variation of τ' with vapor density may be compared with the predictions of Milne's⁴ and Holstein's⁵ theories. Theoretical curves representing the predicted behavior of τ' are plotted in Fig. 4. Both curves have been fitted to the experimental data at one (encircled) point and at the low-pressure limit of τ' . It was also assumed that plane parallel geometry was a suitable approximation to the conditions of the experiment and that the $3^2S_{1/2} \leftarrow 3^2P_{3/2}$ and $3^2S_{1/2} \leftarrow 3^2P_{1/2}$ transitions made equal contributions to the observed radiation, so that the observed value of τ' was an arithmetic mean of $\tau'(\frac{1}{2})$ and $\tau'(\frac{3}{2})$. It may be seen that Milne's theory is in excellent agreement with the experimental results, whereas Holstein's theory agrees only at the higher vapor densities.

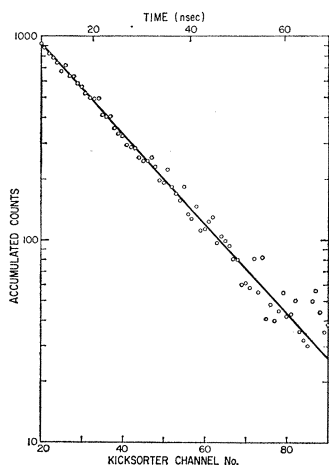


FIG. 3. A semilogarithmic plot of a part of the spectrum shown in Fig. 2. The slope yields $\tau' = \tau = 1.66 \times 10^{-8}$ sec.

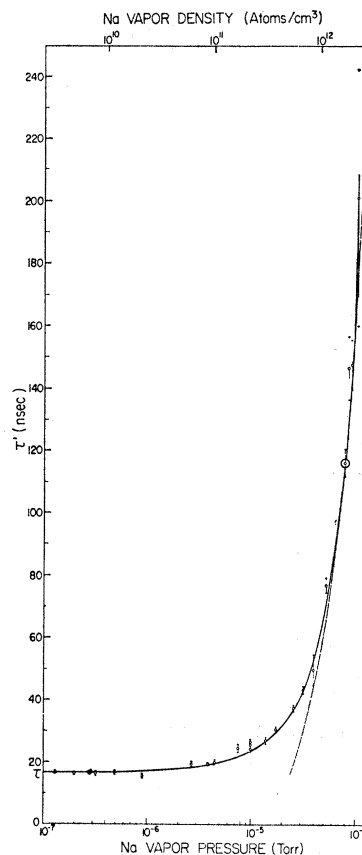


FIG. 4. The variation of the measured lifetime τ' with sodium vapor pressure. Solid line represents Milne's theory; dashed line represents Holstein's theory. The points are experimental. The encircled point and $\tau = 16.3$ nsec were used to fit the theoretical curves to the experimental data.

Milne gives the following representation of the radiation-trapping effect⁴:

$$\tau'(\frac{1}{2})/\tau = 1 + (C_1/\lambda)^2, \quad \tau'(\frac{3}{2})/\tau = 1 + (C_2/\mu)^2,$$

where $\lambda \tan \lambda = C_1$ and $\mu \tan \mu = C_2$. The opacity $C_{1,2} = l\alpha_{1,2}N$. N is the vapor density (obtained from the temperature of the side-arm by a substitution in the Langmuir-Taylor formula¹⁵), l is the thickness of the vapor layer, and α_1, α_2 are the atomic absorption coefficients for the D_1 and D_2 resonance lines ($\alpha_1 = 5 \times 10^{-12}$ cm², $\alpha_2 = 1 \times 10^{-11}$ cm²).¹⁶ A good fit with the experimental results is obtained for $l = 2$ mm, which is consistent with the experimental conditions.

Holstein gives the following expressions for the variation of the lifetimes τ' with the vapor density⁵:

$$\tau'(\frac{1}{2})/\tau = 0.94k_0(\frac{1}{2})l\{\ln[k_0(\frac{1}{2})\frac{1}{2}l]\}^{1/2},$$

$$\tau'(\frac{3}{2})/\tau = 0.94k_0(\frac{3}{2})l\{\ln[k_0(\frac{3}{2})\frac{1}{2}l]\}^{1/2},$$

where $k_0(\frac{3}{2}) = 2\lambda_0^3N/8\pi^{3/2}v_0\tau$, $k_0(\frac{1}{2}) = \frac{1}{2}k_0(\frac{3}{2})$, λ_0 is the mean wavelength of the Na_D lines, N is the atomic density, v_0 is the most probable speed of the atoms, and

¹⁵ J. B. Taylor and I. Langmuir, Phys. Rev. **51**, 753 (1937).

¹⁶ A. C. G. Mitchell and N. W. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, New York, 1934).

TABLE I. Values of the lifetime of the 3^2P states in sodium, from various sources.

τ (10^{-8} sec)	Method of determination
1.63 ± 0.04	Present investigation
1.63 ± 0.05	Hanle effect ^a
1.59 ± 0.015	Phase-shift fluorometry ^b
1.61 ± 0.06	Magnetorotation at the edge of an absorption line ^c
1.63	Calculation by the Bates-Damgaard method ^c

^a Reference 2.

^b Reference 3.

^c Reference 1.

l is the thickness of the plane parallel slab of fluorescing vapor. It is apparent from Fig. 4 that Holstein's expression applies only at vapor pressures greater than 5×10^{-5} Torr. Heron, McWhirter, and Rhoderick⁷ found good agreement with Holstein's theory in their experiments with helium which were performed at considerably higher pressures. In order to fit Holstein's equation to our high-pressure results, it is necessary to assume $l = 8$ mm, which is also in quite good agreement with experimental conditions.

In the low-pressure limit, at pressures below 10^{-6} Torr corresponding to densities of less than 10^{10} atoms/cm³, $\tau' \rightarrow \tau$. The weighted average of the 8 determinations carried out in this pressure region yielded a value of $\tau = (1.63 \pm 0.04) \times 10^{-8}$ sec for the lifetime of the 3^2P states in sodium. {*Note added in proof.* This value, based

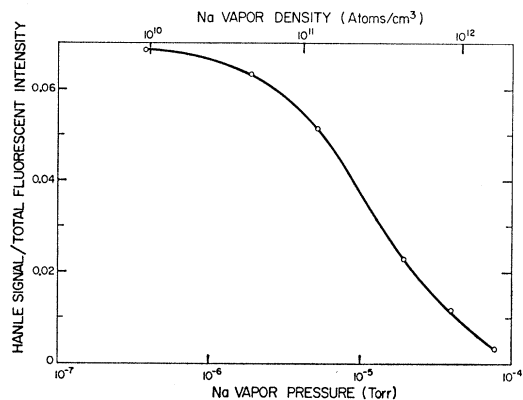


FIG. 5. The relation between the magnitude of the Hanle signal and sodium vapor pressure.

on further experiments, is more accurate than 1.66×10^{-8} sec, which appeared in the original abstract. 1.61×10^{-8} sec was recently obtained by Link [J. Opt. Soc. Am., **56**, 1195 (1966)]. This value is compared in Table I with the results of other authors.

In order to gain additional insight into the process of radiation trapping, the Hanle effect¹⁷ of the 3^2P levels in sodium was investigated over the whole experimental range of vapor pressures. The fluorescing vapor was placed in a magnetic field of a few gauss, uniform to $\pm 1\%$, directed along the axis of the observed light. The Kerr cell was removed and a polarizer was placed in the exciting beam, which polarized the incident radiation in a direction perpendicular to the axis of observation. The steady fluorescent light was analyzed with a polarizer oriented to pass light polarized perpendicularly to the electric field vector in the incident light and was observed, as a function of the magnetic field, by the photomultiplier whose dc output was amplified with an electrometer. The magnitude of the Hanle effect decreased sharply with increasing vapor pressure, as may be seen in Fig. 5, which shows the relative magnitude of the Hanle signal, represented as a fraction of the total fluorescent intensity. There is good correlation between Figs. 4 and 5. τ' becomes very large at the vapor pressure of 10^{-4} Torr, at which the amplitude of the Hanle signal becomes very small. This implies that, at these vapor densities, a photon emitted from a particular atom is unlikely to leave the cell without being multiply scattered and losing its original polarization.

On the other hand, it was found that the width of the Hanle signal, which is a measure of the lifetimes of the atoms producing the effect, remained constant over the whole range of vapor densities to within the experimental accuracy of 5%. The implied conclusion, that coherence is not preserved during multiple scattering in sodium vapor, was to be expected because the hfs of both the $3^2P_{3/2}$ and $3^2P_{1/2}$ levels lie totally within the Doppler widths of the two respective D lines, thus rendering Barrat's theory⁹ inapplicable to the present case.

ACKNOWLEDGMENT

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¹⁷ P. A. Franken, Phys. Rev. **121**, 508 (1961).