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PARAMETRIC FREQUENCY CONVERSION OF RESONANCE RADIATION IN OPTICALLY PUMPED RUBIDIUM-87 VAPOR*

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Parametric frequency conversion of resonance radiation in an optically pumped atomic vapor has been observed for the first time. Conversion efficiencies of 10^{-3} have already been observed in Rb^{87} vapor, and considerably higher conversion efficiencies should be attainable.

In this paper we report the first direct observation of sidebands produced on a beam of rubidium resonance radiation by rubidium-87 atoms, coherently oscillating at the 6.835-GHz ground-state hyperfine frequency. These experiments are of a completely new type for optical pumping work, since they demonstrate that radical changes can occur in the spectral profile of a probing light beam. In addition, such experiments are of considerable interest in the study of parametric frequency conversion, since the properties of an optically pumped vapor are thoroughly understood on an atomic scale, and detailed comparisons of theory and experiment are possible.

Following a suggestion by Dehmelt,[1] Bell and Bloom[2] first demonstrated that precessing, optically pumped atoms could be used to modulate a light beam. In the early "crossed-beam" experiments, a resonant rf magnetic field forced the atoms to precess at the Larmor frequency around a small static magnetic field. The experiments were limited to low precession frequencies ($\lesssim$10 MHz) by a number of technical difficulties. However, optically pumped vapors can provide much higher modulation frequencies if the electronic spin $\vec{J}$ and the nuclear spin $\vec{I}$ are forced to tumble around each other in phase. Recently, Firester and Carver[3] were able to modulate potassium resonance radiation at the 458-MHz hyperfine frequency of K^{39}, and Mathur et al.[4] were able to modulate rubidium resonance radiation at the 6.835-GHz hyperfine frequency of Rb^{87}.

In the case of Rb^{87}, the 6.835-GHz hyperfine frequency exceeds the spectral width ($\approx$1500 MHz) of a typical lamp line, and the light modulation should give rise to well-resolved optical sidebands. Unfortunately, the low index of modulation[4] ($m \approx 3\times10^{-4}$) of the first hyperfine light modulation experiments in Rb^{87} precluded a direct observation of the sideband intensity, since the sideband power would have been about 10^7 times weaker than the carrier power. We have now increased the efficiency for conversion from carrier to sideband power to better than one part in 10^3, which corresponds to modulation indices of 3% or greater. The sidebands can be clearly seen with a Fabry-Perot interferometer, and we report some of our preliminary observations in this Letter.

Hyperfine light-modulation experiments are best understood as a parametric frequency conversion process. A detailed theoretical analysis of these experiments has been carried out,[5] but we will only discuss the main qualitative aspects of the theory here. If the index of refraction of the vapor is modulated at a frequency Ω, an optical carrier wave of frequency ω_0 can be coupled to upper and lower sidebands of frequencies

$$\omega_{\pm 1} = \omega_0 \pm \Omega. \tag{1}$$

In our experiment the index of refraction of the vapor was driven by the microwave fields of a waveguide. Consequently, the index of refraction of the vapor was spatially modulated with a propagation vector of magnitude

$$K = 2\pi/\Lambda, \tag{2}$$

where Λ is the guide wavelength. Then, for efficient conversion from carrier to sideband power, one must satisfy the phase-matching condition

$$\vec{k}n(\omega_0) \pm \vec{K} = \vec{k}_{\pm 1}n(\omega_{\pm 1}). \tag{3}$$

Here $\vec{k}$, $\vec{k}_{+1}$, and $\vec{k}_{-1}$ are the free-space wave vectors of the carrier and of the upper and lower sidebands. The static component of the index of refraction at the frequency ω is denoted by $n(\omega)$.

Because of the gyrotropic nature of the modulated index of refraction, if the carrier is linearly polarized, the sidebands will be linearly polarized at right angles to the carrier.[4,5] The orthogonality of the carrier and sideband polarization affords a convenient way to distinguish between the carrier and the sideband.

In order to generate modulated components of the index of refraction, one can induce the field-independent hyperfine transition $F=1, m=0 \rightarrow F=2, m=0$ with a microwave field. Then modulated components $n_{\pm 1}$ of the index of refraction are produced, and these are proportional to the hyperfine coherences $\rho_{\pm 1}$ of the vapor, i.e.,

$$n_{\pm 1} \propto \rho_{\pm 1}, \tag{4}$$

where

$$\rho_{+1} = \rho_{-1}{}^* = \langle F=1, m=0|\rho|F=2, m=0\rangle, \tag{5}$$

and where ρ is the density matrix for the atomic ground state. Upper and lower sidebands are produced by n_{+1} and n_{-1}, respectively.

The coherence (5) can be maximized by a certain optimum value of the microwave field. The coherence approaches zero at both low and high microwave power; in the latter case, saturation of the transition occurs. As long as the coherence does not become too large, large values of the coherence should give rise to large conversion efficiencies.

The experimental apparatus for our work is shown in Fig. 1. A cylindrical microwave cavity was constructed from a stack of brass plates, spaced 1 cm apart. A hole 6.5 cm in diameter was cut in the center of each plate. The cavity was terminated by plates with a smaller-diameter hole (~3 cm). The total length of the cavity was 45 cm. This cavity had an unloaded Q of about 10^4 for the TE_{01n} modes. A cylindrical quartz cell, filled with a small amount of Rb^{87} metal and nitrogen buffer gas at a pressure of about 12 Torr, was placed in the cavity. The cell temperature was maintained at 65°C. The dimensions of the quartz tube were chosen so that the dielectrically loaded cavity had the proper guide wavelength to satisfy the phase-matching condition (3). The Rb^{87} cell was pumped by resonance radiation from a long Rb^{85} lamp, which was parallel to the cavity axis. An elliptical reflector focused the Rb^{85} light from the long lamp onto the Rb^{87} absorption cell. The Rb^{87} atoms were pumped from the $F=2$ hyperfine level to the $F=1$ hyperfine level of the ground state. Micro-

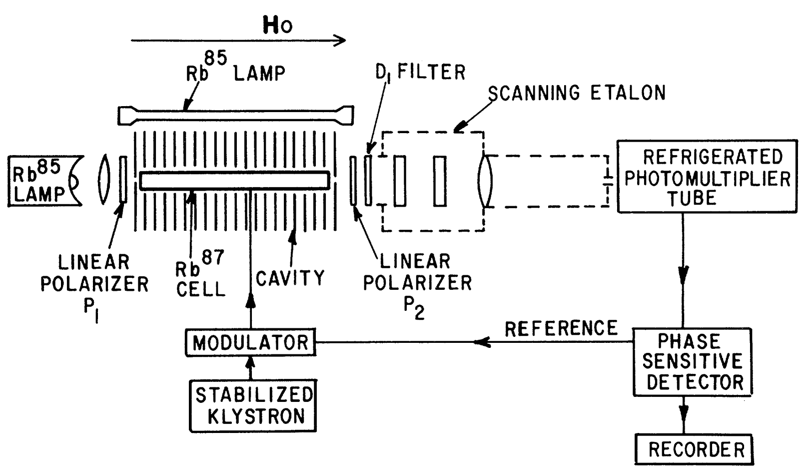

FIG. 1. Schematic diagram of the apparatus used to generate and to observe the sidebands.

waves from a stabilized klystron oscillator were used to induce the 6.835-GHz field-independent transition of the Rb^{87} atoms and to produce the microwave coherence in the vapor. The microwaves were square-wave modulated on and off to enable the accompanying changes in the probing beam intensity to be observed by phase-sensitive detection.

Optical carrier waves were provided by a second Rb^{85} lamp, situated at one end of the cavity. The carrier beam was linearly polarized by the polarizer P_1. After emerging from the microwave cavity, the probing beam passed through a linear polarizer P_2, which was orthogonal to the input polarizer P_1. The polarizer P_2 served to remove the carrier waves. Finally, the D_1 component of the probing beam was frequency analyzed by a scanning Fabry-Perot interferometer. The free spectral range of the interferometer was 1000 mK and the finesse was about 35. The intensity of the central fringe of the interferometer was recorded with a cooled EMI type 9558QA photomultiplier tube, and the photomultiplier signal was detected in synchronism with the 13-Hz microwave modulation frequency.

Some typical observations are reproduced in Fig. 2. Figure 2(a) gives the spectral profile obtained with the apparatus of Fig. 1. The signals of Fig. 2(a) were obtained with a 30-sec integration time and a 20-min scan of the interferometer. Some of the width of the peaks is due to slight drifts of the interferometer during the sweep.

Figure 2(b) shows the spectral profile of the Rb^{85} probing lamp. For comparison, a Rb^{87} spectral profile is shown in Fig. 2(c). Two peaks are visible in Fig. 2(a). One peak is displaced to the high-frequency side of the $F = 3$ component of the Rb^{85} profile by ≈225 mK, which is the microwave frequency, while the other peak is displaced by ≈225 mK to the low-frequency side of the $F = 2$ component of the Rb^{85} profile. The spectral components of Fig. 2(a) do not coincide with the spectral components of either a Rb^{85} lamp profile [Fig. 2(b)] or a Rb^{87} lamp profile [Fig. 2(c)]. The peaks in Fig. 2(a) were not due to fluorescently scattered light from the long, Rb^{85} pumping lamp since they disappeared when the probing lamp was removed. We conclude that the peaks of Fig. 2(a) are upper and lower sidebands, which were generated by the microwave coherence of the vapor. The sideband power is about 10^3 times weaker than the carrier power. This corresponds to an index of modulation of about 3%.

In order to support the interpretation of the peaks in Fig. 2(a) as sidebands, we removed the Fabry-Perot interferometer and recorded the integrated intensity of sidebands as a function of microwave power while the microwave frequency was kept on the center of the resonance [see Fig. 3(a)]. The sidebands attained a maximum intensity at a field strength of about 1.5×10^{-4} G. The sidebands decreased in amplitude for greater or lesser field strengths. Thus, the sidebands of Fig. 2(a) have the same qualitative dependence on microwave power as does the microwave coherence of the vapor. Since the saturation of the microwave transition increases monotonically with

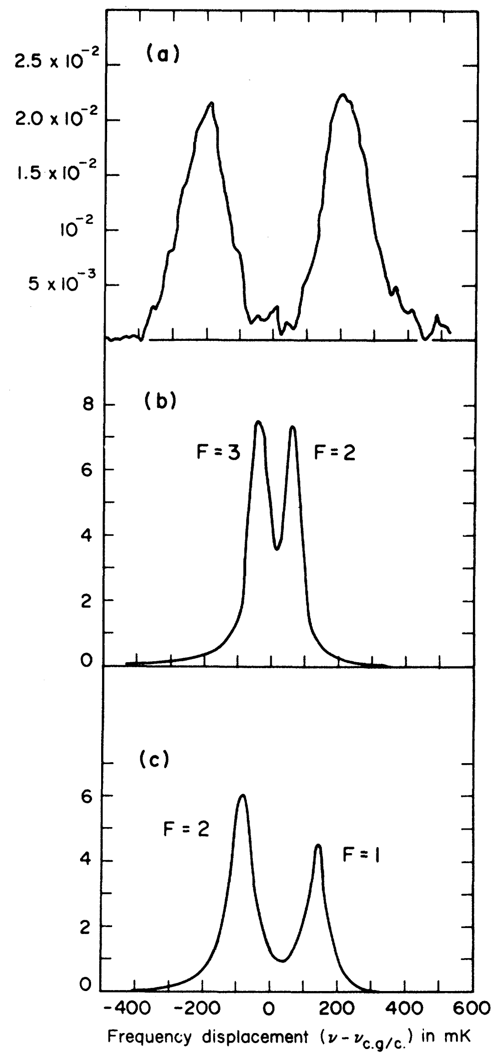

FIG. 2. (a) Spectral profile of the signal observed through the etalon with polarizers P_1 and P_2 crossed. (b) Spectral profile of the D_1-line of a Rb^{85} lamp before passing through the vapor. (c) Spectral profile of the D_1-line of a Rb^{87} lamp.

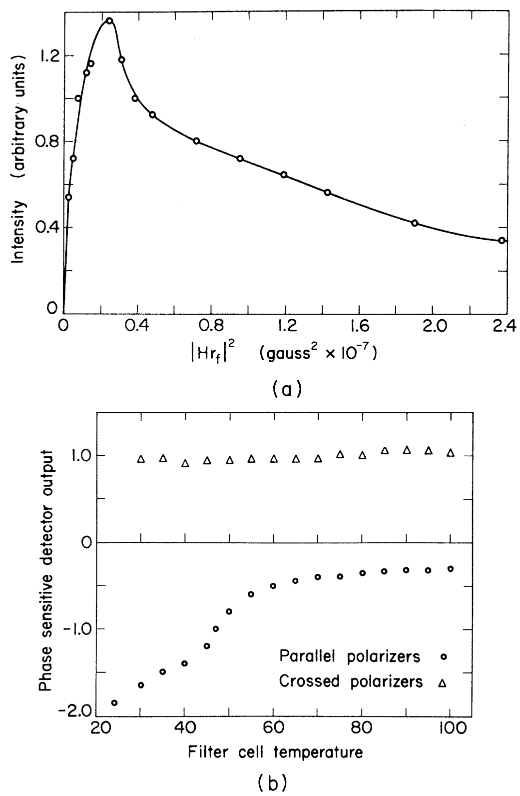

FIG. 3. (a) Total intensity observed with the phototube directly behind the polarizer P_2 and the D_1 filter versus the magnitude squared of the microwave field in the cavity. The polarizers P_1 and P_2 were crossed. (b) Total intensity observed through a Rb^{85} filter cell versus the temperature of the filter cell with the polarizers P_1 and P_2 crossed (sideband intensity) and with P_1 and P_2 parallel (carrier intensity).

microwave power, the signals of Fig. 2(a) are not proportional to the saturation of the transition.

As a final check of the nature of the peaks in Fig. 2(a), we replaced the Fabry-Perot interferometer with a Rb^{85} filter cell,[6] and we recorded the intensity of the probing beam as a function of the filter-cell temperature [see Fig. 3(b)]. There was no noticeable attenuation of the probing beam when the polarizer P_2 was orthogonal to the polarizer P_1, i.e., when only the sidebands passed through the filter cell. The intensity of the probing beam decreased rapidly as a function of filter-cell temperature when the polarizer P_2 was parallel to P_1, i.e., when only the carrier reached the filter cell. This experiment confirms the fact that the peaks of Fig. 2(c) lie well outside the absorption profile of Rb^{85} vapor.

This work is continuing, and we believe that significantly larger conversion efficiencies can be attained with better phase matching and better selection of buffer gas pressure. Similar experiments could be performed with other alkali vapors such as Rb^{85} and Cs^{133}.

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