Since intense proton beams can be produced, and since it is unlikely that the nuclear polarization would be disturbed by the collision of H(2s) with argon, it is probable that a polarized negative-ion source can be built with intensities in the microampere range.

We are grateful for the encouragement offered during the course of this work by J. L. Mckibben of Los Alamos Scientific Laboratory.

*National Science Foundation Undergraduate Research Participant.

[1]W. Gruebler, W. Haeberli, and P. Schwandt, Phys. Rev. Letters 12, 595 (1964).

[2]Although this discussion explicitly mentions only hydrogen, the same processes work for deuterium when it has the same velocity as the hydrogen, and therefore polarized negative deuterium ions can be produced by the same scheme.

[3]B. Donnally, T. Clapp, W. Sawyer, and M. Schultz, Phys. Rev. Letters 12, 502 (1964). L. Madansky and G. Owen, Phys. Rev. Letters 2, 209 (1959), first proposed charge exchange as a means of obtaining H(2s), but the explicit reaction they discussed, charge exchange in H_2, is unsuitable for use in this proposed source because of its low yield of H(2s) and its relatively high yield of H(1s) in the low-energy range.

[4]W. Lamb and R. Retherford, Phys. Rev. 79, 549 (1950); L. Madansky and G. Owen, Phys. Rev. Letters 2, 209 (1959).

[5]A. Abragam and J. Winter, Phys. Rev. Letters 1, 374 (1958); A. Abragam and J. Winter, Compt. Rend. 255, 1099 (1962).

[6]J. B. Hasted, Physics of Atomic Collisions (Butterworths Scientific Publications, Ltd., London, 1964), p. 440.

OBSERVATION OF DIFFERENT LIFETIMES FOR ATOMIC STATES EXCITED WITH LINEARLY AND CIRCULARLY POLARIZED LIGHT*

W. Happer and E. B. Saloman

Columbia Radiation Laboratory, Columbia University, New York, New York

(Received 19 July 1965)

At very low atomic densities all Zeeman sublevels of an excited atomic state decay with the natural radiative lifetime of that state. Under these conditions the widths of the Hanle effect and optical double-resonance signals may be used to measure the lifetime of the state.[1] However, at higher atomic densities the Zeeman sublevels can relax to each other through collision and radiation trapping so that, in general, $(2J+1)^2$ parameters are necessary to specify the decay of the excited atoms. These processes result in a broadening or narrowing of the level crossing or optical double-resonance linewidths and can be ascribed to a change in the lifetime of the excited atomic state.[2,3] In most experiments the requirements of symmetry reduce the number of different relaxation times from $(2J+1)^2$ to $(2J+1)$. Physically, these are the lifetime of the excited-state population, the lifetime (τ_{or}) of the magnetic moment of the excited state (the "orientation"), the lifetime (τ_{al}) of the "alignment" of the excited state (an atomic polarizationwith zero magnetic moment), and the lifetimes of other quantities which describe the angular configuration of the excited atoms. In contrast to classical nmr experiments where only the magnetic moment of the system is observed, an appropriate choice of polarizations in level crossing and optical double-resonance experiments allows one to observe the lifetimes of "orientation" and "alignment" separately.

For computational purposes it is most convenient to treat this problem using the density matrix formalism. Then each separate multipole component $\rho^{[L]}$ of the density matrix decays with a characteristic lifetime.[4-6] The "orientation" of the atoms is proportional to $\rho^{[1]}$, while the "alignment" is proportional to $\rho^{[2]}$ so that level crossing lifetime measurements give τ_{al}. With circularly (elliptically) polarized exciting light, both $\rho^{[1]}$ and $\rho^{[2]}$ are excited, and with an appropriate choice of polarization for the detected light, τ_{or} can be measured.[7]

Under conditions where strong radiation trapping is operative, Omont[8] has recently measured τ_{or} and τ_{al} for the first $^3P_1$ state of mercury. His results are in good agreement with theoretical calculations of Dyakonov and Perel.[6] In this Letter we report on different values for τ_{or} and τ_{al} in the first $^3P_1{}^0$ state of lead where the difference in τ_{or} and τ_{al} is caused by collision effects rather than radiation trapping.

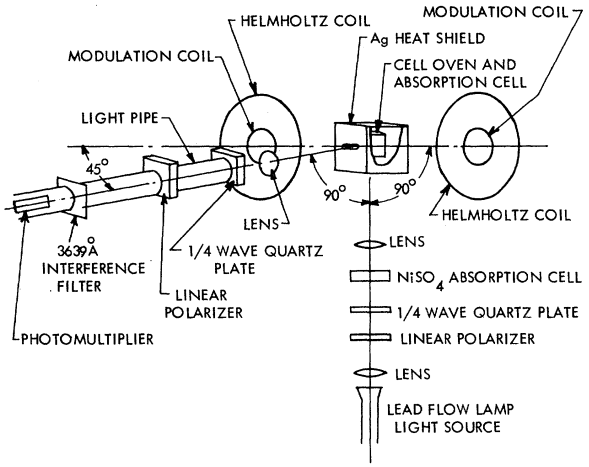

FIG. 1. Schematic of Hanle-effect apparatus.

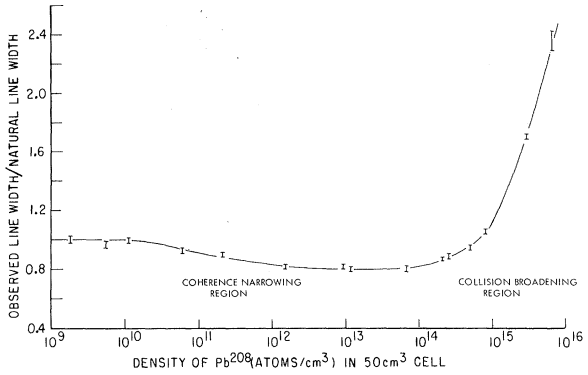

FIG. 2. Linewidth of $\Delta m = 2$ Hanle-effect resonances in the $(6s^2 6p\,7s)\,^3P_1{}^0$ state of Pb^{208} $(1/\tau_{al})$.

In spite of the great qualitative contrast between the short-range, two-body interactions responsible for collision broadening[5] and the long-range coupling of many atoms through the radiation field which causes coherence narrowing,[2] both processes give rise to different lifetimes τ_{al} and τ_{or}.

Lead atoms in a quartz resonance vessel are excited from the $(6s^2 6p^2)\,^3P_0$ ground state to the $(6s^2 6p7s)\,^3P_1{}^0$ excited state by 2833Å resonance light. The level crossing signals were detected in the 3639Å cross fluorescence. A sketch of our apparatus is shown in Fig. 1. A variant of the zero-field level crossing technique[1] (Hanle effect) was used. The widths of the $\Delta m = 1$ zero-field level crossing signals were used since these can originate from both $L = 2$ and $L = 1$ components of the excited-state density matrix. For incident light polarized at 45° to the magnetic field, the $\Delta m = 1$ signal originates from a pure $L = 2$ component of p, while for circularly polarized exciting light the $\Delta m = 1$ signal originates from a pure $L = 1$ component of ρ. Interference from the $\Delta m = 2$ signal can be eliminated by using an analyzer for elliptically polarized light in the detection arm. It can be shown that only $\Delta m = 1$ signals will be detected if the projection of the ellipse on the plane perpendicular to the static field is a circle. Stressed quartz phase-shift plates[9] were used as quarter wave plates in these experiments. After a preliminary alignment of the elliptical analyzer to the correct angles, the phase plate was stressed until the $\Delta m = 2$ signal was eliminated. Usually a slight empirical adjustment of the angle between the quarter wave plate and the linear polarizer was necessary to obtain complete elimination.

The general behavior of the linewidth[10] is indicated in Fig. 2, where the ratio of $\gamma_{al} = 1/\tau_{al}$ to the natural width $\Gamma = 1/\tau$ $[\tau = 5.75(20) \times 10^{-9}$ sec$]$[11] has been plotted as a function of atomic vapor density. The results of measurements of τ_{or} and τ_{al} are summarized in Table I. Because of the fairly large f value for the 2833Å

Table I. Results.

Lead atomic density (cm^{-3})	γ_{or}/Γ	γ_{al}/Γ	$(\gamma_{or}-\Gamma)/(\gamma_{al}-\Gamma)$ Experimental	Theoretical	Reference
2.9×10^{15}	1.99(3)	1.76(3)	1.30(5)	1.30 ↔ 1.67	a
1.3×10^{13}	1.36(2)	1.22(2)	1.64(18)	•••	b
1.3×10^{13}	0.852(18)	0.818(18)	0.8(2)	0.714	c

[a]These results are for the case of resonant self-broadening.
[b]These results are for the case of foreign gas broadening.
[c]These results are for the case of a saturated coherence narrowing effect.

resonance line in lead, the linewidth at high vapor densities should be determined chiefly by resonant collision broadening.[3,5] The results of our measurements are consistent with rough theoretical estimates of Omont[5] for the ratio of the two relaxation times. A marked difference in γ_{al} and γ_{or} was also observed with a gassy resonance cell. The exact constituents and pressure of the foreign gas are unknown, but a spectrographic analysis of the light from a microwave discharge through the cell showed N_2, OH, CO, and NH emission bands as well as strong lines from atomic hydrogen.

Coherence narrowing in lead is much less pronounced than in mercury because of the presence of branch decay modes from the excited state. One can show that the theoretical maximum coherence narrowing is reduced by the branching ratio ($\approx 27\%$) to the ground state. Since the coherence narrowing is so small, our experimentally measured values of γ_{al} and γ_{or} are nearly the same, but within experimental error our ratio of $(\gamma_{or}-\Gamma)/(\gamma_{al}-\Gamma)$ is in agreement with the theoretical ratio of $\frac{5}{7}$.

These experiments demonstrate that under a wide range of experimental conditions the different multipole components of the density matrix describing excited atoms in a vapor relax with measurably different time constants. More detailed studies of this kind should yield considerable information about the mechanisms involved in depolarizing collisions.

The authors would like to thank Mr. Jon Katz for his contributions to these experiments. They are also indebted to Professor R. Novick for his interest in this work.

---

*This work was supported in part by the Joint Services Electronics Program under Contract No. DA-28-043 AMC-00099(E), and in part by the National Aeronautics and Space Administration under Grant No. NsG-360.

[1]A. Lurio, R. L. deZafra, and R. J. Goshen, Phys. Rev. 134, A1198 (1964).

[2]J. P. Barrat, J. Phys. Radium 20, 541, 633 (1959).

[3]F. W. Byron, M. N. McDermott, and R. Novick, Phys. Rev. 134, A615 (1964).

[4]U. Fano, Rev. Mod. Phys. 29, 74 (1957).

[5]A. Omont, J. Phys. Radium 26, 26 (1965).

[6]M. T. Dyakonov and U. T. Perel, Zh. Eksperim. i Teor. Fiz. 47, 1483 (1964) [translation: Soviet Phys.-JETP 20, 997 (1965)].

[7]The initial density matrix for atoms excited with light of polarization vector $\hat{e}$ from a ground state of angular momentum I to an excited state of angular momentum J is

$$\rho = \sum_{LM} (-1)^M E_{-M}^{\ L} T_M^{\ L} \{(-1)^{I+J-1} \sqrt{3}\, W(11JJ;LI)\},$$

where the $T_M^{\ L}$ are Fano's irreducible tensor operators for the level J,

$$T_M^{\ L} = \sum_m |J,m\rangle \langle J,m-M|(-1)^{J+m-M} C(JJL;m,M-m)$$

and

$$E_M^{\ L} = -\sum e_\mu e_{M-\mu}^{\ *} C(11L;\mu,M-\mu)$$

describes the polarization of the incident light. $E_0^{\ 0}$ is a scalar and is the magnitude of the polarization vector. $E_M^{\ 1}$ is simply the spin density of the exciting light and is zero if the light is linearly polarized. $E_M^{\ 2}$ also has a classical interpretation; it is the Maxwell stress tensor for the electric field of the exciting light. Thus while $L=0$ and $L=2$ components of ρ are excited by any light beam, circularly (elliptically) polarized exciting light is required to excite $L=1$ components of ρ. One can also show that the detected optical signal in Hanle-effect experiments and in many optical double-resonance experiments is most lucidly expressed in terms of irreducible tensors. One finds that to detect a signal from an $L=1$ component of ρ, circularly (elliptically) polarized light is required.

[8]A. Omont, Compt. Rend. 260, 3331 (1965).

[9]These phase-shift plates were kindly lent to us by A. Lurio, R. Garwin, and A. Patlach of IBM Watson Laboratory, Columbia University, New York, New York.

[10]E. B. Saloman and W. Happer, Bull. Am. Phys. Soc. 10, 596 (1965).

[11]R. Novick, B. W. Perry, and E. B. Saloman, Bull. Am. Phys. Soc. 9, 625 (1964); E. B. Saloman, Bull. Am. Phys. Soc. 10, 49 (1965).