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Optical Pumping*

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A brief review is given of the basic concepts of optical pumping, followed by description of an optical pumping apparatus suitable for application to a number of experiments in an advanced undergraduate, or graduate laboratory.

I. INTRODUCTION

IN the several years since its discovery, the method for obtaining alignment of atomic spins known as "optical pumping" has received increasing attention because of its usefulness in a variety of lines of research, including extremely precise measurement of atomic hyperfine structure splittings,¹⁻⁴ the effects of collisional processes and other perturbations on atomic energy levels,^{1,2,5,6} the development of compact high-precision frequency standards and sensitive precision magnetometers^{7,8} and the production of free polarized electrons.⁹ Had the development of optical pumping occurred before the development of atomic and molecular beam apparatus, it is safe to assume that it would have played a major role in advancing our knowledge of atomic and nuclear moments. It still offers a remarkably simple means of measuring hyperfine splitting energies and nuclear spins of certain atoms.

Because of the relative simplicity of the technique and equipment, and the elegance of the observations which are possible with it, we feel that optical pumping techniques may also be employed to advantage in a number of interesting applications in the teaching of advanced under-

graduate or first-year graduate physics. In particular, the dependence of magnetic dipole precessional frequency on magnetic field may be easily demonstrated, a straightforward demonstration of the Zeeman effect may be given, and perhaps most important, a direct demonstration of the concept of spin angular momentum in the electron may be given. In this article, we present a brief semiquantitative description of the optical pumping process followed by details of an optical pumping apparatus suitable to any of the applications mentioned.

II. CONCEPTS

Despite use of the word "pumping," no transport of material is involved, but rather a "pumping" of atomic spin orientations from an equilibrium distribution over magnetic substates into a nonequilibrium distribution in which a preponderance of spins are aligned in a given direction.

For simplicity, let us confine our attention to the alkali atoms, which have $^2S_{1/2}$ ground states. Transitions from the first excited state to the ground state involve energies sufficient to give rise to optical radiation. Since the first excited state is a P state (orbital angular momentum $L=1$), and the total electronic spin S is $\frac{1}{2}$, there are in this state two possible values of the total electronic angular momentum, i.e., $J=\frac{3}{2}$ and $\frac{1}{2}$. Due to the spin-orbit interaction, these two states of different total angular momentum differ in energy relative to the ground state, which gives rise to the familiar doublet structure in the spectra of the alkali metals (Fig. 1, first column). Because of the existence of nuclear spin, however, the ground state and all other states will be additionally split into a number of substates, the *hyperfine states* (Fig. 1, second column). Each of these states has slightly different energy,

* This work was supported in part by the U. S. Air Force.

¹ M. Arditi and T. R. Carver, Phys. Rev. 109, 1012 (1958); 112, 449 (1958).

² E. C. Beaty, P. L. Bender, and A. R. Chi, Phys. Rev. 112, 450 (1958).

³ P. L. Bender and E. C. Beaty, Phys. Rev. Letters 1, 311 (1958).

⁴ L. W. Anderson, F. M. Pipkin, and J. C. Baird, Jr., Phys. Rev. 116, 87 (1959); Phys. Rev. Letters 4, 69 (1960).

⁵ M. Arditi, J. phys. radium 19, 873 (1958).

⁶ H. Margeneau, P. Fontana, and L. Klein, Phys. Rev. 115, 87 (1959).

⁷ W. E. Bell and A. L. Bloom, Phys. Rev. 107, 1559 (1957).

⁸ P. Franken and F. D. Colgrove, *Proceedings of the Ann Arbor Conference on Optical Pumping* (University of Michigan Press, Ann Arbor, Michigan, 1959) p. 119.

⁹ H. G. Dehmelt, Phys. Rev. 109, 381 (1958).

arising from the interactions of the electron with the nuclear magnetic dipole moment and electric quadrupole moment. The different energies leading to this hyperfine structure come from the dependence of the interaction energy on the various quantum-mechanically allowed orientations of the nuclear angular momentum I relative to the total electronic angular momentum J . According to the rules of quantization, the allowed combinations are such that the total angular momentum F takes on the values $F=I+J, I+J-1, \dots, I-J$ only. A still further splitting of levels (the Zeeman splitting of the hfs levels) may be produced by application of a *weak external* magnetic field (Fig. 1, third column). This splitting arises from the different energies associated with different orientations of a given F with the external H field and is directly proportional to H in the low field limit. Again, only orientations of F are allowed which give projections m_F along the H direction of $m_F=F, F-1, \dots, -F$. The totality of levels for the ground and first excited states of an alkali atom

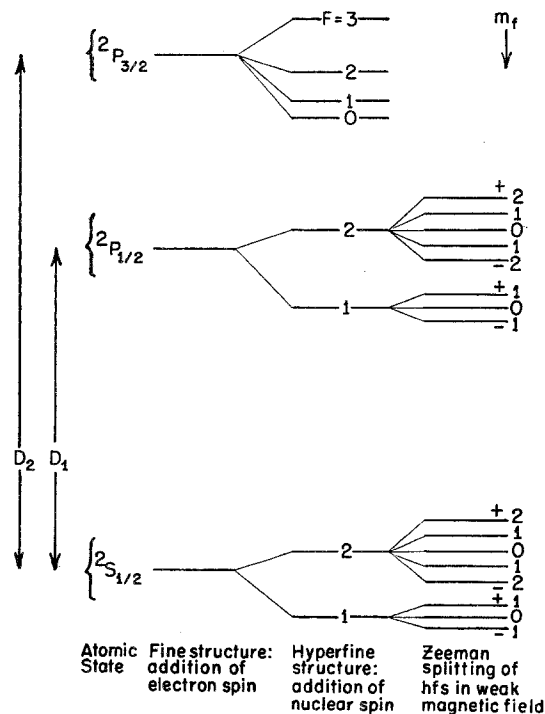


FIG. 1. Atomic energy level splitting produced as various interactions are added to the Bohr model, for an alkali atom having nuclear spin $I=\frac{3}{2}$. The spacing between energy levels is not to scale, and for simplicity the Zeeman splitting of the $2P_{1/2}$ state is not shown.

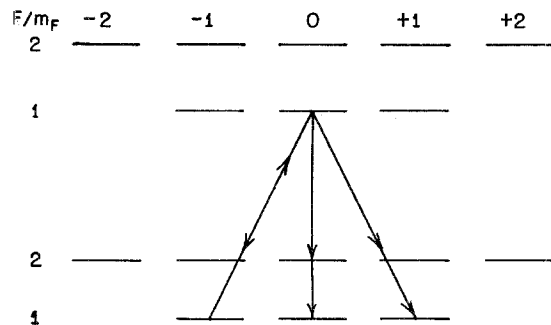


FIG. 2. Possible transitions resulting from the absorption of a photon carrying $+1$ unit of angular momentum, by an atom in the $F=1, m_F=-1$ substate of the ground state.

with nuclear spin $I=\frac{3}{2}$ is shown in Fig. 1. Transitions between levels of different F , the so-called *hyperfine transitions*, will involve absorption or emission of magnetic dipole radiation, with the attendant rule $\Delta F=0, \pm 1$ only. Transitions between different m_F levels, with F remaining constant, comprise the *Zeeman transitions*, with the attendant rule $\Delta m_F=0, \pm 1$ only.

The principle of optical pumping may now be understood. Imagine a vapor of free Rb atoms, for instance, ($I=\frac{3}{2}$ for the isotope Rb^{87}) in a weak magnetic field leading to Zeeman splitting of the hyperfine structure, and consider the effect of shining a beam of circularly polarized resonance radiation through the vapor. For reasons which will become clear later, let us confine our attention to the D_1 , or lower energy member of the doublet comprising the resonance radiation. This radiation will be readily absorbed, leaving an atom in the $2P_{1/2}$ excited state. But if the light is left-circularly polarized (all photon angular momenta parallel to the direction of propagation), then absorption of a photon must lead to a net gain of one unit of angular momentum, if we shine the light in the direction of the external magnetic field vector. The process is illustrated in Fig. 2, where the various Zeeman levels for the ground and $2P_{1/2}$ states are depicted in a somewhat different way than in Fig. 1.

Absorption of a quantum of D_1 radiation by an atom in the $F=1, m_F=-1$ substate, say, can thus only lead to a substate with $m_F=0$ in the $2P_{1/2}$ excited state. (Doppler broadening of the resonance radiation will in general allow transitions to either the $F=2$ or 1 hfs levels of the excited state.) When this state reradiates to the ground state, however, the dipole radiation rules

permit $\Delta m_F = 0, \pm 1$ with equal probability, so that the atom has a probability of $\frac{2}{3}$ to return to the ground state with a greater value of m_F —i.e., a larger component of spin along the reference axis—than it had originally. The atom may absorb and reradiate again and again. If nothing happens to relax the various m_F level populations between these occurrences, it is apparent that in time all atoms would be “pumped” into the $F=2$, $m_F=+2$ ground-state level, giving complete alignment of the atomic spins. Furthermore, whenever an atom reaches this level, it can no longer absorb D_1 radiation, and thus the vapor would in time become transparent to the D_1 radiation.

In fact, however, collisional processes between different atoms of the vapor, and with the walls of the container in which the atoms are held, cause strong mixing of different m_F levels in both the ground and excited states, making the situation less than ideal. The advantages of eliminating the D_2 radiation,¹⁰ as we have hypothetically done in the above discussion, are now clear. If it were allowed to remain, the $^2P_{3/2}$ excited state would contribute a level with $m_F=+3$, to which the $m_F=+2$ ground substate could be raised by D_2 absorption. This would then allow an “escape route” for decreasing the polarization of the sample, at least to the extent that the relaxation processes occur in times short compared to the average lifetime of the excited state. Experimentally, this seems to be the case, and difficulty in obtaining an observable amount of polarization may be encountered unless the D_2 radiation is removed from the incident beam by appropriate filters.

In addition, one may get a better signal by taking steps to minimize the relaxation processes: (a) a noble gas such as argon or neon, which itself has little tendency to produce depolarization during collisions, may be used as a “buffer gas” to increase greatly the time required by an alkali atom to diffuse to the walls of the chamber^{11,12};

¹⁰ W. Franzen and A. G. Emslie, Phys. Rev. **108**, 1453 (1957).

¹¹ The presence of a buffer gas in the proper pressure range also results in considerable reduction in the Doppler broadening of resonance signals—an important feature in making high-precision measurements of energy splittings [R. H. Dicke, Phys. Rev. **89**, 472 (1953), and footnote references 1, 2, and 5].

¹² F. Hartman, M. Rambosson, J. Brossel, and A. Kastler, Compt. rend. **246**, 1522 (1958).

(b) the incident light intensity may be increased, so as to decrease the pumping time relative to the mean relaxation time; (c) various compounds, generally heavy paraffinlike hydrocarbons such as eicosane or dotriacontane, may be used as wall coatings, since these show much the same neutral effect in collisions as the noble gases.^{13,13a}

The formal equations describing the optical pumping process consist of a set of simultaneous differential equations describing the rate of pumping to and from each possible substate. The general form for a system having n substates in the ground state is given by the n equations

$$\dot{P}_k = - \sum_{j=1}^n (b_{kj} + w_{kj}) P_k + \sum_{i=1}^n (b_{ik} + w_{ik}) P_i; \quad k=1, 2, \dots, n, \quad (1)$$

where P_k is the occupation probability of the k th substate (thus $\sum_k P_k = 1$, i.e., the probability of being in *some* substate equals unity), b_{ij} represents the probability per unit time of a transition from substate i to substate j of the ground state by absorption and re-emission of a photon, and w_{ij} represents the probability per unit time of the corresponding ground state transition induced by any relaxation mechanisms present. Thus for a given state k the first summation term on the right represents the rate of *loss* from state k to all other states $j \neq k$ (signified by the prime on the summation) due to photon-induced transitions and to transitions induced by the relaxation mechanism, and the second summation represents the rate of *transfer to* state k from all other states $i \neq k$ due to the same causes.

If no relaxation mechanism is assumed to

¹³ H. G. Robinson, E. S. Ensberg, and H. G. Dehmelt, Bull. Am. Phys. Soc. **3**, 9 (1958); W. Franzen, Phys. Rev. **115**, 850 (1959); W. B. Hawkins, W. R. Bennett, L. Y. Chow, and H. Robinson, Bull. Am. Phys. Soc. **3**, 372 (1958).

^{13a} Note added in proof.—It has been pointed out to the author by C. Alley (private communication) that in a properly wall-coated and evacuated bulb, a D_2 filter is not necessary, since the wall coating tends to preserve spin direction during collisions, and removal of the buffer gas reduces the number of collisions per second experienced by an alkali atom to the point where the relaxation time due to collisional mixing proves long compared to the reradiation time from the excited state. Recent experiments have shown that probably the best, and certainly the most easily applied wall coatings, are certain silicon compounds, such as SC-87 manufactured by the General Electric Company. The author will be glad to supply instructions for their application.

operate on the excited states, then determination of the b_{ij} can be made from a straightforward application of the standard formulas for electric dipole excitation.¹⁴ In the more general (and realistic) case, however, the b_{ij} thus determined must be modified to account for the presence of relaxation in the excited state. The general solution of Eq. (1) has been treated for the limiting cases of no mixing, and complete mixing in the excited and ground states, by Bell and Bloom⁷ and by Franzen and Emslie.¹⁰

Before describing the method of detecting the attainment of a net polarization in the sample, and of detecting Zeeman and hfs transitions, we wish to point out that to obtain optical pumping, there *need* be no magnetic field involved, as the direction of propagation of the circularly polarized light itself defines an axis of quantization. In the discussion preceding, we have chosen the situation in which a magnetic field does exist, because it seems easiest to give a lucid description in this case, and it is the one almost always encountered experimentally. In the absence of any H field one obviously cannot observe *Zeeman* transitions, as all the m_F levels are degenerate in energy, although not in angular momentum. In this case orientation of the sample may still be observed through the medium of hyperfine transitions.

The detection of optical pumping may be accomplished quite simply,¹⁵ since the light beam responsible for the pumping also will indicate that pumping has taken place. For, as noted above, the vapor will become to a certain extent transparent to the incident resonance radiation when a time has elapsed sufficient to allow a nonequilibrium population to be built up in the higher m_F substates—typically a few tens of milliseconds. If anything is now done to disturb this new population distribution, the absorption of the vapor will again increase. The simplest way to achieve this is to reverse the external magnetic field, carrying the atomic spins with it.¹⁵ Then the states of negative m_F , relative to the axis defined by the light beam, will suddenly become the most heavily populated, and strong

light absorption will occur as these are pumped back toward positive values. A simple phototube detector connected to an oscilloscope and monitoring the transmitted light intensity serves as the detector of optical pumping. This method, although sufficient to illustrate the presence of atomic alignment, does not reveal information of much interest. If, however, an rf or microwave signal of a proper frequency to cause Zeeman or hyperfine transitions is applied to the sample, the light absorption of the sample will again increase as long as the signal lasts, and after that will immediately start to decrease until the upper m_F states again attain their maximum populations under the influence of the pumping radiation. Thus *optical* detection of Zeeman and hfs transition frequencies is made possible, which gives the optical pumping method its considerable advantage and simplicity compared to atomic beam experiments or other types of resonance techniques.

Only a brief discussion of the most straightforward features of optical pumping has been given here. The literature contains many detailed theoretical discussions and specific applications of optical pumping techniques, and gives further details of experimental refinements which may be applied to the apparatus as it is outlined in the following section. We wish to emphasize, however, that the system as described in the following is fully capable of illustrating a number of fundamental concepts in modern atomic physics.

III. EXPERIMENTAL APPARATUS

A schematic diagram of the required apparatus is shown in Fig. 3. The two alkali elements on which optical pumping may most easily be accomplished are sodium and rubidium. Of these, Rb pumping is hampered by the lack of intense light sources, but otherwise presents distinct advantages over Na pumping. These are (a) the low melting point of Rb (39°C) which allows sufficient vapor pressure for good pumping at a temperature of 35–45°C, in contrast to the 100–140°C needed for Na, and (b) the large separation of the D_1 and D_2 lines (7947 Å and 7800 Å, respectively) allowing relatively inexpensive and efficient removal of the D_2 line by the use of interference filters. In the following discussion, we

¹⁴ E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, New York, 1935).

¹⁵ H. G. Dehmelt, *Phys. Rev.* **105**, 1487 (1957).

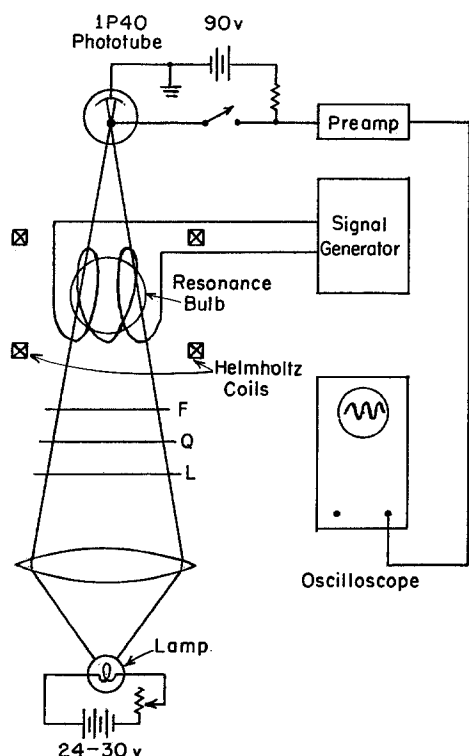


FIG. 3. Schematic of the optical pumping apparatus. The ac starting circuit for the lamp is not shown. *F*, *Q*, and *L* are the D_2 filter, quarter-wave plate, and linear polarizing plate, respectively.

shall consider a Rb pumping system, although the basic elements are of course the same for Na pumping.

A. General Apparatus

Osram lamps are probably the most suitable commercial sources of resonance radiation. They are designed to start on 220-v ac, but after warmup they can, and must, be switched to dc to eliminate 60-cycle modulation. They can be readily driven by 24- to 30-v dc and generally require a little less than the maximum rating of 1.5 amp to obtain the best light quality. The bulbs need not be run in a vertical position, despite the manufacturer's instructions. Some bulbs may show low-frequency oscillations even under dc operation. The frequency and amplitude of these oscillations seems to be a function of bulb temperature, tending to be worse at low temperature. For ease in controlling the bulb temperature, the outer vacuum jacket may be removed, and this is often enough to improve

operation. A bath of heated silicone oil around the bare bulb may be required for optimum temperature control, and in any case will usually result in a severalfold increase of resonance light output. The temperature and lamp current should both be kept below the point where the light begins to take on a whitish color—a uniform raspberry color seems to be best.

Almost any lens will do to focus light on the resonance bulb. Since an interference filter must be used to remove the D_2 line,^{18a} the divergence of the beam passing through the filter must be kept small (less than about 8° half-angle). The best compromise between light gathering power and low divergence is obtained by using a 2- or 3-in. diam lens of short focal length, with the lamp placed approximately at the focus.

High quality interference filters are available from various optical suppliers¹⁶ at a cost of about \$250 for a $3\frac{1}{4}$ -in. diam filter. Filters not meeting stringent test requirements but suitable for use here may occasionally be picked up for considerably less. The circular polarizer consists of a linear polarizer (Polaroid type *J* or similar is suitable) and a quarter-wave plate. The latter is available at low cost from the Polaroid Corporation in thicknesses suitable for Na or Rb wavelengths.

We shall pass over discussion of the resonance bulb for the moment, to consider the detector next. This consists of a simple phototube—the RCA 1P40 is suitable, or any other with a comparably high gain and low dark current. The detection system should be shielded from extraneous light, particularly that from incandescent room lighting as this shows considerable 60-cycle modulation. The signal is taken from the phototube through shielded cable to a low noise preamplifier, such as the Tektronix 122 or 123, having a gain of about 100. The preamp output is then fed to an oscilloscope. The latter should have a sensitivity of at least 0.1 v/cm for a preamp gain of 100.

A set of Helmholtz coils about 15 or 16 in. in diameter will produce a field of enough uniformity to give reasonably narrow resonance lines. Depending on local magnetic field conditions, it may be desirable to place a second

¹⁸ For example, Spectrolab, Inc., 7423 Varna Avenue, North Hollywood, California.

Helmholtz pair at right angles to the first to eliminate any appreciable transverse field components. Alternatively, the whole apparatus may be tilted so that its axis lies along the local external field direction. One or two gauss of applied field is sufficient to give good resonances at convenient frequencies, although generally not enough to allow resolution of individual Zeeman transitions arising from nonlinear terms in the field dependent expression for the Zeeman splitting. It is quite possible to observe resonances with the apparatus aligned in the earth's field alone, but some applied field is recommended, because of the flexibility thus allowed.

If Rb is used, the resonance bulb may conveniently be warmed by directing a low velocity stream of warm air at the bulb. Heating coils, blower motors, etc. should be kept from the immediate neighborhood of the resonance bulb to avoid modulation or distortion of the magnetic field. It will often be found that sufficient Rb vapor will be present at room temperature to observe a good resonance signal, although a bulb which has stood for some time, or a new bulb, may require preliminary heating to a temperature of at least 50 or 60°C. It has often been observed, particularly with new bulbs, that a satisfactory resonance cannot be obtained unless some Rb has been deposited on the walls of the bulb itself and then cleared off by using a gentle flame. A deposit of Rb in the filling neck alone is often not sufficient. This does not apply if a wall coating is to be used.

B. Resonance Bulbs

Spherical bulbs with approximately 200–300 cm³ capacity are handiest, as they are large enough to provide long wall diffusion times and small enough so that a relatively uniform H field may be provided over the bulb volume without use of excessively large Helmholtz coils. Bulbs much larger than the cross section of the resonance light beam passing through them should be avoided. Proper preparation of resonance bulbs, although straightforward, is rather tedious and may require some practice. The bulbs should be cleaned with chromic acid or a similar cleaning solution and thoroughly rinsed and dried before attachment to a vacuum system for evacuation.

They should be pumped to at least 5×10^{-6} mm Hg while being simultaneously baked at 250–300°C for about 24 hr. A small oven of asbestos board heated with a glow coil is suitable for baking. Free alkali metals may be obtained by reduction, in vacuum, of their chloride salts by heating with finely divided calcium metal,¹⁷ but this method is quite troublesome and may sometimes fail to yield sufficient free metal. It is generally easier to introduce the pure metal into the vacuum system. Metallic Rb may be obtained¹⁸ in sealed ampules which may be stored under toluene after opening. The latter, being quite volatile, is easily pumped from the sample after it is sealed into the vacuum system. A small piece of the metal is placed in a constricted sidearm on the resonance bulb and after a pressure of 10^{-2} or 10^{-3} mm has been reached, a preliminary distillation should be made to get rid of volatile impurities.¹⁷ Pumping and baking of the bulb are then continued until the lowest possible pressure has been reached, whereupon the bulb is allowed to cool and a thin visible film of Rb is distilled into it. If a wall coating is to be used, this should be distilled into the bulb from another sidearm before the Rb is introduced, or if a buffer gas is to be used, this is let into the bulb to a pressure of 2 to 5 cm of mercury after the introduction of the Rb. Only spectroscopically pure gas¹⁹ should be used. After sealing off and removing the bulb from the vacuum system, as large an area of the walls as possible should be cleared of any visible Rb film by gentle heating.

C. Detection

An rf or microwave signal of the proper frequency applied to a bulb being optically pumped will induce Zeeman or hyperfine transitions between various energy levels. The resonant frequency depends on the magnetic field present and on the particular transition to be observed. Rules allowing a calculation of the Zeeman splitting of the hyperfine levels ($\Delta F=0$, $\Delta m_F = \pm 1$) and the hyperfine level splitting ($\Delta F = \pm 1$)

¹⁷ J. Strong, *Procedures in Experimental Physics* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1938), Chap. 13.

¹⁸ Available from various suppliers, such as A. D. Mackay, Inc., 198 Broadway, New York, New York.

¹⁹ Available commercially from the Linde Division of the Union Carbide Corp., in 1-liter sealed flasks.

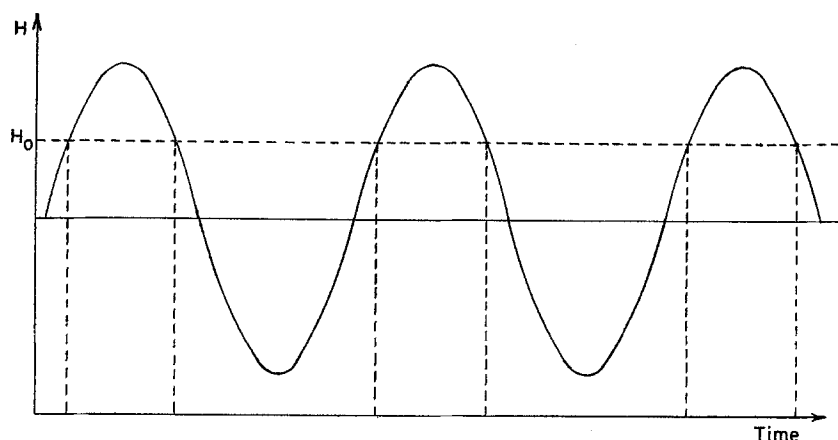


FIG. 4. Spacing of resonance signals as a function of magnetic field. The solid horizontal line represents the steady magnetic field value. The sinusoidal line represents the superimposed wobbling field, and the dotted line, the proper magnetic field to cause resonance at the particular rf frequency being applied to the sample.

in the low field limit may be found in any standard text on atomic structure. For fields of 1 or 2 gauss, the frequencies for the former are usually in the range of several hundreds of kilocycles to a few megacycles, and for the latter, a few thousand megacycles. Unless a long time-constant detection device such as a pen recorder is to be used, it is most convenient to keep the frequency fixed and cycle the magnetic field about the resonance value so as to obtain a continuous series of resonance signals for presentation of an oscilloscope. A field wobbling frequency as high as 60 cycles may be used, and has the advantage of convenience. Unless a light source of more than average intensity is to be used so as to give rapid repumping, higher wobbling frequencies should be avoided. Modulation of a few percent of the applied field is sufficient, although if the dc field is particularly homogeneous, a modulation amplitude of much less than this will be adequate, and may even be provided by stray 60-cycle fields in the laboratory.

Sufficient rf field to induce Zeeman resonance can be obtained with 5 or 6 turns of wire in a 4-in. diam helix around the bulb at right angles to the optical axis. The generator should be capable of delivering a voltage of roughly 0.5 v across a 100-ohm carbon resistor in series with such a coil, as measured with a vacuum-tube voltmeter.

The signal will appear as a spike on the oscilloscope trace with an amplitude, under optimum conditions, at least 100 times greater than noise level, and a repetition rate of 120 cps, since the H field sweeps through the resonance frequency twice on each full cycle. The spacing between

successive pairs of peaks on the oscilloscope trace will be found to vary as either the dc magnetic field or the frequency is changed, and successive pairs of peaks may in fact be made to merge with each other so that a repetition of only 60 cycles is observed. The reason for this may be seen by reference to Fig. 4. The dotted line represents the field H_0 required for resonance with a fixed rf signal. The solid horizontal line represents the dc magnetic field value, and the sinusoidal line the superimposed wobbling field. Whenever the wobbling field cuts the line H_0 , resonance occurs. As the dc field is varied the wobbling field is of course carried along and so cuts the H_0 line at varying times. The argument is similar if one holds the dc field fixed and slowly varies the frequency.

Natural rubidium consists of a mixture of two isotopes, Rb^{85} and Rb^{87} , in the ratio 72 to 28%, with nuclear spins of 5/2 and 3/2, respectively. Each isotope thus has a different Zeeman and hfs structure. If the sweep of the wobbling field is too large, these resonances may overlap on the oscilloscope trace, leading to some confusion. In addition various rf harmonics may be present in

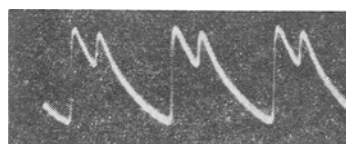


FIG. 5. Resonance signals from Rb with free running oscilloscope trace and 60-cycle field modulation. Adjacent close-spaced peaks are explained by Fig. 4. A positive going trace represents a decrease in light intensity. The "exponential" decay of each signal is caused by the time needed to repump the sample.

the rf signal and this will lead to resonances at several magnetic field values.

The appearance of a typical Zeeman resonance signal in Rb is presented in Fig. 5. Here the external magnetic field is sufficiently weak, and the resonance is sufficiently broadened by field inhomogeneities, that the individual Zeeman transition frequencies between different m_F levels are not resolved, although in principle their resolution is quite feasible.

IV. CONCLUSION

The basic theory and constructional features for an optical pumping apparatus suitable for observing Zeeman splitting of hyperfine levels, and hyperfine transition frequencies have been outlined above. At the cost of slightly more complexity, several other interesting experiments may be performed, a few of which are mentioned in the following.

In addition to Zeeman and hyperfine structure resonance frequencies in the limited number of atoms susceptible to direct optical pumping, resonances may be observed in other atoms and in free electrons by "secondary" optical pumping.^{9,20} If certain types of foreign atoms are present in the alkali vapor being optically pumped, some orientation will be imparted to these by spin exchange collisions with oriented alkali atoms. Appropriate resonance frequencies may then be applied to study transitions in this new system of oriented atoms. At resonance, the orientation of the foreign atoms will be destroyed and therefore alignment will be taken from the alkali system in subsequent collisions until the system again reaches equilibrium. During the interval before equilibrium is again reached, the alkali system will show increased opacity to the resonance light, and thus acts not only to orient the foreign atoms, but also to detect their subsequent disorientation by an rf resonance signal.

Instead of foreign atoms, one may inject free electrons and detect their spin flip resonance frequency, ν_s , from which the electron gyromag-

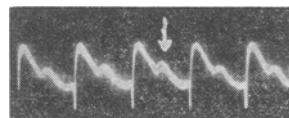


FIG. 6. An electron resonance signal. The large amplitude negative signals are caused by 60-cycle ionization pulsing of about 3-msec duration per pulse. The slope of the line upon which the electron signal is superimposed is caused by the slowly increasing transparency of the vapor as it is repumped after the disorientation resulting from the ionization pulse.

netic ratio g_s may be determined according to the relation $g_s = h\nu_s/\mu_0 H$, where μ_0 is the Bohr magneton and H the applied magnetic field. Since in practice the latter is difficult to determine accurately, ν_s may be compared with the frequency of an alkali Zeeman resonance by alternately observing the two resonance signals in the same field. One thus determines g_s in terms of the g factor of the alkali atom involved. Since the latter can be readily calculated with good precision, one ultimately may obtain g_s itself.⁹ One may easily obtain free electrons by applying a strong rf signal of a few megacycles for a few milliseconds duration to a tuned coil surrounding the projecting neck of a resonance bulb, or more easily, directly to an electrode sealed through the wall of the bulb. A momentary discharge in the buffer gas provides a large number of free electrons. If the ionization pulsing is synchronized to the magnetic wobbling frequency in such a way as to allow at least several milliseconds to elapse between pulsing and the time the field reaches the electron resonance value, so that the electron-alkali system will have come to equilibrium, a weak electron resonance signal (~ 10 to 100 times less than the alkali resonance signal) can be detected. Such a resonance is shown in Fig. 6. The ratio of bound to free electron g factors may be measured to at least three significant figures by this method with quite a crude setup, and to better accuracy if more pains are taken.

Several other variations and/or extensions of the technique of optical pumping are possible and have recently appeared in the literature.²¹

²⁰ R. Novick and W. W. Holloway, Jr., *Bull. Am. Phys. Soc.* **3**, 371 (1958); N. Knable, W. W. Holloway, Jr., and R. Novick, *Bull. Am. Phys. Soc.* **4**, 259 (1959); P. Franken, R. Sands, and J. Hobart, *Phys. Rev. Letters* **1**, 52 (1958); and Anderson *et al.*, footnote reference 4.

²¹ See in particular, Vol. 19, *Journal de physique et Le radium*, and *Proceedings of the Ann Arbor Conference on Optical Pumping* (University of Michigan Press, Ann Arbor, Michigan, 1959).

We wish again to emphasize the simplicity of the fundamental concepts and apparatus, however, and the fact that the technique allows a basic demonstration to be given of the concepts of quantization of angular momentum and the vector addition of angular momenta and associated magnetic moments in atomic structure. The demonstration is particularly forceful if the basic apparatus is extended to make possible the observation of free electron spin.

V. ACKNOWLEDGMENT

No consistent attempt has been made in this article to refer in the footnotes to the earliest work in any given facet of optical pumping. Rather, I have attempted to list what I hope may be the most helpful references in each given case. For any sins of omission, I beg the pardon of those involved. I also wish to express gratitude to Dr. A. K. Mann for helpful suggestions in the preparation of this manuscript.

Lecture Demonstration of Optical Properties of Thin Films of Dielectrics and Metals

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Apparatus for demonstrating some optical properties of thin films to large audiences is described. In addition to all the thin film interference effects, one can demonstrate (a) the phase shift experienced by the electric vector parallel to the plane of incidence when passing through the Brewsterian angle and (b) the phase shift at a metal-dielectric reflection. The apparatus depends for its success upon the high brilliance and saturation of the reflected interference colors from a composite metal-dielectric film. Its construction and properties are described.

I. INTRODUCTION

THE demonstration of optical interference in thin dielectric films to a large audience is limited by the relatively low intensity of reflected light and the lack of saturation of the interference colors in transmission. Brilliant and highly saturated interference colors sufficiently brilliant for use in a large lecture hall, with usual white light sources, are obtained when one electrolytically oxidizes completely a front-surfaced aluminum mirror on glass to form a transparent aluminum oxide film.

II. PREPARATION OF INTERFERENCE FILMS

The author has prepared his films by complete oxidation of the aluminum of a front-surfaced mirror, employing a 2% aqueous solution of oxalic acid. This solution was placed in a large beaker and covered with about one-half inch of kerosene. This kerosene layer increases the contact angle between the solution and the metal and so prevents high current densities in the meniscus

and consequent too rapid oxidation in this region. Hass and Scott¹ have described the electrolytic oxidation of front-surfaced mirrors. The interested reader may find it profitable to study their discussion before attempting to produce his own mirrors.

When the aluminum is properly oxidized, one obtains a glass plate coated with an adherent transparent layer of aluminum oxide of interference color thickness. The interference color by reflection is very brilliant and highly saturated. But the interference color observed by reflection from the oxidized side (obverse) is the complement of that reflected from the glass side (reverse). As will be seen later, these effects are related to a discontinuous layer of unoxidized aluminum at the glass surface.

A typical specimen has the following properties. The mirror has a transmittance of 84%. The reflected intensity near normal incidence from the obverse side (oxide-air interface) will be approxi-

¹ G. Hass and N. W. Scott, *J. phys. radium* 2, 394 (1950).