MICROWAVE LIGHT MODULATION BY AN OPTICALLY PUMPED Rb^{87} VAPOR*

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We have observed appreciable light modulation at the 6.835-GHz hfs transition frequency of optically pumped Rb^{87} vapor. The frequency separation between the levels involved in the transition exceeded the total spectral width of the initial lamp profile. These observations indicate that persistent doubts as to the feasibility of a transmission-light-modulation experiment under such conditions are groundless.

We have observed intensity modulation of light which has passed through a vapor of Rb^{87} atoms, coherently oscillating on the 0-0 ground-state hyperfine structure transition. The 1.5-GHz spectral widths of our lamps and the 0.01-GHz decay rates of the excited states were considerably smaller than the 6.835-GHz modulation frequency (see Fig. 1). Although Firester and Carver[1] have recently observed similar light modulation at 0.458 GHz in K^{39}, widespread uncertainty has remained as to whether light modulation could be observed in Rb^{87} or in Cs^{133}, where the hfs frequencies may not only exceed the decay rate of the excited state but also the width of typical lamp spectral profiles.[2,3] Because of the controversy which has surrounded the question, this Letter presents a brief discussion of the theory of transmission light modulation and a description of our observations of the modulation in Rb^{87}.

In early theories of optical pumping,[3-5] a rather one-sided emphasis was placed on the evolution of the atomic system as a result of its interaction with light. The equally important problem of the propagation of a light beam through an optically pumped vapor was not examined as critically, so that it was difficult to understand the physical mechanism of transmission light modulation. One can develop either a semiclassical or a fully quantum mechanical theory of the propagation of light, but since both give the same result for the forward-scattered light, we shall discuss the light modulation in terms of semiclassical radiation theory. More details of the theory may be found in a recent paper by Happer and Mathur.[6]

Let us represent the electric field, $\vec{E}$, of a quasimonochromatic light wave by

$$\vec{E} = \vec{\mathcal{E}} \exp i(\vec{k}\cdot\vec{r} - \omega t) + \text{c.c.} \qquad (1)$$

Here c.c. denotes "complex conjugate." The amplitude $\vec{\mathcal{E}}$ will vary very little over the distance of an optical wavelength or during the period of an optical oscillation. The electric field will induce a dielectric polarization, $\vec{P}$, in the medium:

$$\vec{P} = \vec{\mathcal{P}} \exp i(\vec{k}\cdot\vec{r} - \omega t) + \text{c.c.} \qquad (2)$$

The polarizability amplitude, $\vec{\mathcal{P}}$, is proportional to the electric field amplitude, $\vec{\mathcal{E}}$:

$$\vec{\mathcal{P}} = \langle\chi\rangle\vec{\mathcal{E}}. \qquad (3)$$

Here the susceptibility dyadic $\langle\chi\rangle$ is the expectation value of a susceptibility dyadic operator, χ.[6] If hfs coherence has been produced by a microwave field of frequency ω_m, the hfs component of the oscillating magnetic dipole moment of the

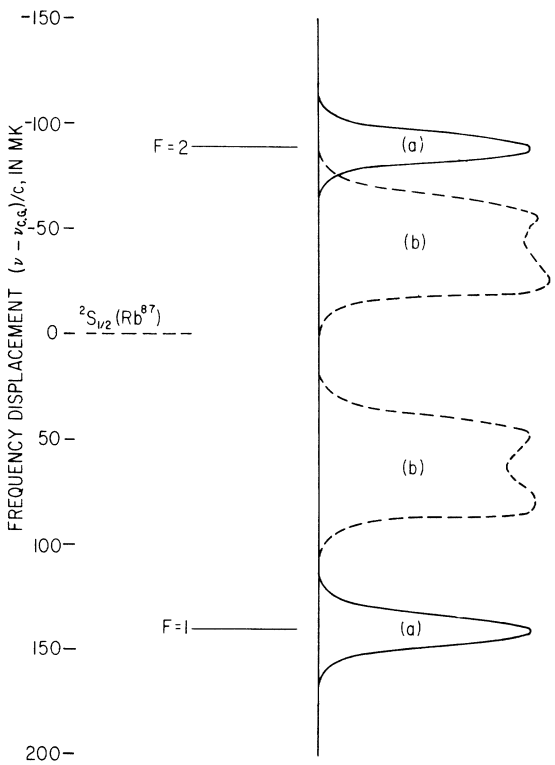

FIG. 1. (a) The optical absorption lines of Rb^{87} and (b) a typical spectral profile of a Rb^{85} lamp. More detailed experimental studies of spectral profiles in the rubidium isotopes can be found in H. M. Gibbs and R. J. Hull, Phys. Rev. 153, 132 (1967).

atoms will be of the form

$$\langle\mu\rangle_{\text{hfs}} = \vec{\mu}_1 \exp(-i\omega_m t) + \text{c.c.} \quad (4)$$

The associated susceptibility will be

$$\langle\chi\rangle_{\text{hfs}} = f_a \exp(-i\omega_m t)\vec{\mu}_1 \times + f_b \exp(i\omega_m t)\vec{\mu}_1{}^* \times. \quad (5)$$

Suppose that an initially monochromatic light beam propagates along the z axis of a coordinate system and passes through a slab of vapor which fills the space $0 \leq z \leq l$. Assume that the moments $\vec{\mu}_1$ of the vapor are all equal and parallel to the z axis, and assume that the atomic density is low enough that the vapor slab is optically thin. After emerging from the vapor, the light beam will be of the form

$$\vec{E} = \vec{\mathcal{E}}_0 \exp[i(2\pi lk\langle\chi\rangle_{\text{sc}})] \exp[i(kz-\omega t)] + 4\pi i(\lambda_m/\lambda)\sin(\pi l/\lambda_m)\{f_a(\omega)\vec{\mu}_1 \times \vec{\mathcal{E}}_0 \exp\{i[(k+k_m)z-(\omega+\omega_m)t]\} + f_b(\omega)\vec{\mu}_1{}^* \times \vec{\mathcal{E}}_0 \exp\{i[(k-k_m)z-(\omega-\omega_m)t]\}\}. \quad (6)$$

The first term in (6) describes the carrier beam, which has been attenuated and phase shifted by an amount $\exp(2\pi ikl\langle\chi\rangle_{\text{sc}})$. Here $\langle\chi\rangle_{\text{sc}}$ is the scalar susceptibility of the vapor. The remaining terms represent a sideband at the frequency $\omega+\omega_m$ and a sideband at the frequency $\omega-\omega_m$. The sideband amplitudes need not be the same since $f_a(\omega) \neq f_b(\omega)$. Because of the factor $\sin(\pi l/\lambda_m)$, the sidebands will be of maximum amplitude when the thickness l of the vapor is $\frac{1}{2}\lambda_m$, one-half a hfs frequency wavelength. The sinusoidal dependence of the sideband amplitude on the length of the active region is a phenomenon well known in parametric transmission lines.[7]

From (6) one can see that the light intensity $\vec{E}\cdot\vec{E}$ will be modulated at the hfs frequency ω_m. The amplitude of the modulation will be proportional to $\vec{\mu}_1\cdot(\vec{\mathcal{E}}_0\times\vec{\mathcal{E}}_0{}^*)$. Since $(\vec{\mathcal{E}}_0\times\vec{\mathcal{E}}_0{}^*)$ is proportional to the average photon spin, intensity modulation can occur only if the light has some degree of circular polarization. The intensity of initially linearly polarized light is not modulated because the sidebands are polarized at right angles to the carrier polarization, and interference cannot occur. Unpolarized light may be thought of as independent right and left circularly polarized components of equal intensity. These beams will be modulated with equal intensity but 180 degrees out of phase. Hence, no modulation will be detected with unpolarized light unless a circular analyzer is inserted before the photodetector to select one sense of circular polarization.

Light with a broad spectral profile may be thought of as an ensemble of many monochromatic waves of random phases. The modulated light intensity is then proportional to the convolution of the spectral profile of the light with appropriate combinations of the frequency response functions $f(\omega)$. Following the preceding line of reasoning, we estimated that under our experimental conditions the index of modulation should be about 10^{-3}. Experimentally observed indices of modulation were of this order of magnitude.

A schematic diagram of the apparatus is shown in Fig. 2. A glass absorption cell contained a small amount of Rb^{87} and about 10 Torr of neon buffer gas. The cell temperature was maintained at 45°C. The cell, which was about 4 cm in length, was placed in a microwave cavity, tuned to resonant at 6.835 GHz in the TE_{011} mode. A beam of Rb^{85} resonance radiation from a Varian λ49-609 spectral lamp passed through the vapor and pumped the Rb^{87} atoms into the lower hfs level (see Fig. 1). A small static magnetic field along the axis of the cavity removed the degeneracy of the hfs transitions. The $F=2, m=0$ and the $F=1, m=0$ sublevels of the Rb^{87} ground state were coupled by the microwave field of the cavi-

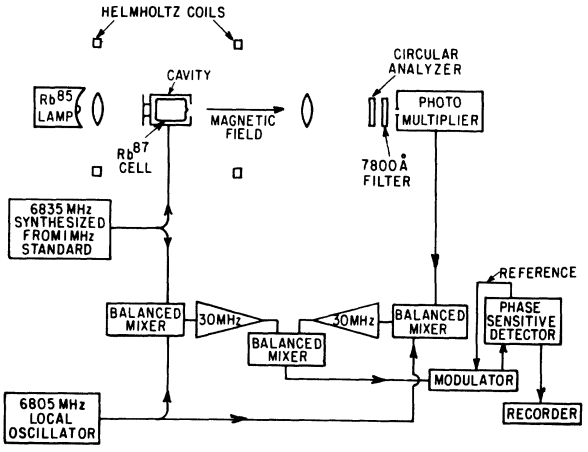

FIG. 2. The experimental apparatus.

ty. The microwaves were generated by harmonic multiplication of the 999 225-Hz signal from a crystal-controlled oscillator. The microwave frequency could be tuned through the hfs resonance frequency by varying the capacitative loading of the crystal. After emerging from the cavity, the light beam passed through a circular analyzer and a D-line interference filter. The light intensity was detected with a crossed-field photomultiplier tube.[8] The tube had a current gain on the order of 50 dB and a bandwidth on the order of 10 GHz. The useful active area of the photocathode was on the order of 0.1 cm^2, and the quantum efficiency was about 10^{-4} at 7800 Å.

The 6.835-GHz signal from the photomultiplier tube was mixed with a 6.805-GHz signal from a klystron local oscillator. The 30-MHz difference frequency was amplified and mixed with a 30-MHz reference signal, which was the difference frequency between the 6.805-GHz local oscillator frequency and the 6.835-GHz cavity excitation frequency. The dc output of the 30-MHz balanced mixer could be observed directly on a millivoltmeter, or it could be further narrow-banded with a commercial lock-in amplifier. The gain of the photomultiplier was adjusted until the shot noise from the light beam was approximately equal to the thermal and mixer noise of the detection system.

A typical signal for 7800-Å D_2 light is shown in Fig. 3. Similar signals were observed with 7947-Å D_1 light. It was also possible to observe light modulation with an isotope-filtered[9] Rb^{87} lamp, although the signal-to-noise ratios were poorer, presumably because of the lower light intensity. The shape of the resonance curve could be charged from dispersion to absorption by adjusting the phase of the reference signal. The index of modulation was measured experimentally to be between 10^{-3} and 10^{-4}. The signal in Fig. 3(a) was recorded with the circular analyzer in place, and the signal in Fig. 3(b) was recorded with the circular analyzer removed, i.e., for unpolarized light. Unpolarized light, in agreement with theory, was not intensity modulated.

In summary, the observation of Rb^{87} hfs light modulation with a Rb^{85} lamp demonstrates that the initial spectral profile of the lamp need not overlap both hfs components of the atomic absorption line, as was sometimes thought to be necessary. The observed index of modulation and the observed dependence of the modulation on the polarization of the light are in agreement with theory. It may be possible to detect the sidebands directly with a Fabry-Perot interferometer, and we intend to begin such studies in the near future. Much better signal-to-noise ratios can be anticipated for photdetectors with larger active areas and higher quantum efficiencies. No particular efforts have yet been made to make the active length of the vapor one-half a microwave wavelength, although the inhomogeneity of the hfs magnetization within the cavity probably helps to satisfy this criterion. From a practical viewpoint, one might be able to use the light modulation to construct microwave spin generators. The modulation could also be used to monitor the phase of the atomic magnetization with respect to the phase of the microwave field in a Rb^{87} maser so that cavity pulling could be eliminated.[10]

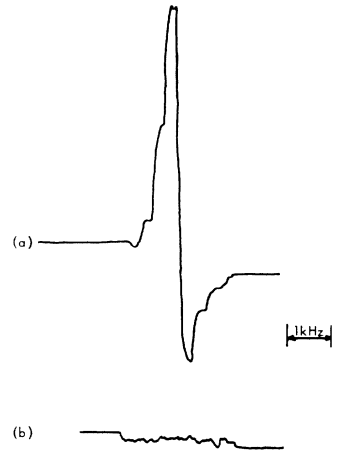

FIG. 3. Typical 6.835-GHz light-modulation signals for 7800-Å D_2 Rb^{85} light; (a) with a circular analyzer; (b) without a circular analyzer. The vertical scale is the output of the 30-MHz balanced mixer in arbitrary units. The detection-system bandwidth was 1 Hz.

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ELECTRON PARAMAGNETIC RESONANCE ABSORPTION IN OXYGEN WITH THE HCN LASER

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Paramagnetic resonance absorption between the $N=3, J=4, M=-4$ and $N=5, J=5, M=-4$ levels of ground state O_2 was observed at the HCN laser frequency of 890 759 ± 3 MHz in a magnetic field of 16 418 ± 1 G. This is the first observation of absorption in a gaseous sample in which laser electron-paramagnetic-resonance techniques were used.

In an extension of previous work,[1-4] a calculation of the effect of magnetic fields on molecular oxygen has shown that the difference between the $N=3, J=4, M=-4$ and $N=5, J=5, M=-4$ levels of ground-state molecular oxygen equals the HCN laser frequency of 890 GHz in a magnetic field of about 16.4 kG as is shown in Fig. 1. This note reports the first paramagnetic resonance absorption between these levels and the first laser electron paramagnetic resonance (LEPR) absorption[5] in a gaseous sample. The observations were made utilizing a specially constructed LEPR spectrometer.

An HCN laser 7.5 cm in diameter and 2 m long was tuned to the 337-μ line. The center frequency of this line was measured to be 890 759.4 ± 0.1 MHz[6] in agreement with Hocker et al.[7] Significant improvement in the short term (0.1-sec) stability of the laser was obtained by using a current-regulated power supply and 120-mA neon-sign hot hollow cathodes operating at 600 mA. Laser-frequency drift during the 5- or 10-min scan was less than 3 MHz at maximum power output.

The laser beam was focused into a Fabry-Perot interferometer consisting of a 10-cm long cylinder 1.7 cm in diameter with one flat mirror and a 15-cm focal length mirror. Each mirror had a 0.75-mm hole in order to operate the Fabry-Perot interferometer as a transmission cavity whose Q was approximately 5×10^4. The interferometer was centered in the 2-in. gap of a 15-in. magnet with 5-in., tapered, Rose-shimmed pole faces. Radiation transmitted through the cell was directed by a $\frac{1}{4}$-in. copper pipe to a Golay cell placed outside the magnetic field 75 cm from the center of the magnet. The output of the Golay cell was connected to a 13.5-Hz phase detector referenced to the modulation of the magnetic field.

Figure 2 shows the derivative of the absorption observed at a pressure of 7.5 Torr with approximately a 10-G modulation field. The maximum absorption in Fig. 2 corresponded to about one part in 10^4 of the radiation transmitted through the Fabry-Perot cavity; this high sensitivity was largely due to the extreme stability of the laser discharge. Linewidths from 3 to 25 G were observed at pressures between 2 and 19 Torr, yielding values of the linewidth parameter of about 1.8 MHz/Torr in agreement with Zimmerer and Mizushima.[2] An asymmetrical line shape was observed even though linewidths were increased by an order of magnitude by pressure broadening and the modulation reduced to about one-fourth the linewidth.

Since the ground state of the oxygen molecule is $^3\Sigma_g^-$, the rotational quantum number N of the