Lifetime of the First Excited Atomic States of Rb^{87}†

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The Hanle effect on the resonance lines of the Rb^{87} atom has been observed. The line shape of the signal for the $5^2S_{1/2} \leftrightarrow 5^2P_{3/2}$ is found to be Lorentzian, while that for the $5^2S_{1/2} \leftrightarrow 5^2P_{1/2}$ is dispersion-type. The half-width of these lines was observed to be 3.146±0.13 G and 23.0 G±5%, respectively. These values yield the lifetime of $(2.71\pm0.14)\times10^{-8}$sec and $(3.0\pm0.3)\times10^{-8}$ sec for the $5^2P_{3/2}$ and $5^2P_{1/2}$ states, respectively. The oscillator strengths for the corresponding transitions are found to be 0.673±0.03 and 0.32±0.03, respectively. When the temperature of the scattering cell was changed from 0 to 36°C, an apparent increase of the linewidth at higher temperature was observed. This effect has been explained as a magnetic-field-dependent radiative transfer effect at optical depth of the order of the scattering cell dimensions. When the data were analyzed with such an effect included, the reduced Hanle signal showed no temperature dependence of the half-width.

I. INTRODUCTION

BECAUSE of the recent developments in photon detection techniques, the zero-field level crossing experiment, or the Hanle effect,[1] has again become a popular, accurate method for determining the lifetime of resonance states of atoms.[2] This method is better than the Hook method [2,3] for the determination of the oscillator strengths since one does not need to know the density of atomic scatterers provided there is no multiple scattering.

The theory of the Hanle effect has been given by Breit[4] and Weisskopf,[5] and has been developed more recently by Franken[6] and by Rose and Carvillano.[7] Recent experimental data can be found mostly in papers by Lurio, Novick, and their collaborators,[8] but no Hanle effect has been measured on the first excited state of the rubidium atom. Experiments similar to the present one are the level crossing experiments on lithium[9] and on sodium and cesium.[10] The measurements of the oscillator strengths in the Rubidium atom have been done by the magnetic rotation method[11] and by the Hook method.[12] The comparison of the present results with the results of these and other experiments, as well as theoretical calculations, will be given in Sec. IV of this paper.

II. THEORY

The energy levels of Rb^{87}, which are of interest here, are the ground-state $5^2S_{1/2}$ and the first excited states $5^2P_{1/2}$ and $5^2P_{3/2}$. The hyperfine structure of these states is shown in Fig. 1.

The rate $R(\hat{f},\hat{g})$ at which incident photons of polarization $\hat{f}$ are scattered by an atomic vapor and become

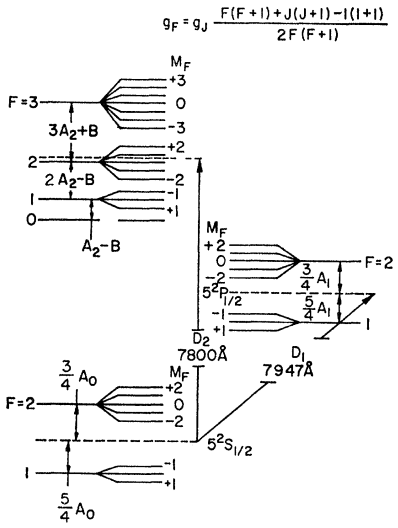

FIG. 1. Level scheme (not to scale)-Rb^{87}. ($I=\frac{3}{2}$). $A_0=3417.34$ MHz [see L. Essen, E. G. Hope, and D. Sutcliffe, Nature **189**, 298 (1961); S. Penselin, T. Moran, V. W. Cohen and G. Winkler, Phys. Rev. **127**, 524 (1962)], $A_1=409$ MHz [see B. Senitzky and I. I. Rabi, Phys. Rev. **103**, 315 (1956)], $A_2=84.85$ $B=12.61$ MHz [see H. A. Schussler, Z. Physik **182**, 289 (1965)].

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scattered photons of polarization $\hat{g}$ is[6]

$$R(\hat{f},\hat{g})=K\sum_{m,m';\mu,\mu'}\frac{f_{\mu m}f_{m\mu'}g_{\mu' m'}g_{m'\mu}}{\Gamma-(i/\hbar)(E_\mu-E_{\mu'})}, \quad (1)$$

where μ and μ' denote the magnetic sublevels of the atomic resonance state, while m and m' denote those of the atomic ground state, $\hat{f}$ and $\hat{g}$ are the unit vectors in the polarization directions of the incident and scattered photons, respectively, $\Gamma=1/\tau$ is the width of the excited state, and $E_\mu-E_{\mu'}$ is the energy difference between magnetic sublevels of the excited state. The electric dipole matrix elements are given, for example, by

$$f_{\mu m}=\langle\mu|\hat{f}\cdot\mathbf{r}|m\rangle.$$

The coefficient K contains the intensity of the incident radiation and the density of atoms in the scattering vapor.

The existence of a nuclear spin $I>0$ complicates the level crossing signal given by Eq. (1), because the energy difference

$$E_\mu-E_{\mu'}\equiv E_{M_F}-E_{M'_F}$$

between the magnetic sublevels of the excited states depends, in general, on F. Thus, the crossing signal is sometimes difficult to analyze. If the magnetic field is sufficiently small so that the atoms are in the linear Zeeman region, we may write

$$|E_{M_F}-E_{M'_F}|=g_F\mu_0B|M_F-M'_F|, \quad (2)$$

where

$$g_F=g_J\frac{F(F+1)+J(J+1)-I(I+1)}{2F(F+1)}. \quad (3)$$

In those excited states of an isotope in which $J=I$, g_F reduces to

$$g_F=g_J/2, \quad (4)$$

which is independent of F. Such is the case for the $^2P_{3/2}$ states of Rb^{87}, and, in fact, for $^2P_{3/2}$ states in many common isotopes of the alkalies. This F independence of the g_F factor yields, for the experimental configuration of linearly polarized light used here, a simple Lorentzian form for the Hanle effect signal of

$$R(^2P_{3/2})\propto\left\{1-\frac{27}{200}\left[1+\left(\frac{g_J\mu_0B}{\hbar\Gamma}\right)^2\right]^{-1}\right\}, \quad (5)$$

where $g_J=\frac{4}{3}$ for the $^2P_{3/2}$ state. For $^2P_{1/2}$ state, Eq. (4) does not hold, and the resulting form is $g_F=\pm g_J/4$, where the + is for the $F=2$ and the − for $F=1$. For the experimental configuration of circularly polarized light,[13] the signal is no longer a simple Lorentzian, but is a dispersion curve of the form

$$R(^2P_{1/2})\propto\left\{1-\frac{10}{128}\left(\frac{\Gamma\mu_0B}{\hbar}\right)\left[\Gamma^2+\left(\frac{g_J\mu_0B}{4\hbar}\right)^2\right]^{-1}\right\}, \quad (6)$$

where $g_J=\frac{2}{3}$ for the $^2P_{1/2}$ state. A cancellation of terms in Eq. (1) results in a null signal from the $F=1$ states of $^2P_{1/2}$, thus, the final form (6) is due to the $F=2$ states only. It must be noted that Eqs. (5) and (6) are rigorously valid only at small magnetic fields where the linear Zeeman approximation is valid, that is, $g_J\mu_0B$ is much less than the hyperfine splittings of the corresponding excited state.

III. EXPERIMENT

A Pyrex scattering cell containing enriched Rb^{87} ($\sim$2%Rb^{85}, 98% Rb^{87}) was partially enclosed in a water-filled Lucite box which also contained a heater, stirrer, thermocouple and thermometer. This scattering apparatus was placed between a pair of Helmholtz coils of 30 cm radius which supplied a magnetic field of 0–50 G, normal to the plane of Fig. 2. Surrounding this were three sets of mutually perpendicular Helmholtz coils of about 1 m diam which were used to "buck-out" the earth's field. Light from a radiofrequency excited lamp containing natural rubidium passed through a single lens and appropriate polarizers and entered the scattering cell at right angles to the magnetic field direction. The light which was scattered at right angles to the incident beam and to the magnetic field was gathered by a single lens, analyzed by an appropriate polarizing element and filter(s), and taken through an aluminized light pipe to a μ-metal shielded photomultiplier, a RCA 7102.

The signal from the photomultiplier at zero magnetic field was balanced through a cathode-follower difference amplifier against a constant dc voltage provided by a bank of mercury cells. Thus, changes in the light signal

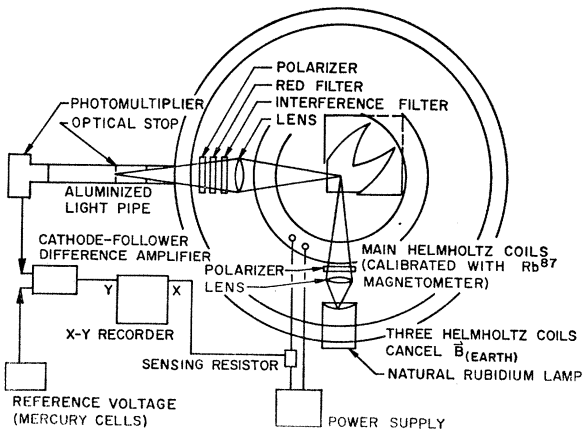

FIG. 2. Experimental setup. Polarizers are linear for $P_{3/2}$, and circular for $P_{1/2}$ resonances.

[13] Atomic states with $J<1$ exhibit no Hanle effect using linearly polarized light.

were easily plotted continuously versus magnetic field on an X-Y recorder.

The current-regulated supply for the main field was calibrated against a Rb^{87} magnetometer which uses the optical pumping technique.[14,15] The bucking fields were adjusted to give a minimum field of about 2 mG, and then the voltage across a small sensing resistor in series with the main field coils was calibrated against the magnetic field measured by the rubidium magnetometer. The voltage across the sensing resistor was used to drive one arm of the X-Y recorder.

Several changes were made in the experimental apparatus after the original measurements were taken by one of us (JDF)[16]. The most important changes were made on the following pieces of apparatus: (a) the cell holder was originally an air-filled, styrofoam covered, aluminum box; (b) the main magnetic field could go as high as 120 G, but had a small region of uniform field; (c) the data-display system was originally a chart recorder that could only record the intensity versus magnetic field at discrete points.

IV. RESULTS AND DISCUSSION

Using the earlier experimental configuration, data from several runs at 0°C for the $^2P_{1/2}$ level were fit using least-squares techniques with Eq. (6). The results of typical runs for the $P_{1/2}$ state are shown in Fig. 3. The best value for the half-width found in this manner is

$$B_{1/2}(^2P_{1/2}) = 23.0\text{G} \pm 5\%.$$

The uncertainty, here, of 5% is largely due to the influence of noise on the data taking method. This value yields, after calibration uncertainties and systematic errors are considered,

$$\tau(^2P_{1/2}) = (3.0 \pm 0.3) \times 10^{-8} \text{ sec},$$

and

$$f(5^2P_{1/2} - 5^2S_{1/2}) = 0.32 \pm 0.03.$$

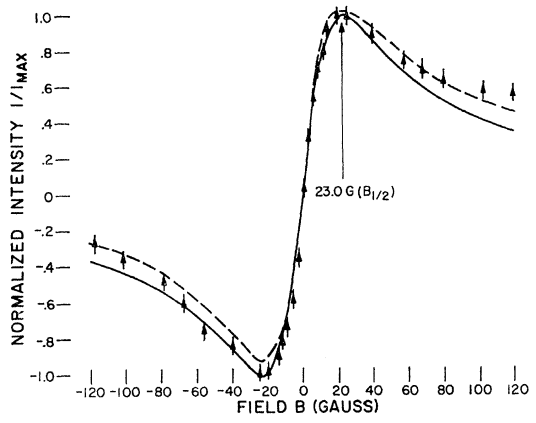

FIG. 3. Hanle effect signals for $5^2P_{1/2}$ state. The solid line is the best theoretical curve based on Eq. (6). The dashed-line is a theoretical curve which includes an estimated 10% admixture of the Lorentzian signal [Eq. (5)], due to the fact that the interference filter and circular polarizer for the D_1 line (7947 Å) could not completely eliminate the stronger D_2 line (7800 Å).

Data were also taken for the $^2P_{3/2}$ state at cell temperatures up to 40°C. When the data were fit by least-squares techniques to Eq. (5), a large anomalous, apparent temperature dependence of the half-width, and thus the lifetime, was observed. When the experiment was repeated using the improved experimental configuration outlined in Sec. III, the apparent temperature dependence was explained[17] as a magnetic scanning effect at intermediate optical depth, which is similar to that observed previously by Lurio[18] and reported by Mitchell and Zemansky.[19] By a careful analysis of the experimental data which "normalizes" the Hanle curves to remove the effect of the variation of the incident light intensity in Eq. (1), we saw easily the expected decrease (due to multiple scattering processes) in the Hanle signal as a fraction of the total scattered light. The amount of decrease, however, was smaller than expected, indicating a complicated effect involving different optical depths for different hyperfine components. The Hanle signal decreased from about 6 to 3% of the total signal over the range of temperatures. We also clearly saw that the deviation from the Lorentzian shape of the Hanle signal depended on temperature and thus on optical depth. While it is true that at higher fields (10 G and higher) the theoretical scattering signal is an extremely complicated sum of many Lorentzian shapes, this nonlinear Zeeman effect (which is smaller or of the order of the Hanle signal itself) is completely dominated by changes in the background signal. A 10% change with magnetic field in the background signal would be two to three times larger than a reasonable sized change caused by the nonlinear Zeeman effect. Exact theoretical calculations done by Khadjavi[20] indicate that the non-linear Zeeman effect contributes a 6% change in the Hanle signal or about a 1% change in the total signal at a field of 50 G. The inclusion of a first-order correction term due to the optical-depth effect gives a suitable form for the scattering rate for the $^2P_{3/2}$ state as[17]

$$R(^2P_{3/2}) = F'[\rho(T)] + F[\rho(T)] \times \left\{1 - \frac{27}{200}\left[1 + \left(\frac{g_J \mu_0 B}{\hbar\Gamma}\right)^2\right]^{-1}\right\} + G[\rho(T)]B^2, \quad (7)$$

where F, F', and G are independent of the magnetic

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TABLE I. Comparison of lifetime and oscillator strength.

Investigators	Methods	Lifetime (10^{-8} sec.) $5^2P_{1/2}$	Lifetime (10^{-8} sec.) $5^2P_{3/2}$	Oscillator strengths $5^2P_{1/2}$	Oscillator strengths $5^2P_{3/2}$
This paper	Hanle effect	3.0 ±0.3	2.71±0.14	0.32 ±0.03	0.673±0.03
a	Magnetic rotation	2.89±0.09	2.78±0.09	0.335±3%	0.661±3%
b	Phase shift	2.81±0.05	2.70±0.05	0.335±2%	0.675±2%
c	Hanle effect	3.0 ±7%	2.7 ±7%	0.32 ±7%	0.675±7%
d	Hook method	2.33	2.27	0.399±0.006	0.804±0.011
d	Theoretical (SCF)	2.62	2.48	0.362	0.734
e	Theoretical (Bates-Damgaard)	2.81	2.67		
f	Theoretical				sum 1.09

[a] See Ref. 11.
[b] J. K. Link, J. Opt. Soc. Am. **56**, 1195 (1966).
[c] A. Gallagher, Phys. Rev. **157**, 68 (1967).
[d] See Ref. 12.
[e] O. S. Heavens, J. Opt. Soc. Am., **51**, 1058 (1961).
[f] E. M. Anderson and V. A. Zitilis, Opt. i Spektroskopiya **16**, 382 (1963), [English transl.: Opt. Spectry. (USSR) **16**, 211 (1963)].

field B, but depend critically on the line profile of the exciting radiation and on the optical depth ρ (and thus the temperature T) in the scattering cell. When the data from 27 run over the temperature range 1.4 to 36.0°C were fit by least-squares techniques with Eq. (7), no temperature dependence in the true half-width was observed. It is thus concluded that no observable coherence narrowing took place even though the pressure in the scattering cell changed by a factor of about 100 from about 10^{-8} to 10^{-6} Torr. For a pressure of 10^{-6} torr, the optical depth for an ideal structureless resonance line in rubidium is about 5. At this optical depth, one expects to be able to observe coherence narrowing. The coherence narrowing at large optical depths is a function of the spin quantum number J of the resonance state.[21] For a $J=\frac{3}{2}$ state with a nuclear spin of $\frac{3}{2}$, the narrowing is small but should be observable. Due to the complex hyperfine structure of the ground and excited states, the wings of the line dominate (with single scattering processes) the center of the complex resonance line since the latter is almost completely absorbed (by multiple scattering processes). Thus at higher temperatures, different components have different optical depths. Single scattering processes at low optical depth dominate and mask any coherence narrowing effect. Two typical data runs are shown in Fig. 4. The value for the half-width of the Lorentizian is found to be

$$B_{1/2}(^2P_{3/2})=3.146\pm0.13 \text{ G}.$$

This value of the half-width gives for the lifetime and oscillator strength

$$\tau(^2P_{3/2})=(2.71\pm0.14)\times10^{-8} \text{ sec};$$
$$f(5^2P_{3/2}-5^2S_{1/2})=0.673\pm0.03,$$

where the estimated error is one standard deviation. A comparison with other measurements of lifetime and oscillator strengths is given in Table I.

V. MAGNETIC SCANNING EFFECT

In most experiments on the Hanle effect, multiple scattering effects and violation of the linear-Zeeman assumption are the most important nuisance errors encountered. In this experiment, however, the dominant deviation from the expected Hanle signal arose because the intensity of absorbed light was magnetic field dependent. Atoms in the front of the scattering cell (near the entrance window) absorbed the incident light as a function of the magnetic field since the atomic levels of the atoms were Zeeman split. This Zeeman absorption pattern affected the light incident upon the atoms taking part in the scattering to the detector. The spectrum of the R-F excited natural rubidium lamp was unknown, but it certainly must have had a complicated structure. The deviation from a "white light" spectrum enhanced the magnetic field dependent

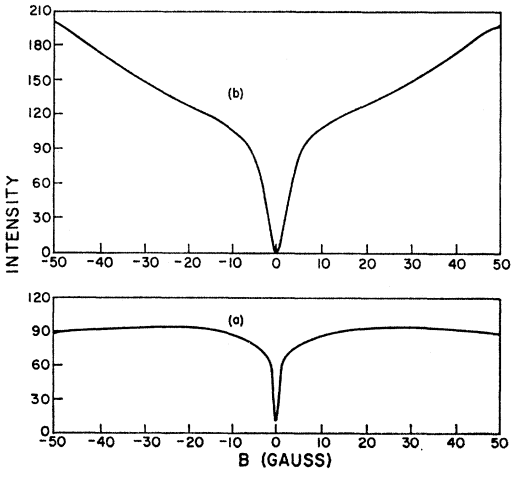

FIG. 4. Typical Hanle effect signals for $5^2P_{3/2}$ state. (a) Cell temperature 19°C. (b) Cell temperature 34°C.

[21] M. I. D'Yakonov and V. I. Perel', Zh. Eksperim. i Teor. Fiz. **47**, 1483 [English transl.: Soviet Phys.—JETP **20**, 997 (1965)]; J. P. Barrat, J. Phys. Radium **20**, 541, (1959); **20**, 633 (1959); **20**, 657 (1959).

absorption effect. Thus, it appears as though the parts of the spectrum that absorb, scan the light profile as a function of the magnetic field. When Lurio[18] encountered this effect, he applied a magnetic field to his lamp to flatten and broaden the incident light profile, and thus decrease this kind of effect.

It is easily seen that correction terms to the Hanle scattering rate must depend on an even power of the magnetic field, and that the dominant term is most likely proportional to B^2. The curves of Figs. 4(a) and 4(b) indicate that a B^2 correction term whose coefficient is temperature (optical depth) dependent gives a good first-order correction. The optical-depth dependence of the coefficient of the B^2 term as well as the dependence of the coefficients for the Hanle term and the background are to be explained in a separate paper.[17] The form of Eq. (7) fits the experimental data well.

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Relativistic Effects in the Excitation of Triplet Helium States by Electrons*

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The excitation of triplet He states is a pure rearrangement collision provided that spin-dependent potentials are neglected. The high-energy behavior of this rearrangement cross section is proportional to E^{-3}. If relativistic spin–other-orbit terms are admitted, they produce contributions to the cross section which go to a constant at moderately high energies. These new terms are incoherent with the old ones and have a relative coefficient of α_F^4, where α_F is the fine-structure constant. They become significant at an incident energy of about 8 keV and so are not presently observable. Part of the result obtained here also applies to the case where the incident particle is a proton.

I. INTRODUCTION

THE theory of rearrangement collisions is not completely understood even at high energies. To further this understanding various first Born approximations to the cross section for the excitation of triplet states of helium by electrons have been calculated and compared with experiment with somewhat inconclusive results.[1] These reactions are pure rearrangement collisions in the approximation where spin-dependent interactions are neglected. In that case, the singlet-triplet reaction can only come about through the process of the incident electron colliding with and ejecting a bound electron and then becoming bound itself. In the references cited,[1] eight different forms of the transition matrix element were tested. They gave different results but for high enough energy it can be shown that they all converge to the same results. This cross section at high energies behaves as E^{-3}.

When spin-dependent potentials are admitted, the reaction can proceed via spin flip and not by rearrangement. Spin-dependent potentials are small, but because the reaction is a direct one and not a rearrangement reaction, we may expect that this part of the cross section will not fall off as rapidly at high energies and may well be important. Indeed, we shall see that the spin-flip cross section goes to a constant at moderately high energies. This comes about from the interaction of the spin of a bound electron with the orbital motion of the projectile electron. This force is proportional to the incident velocity so that the impulse which causes the transition is energy-independent at high energies and consequently the cross section is also. There are other spin-dependent potentials (besides the spin-other orbit one) but these give contributions to the cross section which fall off with energy more rapidly than the one considered here, and so they will be neglected.

II. CALCULATION

As an example, we deal with the excitation of He (2^3S). Our starting point is the expression for the Born approximation for the T matrix for this reaction

$$T_{fi}=\langle\lambda_f(1)V_f(1)A_0\lambda_i(0)\rangle, \quad (1)$$

where the notation is that of Ref. 1. The potential may be decomposed into a spin-independent part $V_f^{(0)}(1)$ and a spin-dependent part $V_f^S(1)$. The contribution from $V_f^{(0)}(1)$ is just that obtained in Ref.

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[1] Charles J. Joachain and Marvin H. Mittleman, Phys. Rev. 140A, 432 (1965); 151, 7 (1966).