Orientation and Alignment of Sodium Atoms by Means of Polarized Resonance Radiation*

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Sodium atoms have been oriented by means of circularly polarized resonance radiation. The amount of orientation agrees with that calculated from the intensity of the light source used. The polarization of the light scattered from the sodium sample varies in the expected manner when the light intensity and applied magnetic field are varied. Illumination with unpolarized resonance radiation is shown to result in alignment of the sodium atoms. The largest degree of orientation achieved corresponds to an average value for the nuclear, electronic, and total angular momenta of $\bar{M}_I=0.180=0.120I$, $\bar{M}_S=0.035=0.070S$, $\bar{M}_F=0.216=0.108F$, respectively.

INTRODUCTION

KASTLER[1] has proposed that atoms could be oriented by means of circularly polarized resonance radiation. I shall use the word orientation to denote a state in which an ensemble of atoms has an average atomic or nuclear magnetic moment not equal to zero; I prefer this to the more commonly used word polarization in order to avoid confusion with light polarization. I shall retain the common usage of the word alignment to denote a condition in which, while the ensemble average magnetic moment is zero, atomic states with $|M_F|=F$ are more probable than those with $M_F=0$. A substantial quantity of oriented atoms could be used as a source of oriented nuclei for such experiments as the study of angular distribution of radioactive decay products, as a source of polarized electrons, or as a means of enhancing the signal in nuclear magnetic resonance experiments.

Bitter and Brossel[2] have unsuccessfully attempted to induce orientation in mercury by this method. More recently, Brossel, Kastler, and Winter[3] have achieved a positive result with sodium, while Bitter, Lacey, and Richter[4] also report failure with mercury. The present experiment using sodium has already been briefly described.[5,6] Brossel, Cagnac, and Kastler[7] have induced radio-frequency transitions between the Zeeman sublevels of oriented sodium atoms. They observe transitions involving two rf photons corresponding to a change $\Delta M_F=2$ in the axial angular momentum as well as the usual single-photon $\Delta M_F=1$ transitions. An excellent review of the whole subject has been given by Kastler in his recent Holweck lecture.[8]

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[1] A. Kastler, J. phys. et radium **11**, 255 (1950).
[2] F. Bitter and J. Brossel, Phys. Rev. **85**, 1051 (1952).
[3] Brossel, Kastler, and Winter, J. phys. et radium **13**, 668 (1952).
[4] Bitter, Lacey, and Richter, Revs. Modern Phys. **25**, 174 (1953).
[5] W. B. Hawkins and R. H. Dicke, Phys. Rev. **91**, 1008 (1953).
[6] W. B. Hawkins, Phys. Rev. **96**, 532 (1954).
[7] Brossel, Cagnac, and Kastler, J. phys. et radium **15**, 6 (1954).
[8] A. Kastler, Proc. Phys. Soc. (London) **A67**, 853 (1954).

THEORY OF ORIENTATION

General

The simplest atomic system which can be oriented by means of circularly polarized resonance radiation is one having a ground state of total angular momentum $F=\frac{1}{2}$ and an optical first excited state having $F=\frac{3}{2}$. If such an atom is in the ground state sublevel with axial angular momentum $M_F=+\frac{1}{2}$ and absorbs a circularly polarized photon with axial angular momentum $+1$, it will go into the $M_F=+\frac{3}{2}$ substate of the excited level. By virtue of the electric dipole selection rules which are assumed for the transition, this excited atom must return to the substate $M_F=+\frac{1}{2}$ when it radiates. An atom originally in the $M_F=-\frac{1}{2}$ ground state absorbing such a photon can return to either ground state sublevel when it radiates. Thus the result of the scattering of this kind of light by an ensemble of such atoms is that all atoms originally in the $M_F=+\frac{1}{2}$ state will return there, while some of those with $M_F=-\frac{1}{2}$ will be transferred to the $M_F=+\frac{1}{2}$ state by the scattering process. The ensemble has been partially oriented.

More complicated atoms having larger angular momenta or hyperfine structure behave in essentially the same way, the ground-state sublevel of largest (or smallest if the sense of the light polarization be reversed) magnetic quantum number gaining atoms at the expense of the others. Figure 1 shows how the orientation process works for one of the hyperfine transitions in Na^{23} together with the complete energy

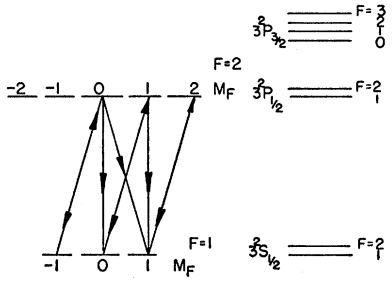

FIG. 1. Na^{23} energy level diagram with a sample of the transitions involved in orientation, in this case transitions via the $3\,^2P_{\frac{1}{2}}$, $F=2$ states.

level diagram. Although the magnetic quantum number can change by at most two units during a single scattering process, the scattering of further photons will increase the orientation if the partially oriented ensemble remains undisturbed. However, its members may be disturbed between photon-scattering events by collisions with other atoms or by the presence of magnetic fields.

A magnetic field perpendicular to the axis of quantization defined by the direction of the incident light will cause a mixing of all the M_F states for a particular F. Although the resulting state is not one of thermal equilibrium, it may be treated as such for present purposes. A field of about 10^{-3} gauss is sufficient for thorough mixing in a time of the order of 10^{-4} second, the time of observation in this experiment. However, the situation is quite different if in addition to this transverse component a larger axial field component is present, for a field nearly parallel to the axis of quantization mixes the states only slightly. The angle between the axis and whatever magnetic field is present is more important than the size of the field, provided that it is not large enough to seriously alter the atomic wave functions. The fields normally applied in this experiment did not appear to be large enough to cause significant alteration. Such changes in wave functions, while rendering the calculations for zero field invalid, do not interfere with the basis process of "optical pumping." Their avoidance is thus a matter of convenience in the interpretation rather than an essential of the experiment.

Collisions with other atoms of suitable kinds can result in disorientation either by electron exchange or by mutual spin-flipping. This source of disturbance can be eliminated by keeping the sodium mean free path large compared to the dimensions of the apparatus, which is equivalent in this experiment to keeping the pressure below 10^{-2} mm of Hg. The normal pressure was 10^{-6} to 10^{-4} mm of Hg.

Sodium Resonance Fluorescence

The electronic ground state of sodium is a $3\,^2S_{\frac{1}{2}}$ state which combines with the nuclear spin of the stable isotope, $I=\frac{3}{2}$, in two hyperfine states having $F=1$ and 2. The first excited state is a doublet having term values $3\,^2P_{\frac{1}{2}}$ and $3\,^2P_{\frac{3}{2}}$, the lower of which ($J=\frac{1}{2}$) has the same hyperfine structure as the ground state, while the upper has four hyperfine states with F ranging from 0 to 3 (see Fig. 1).

Since the smallest hyperfine splitting is only 2.6 times the natural level breadth, the treatment of resonance fluorescence given in Heitler[9] must be extended to include the effect of the overlapping of levels. The derivation is very similar to Heitler's, and only the result will be given. The probability of an atom being in a given final state (at $t=\infty$ when absorption is known to have taken place) is

$$|b_m(\infty)|^2=\frac{4\pi^2\rho_\omega^2}{\Gamma\hbar^4}\sum_{n,n'}\frac{H_{mn}H_{n0}H_{mn'}{}^*H_{n'0}{}^*}{\gamma-i\omega_{nn'}}, \quad (1)$$

where ρ_ω is the density of radiation oscillators at the resonance frequency, $1/\Gamma$ is the lifetime of the initial state, $1/\gamma$ that of the intermediate state, and $\omega_{nn'}$ the energy difference between the two indicated intermediate states. The index 0 refers to the initial state, n and n' to intermediate states, and m to the final state. This result has been averaged over the frequency distributions of the emitted and absorbed photons, assuming a continuous spectrum incident. This assumption is justified for a light source emitting lines much broader than the absorption Doppler breadth such as the one used in this experiment.

The matrix elements H_{mn} and H_{n0} for the emission and absorption processes may be obtained from the formulas in Condon and Shortley,[10] page 63, although a unitary transformation must be applied to obtain the values in the desired representation having the quantum numbers I, J, F, and M_F. The matrix of this unitary transformation may be constructed from the tables in Condon and Shortley, page 76. Applying this transformation, one obtains the matrix elements of electron momentum in the desired representation. The elements H_{mn} and H_{n0} of the Hamiltonian are derived from these by taking the inner product of the electron momentum and the vector potential of the radiation field in the dipole wave approximation, care being taken to appropriately restrict the polarization of the incident wave.

It may be seen from Eq. (1) that a pair of intermediate levels will contribute an interference term whose ratio to the direct transition terms is $2/(\omega_{nn'}{}^2+\gamma^2)$. The natural line breadth γ is[11] 6.1×10^7 radians/sec, while the total hyperfine splittings used in the calculations are[12] 1.5×10^9 radians/sec for the $3\,^2P_{\frac{3}{2}}$ state and 9.4×10^8 radians/sec for the $3\,^2P_{\frac{1}{2}}$ state. Applying the Landé interval rule, it is found that only the levels $F=0$ and $F=1$ of the latter state ($\omega_{01}=2.6\gamma$) are close enough to contribute interference terms as large as 10 percent of the direct transition terms. More recent measurements[13] give a smaller total splitting in the $3\,^2P_{\frac{3}{2}}$ state, as well as a small deviation from the interval rule, but the difference does not significantly affect the results of the present calculations.

The transition probabilities also depend on the intensity ratio in the light source of the two fine structure components of the sodium D-line. The source used yielded equal peak intensities of the two

[9] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1944), second edition, pp. 137–142.

[10] E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, London, 1951).

[11] L. Larrick, Phys. Rev. **46**, 581 (1934).

[12] L. P. Granath and C. M. Van Atta, Phys. Rev. **44**, 935 (1933).

[13] P. Sagalyn, Phys. Rev. **94**, 885 (1954).

TABLE I. Probabilities of transitions induced by resonance radiation between substates of the sodium $3\,^2P_{\frac{1}{2}}$ ground state complex for equal incident fluxes of the two D-line components. Column one gives the quantum numbers F and M_F of the initial states, labelling the rows; and the other columns are headed by the quantum numbers F and M_F of the final states. The probabilities are so normalized that each initial state has a total absorption probability of one.

Initial F, M_F	Final F, M_F 1,−1	1,0	1,1	2,−2	2,−1	2,0	2,1	2,2
1,−1	0.343	0.167	0.268	0	0.085	0.056	0.082	0
1, 0	0	0.312	0.299	0	0	0.140	0.085	0.164
1, 1	0	0	0.500	0	0	0	0.167	0.333
2,−2	0.169	0.164	0	0.321	0.193	0.154	0	0
2,−1	0.085	0.167	0.082	0	0.229	0.207	0.229	0
2, 0	0	0.140	0.138	0	0	0.292	0.276	0.154
2, 1	0	0	0.167	0	0	0	0.500	0.333
2, 2	0	0	0	0	0	0	0	1.000

lines; and therefore, because of the difference in statistical weights of the two levels, the total absorption probability leading to the $3\,^2P_{\frac{3}{2}}$ state is twice that for the $3\,^2P_{\frac{1}{2}}$ level. This having been taken into account, Table I gives the total relative probabilities of resonance fluorescence between given initial and final states via all possible intermediate states. The columns are labeled by the quantum numbers of the final states, and the rows by those of the initial states. Both initial and final states are all members of the $3\,^2S_{\frac{1}{2}}$ ground state complex. The total absorption probabilities of all the initial states are the same, which is only true for equal intensities in the source of the two components of the doublet.

Table I may be regarded as a matrix operator. The vector on which it operates has components proportional to the occupation probabilities of the various initial states for whatever ensemble of atoms is under consideration. Similarly, the vector resulting from the operation gives the occupation probabilities for the same ensemble after each of its members has scattered one and only one photon. The initial ensemble is normally taken as being in thermal equilibrium, and by n applications of the operator one may find the occupation probabilities of an ensemble each of whose members has scattered exactly n photons.

One may simplify the computation of the polarization of the scattered light by observing that each element in the matrix operator just described corresponds to a definite polarization component, σ or π, in the scattered radiation. Summing the elements of like polarization in each row of the matrix produces a pair of vectors whose components are labeled by the quantum numbers of initial states. The inner product of one of these vectors with the occupation vector of an ensemble is proportional to the amount of the corresponding polarization emitted when each member of that ensemble scatters one photon. For example, applying this process to an ensemble whose members have each scattered two photons, one can compute the polarization emitted when a third photon is scattered.

Table II gives the state occupation probability distributions of ensembles whose members have each scattered a definite number, n, of photons. It also gives the expectation values for these ensembles of the observables M_F, M_I, and M_S, the axial projections of the total, nuclear, and electronic angular momenta, as well as the polarization ratio $R_n=(\sigma-\pi)/(\sigma+\pi)$ of the light scattered at 90° to the axis of quantization. π radiation has its electric vector parallel to the axis, σ, perpendicular, both being plane polarized at this scattering angle.

Of course, no physically realizable ensemble is composed of members all of which have scattered the same number of photons. However, the expectation value of an observable for a physical ensemble may be computed by taking a weighted average of the expectation values for the several such idealized ensembles. The value of the observable for the ith ensemble is weighted by the probability P_i of an atom in the physical ensemble scattering i photons.

To compute P_i, consider a beam of sodium atoms passing through an illuminated region such that each atom scatters on the average N photons in the L cm of illuminated beam. The probability of absorbing a photon in any interval of path dl is $(N/L)dl$, and the probability of not absorbing any photon at all is

$$P_0=(1-Ndl/L)^{L/dl}=e^{-N}, \tag{2}$$

of absorbing one and only one photon is

$$P_1=\int_0^L \frac{N}{L}e^{-N}dl=Ne^{-N}, \tag{3}$$

and of absorbing exactly i photons is

$$P_i=N^ie^{-N}/i!. \tag{4}$$

EXPERIMENTAL PROCEDURE

The apparatus consisted of a source of sodium resonance radiation, a sodium sample in the form of a

TABLE II. Expectation values of various quantities for ensembles each of whose members has scattered n photons. R_n is the polarization ratio of photons scattered at 90° for the nth scattering process.

	Occupation numbers											
	$F=1$					$F=2$						
n	$M_F=1$	0	−1	2	1	0	−1	−2	M_F	M_I	M_S	R_n
1	0.182	0.119	0.075	0.247	0.168	0.106	0.063	0.040	0.626	0.523	0.103	0.070
2	0.194	0.082	0.038	0.399	0.174	0.071	0.029	0.012	1.075	0.884	0.191	0.173
3	0.173	0.049	0.018	0.546	0.156	0.042	0.012	0.004	1.384	1.116	0.268	0.313

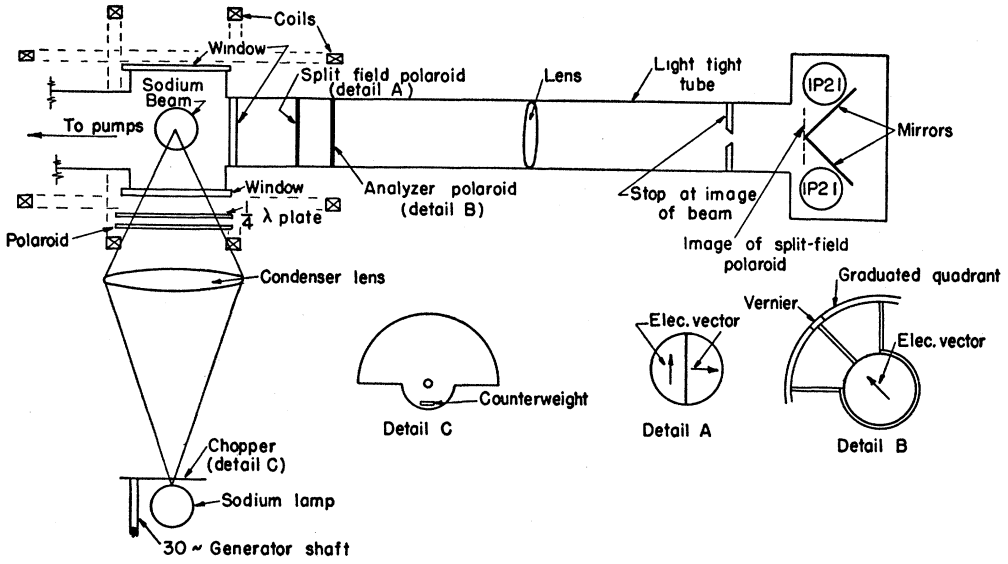

FIG. 2. Optical system used to orient sodium and to detect the orientation when produced.

beam, coils to control the magnetic field seen by the sample, and equipment to detect the polarization of the light scattered by the sample. A view of the optical system is shown in Fig. 2.

A Phillips sodium spectral lamp No. 93122E was used, together with a sheet of polaroid and a quarter wave plate, to produce circularly polarized sodium resonance radiation. A lens formed an image of the lamp at the beam position, and the light was interrupted at 30 cycles per second by a semicircular metal segment on the shaft of a small motor generator. The 30-cycle/sec output from the generator was used as the reference signal for the lock-in amplifier that formed part of the polarization detector.

A sodium beam was used as a simple means of insuring relative freedom from foreign gas contamination. The beam source was a stainless steel oven heated to 100°–200°C by 60-cycle/sec ac. The beam was collimated into a cone of 9° half-angle and caught by a liquid nitrogen trap after leaving the illuminated region. The beam chamber was evacuated by an oil diffusion pump provided with a solid CO_2 cooled trap to intercept oil vapor. With both traps cold, a vacuum of between 10^{-4} and 10^{-6} mm of Hg could be obtained, depending on the beam source conditions.

In the early stages of the experiment, the magnetic field applied was quite inhomogeneous, especially the axial component (that parallel to the direction of the incident light), since it was supplied by two solenoids separated axially by a distance equal to their diameter, the diameter being no larger than the region in which the sodium was observed. When it was found that this inhomogeneity increased the difficulty of interpreting the data, the first set of coils was replaced by three mutually perpendicular Helmholtz pairs, each producing a field of 4 gauss/ampere within the accuracy of mounting them to fulfill the Helmholtz condition. No refined technique of mounting them was used, neither the field calibration nor the homogeneity being sufficiently critical to the experiment to require it. The smallest of the three pairs had a radius of 11.4 cm and the largest 19.0, while the length of sodium beam observed was 3.8 cm.

The polarization ratio $(\sigma-\pi)/(\sigma+\pi)$ of the light scattered by the sodium beam was determined by an optical bridge whose parts are indicated in Fig. 2. All observations were made at a scattering angle of 90°. The light passed through a polaroid filter consisting of two semicircular segments each transmitting only σ or only π radiation. At 90° to the axis of quantization, each component is plane polarized with their planes of polarization perpendicular to each other. Each half of this split-field polaroid was imaged by a lens on one of two 1P21 photomultiplier tubes. Between the split-field polaroid and the lens was a rotatable polaroid (hereafter called the analyzer) provided with a graduated quadrant and vernier. Its function was to produce known changes in the intensity ratio of the two light polarization components and thus calibrate the detector. It was normally set at 45° in order to transmit equal amounts of the two components. All this equipment was enclosed in a light-tight tube. To reduce stray light to a minimum, the detector was placed opposite the vacuum pumping lead; and a stop was placed at the image of the beam (see Fig. 2) to intercept light scattered from the edges of the observation window.

Since the incident light was modulated at 30 cycles/sec, ac detection could be used. The output of one 1P21 was inverted in phase and added to that of the other, the combined signal being fed into a 30-cycle/sec lock-in amplifier. The output of this amplifier appeared on an Esterline-Angus recording milliammeter. The

system thus formed an optical bridge giving a null signal for equal outputs from the two photomultiplier tubes. Since the gains of the two 1P21's could not be depended on to be the same, absolute light polarization ratios could not be measured. All polarization measurements were therefore referred to a standard condition, that in which sodium orientation was destroyed by application of a magnetic field perpendicular to the axis. The field of the earth was found to serve this purpose excellently. With unoriented sodium, the bridge was approximately balanced by adjusting the applied voltage, and thus the gain, of one photomultiplier. The difference between the resulting signal and that for any other sodium sample condition was reduced to a light polarization difference by comparing it with the signal change produced by rotating the analyzer polaroid a known amount.

RESULTS AND DISCUSSION

The sodium lamp used was compared with a white lamp of known intensity by means of visual observation with a flicker photometer (average of 92 comparisons) and found to emit 2.72×10^{16} photons/(cm^2 sec sterradian) perpendicular to its surface. If an image of this source is formed by a lens, it can be shown that the photon flux through the image is simply $I\Omega$, where I is the surface intensity of the source as given above and Ω is the solid angle subtended by the lens at the image. For a given lens, the maximum solid angle is determined by the requirement that the image of the light source be at least as big as the region occupied by the sodium beam. In practice, the solid angle was limited not by the lens, but by the window admitting light to the vacuum system, which subtended an angle at the sodium beam of $\pi/16$ steradian; since the polaroid had a transmission of 0.365, the total photon flux at the beam was therefore 1.95×10^{15} photons/cm^2 sec.

To determine the intensity ratio and line widths of the two fine structure components, the light source was photographed through a Lummer-Gehrcke plate (crossed with a prism), for the loan of which I am indebted to Professor A. G. Shenstone. The density of the photographic images was measured with a recording densitometer owned by the R. C. A. laboratory in Penn's Neck, New Jersey. I am grateful to Dr. Hillier of that laboratory for permission to use the instrument and to Dr. H. Halma for making the measurements with it. The two components of the doublet were found to have approximately the same peak intensities; their widths were 10^{10} cycles/sec for the component radiated by atoms in the $3\,^2P_{\frac{1}{2}}$ level and 1.4×10^{10} cycles/sec for the $3\,^2P_{\frac{3}{2}}$ component. This equality of peak intensities, together with the extreme breadth (20 times Doppler breadth), suggests that the source is so self-reversed as to be effectively a blackbody at the center of the lines. This is quite reasonable since we shall see that approximately 10^{10} atoms cm^{-2} completely absorb all light at the resonance frequency. For a thickness of 1 cm, this corresponds to a pressure of 2.5×10^{-7} mm of Hg, and the sodium partial pressure in the lamp was certainly much higher than this. From these observations, the common peak flux per unit frequency at the sodium beam is

$$I_0=1.29\times10^4 \text{ photons/cm}^2 \text{ radian}$$

for each component.

In sodium the transition to the first excited state complex accounts for 0.975 of all the radiation absorbed by atoms in the ground state (see Condon and Shortley,[10] p. 149). For present purposes the oscillator strength of the line may therefore be taken as one, and the number of photons absorbed per atom is thus given by the formula (Heitler,[9] p. 108) for a classical oscillator,

$$n=2\pi^2 r_0 c I_0, \tag{5}$$

where r_0 is the classical radius of the electron. Thus for the given light flux,

$$n=2.14\times10^3 \text{ photons/sec atom.}$$

A sodium beam source temperature of 200°C corresponds to a velocity of 4.8×10^4 cm/sec. Since the illuminated length of beam was 3.8 cm, each atom might be expected to absorb on the average 0.17 photon.

Clearly the number of sodium atoms which can be oriented is no larger than that number which absorbs all the incident light. A larger sample will result in multiple scattering of photons and a lower orientation per atom. For a sodium sample (not necessarily a beam) at a temperature in the neighborhood of 200°C, the Doppler breadth of the absorbed line is approximately 2×10^9 radians/sec. The total number of photons which can be absorbed is roughly equal to the line breadth times the peak intensity, or 2.4×10^{13} photons/cm^2 sec. Since each atom absorbs 2.1×10^3 photons/sec, 10^{10} atoms/cm^2 will absorb all photons in the usable range. Other considerations, based on the tendency of a gas to radiate coherently,[14] thus reradiating photons of the same polarization as those absorbed, limit the sample to about one-fifth this number.

Although the fact is obscured by the method of derivation, the sample size is independent of the light intensity; for as the intensity increases, each atom absorbs more photons. In other words, the absorption cross section is independent of the light intensity.

This limitation of the sample size is naturally a serious limitation on any experiments it is desired to perform with atoms or nuclei oriented by this technique. As an example, if it were desired to study the angular distribution of radioactive emissions, an isotope with a half-life of one year would produce 130 counts/min in a perfectly efficient counter subtending a solid angle of $4\pi\times10^{-2}$ steradian if the radiation were isotropic. Again, the hyperfine structure resonance of

[14] R. H. Dicke, Phys. Rev. **93**, 99 (1954).

Na^{23} could be observed with a signal-to-noise ratio of about 500 assuming an orientation of 10 percent. The signal-to-noise ratio varies as the square of the amount of orientation.

It was found that the sodium source temperature (and beam density) could be allowed to vary over wide limits without changing the experimental results. This is interpreted to mean that the density was at all times well below the allowable maximum. No attempt was made to observe the actual source temperature beyond rough estimates.

Since the rate of light absorption is now known, the probability of an atom scattering any given number of photons i may be computed from Eq. (4). The average polarization of the light scattered is given by

$$\bar{R}=\sum_{i=1}^{\infty} R_i \sum_{j=i}^{\infty} P_j \Big/ \sum_{i=1}^{\infty} iP_i, \tag{6}$$

where the R_i are taken from Table II. For the given absorption probability, P_3 is only 5 percent of P_2, and all higher P_i's may be neglected. The average polarization is then

$$\bar{R}=0.0789.$$

When orientation is destroyed by application of a transverse magnetic field, the polarization of the scattered light is not quite equal to R_1, as one would expect if true thermal equilibrium were produced. It can be shown that a slight degree of *alignment* remains and that the average scattered polarization in this case is

$$\bar{R}=0.0710,$$

while the value of R_1 is 0.0700. The change in light polarization expected upon destruction of orientation by a transverse field is therefore

$$\Delta\bar{R}_{\text{th}}=0.0079.$$

Twenty-one observations of this change were made over a period of five months, and their average was

$$\Delta\bar{R}_{\text{obs}}=0.0097.$$

The largest and smallest values observed were

$$\Delta\bar{R}_{\text{min}}=0.0058,$$
$$\Delta\bar{R}_{\text{max}}=0.0129.$$

Since the intensity of illumination may easily be affected by changes in position of the light source, line voltage changes, and other effects not under experimental control, this variation is not considered alarming.

The agreement with the predicted value of ΔR is considered good in view of the uncertainties involved in the use of the flicker photometer and the photographic measurement of the line width, especially since the latter was not done with as much skill as might be desirable.

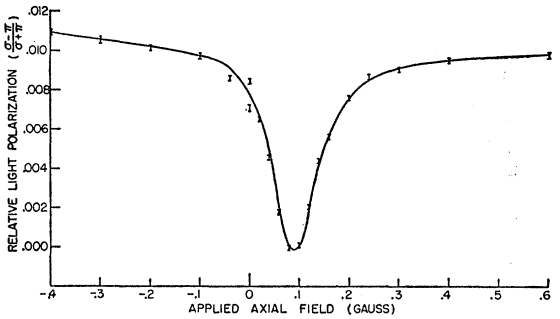

FIG. 3. Polarization ratio of the scattered resonance radiation as a function of the applied axial field component, incident light circularly polarized.

Although the transverse components of the ambient magnetic field were compensated as well as possible, one could not hope to reduce them below the 10^{-3} gauss necessary to avoid disturbing the sodium orientation. In addition, no attempt was made to compensate ac fields. Thus one might expect that orientation would disappear for insufficiently large values of the axial field. This was indeed the case, and a representative curve of light polarization *versus* applied axial field is shown in Fig. 3. The minimum is displaced from zero by the presence of a terrestrial component of 0.1 gauss in the axial direction. It should be stressed that ideally the incident light is sufficient to define the axis of quantization, and it is only necessary to apply a field to overcome the disturbing effects of fields already present.

When the measurements are extended to axial fields of several gauss, the polarization at first increases for one field direction and decreases for the other as the absolute value of the field is increased. In both directions it eventually goes through a minimum and then increases steadily. This is believed to be the result of the alteration of atomic wave functions by the magnetic field. The asymmetry is a result of the asymmetry of the light polarization and disappears for unpolarized light although the changes in polarization with increasing field remain.

The measurements of $\Delta\bar{R}$ discussed above were all taken from curves similar to Fig. 3. at about -0.2 gauss applied where the axial field is sufficiently large to allow orientation to proceed undisturbed but not large enough to change the wave functions appreciably.

The vacuum system was later modified to increase by a factor of 2.5 the illuminated solid angle seen by the sodium beam. The change in light polarization upon destruction of orientation then became $\Delta\bar{R}=0.0175$, an increase by a factor of 1.8. This increase could reasonably be expected to be less than the increase in solid angle because the additional light is all moving at an angle to the axis and is therefore less effective in producing orientation.

This polarization change of 1.75 percent corresponds theoretically to an effective average of 0.365 photon

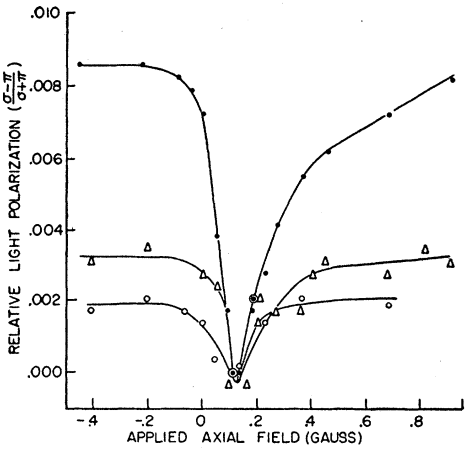

FIG. 4. Polarization ratio of the scattered resonance radiation as a function of incident intensity of circularly polarized radiation. Incident light intensities in ratio 1.00:0.39:0.17 for upper, middle, and lower curves, respectively.

scattered by each atom. For this probability of photon scattering, the ensemble averages of the expectation values of nuclear, total, and electronic angular momenta are, respectively,

$$\bar{M}_I = 0.180 = 0.120I,$$
$$\bar{M}_F = 0.216 = 0.108F,$$
$$\bar{M}_S = 0.035 = 0.070S.$$

In the case of F, the fractional figure is given relative to the larger F value of the two hyperfine states, for if orientation were complete, all atoms would be in the state $F=2$.

In order to obtain an experimental check on the polarization of the first photon scattered by an unoriented atom, the absolute polarization of light scattered by sodium atoms in the earth's magnetic field was measured. An attempt was made to do this by using the electronic detector already described and a modified scheme of analyzing polaroids, but the results were not self-consistent. Visual methods were therefore used. A Babinet compensator followed by a Nichol prism shows dark bands crossing the field of view if the polarization ratio of the light incident on the arrangement is greater than about 0.01. A glass plate between the light source and the Babinet compensator may be used to introduce an opposite polarization by rotating it in a plane containing the electric vector of the observed polarization. The polarization of the light may be calculated from the angle at which the bands disappear.[15] The angle observed was $34.2\pm1.5°$ (average of ten measurements) when a glass plate of refractive index 1.52 was used. The index was not known exactly and limits of ±0.02 were assigned which account for about one-quarter of the error quoted below for the polarization. The resulting polarization ratio is

$$R = 0.049 \pm 0.008,$$

which differs from the predicted value of 0.071 by 2.8 times the probable error. An error of this size has a probability of 6 percent, viewed as a random deviation.

To explain this result by an error in the measurement of relative intensities of the two components emitted by the source would require the assumption that the $3\,^2P_{\frac{3}{2}}$ component had only one-half the intensity of the other. Not only does this large an error in the photometric measurement seem highly unlikely, but it contradicts the very plausible assumption of blackbody emission at the line centers. An incorrect value for the hyperfine splittings would not affect the polarization by such a large amount, for the predicted polarization is decreased by wider separation of the levels, and with evels completely separated the polarization is only decreased to 0.069. Moreover, the more recent values of the splitting lead to a smaller level separation than the value used.

The intensity of the incident light was varied by means of neutral density filters. By a simple algebraic manipulation of the probabilities P_i of Eq. (4), it can be shown that if P_3 is neglected, the polarization of the scattered light is proportional to $N/(N+1)$ where N is the average number of photons scattered per atom. Since N is small compared to 1 ($N \leqslant 0.2$ in Table III), the polarization can be considered proportional to the light intensity. Figure 4 shows a sample set of curves taken with two filters of different density and without any filter. The asymmetry in these curves was a result of the inhomogeneity of the field produced by the first set of coils used. Table III gives the change in light polarization when orientation is destroyed for two sets of observations similar to those in Fig. 4. The light intensities were determined from the size of the signal produced by rotating the analyzer polaroid a known amount and agree with the filter densities given by the manufacturer. The variation is seen to be linear as expected.

Representative data are given in Fig. 5 and Fig. 6 showing the effect of varying a transverse component of the magnetic field keeping the axial component fixed. The axial components are different for the two figures. The solid curves are theoretical curves computed by assuming that many cycles of Larmor precession occur between successive photon scattering events for any one atom. Since these events are random, only time

TABLE III. Change in scattered light polarization ($\Delta\bar{R}$) when orientation is destroyed as a function of incident light intensity.

Light intensity (arbitrary units)	$\Delta\bar{R}$ (arbitrary units) run 1	run 2
1.00	1.00	1.00
0.39	0.36±0.1	0.38±0.1
0.17	0.20±0.1	0.18±0.1

[15] R. W. Wood, *Physical Optics* (Macmillan Company, New York, 1934), third edition, pp. 341–343.

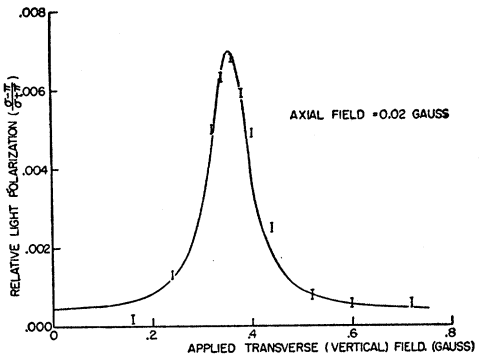

FIG. 5. Polarization ratio of the scattered resonance radiation as a function of one applied transverse field component; the solid curve is computed theoretically.

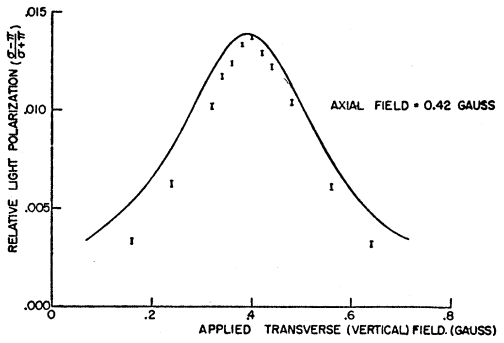

FIG. 6. Polarization ratio of the scattered resonance radiation as a function of one applied transverse field component; the solid curve is computed theoretically.

average results are required. Time-average projection operators of the z-component of angular momentum in a rotating coordinate system were derived. These were used to find the time-average occupation probabilities of the precessing atoms in eigenstates of M_F referred to stationary axes. The calculation is straightforward but too lengthy to be reproduced here. It results in the following expression for the polarization of the second-scattered photon (R_2) as a function of the angle between the magnetic field and the axis of quantization defined by the incident light:

$$R_2(\theta)=\frac{9.8\times10^{-4}\cos^8\theta-0.115\cos^6\theta+4.14\cos^4\theta+4.58\cos^2\theta+7.0}{-10.8\times10^{-4}\cos^8\theta+3.86\times10^{-2}\cos^6\theta-1.28\cos^4\theta-1.50\cos^2\theta+93.4}. \quad (7)$$

This reduces to the correct value of R_2 for $\theta=0$.

The data shown in the figures are typical in that the agreement with theory is good for small axial fields and not for larger ones, the experimental points always falling below the theoretical curve. The only adjustable parameter is the value of the polarization at the maximum of the curve. The disagreement may well be a result of the alteration of atomic wave functions by the larger total fields.

When the incident radiation is unpolarized, the curve of scattered light polarization *versus* axial magnetic field is similar to Fig. 3 in every respect except the light polarization scale. The total change in light polarization is only $\Delta\bar{R}=0.0048$. This may be explained by considering the incident light as composed of equal parts of the two senses of circular polarization. Table II shows that scattering of a circularly polarized photon reduces the population in the $M_F=0$ states independently of its sense of rotation. Consequently, scattering of unpolarized light will increase the populations of the states with $|M_F|=F$ relative to those with $M_F=0$. The atomic ensemble is then in a condition of partial alignment. Among the atoms which scatter two photons there are some which scatter two with the same sense of rotation and contribute to the alignment of the ensemble as well as others which scatter photons of opposite senses and do not contribute significantly. Since the effect of unpolarized resonance radiation on a completely aligned ensemble is to reduce the alignment, the process does not proceed to complete alignment for very large light intensity but rather reaches an equilibrium partial alignment. This possibility of alignment by unpolarized light was briefly mentioned by Kastler in the article already cited.[1]

Table IV gives the probability of an atom being in various angular momentum states after scattering one to four photons, as well as the values at equilibrium ($n=\infty$). These were computed by a straightforward application of Table I considered as a matrix operator. Although the approach to equilibrium is rapid, the equilibrium alignment is small. However, the difference in population of corresponding magnetic substates of the two hyperfine levels is still many times that resulting from the energy separation in thermal equilibrium at room temperature.

Combining the photon polarizations in Table IV with the figure (obtained from the degree of orientation for the same amount of polarized light) of 0.365 photon absorbed per atom, the total change in light polarization expected upon destruction of alignment is calculated to be $\Delta\bar{R}=0.0034$, compared to an observed value of 0.0048. This indicates that the incident light may not have been completely unpolarized.

There is no reason to believe that the effect should be confined to sodium, since the argument for the produc-

TABLE IV. Occupation probabilities for ensembles each of whose members has scattered n photons of unpolarized light, including the equilibrium state ($n=\infty$). P_1 and P_2 are the probabilities of atoms being in states with F equal to 1 and 2. R_n is the polarization ratio of photons scattered at 90° in the nth scattering process.

	$F=1$		$F=2$					
n	$M_F=\pm1$	0	±2	±1	0	P_1	P_2	R_n
1	0.128	0.119	0.144	0.115	0.106	0.375	0.624	0.0700
2	0.128	0.116	0.154	0.110	0.100	0.372	0.628	0.0905
3	0.126	0.115	0.161	0.108	0.099	0.367	0.637	0.0997
4	0.126	0.115	0.162	0.108	0.099	0.367	0.639	0.1049
∞	0.125	0.115	0.163	0.106	0.098	0.365	0.636	···

tion of alignment, like that for orientation, is quite general. The argument applies, however, only when the light is incident from a restricted part of a sphere, for isotropic unpolarized light defines no axis with respect to which alignment can occur.

I wish to express my gratitude to Dr. R. H. Dicke, without whose inspiration and invaluable counsel this research could not have been undertaken or completed. I am also indebted to Dr. D. R. Hamilton for assistance in the early part of the work.

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Angular Correlation of Photons from Positron Annihilation in Light Metals*

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The angular correlation of gamma rays from positron annihilation in the light metals, Li, Be, Na, Mg, Al, K, and Ca, has been measured. The deviation of the two photons from colinearity is a measure of the total momentum of the annihilating electron-positron pair. If it is assumed that the positrons are thermalized before annihilation and that they annihilate only with electrons of a Sommerfeld free electron gas, the experimental results may be used to determine the Fermi energy of these electrons. It is interesting that the Fermi energies so determined are in good agreement with calculated values.

I. INTRODUCTION

THE possibility of determining electron velocities in metals from positron annihilation radiation was first pointed out by DuMond *et al.*[1] In their examination of the 0.511-Mev annihilation radiation these authors showed that the measured line width exceeded the instrumental width and ascribed the excess to motion of the electrons with which the positrons annihilate. Since that time DeBenedetti *et al.*[2] and other experimenters[3,4] have measured angular correlation of the two annihilation photons to determine the motion of the center of mass of the annihilating pair. The results of all these experimenters indicate that the center of mass of the annihilating pair has a momentum which is comparable with the momentum of conduction electrons in metals.

This paper reports an experiment in which the angular correlation of the annihilation photons was measured with slightly better angular resolution than had been previously used. Whereas other experimenters have attempted to relate the shape of the "tail" of the angular correlation curve to electron velocity, in this paper it is shown that the width of the observed curve is directly related to the momentum of the annihilating electrons.

Measurements in several metals have been made and the results are compared with theory.

II. EXPERIMENTAL

A schematic diagram of the apparatus is shown in Fig. 1. A source of annihilation radiation was placed at the middle of the horizontal line joining the two scintillation detectors. The rate of coincidences in the two detectors was measured as a function of the vertical displacement of the source. When corrected for background, this coincidence rate curve gives the angular correlation of the gamma-ray pairs.

The positron emitter was made by evaporating an aqueous solution of sodium chloride containing about 0.4 mC of Na^{22} on a thin (2 mg/cm²) aluminum foil. The approximately 0.5-in. diameter deposit and supporting foil were covered on both sides by another piece of the same foil. A sheet of the specimen metal

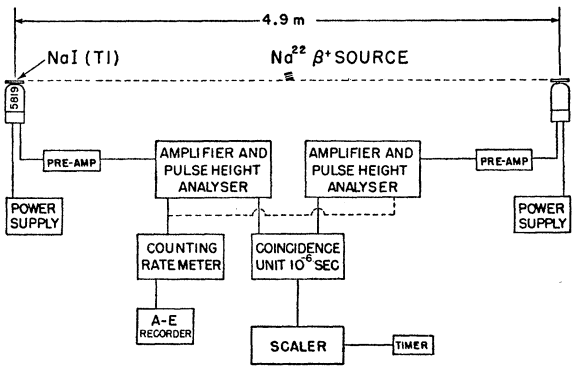

FIG. 1. Schematic diagram of apparatus.

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[1] DuMond, Lind, and Watson, Phys. Rev. **75**, 1226 (1949).

[2] DeBenedetti, Cowan, Konneker, and Primakoff, Phys. Rev. **77**, 205 (1950).

[3] P. E. Argyle and J. B. Warren, Can. J. Phys. **29**, 32 (1951), and J. B. Warren and G. M. Griffiths, Can. J. Phys. **29**, 325 (1951).

[4] H. Maier-Leibnitz, Z. Naturforsch. **6a**, 663 (1951).