mann for suggesting this experiment and for many useful discussions. We are grateful to Dr. D. T. Phillips for much help with the electronics.

*Work supported in part by the Alfred P. Sloan Foundation and the U. S. Atomic Energy Commission.

†National Science Foundation Predoctoral Fellow.

‡Alfred P. Sloan Fellow.

[1]C. N. Yang, Phys. Rev. 77, 242 (1950).

[2]E. Bleuler and H. L. Bradt, Phys. Rev. 73, 1398 (1948).

[3]C. S. Wu and I. Shaknov, Phys. Rev. 77, 136 (1950).

[4]A. Einstein, B. Podolsky, and N. Rosen, Phys. Rev. 47, 777 (1935).

[5]N. Bohr, Phys. Rev. 48, 696 (1935).

[6]D. R. Bates and A. Damgaard, Phil. Trans. Roy. Soc. London Ser. A 242, 101 (1949).

[7]W. W. Smith and A. Gallagher, Phys. Rev. 145, 26 (1966).

[8]R. D. Kaul, J. Opt. Soc. Am. 56, 1262 (1966).

[9]H. Frauenfelder and R. M. Steffen, in Alpha-, Beta-, and Gamma-Ray Spectroscopy, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), Vol. II, Chap. XIX, Sec. 9.

OFF-RESONANT LIGHT AS A PROBE OF OPTICALLY PUMPED ALKALI VAPORS*

W. Happer and B. S. Mathur
Columbia Radiation Laboratory, Columbia University, New York, New York
(Received 5 December 1966)

Vapors of optically pumped atoms are usually studied by observing the interaction of resonant light with the atoms. Experiments using off-resonant light have been proposed and have been carried out with different isotopes of mercury.[1] In this Letter we discuss the fundamental criteria which determine the feasibility of experiments using off-resonant light, and we show explicitly which observables can be measured in the important case of the alkali atoms. The light of a Sr II line is shown to be a good nonperturbing probe for optically pumped rubidium atoms.[2]

Consider a vapor of alkali atoms with nuclear spin I, traversed by a beam of monochromatic light whose frequency ω almost coincides with a resonant absorption frequency ω_F of the vapor. We will assume that the hyperfine structure (hfs) in the optical line of the alkali atom is chiefly due to the splitting of the ground-state $^2S_{1/2}$ level into the two hfs sublevels $F = I \pm \frac{1}{2}$. Then one can show that the indices of refraction for right (+) and left (−) circularly polarized light, propagating in the direction of the unit vector $\hat{\imath}$, are

$$n_\pm = 1 + \delta n_0 \pm \delta n_1, \tag{1}$$

where

$$\delta n_0 = \sum_F \frac{Ne^2\lambda_F f_J(\omega-\omega_F)}{2mc[(\omega-\omega_F)^2+\Gamma^2]} p_F, \tag{2}$$

and

$$\delta n_1 = \sum_F \frac{Ne^2\lambda_F f_J(\omega-\omega_F)}{2mc[(\omega-\omega_F)^2+\Gamma^2]} \times \left[\frac{11-4J(J+1)}{4}\right] \frac{\langle\vec{\mu}\rangle_F \cdot \hat{\imath}}{g_J \mu_0}. \tag{3}$$

Here N is the atomic density, Γ is the collision-broadened width of the excited state, f_J is the absorption oscillator strength for the ^2P_J resonance line, and λ_F is the wavelength of the transition. The index of refraction $n_\pm$ depends on the ground-state observables p_F and $\langle\vec{\mu}\rangle_F$, where p_F is the probability that the atom is in the hyperfine sublevel F, and $\langle\vec{\mu}\rangle_F$ is the expectation value of the electron magnetic moment within that sublevel.[3] Thus all of these observables may be detected using off-resonant light.

An examination of Eqs. (2) and (3) shows that the wavelength λ of the off-resonant probing light must fulfill the following criteria. In order to detect the relative hfs populations p_F, it is necessary that the wavelength of the probing light be much closer to one hfs component, $\lambda(F)$, of an absorption line than to the other, $\lambda(F')$; i.e., $|\lambda-\lambda(F)| \ll |\lambda-\lambda(F')|$. If this is not the case, δn_0 [Eq. (2)] may be expanded in a power series $\delta n_0 \propto 1+\beta_1(p_F-p_{F'})[\lambda(F)-\lambda(F')]/\{\lambda-\frac{1}{2}[\lambda(F)+\lambda(F')]\}+\cdots$ in which the terms containing $p_F-p_{F'}$ are negligibly small. However, the net magnetization $\langle\vec{\mu}\rangle_F+\langle\vec{\mu}\rangle_{F'}$ can still

be detected if the wavelength of the probing light is much closer to one component, $\lambda(^2P_J)$, of a resonance line doublet than to the other, $\lambda(^2P_{J'})$; i.e., $|\lambda-\lambda(^2P_J)| \ll |\lambda-\lambda(^2P_{J'})|$. If $|\lambda-\lambda(^2P_J)| \approx |\lambda-\lambda(^2P_{J'})|$, the contributions of the magnetization to the index of refraction δn_1 (Eq. 3) almost cancel, and the off-resonant light no longer serves as a sensitive probe of population differences within the alkali ground-state sublevels.[4]

A very simple relationship exists between the index of refraction n [Eq. (1)] and the light shift[5] δE of the ground-state atomic-energy levels. Both n and δE can be represented as the expectation value of certain operators, i.e., $n = \mathrm{Tr}(\rho\hat{n})$ and $\delta E = \mathrm{Tr}(\rho\,\delta\hat{E})$, where ρ is the density matrix for the ground-state atoms. Then

$$N\delta\hat{E} + (\hat{n}-1)u_0 = 0, \tag{4}$$

where u_0 is the time-averaged energy density of the probing light. Equation (4) is just an expression for the law of conservation of energy since $N\delta E$ is the increase in the average energy density of the atoms, while $(n-1)u_0$ is the increase in the energy density of the probing light beam.

The light-shift operator $\delta\hat{E}$ [Eq. (4)] may be expressed as the sum of a scalar operator $\delta\hat{E}_0$, which shifts all of the Zeeman sublevels of a given hfs multiplet by an equal amount, plus an effective magnetic field[6] $\delta\vec{\mathrm{H}}$ which interacts with the magnetic moment of the electron, $\vec{\mu} = g_J\mu_0\vec{\mathrm{J}}$; i.e.,

$$\delta\hat{E} = \delta\hat{E}_0 + \delta\vec{\mathrm{H}}\cdot\vec{\mu}, \tag{5}$$

where

$$\delta\hat{E}_0 = \frac{\lambda^2 u_0}{4\pi}\frac{e^2}{mc^2} f_{ge}\frac{(\omega-\omega_F)\omega}{(\omega-\omega_F)^2+\Gamma^2}, \tag{6}$$

and

$$\delta\hat{H} = \frac{\delta\hat{E}_0}{g_J\mu_0}\left[\frac{11-4J(J+1)}{4}\right]\left[\frac{\hat{e}\times\hat{e}^*}{i}\right]. \tag{7}$$

Here $\hat{e}$ is the normalized complex polarization vector of the incident light.

The effective magnetic field $\delta\vec{\mathrm{H}}$ [Eq. (7)] will cause the atoms of an oriented vapor to precess, thus changing the angular momentum of the vapor. Since no permanent absorption of off-resonant light can occur, it is interesting to ask how angular momentum is conserved. This aspect of the interaction of off-resonant light with free atoms can be understood with reference to Fig. 1. A beam of σ_+ light of cross sectional area A passes through a thickness l of alkali atoms which have been optically pumped so that their spins lie in the direction of the y axis; i.e., $\langle\vec{\mathrm{F}}\rangle = \langle F_y\rangle\hat{\imath}_y$. For simplicity we shall assume that all of the atoms are in one hfs level, so that the subscript F may be dropped. Because of the presence of the σ_+ light, the atoms within the beam experience a small magnetic field $\delta\vec{\mathrm{H}}$ along the z axis, which rotates the atoms by an angle $\omega_L dt = (1/\hbar)(\langle\mu_y\rangle/\langle F_y\rangle)|\delta H|dt$ around the z axis during the time interval dt. The net change in the angular momentum of the NAl atoms illuminated by the beam is

$$|dL| = NAl\langle F_y\rangle\hbar\omega_L dt = NAl\langle\mu_y\rangle|dH|dt. \tag{8}$$

Now consider what happens to the light beam. Equation (1) indicates that the alkali vapor acts like a doubly refracting crystal when $\langle\vec{\mu}\rangle \neq 0$. There are two wave front surfaces, one for σ_+ and one for σ_- light emanating from a common source point. The surfaces are congruent ellipsoids of revolution with the source point at one focus for σ_+ light and at the other focus for σ_- light. The major axes of the ellipsoids are parallel to the direction of $\langle\vec{\mu}\rangle$. In Fig. 1 Huygens' construction has been used to show that a beam of σ_+ light propagates obliquely through the oriented alkali vapor.[7] Upon emerging from the vapor the beam has been

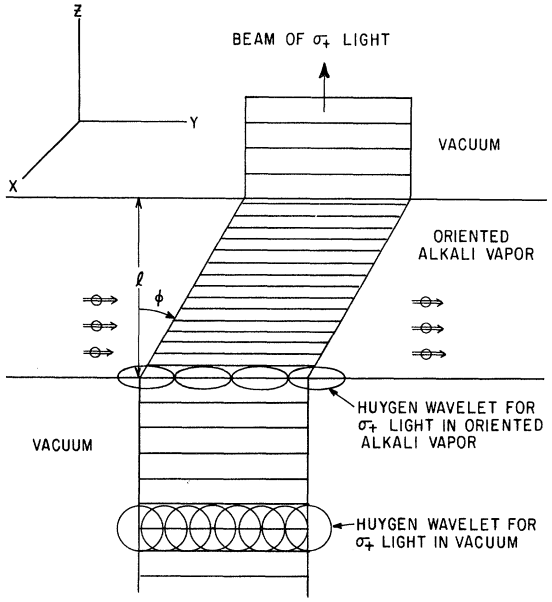

FIG. 1. Light propagation through an oriented alkali vapor, illustrating conservation of angular momentum.

displaced by a distance $l\tan\varphi \approx l\varphi$ parallel to itself along the y axis. From Eqs. (3) and (7), one can easily show that $\varphi = (N|\delta H|/u_0)\langle\mu_y\rangle$. During the time interval dt, $u_0 A\,cdt/h\nu$ photons pass through the vapor, and each photon's angular momentum is increased by an amount $(h/\lambda)l\tan\varphi$, so that the total increase in the angular momentum of the light is

$$|dL'| = \frac{u_0 A\,cdt}{h\nu}\frac{h}{\lambda}l\tan\varphi = NAl\langle\mu_y\rangle|\delta H|dt. \quad (9)$$

The magnitudes of the angular momentum increments in Eqs. (8) and (9) are equal, and it can be shown that the directions are opposite, so that the angular momentum gained by atoms is just compensated for by the angular momentum lost by the light beam.

Off-resonant light can also impart linear momentum to the center-of-mass motion of the illuminated atoms. However, since the linear momentum of the center of mass is not an interesting quantum number for atomic vapors, we will not discuss this aspect of the interaction of off-resonant light with atoms.

A near coincidence[8] between the $5^2S_{1/2}$-$6^2P_{1/2}$ 4215-Å second resonance line of rubidium and the 4215-Å $5^2S_{1/2}$-$5^2P_{1/2}$ first resonance line in Sr II has been exploited to demonstrate the use of off-resonant light to detect the magnetization of an optically pumped alkali vapor. The experimental arrangement (Fig. 2) is very similar to that of the original crossed-beam experiment suggested by Dehmelt.[9] Linearly polarized Sr II light from a microwave-excited flow lamp passes through an optically pumped Rb vapor contained in a 300-ml Paraflint-coated spherical flask. Interference filters are used to isolate the appropriate optical lines. A transverse oscillating magnetization is induced in the Rb vapor with an rf magnetic field at the Zeeman frequency. The vapor then behaves like an optically active crystal which rotates the plane-polarized light either to the right or to the left depending on the direction of the magnetization. After traversing the Rb vapor, the probing light passes through a linear polarizer whose axis is rotated by 45° to the initial polarization axis of the probing beam. The light transmitted by this polarizer is intensity modulated at the Zeeman frequency. The modulated light is detected by a photomultiplier tube and associated electronics. A typical Zeeman resonance detected with Sr II probing light is shown in Fig. 3. The bandwidth of the detection system was 1 cps.

Zeeman resonances in both Rb^{85} and Rb^{87} were observed in this way. The Rb^{85} signals were about twice as large as the Rb^{87} resonance; this is to be expected since one of the Rb^{85} hfs components is very close to the Sr II line.[8]

The Sr II light was filtered with a heated natural-rubidium filter cell in order to absorb any of the Sr II lamp profile which coincided with the Rb absorption profile. The Zeeman resonances in the transmitted rubidium pumping light were monitored with a silicon photodiode. No change in the amplitude of these resonances was observed as the Sr II probing light was turned on and off. Furthermore, the two linear polarizers were removed from the path of the probing beam, and, just as in the original crossed-beam experiments,[9] circularly

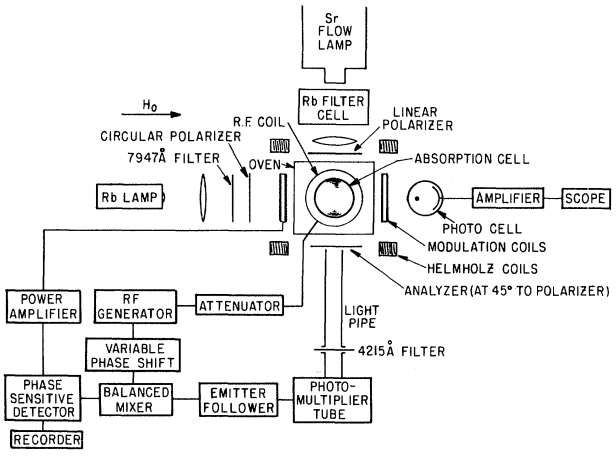

FIG. 2. A schematic diagram of the apparatus.

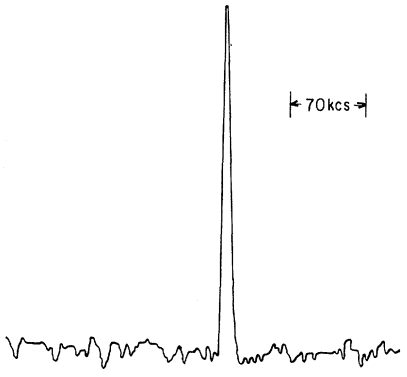

FIG. 3. A recorder trace of the phase-sensitive detector output as the magnetic field is swept through the resonance. The phase-sensitive detector time constant was 1 sec.

polarized Sr II light was used as a probe. No Zeeman modulation could be detected on the probing beam, even when the natural-Rb filter cell was removed from the path of the Sr II light. We conclude that no detectable absorption of the probing beam occurred, and that the populations of the rubidium ground-state sublevels were not affected by the Sr II light.

The fundamental source of noise in experiments of this type is the shot noise due to the light of the probing beam. One can show that the power signal-to-(shot) noise ratio is

$$\frac{S}{N}=\frac{\eta\Phi_0}{\Delta f}\left[\frac{2\pi l}{\lambda}|\delta n_1|\right]^2. \qquad (10)$$

Here η is the quantum efficiency of the photodetector, Φ_0 is the light flux reaching the photodetector in photons/sec, Δf is the effective bandwidth of the detection system, λ is the wavelength of the probing light, l is the path length traversed by the probing beam through the vapor, and $|\delta n_1|$ is the amplitude of the oscillating index of refraction induced by the oscillating transverse magnetization $\langle\mu\rangle_F$ [Eq. (3)]. The signal-to-noise ratios measured in this experiment were in good agreement with the value predicted in Eq. (10).

Experiments using off-resonant light have several advantages over experiments using resonant light. There is no depumping of the optically pumped samples, and conversely, there is no attenuation of the probing beam. Optically thick samples can be studied as easily as optically thin samples. The probing lamp profile and the absorption profile of the pumped vapor are of minor importance when these profiles are well resolved. Broad, self-reversed lines from resonance lamps or narrow laser lines with single or multimode operation are equally suitable as probing light soucrces. The close connection between the index of refraction n and the light shift δE induced by the probing beam [Eq. (4)] can be utilized to deduce atomic parameters in situations where both δE and $n-1$ can be measured. Alternatively, δE can be deduced from measurements of δn, and for practical work with optically pumped frequency standards it may be considerably more convenient to measure δn since no microwave equipment is necessary.

The authors gratefully acknowledge the enthusiastic help of Mr. Richard Brignoli in the experimental work. They would like to thank Professor E. B. Saloman for his help in the early stages of the work. They are also grateful to Professor A. Lurio, Professor W. W. Smith, and Professor L. R. Wilcox for helpful discussions. Finally they wish to thank Professor Robert Novick for his encouragement and advice, and for allowing them to use the facilities of the Columbia Radiation Laboratory.

---

*This work was supported wholly by the Joint Services Electronics Program under contract No. DA-28-043 AMC-00099(E).

[1]A. Gozzini, Compt. Rend. 225, 1905 (1962); A. Kastler, Compt. Rend. 232, 953 (1951); W. Opechowski, Rev. Mod. Phys. 25, 264 (1953); and J. Manuel and C. Cohen-Tannoudji, Compt. Rend. 257, 413 (1963).

[2]The fortuitous near coincidence of the wavelength of the strong 4215-Å Sr II resonance line with that of the 4215-Å second resonance line of rubidium was pointed out to the authors by Professor Robert Novick.

[3]Hfs coherence is assumed to be absent in Eq. (1). If such coherence is present, additional terms appear which contain the components of $\langle\mu\rangle$ which oscillate at the hyperfine frequency.

[4]Compare M. A. Bouchiat, J. Phys. Radium 26, 415 (1965), for a discussion of the observables which can be measured by monitoring the absorbed light.

[5]J. P. Barrat and C. Cohen-Tannoudji, J. Phys. Radium 22, 329 (1961); 22, 443 (1961).

[6]In general, the light-shift operator also contains a term which looks like the interaction of the electric quadrupole moment of the atom with an effective electric-field gradient, i.e., the energy shift can be represented in terms of a scalar, vector, and tensor polarizability. However, no significant tensor polarizability is present in the ground state of the alkali atoms because the atomic spin is $J=\frac{1}{2}$ and hfs effects are too small to give rise to a significant tensor polarizability.

[7]For instance, see F. A. Jenkins and H. E. White, Fundamentals of Physical Optics (McGraw-Hill Book Company, Inc., New York, 1937), p. 354. There are distinct wave-front surfaces for σ_+ and σ_- light as long as the index of refraction does not differ significantly from unity. It is interesting to notice that the dielectric tensor ϵ is not symmetric for oriented alkali vapors.

[8]J. Winocur and R. V. Pyle, J. Appl. Phys. 36, 2740 (1965).

[9]H. G. Dehmelt, Phys. Rev. 105, 1924 (1957); W. E. Bell and A. L. Bloom, Phys. Rev. 107, 1559 (1957).