# Optical-pumping noise in laser-pumped, all-optical microwave frequency references

### J. Kitching,\* H. G. Robinson, and L. Hollberg

Time and Frequency Division, The National Institute of Standards and Technology, M.S. 847.10, 325 Broadway, Boulder, Colorado 80303

#### S. Knappe and R. Wynands

Institut für Angewandte Physik, Universität Bonn, Wegelerstraße 8, D-53115 Bonn, Germany

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We demonstrate that optical pumping plays a significant role in determining the noise in certain types of laserpumped vapor-cell microwave frequency standards by changing the way in which the laser's FM noise is converted to AM noise by the optical-absorption profile. When this FM–AM conversion is the dominant noise source, the noise spectrum of the transmitted intensity can be dramatically altered by the optical-pumping process. FM noise at Fourier frequencies larger than the optical-pumping time is converted to AM noise differently from noise at lower Fourier frequencies. This effect can modify the optimum design of vapor-cell frequency references and adds an additional FM–AM-related noise source that cannot be eliminated with laser tuning. © 2001 Optical Society of America

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## 1. INTRODUCTION

Optical pumping<sup>1</sup> has long been known to affect the optical-absorption properties of alkali atoms confined in a cell with a buffer gas.<sup>2</sup> Because the alkali atoms' motion is restricted by collisions with buffer-gas atoms, the alkali atoms spend a much longer time interacting with the incident light than they would in a low-pressure vapor. When this optical interaction time becomes comparable with the optical-pumping time between the two hyperfine ground states, the optical pumping can alter the ground-state populations and therefore the absorption spectrum of light by the atoms.

In this paper we present some considerations of hyperfine optical pumping in alkali atoms within the context of laser-pumped microwave frequency references. Optical pumping plays a critical role in all vapor-cell microwave atomic frequency references. In conventional frequency references based on optical-microwave double resonance,<sup>3</sup> for example, the change in ground-state populations induced by this process gives rise to the error signal that is used to lock the external microwave oscillator to the narrow atomic resonance. In a diode-laser-pumped system we have recently found that optical pumping can dramatically affect the way in which frequency modulation on the laser is converted into intensity modulation by the atomic absorption line.<sup>4</sup> Since FM-AM noise conversion can dominate the short-term stability of microwave frequency references, the optical-pumping process can significantly alter the design parameters that optimize the atomic standard. The results presented here appear to be general enough to encompass all laser-pumped frequency references in which the laser's frequency noise contributes significantly to the frequency-reference instability.

Laser-pumped frequency references can be divided into two main types. Conventional frequency references<sup>3</sup> use a microwave field to excite the atomic hyperfine transition. A laser resonant with an optical transition in the atom provides some initial optical pumping to change the populations of the ground-state levels from their equilibrium value. When the microwave frequency coincides with the ground-state hyperfine frequency, the level populations change back toward their equilibrium values, and this change is measured through the resulting change in absorption of the optical field. In all-optical frequency references, 5-8 dark-line or coherent population-trapping resonances 9-11 are used: two laser fields separated in frequency by the ground-state hyperfine frequency excite the microwave transition through an optical multi-photon process. The absorption of those same laser beams is used to determine when the difference frequency between the lasers coincides with the resonance. Although this paper focuses mainly on the all-optical systems, many of the arguments and conclusions may apply equally well to conventional references.

The short-term stability of laser-pumped frequency references is determined by noise from a number of sources: shot noise in the detection process, excess intensity noise on the laser, FM-AM conversion in the vapor, and electronic noise. When lasers with large linewidths (for example, vertical-cavity surface-emitting lasers) are used, the FM-AM conversion noise can dominate and almost exclusively determine the stability (Allan standard deviation) of the frequency reference.

FM-AM conversion in resonant media has been studied previously as a tool to perform broadband, highresolution spectroscopy with a very simple experimental configuration.<sup>12-15</sup> In that context, fluctuations in amplitude of the optical field exiting the cell are related to fluctuations in polarization of the atomic medium and therefore contain information about the spectral structure of the atoms. Complicated structure in the intensity noise spectrum of light propagated through an optically thick atomic medium has been predicted theoretically and observed experimentally.<sup>12–17</sup> More recent studies of FM–AM conversion have focused on nonlinear effects in optically thick media that affect the performance of conventional vapor-cell atomic clocks.<sup>18,19</sup>

We present here a detailed investigation of the FM-AM conversion process combined with optical pumping near a resonant transition in an optically thin vapor of alkali atoms. It is found that the FM-AM conversion at frequencies up to 100 kHz can be well explained with a simple. linear theory that includes the effects of optical pumping. In this theory the FM laser noise is translated into AM noise by the slope of the atomic absorption spectrum. It is found experimentally, however, that when the noise at Fourier frequencies near 500 Hz is measured as a function of optical detuning from the resonance peak, the noise minimum does not exactly coincide with the peak of the dc absorption profile but is instead shifted by several tens of MHz.<sup>20</sup> This shift can be explained<sup>4</sup> by taking into account optical pumping, which causes the atoms to respond to noise at higher frequencies in a manner different from that to the dc optical power. In addition, the optical pumping adds a noise component out of phase with the original frequency noise, in which the component cannot be eliminated with an appropriate choice of laser tuning. A simple theory is presented that includes the optical-pumping process as well as the excited state's hy-This theory explains semiperfine structure. quantitatively the shift observed in the experimental data as well as certain features in the AM noise spectra.

### 2. COMPACT ATOMIC FREQUENCY REFERENCE

The measurements presented in this paper were carried out with a compact frequency reference shown schematically in Fig. 1 and described more thoroughly in previous publications.<sup>4,20,21</sup> This frequency reference uses an all-



Fig. 1. Schematic of the experimental setup. The inset shows the tunings of the relevant laser fields with respect to the atomic energy levels.

optical scheme<sup>5-8,22</sup> to excite the microwave clock transition, in which the laser's injection current is modulated to produce optical sidebands on the laser. Two sidebands couple the two atomic ground states to an excited state in a  $\Lambda$  configuration, creating a narrow, dark-line resonance to which an external microwave oscillator can be locked. Although this system was developed as a compact clock, of primary interest here is the way in which the FM noise is converted into AM noise upon passing through the vapor and the extent to which this determines the stability of the frequency reference.

The light from a vertical-cavity surface-emitting laser (VCSEL) emitting at 852 nm in a single transverse and longitudinal mode is passed through a cell 2 cm long and containing Cs along with several kPa of Ne or Ar buffer gas. The laser's power is attenuated before the cell such that roughly 10  $\mu$ W of optical power in a beam 4 mm in diameter is incident on the cell. The laser's injection current is modulated at 4.6 GHz, resulting in the two firstorder optical sidebands being separated by 9.2 GHz, which is equal to the ground-state hyperfine splitting of the Cs atom. Roughly 40% of the optical power was contained in the blue sideband, whereas roughly 20% of the power was in the red sideband; the remainder of the optical power was distributed between the carrier (13%) and the higher-order sidebands. The laser was tuned such that the two first-order sidebands were resonant with the transitions from the two atomic ground states to the  $6P_{3/2}$ excited state, as shown in Fig. 2. Because two optical fields are present, the atoms are not completely optically pumped into a single ground-state hyperfine level. They instead come to some equilibrium distribution, with nonzero population in each level; the populations depend on the relative intensities and detunings of the two optical fields. The atoms therefore fluoresce and scatter a significant amount of laser light, resulting in nearly 43% of the laser's sideband power being absorbed. A typical absorption profile as the modulated laser is scanned over the atomic lines is shown in Fig. 3.

With 9.7 kPa of Ne buffer gas in the cell, the opticalabsorption linewidth is measured to be 1.3 GHz, far above the natural linewidth (~5 MHz) of the transition in question. The broadening comes from a number of sources and is a combination of both homogeneous and inhomogeneous mechanisms. Collisions with buffer-gas atoms dominate the broadening and contribute a linewidth of roughly 1 GHz. The Doppler broadening for this Cs transition at the cell temperature of 29 °C is 380 MHz. In addition, the excited-state hyperfine structure is not resolved; the hyperfine states of lowest and highest energy are split by 600 MHz, leading to some additional broadening.

The power spectrum of the FM noise of the unmodulated laser is shown in Fig. 4(a) and was obtained by tuning the laser onto a fringe of a Fabry–Perot etalon and measuring the transmitted intensity fluctuations. The increasing noise at low Fourier frequencies is typical of all types of semiconductor lasers. The laser with the RF modulation applied was then tuned onto the side of the largest absorption peak shown in Fig. 3 so that the two first-order sidebands were resonant with the atomic transitions from the two ground states. Figure 4(b) compares



Fig. 2. Tuning of the laser with respect to the atomic transitions. The first-order sidebands on the optical field are resonant with the two atomic transitions from the F = 3 and F = 4ground states.



Fig. 3. Fraction of total optical power transmitted through the cell as the RF-modulated laser is scanned over the  $Cs D_2$  line. The largest peak occurs when the two first-order laser sidebands are resonant with the two hyperfine absorption lines, as in Fig. 2. The remaining peaks occur when the carrier or higher-order sidebands are resonant.

the AM noise measured with a photodiode at the output of the Cs cell under these conditions (trace A) with the projected AM noise calculated from the laser's FM noise spectrum and the slope of the absorption line (trace B). It is clear that although the agreement is very good at low Fourier frequencies the two sets of data deviate significantly from each other at frequencies >1 kHz, suggesting that some additional process is occurring.

Typical spectra taken near the line peak, on the side of the line, and off the line entirely are shown in Fig. 4(c). It is clear from the data that both the amplitude of the noise and the shape of the spectrum change dramatically as a function of the laser detuning. Traces B and C, for example, cross near 100 Hz. This is not consistent with a simple linear theory that predicts that the noise will change uniformly throughout the spectrum as the absorption slope changes. It is important to note that the intrinsic AM noise limit is well below the FM–AM conversion noise level. The actual measured AM noise level is shown in Fig. 4(c) (trace D).

The noise at a frequency of 530 Hz, extracted from spectra similar to those in Fig. 4(c), is shown in Fig. 5 as a function of the laser detuning from the peak of the dc

absorption spectrum. It is clear that the noise increases with detuning but that the noise minimum does not occur at the peak of the dc absorption spectrum, as might initially be expected. The experimental data points agree well with the prediction of the linear FM–AM conversion theory shifted by 40 MHz from the dc absorption line center. When this information is combined with the spectral data in Fig. 4, it appears that the noise at higher frequencies exhibits a behavior as a function of laser detuning



Fig. 4. (a) Laser FM noise spectrum measured by use of an etalon fringe. (b) Comparison of the AM noise measured at the output of the Cs cell with the laser detuned by -217 MHz from the peak of the dc atomic absorption line (trace A) with the projected AM noise calculated from the laser FM noise spectrum and the slope of the atomic absorption line (trace B). (c) AM noise at the cell output with the laser detuned by -217 MHz (trace A), -9.5MHz (trace B), and +17 MHz (trace C). The noise floor (trace D), determined by the intrinsic AM noise of the laser and measured with the laser tuned away from the absorption lines, is also shown. Strong peaks related to the 60-Hz line frequency have been removed from all spectra for clarity of presentation.



Fig. 5. Noise at the detector output at 530 Hz, normalized to the dc level, measured as a function of the detuning of the laser from the peak of the dc optical-absorption spectrum. The points are the experimental data, and the solid curve is the prediction of the linear FM-AM conversion model. The 40-MHz shift of the noise minimum from the line peak can be explained with the optical-pumping process.

somewhat different from that of the noise at lower frequencies. The crossover frequency for this effect is 1 kHz, which is roughly the inverse of the 1 ms we estimate to be the optical-pumping time for this laser intensity and buffer-gas pressure. We have therefore developed a theoretical model that includes the effects of optical pumping and excited-state hyperfine structure to explain the measured data.

### 3. THEORETICAL MODEL

The model described here is shown in Fig. 6. Two ground-state levels are coupled to a single excited state by a pair of optical fields in a  $\Lambda$  configuration. The transition rates from state 1 to state 3 and from state 2 to state 3 are denoted  $R_1(\omega)$  and  $R_2(\omega)$ , respectively, and are functions of the detuning from resonance,  $\omega$ . It is also assumed for simplicity that the excited state decays to both ground states with equal branching ratios and with a total decay rate equal to  $\gamma$ . Finally, a phenomenological population decay rate between the two ground states is included, equal to  $1/2\tau$ , which accounts for the effects of Cs–Ne spin-exchange collisions on the level populations.

The key aspect of this model is that the ground-state populations change as a function of laser detuning. Physically, the reason this happens is that the relative absorption of the optical fields through the cell changes with detuning, causing the optical-pumping rates to change as well. State 1 in the model represents the F = 3 ground state of the Cs atom. This state couples to the F = 2, 3, and 4 excited hyperfine levels. State 2 in the model represents the F = 4 ground state in Cs, which couples to the F = 3, 4, and 5 excited states. Since the mean energies of the two sets of excited states are separated by roughly the excited states' hyperfine splitting energy,  $2\hbar\Delta$ , the absorption profiles of the two optical fields will be different as they are scanned together over the optical transition. Although the model has only a single excited state, state 3, it is assumed that the two absorption profiles peak at slightly different optical detunings. As a result, the average intensities of the fields, and therefore also the average transition rates, will change relative to each other as a function of detuning.

The fractional population of each state i in Fig. 6 is denoted  $N_i$ . Coherence between the ground states is important only when the optical fields are within a ground-state transition linewidth of Raman resonance. In the buffer-gas cells used in the experiment this linewidth is of order 100 Hz. It is therefore possible to tune the optical fields substantially away from Raman resonance while still keeping them simultaneously on optical resonance. The ground-state coherence can then be neglected, and only population transfer between the levels need be considered. In addition, it is assumed that the optical fields coupling the levels are weak and therefore that the excited state is almost empty. The rate equations for this system can be written as

$$\begin{split} \dot{N}_1 &= -R_1(\omega)(N_1 - N_3) + \frac{\gamma}{2}N_3 \\ &- \frac{(N_1 - N_2) - (N_1^{(0)} - N_2^{(0)})}{2\,\tau}, \end{split} \tag{1}$$



Fig. 6. Theoretical model including optical pumping and excited-state hyperfine structure.

$$\dot{N}_{2} = -R_{2}(\omega)(N_{2} - N_{3}) + \frac{\gamma}{2}N_{3} - \frac{(N_{2} - N_{1}) - (N_{2}^{(0)} - N_{1}^{(0)})}{2\tau}, \qquad (2)$$

$$\dot{N}_3 = R_1(\omega)(N_1 - N_3) + R_2(\omega)(N_2 - N_3)$$

$$- \gamma N_3.$$
(3)

Here  $N_1^{(0)}$  and  $N_2^{(0)}$  are the fractional ground-state populations in the absence of the optical fields.

The system of equations can be solved for the steadystate ground-state populations  $N_1$  and  $N_2$ . The scattering rate of photons out of the optical field by a single atom is then calculated from the expression  $S = R_1(\omega)N_1$ +  $R_2(\omega)N_2$  to be

$$S = \frac{2}{\tau} \frac{(\beta_1 N_1^{(0)} + \beta_2 N_2^{(0)}) + 2\beta_1 \beta_2}{1 + \beta_1 + \beta_2}, \qquad (4)$$

where  $\beta_i(\omega) = R_i(\omega) \tau/2$ . The maximum dc absorption occurs at a detuning,  $\omega$ , that is the solution of  $\partial S/\partial \omega = 0$ .

We now consider the effects of some small laserfrequency modulation about the mean frequency. This is equivalent to the situation in which FM noise is present on the laser. Since the modulation is assumed to be small, the expression for the dc absorption, Eq. (4), is unaffected. The rate equations (1)–(3) are solved under conditions of small modulation,  $\delta\beta_1$  and  $\delta\beta_2$ , of the normalized optical transition rates. In terms of the Fourier transforms  $\delta\tilde{\beta}_i(\Omega)$ , the Fourier transform of the modulation in the scattering rate, as a function of the modulation frequency  $\Omega$ , is

$$\delta \tilde{S}(\Omega) = \frac{2}{\tau} \left[ \frac{\beta_2 + N_1^{(0)}}{1 + \beta_1 + \beta_2} \left( \frac{1 + i\Omega\tau + 2\beta_2}{1 + i\Omega\tau + \beta_1 + \beta_2} \right) \delta \tilde{\beta}_1(\Omega) + \frac{\beta_1 + N_2^{(0)}}{1 + \beta_2 + \beta_1} \left( \frac{1 + i\Omega\tau + 2\beta_1}{1 + i\Omega\tau + \beta_2 + \beta_1} \right) \delta \tilde{\beta}_2(\Omega) \right].$$
(5)

Equation (5) shows generally how fluctuations in the photon-scattering rate  $\delta \tilde{S}(\Omega)$  depend on small modulation of the normalized optical-pumping rates,  $\delta \tilde{\beta}_i$ . In order to proceed further we now assume a specific form for

the atomic absorption profile: that of a Lorentzian with width  $2\Gamma$ . The absorption coefficients for the two optical fields can then be written

$$\alpha_{1,2}(\omega) = \alpha_0 \frac{\Gamma^2}{(\omega \pm \Delta)^2 + \Gamma^2},$$
(6)

where  $\alpha_0$  is the peak absorption coefficient, and  $2\Delta$  is the optical frequency separation between the two profiles. It is these frequency-shifted absorption profiles that give rise to the changing ground-state level populations that in turn result in an alteration of the FM–AM noise conversion process.

Integrating over the length L of the cell, and assuming exponential absorption, we find the average intensity of the two fields,  $I_1$  and  $I_2$ , as a function of optical detuning  $\omega$  to be

$$\langle I_{1,2}(\omega) \rangle = \frac{I_{10,20}}{\omega_{1,2}(\omega)L} \{ 1 - \exp[-\alpha_{1,2}(\omega)L] \}$$

$$\approx I_{10,20} \bigg[ 1 - \frac{\alpha_{1,2}(\omega)L}{2} \bigg],$$
(7)

where  $I_{i0}$  is the incident intensity,  $\alpha_i(\omega)$  is the absorption coefficient, and where it has been assumed that  $\alpha_i(\omega)L \ll 1$  (optically thin cell).

The parameters  $\beta_i$  are related to the average field intensities and absorption profiles given by expressions (6) and (7). We find that

$$\beta_{1,2}(\omega) = R_{1,2}(\omega) \frac{\tau}{2} = \left(\frac{\gamma}{2} \frac{\langle I_{1,2} \rangle}{I_{\text{sat}}}\right) \frac{\tau}{2} \frac{\Gamma^2}{(\omega \pm \Delta)^2 + \Gamma^2}$$
(8)  
$$= \frac{\gamma \tau}{4} \frac{I_{10,20}}{I_{\text{sat}}} \left[1 - \frac{\alpha_0 L}{2} + \frac{\Gamma^2}{(\omega \pm \Delta)^2 + \Gamma^2}\right]$$
$$\times \left[\frac{\Gamma^2}{(\omega \pm \Delta)^2 + \Gamma^2}\right],$$
(9)

where  $I_{\rm sat}$  is the saturation intensity of the optical transition and where  $I_{10,20} \ll I_{\rm sat}$ . For small fluctuations in frequency,  $\delta \omega$ , the normalized optical-pumping rates  $\delta \beta_i$ can be calculated from Eq. (9) to be

$$\delta\beta_{1,2} = \frac{\gamma\tau}{2} \frac{I_{10,20}}{I_{\text{sat}}} (1 - \alpha_0 L) \frac{\omega \pm \Delta}{\Gamma^2} \delta\omega, \qquad (10)$$

where it has been assumed that the detunings are small compared with the optical width of the absorption line,  $|\omega \pm \Delta| \ll \Gamma$ .

Substituting the above expressions into Eq. (5) and assuming that the optical-pumping rates are both much larger than the ground-state population decay rate ( $\beta_i \ge 1$ ), the scattering-rate modulation resulting from  $\delta \omega$  is found to be

$$\delta \tilde{S}(\Omega) = \frac{-2\gamma}{\Gamma^2} (1 - \alpha_0 L) \frac{I_{10}I_{20}}{(I_{10} + I_{20})I_{\text{sat}}} \\ \times \left[ \omega - \frac{\beta_{10} - \beta_{20}}{(\beta_{10} + \beta_{20} + i\Omega\tau)} \Delta \right] \delta \tilde{\omega} \quad (11)$$

$$= \frac{-2\gamma}{\Gamma^2} (1 - \alpha_0 L) \frac{I_{10}I_{20}}{(I_{10} + I_{20})I_{\text{sat}}} \\ \times \left[ \omega - \frac{\beta_{10}^2 - \beta_{20}^2}{(\beta_{10} + \beta_{20})^2 + (\Omega\tau)^2} \Delta + i \frac{\Omega\tau(\beta_{10} - \beta_{20})}{(\beta_{10} + \beta_{20})^2 + (\Omega\tau)^2} \Delta \right] \delta \tilde{\omega}.$$
(12)

Here  $\beta_{10}$  is the peak normalized optical-pumping rate. The FM–AM, conversion process is therefore dependent on the Fourier frequency,  $\Omega$ , of the noise. Equation (12) can be solved for the detuning at which the scatteringrate modulation, and therefore the FM–AM conversion, is a minimum:

$$\omega_{\delta S_{\min}}(\Omega) = \frac{\beta_{10}^2 - \beta_{20}^2}{\Omega^2 \tau^2 + (\beta_{10} + \beta_{20})^2} \Delta.$$
(13)

At high modulation frequencies  $(\Omega \ge 1/\tau)$ , therefore, the FM–AM conversion is a minimum when the lasers are tuned exactly halfway between the two individual absorption-profile peaks. For lower modulation frequencies  $(\Omega \ll 1/\tau)$  the FM–AM conversion is minimized at a different detuning as long as  $\beta_{10} \neq \beta_{20}$ . The expression for  $\omega \delta S_{\min}$  at low modulation frequencies is, in fact, identical to the detuning at which the dc absorbance is a maximum.

The following general conclusions can be drawn from an examination of Eq. (13). First, if the transition rates induced by the two optical fields are equal, then  $\omega \delta S_{\min}$ = 0 and the minimum high-frequency FM-AM conversion occurs at the peak of the dc absorption spectrum. In this case the AM noise spectrum would change identically at all Fourier frequencies as the laser was detuned from resonance. This is what would be expected without considering the effects of optical pumping. However, if the transition rates differ significantly (due to unequal powers in the two optical fields, for example), then  $\omega_{\delta S_{\min}}(\Omega)$  $\neq 0$ )  $\neq \omega_{\delta S_{\min}}(\Omega = 0)$  and the minimum FM–AM conversion occurs away from the peak of the dc absorption spectrum. The difference between the two frequencies  $\omega_{\text{diff}}$  $= \omega_{\delta S_{\min}}(\Omega=\infty) - \omega_{\delta S_{\min}}(\Omega=0) \ \text{is of the order of the}$ excited-state hyperfine splitting,  $2\Delta$ . For a cell containing a buffer-gas pressure similar to that used in the experiment, the homogeneous broadening  $\Gamma$  due to collisions is in fact larger than the excited-state hyperfine structure,  $\Delta$ , satisfying the assumption that  $|\Delta - \omega| < \Gamma$  for small detunings  $\omega$ .

Equation (12) indicates that even when  $\omega = \omega_{\delta S_{\min}}$ , some residual modulation of the scattering rate is still present due to the imaginary part of the term in brackets. This term occurs because the modulation of the groundstate population induced by the optical pumping is phase shifted from the original frequency modulation at Fourier frequencies above the optical-pumping rate. The modulation of the scattering rate induced by the absorption line at constant ground-state population [the first term in brackets in Eq. (12)] is always in phase with the frequency modulation and can therefore never cancel this out-of-phase term. The residual modulation is small at very low and very high modulation frequencies and peaks at a frequency equal to the optical-pumping rate. If FM noise is present in the laser with a power spectrum given by  $S_{\delta\omega}(\Omega)$ , then the resulting minimum fluctuation in the optical power exiting the cell is

$$\begin{split} S_{\delta P}(\Omega) \\ = & \left(\frac{2\Delta}{\Gamma^2}\right)^2 (\alpha_0 L)^2 P_0^2 \bigg[\frac{\Omega_{\tau}(\beta_{10} - \beta_{20})}{(\beta_{10} + \beta_{20})^2 + (\Omega_{\tau})^2}\bigg]^2 S_{\delta \omega}(\Omega), \end{split}$$
(14)

where  $P_0$  is the power (in both sidebands) incident on the cell. The FM noise of the laser used here at a Fourier frequency of 500 Hz is  $\sim 10^{10}$  Hz<sup>2</sup>/Hz, which results in a peak AM noise of roughly four times the shot-noise level for an input power of 10  $\mu$ W. Thus the excess noise is of the right order of magnitude to affect somewhat the noise performance of the frequency reference. We believe that this noise accounts for the discrepancy between experiment and theory mentioned in Ref. 20.

# 4. FM–AM CONVERSION WITH OPTICAL PUMPING

We can now examine the experimental data in Figs. 4 and 5 in light of the theoretical considerations of Section 3. The asymmetry in the sideband power in the experiment is approximately 2:1, leading to a frequency shift  $\omega_{\text{diff}}$  $\approx \Delta/3$  for very fast modulation compared with slow dc absorption. With an average sideband intensity of 15  $\mu$ W/cm<sup>2</sup> incident on the atoms and a collisional broadening of 1 GHz, the optical-pumping rate is approximately  $2\pi(36 \text{ Hz})$ . We therefore expect that the noise spectrum of light passing through the cell will deviate significantly from the laser's FM noise spectrum at frequencies 36 Hz. This is indeed the case [see Fig. 4(b)]. In addition, it would be expected that the noise minimum in Fig. 5 would be shifted from the dc absorption peak by something of the order of 50 MHz. The slightly smaller experimental value of 40 MHz can be explained by the fact that the noise frequency considered in Fig. 5 is 530 Hz and therefore is not sufficiently far above the opticalpumping rate to realize the full shift.

Further details of the spectra in Fig. 4(c) can be explained on a more qualitative level. Trace A shows a spectrum that agrees well with the laser's FM noise spectrum at low frequencies but that increases above the laser's FM noise at higher frequencies. Because the highfrequency spectrum is shifted with respect to the lowfrequency spectrum by several tens of MHz (Fig. 7), FM noise at high frequencies will be converted by a different atomic absorption slope, and hence the resulting AM noise can appear to increase. Trace B is taken on the same side of the dc absorption line as trace A but closer to the line center. For this trace the FM-AM conversion slope at high frequencies should be very close to zero, resulting in a somewhat stronger suppression of the noise at high frequencies than at low frequencies, as observed in the data. Finally, trace C is taken again close to the peak of the dc absorption profile but on the other side. Here, the low-frequency noise is strongly suppressed, but the increasing conversion slope at higher frequencies leads to an increase in the transmitted AM noise there.

One additional test was carried out to confirm the optical-pumping mechanism and is partly described in Ref. 4; it is included here for completeness. The laser current was modulated at two different frequencies  $f_1$  and  $f_2$ .  $f_1$  was set to a fixed value of 50 kHz, and  $f_2$  was varied between 50 Hz and 12 kHz. The light was passed through a cell containing a combination of buffer gases (1.8 kPa Ne along with 2.4 kPa Ar), which resulted in a total broadening of 970 MHz. The detector's output at each of these frequencies was demodulated with a lock-in amplifier, and the magnitude of the demodulated signal  $(R-\theta \text{ mode})$  was monitored as a function of laser detuning. When  $f_1 \neq f_2$ , the lock-in outputs,  $R_{f_1}(\omega)$  and  $R_{f_2}(\omega)$ , were found to reach a minimum at different laser detunings,  $\omega$ ; this difference in detunings,  $\omega_{\text{diff}} = \omega_{\delta S_{\min}}(2\pi f_1)$  $- \omega_{\delta\!S_{\rm min}}(2\pi\!f_2),$  and the lock-in output minimum value for  $f_2,\,R_{f_2}[\,\omega_{\delta\,S_{\rm min}}(2\pi\!f_2)],\,{\rm was}$  measured as a function of the frequency  $f_2$ .

The results are shown in Fig. 8 for three different optical intensities (and therefore three different opticalpumping rates). The theory in Section 3 predicts that for a given Fourier frequency  $\Omega$ , the noise should be minimum at some particular detuning,  $\omega_{\delta S_{\min}}(\Omega)$ . The expression for  $\omega_{\text{diff}}$  can then be calculated: when  $f_2$  is very low (and  $f_1$  very high),  $\omega_{\text{diff}}$  is at its maximum value of

$$\omega_{\rm diff}^{(\rm max)} = \frac{\beta_{10} - \beta_{20}}{\beta_{10} + \beta_{20}} \Delta.$$
(15)

Thus a large value for  $\omega_{\text{diff}}$  is expected and indeed measured, as can be seen in Fig. 8(a). As  $f_2$  increases,  $\omega_{\text{diff}}$  drops to zero as the two frequencies become almost equal. The corner frequency at which  $\omega_{\text{diff}}$  starts rolling off toward zero should be roughly equal to the optical pumping rate. It can be seen from the figure that as the laser intensity is decreased, the corner frequency gets lower, as expected. The solid curves in Fig. 8(a) are fits to the data that use the functional form [chosen according to Eq. (12)]

$$\omega_{\text{diff}}(f) = \frac{\omega_{\text{diff}}^{(\text{max})}}{1 + \left(\frac{f}{\phi_c I_{\text{opt}}}\right)^2},$$
(16)

where  $\omega_{\text{diff}}^{(\text{max})}$  is the maximum frequency shift, and  $2 \pi \phi_c$  is the optical-pumping rate normalized to optical intensity,  $I_{\text{opt}}$ . The fits give parameter values of  $\omega_{\text{diff}}^{(\text{max})}$ 



Fig. 7. Detunings from the optical-absorption line at which the data in Fig. 4(c) were taken. At detuning A the high-frequency noise slope is larger than the slope of the low-frequency noise. At detuning B the high-frequency slope is near zero, and the low-frequency slope is nonzero but small. At detuning C the low-frequency slope is near zero but the high-frequency slope is nonzero.



Fig. 8. (a) Optical frequency difference between the FM–AM conversion minima  $\omega_{\rm diff} = \omega_{\delta S_{\rm min}}(2\pi f_1) - \omega_{\delta S_{\rm min}}(2\pi f_2)$  and (b) minimum FM–AM conversion at frequency  $f_2$ , normalized to the light intensity, as a function of modulation frequency  $f_2$ . The average optical intensity in each of the first-order sidebands was 170  $\mu$ W/cm<sup>2</sup> (circles), 47  $\mu$ W/cm<sup>2</sup> (triangles), and 15  $\mu$ W/cm<sup>2</sup> (squares). The solid curves are fits to Eq. (16) with  $\omega_{\rm diff}^{\rm max}$  = 69.4 MHz and  $\phi_c = 2.6$  Hz/( $\mu$ W/cm<sup>2</sup>).

= 69.4 MHz and  $\phi_c = 2.6 \text{ Hz}/(\mu W/\text{cm}^2)$ . The value obtained for the optical-pumping rate,  $2\pi\phi_c \times I_{\text{opt}}$ , is within a factor of 2 of that calculated theoretically from the intensity incident on the atomic sample, and the value of  $\omega_{\text{diff}}^{(\text{max})}$  is within 20% of the theoretically predicted value. Reasonable semi-quantitative agreement between experiment and theory is therefore found.

Figure 8(b) shows the excess modulation,  $R_{f_2}(\omega)$ , present when  $\omega = \omega_{\delta S_{\min}}(2\pi f_2)$ . As expected from theory, this quantity peaks when the modulation frequency,  $2\pi f_2$ , is equal to the optical-pumping rate. The solid curves are a fit to the functional form

$$R_{f_2}(2\pi f) = M_{\max} \frac{\left(\frac{f}{\phi_c I_{\text{opt}}}\right)}{1 + \left(\frac{f}{\phi_c I_{\text{opt}}}\right)^2},$$
(17)

where  $M_{\rm max}$  is a single scale factor for all three curves and  $\phi_c$  takes on the value stated above. The existence of the additional FM–AM conversion is thus verified experimentally.

# 5. IMPLICATIONS FOR PRACTICAL FREQUENCY REFERENCES

In designing a practical atomic-frequency reference, several issues are important in defining the performance of the standard. The short-term stability is determined by the size and shape of the resonance signal and by the amount of noise at the RF-source modulation frequency. As shown in Fig. 4, this noise level depends critically on the Fourier frequency and also on the laser detuning from resonance. High modulation frequencies (1 kHz) will clearly be optimal since the noise is lower there. In this case the optimal laser detuning will not be at the peak of the dc absorption profile, as might be expected, but instead will be shifted by several tens of MHz due to the optical-pumping effect. In addition, excess noise may be present due to the residual FM-AM conversion in the frequency range of a few hundred Hz to a few kHz. Since the modulation frequency of the external oscillator in the frequency reference is often in this range, the excess noise may affect the performance of the reference.

Another consideration is the dependence of the ac Stark shift on the optical detuning.<sup>23–25</sup> Ideally, one wants to operate at a detuning for which the ac Stark shift is a minimum in order to obtain good long-term stability. In general, this detuning does not correspond to the detuning for which the short-term stability is optimal. But with a careful choice of laser power, modulation frequency, and sideband amplitude, it may be possible to optimize the frequency-reference design from both viewpoints.

In the experiment described above an all-optical excitation of the microwave clock transition is used. This differs somewhat from a conventional frequency reference in that two, rather than one, optical fields are present that optically pump the atoms. However, even when only a single field is present, the ground-state population distribution will be affected by optical pumping. The opticalpumping rate of a conventional frequency reference is comparable with that of the reference described here, and so a similar difference between the low- and highfrequency parts of the FM-AM conversion spectra might be expected. An equivalent optimization of the reference modulation frequency and detuning might therefore be possible. The excess FM-AM conversion noise due to the out-of-phase component of the ground-state population change would also be expected to be present.

# 6. CONCLUSIONS

We have shown that optical pumping dramatically affects the way in which laser FM noise is converted into AM noise in a buffer-gas-confined atomic vapor. Noise at low Fourier frequencies causes the atomic-population distribution to change adiabatically with the field fluctuations, while the limited optical-pumping rate prevents this at higher Fourier frequencies. The AM noise spectrum at the output of the cell therefore has a somewhat complicated structure that depends on both the optical power and the laser detuning but that is well explained by our simple theoretical model. It is possible that this structure could be exploited to improve the performance of laser-pumped frequency references.

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\*Also at JILA, The University of Colorado, Colorado 80309.

#### REFERENCES

- A. Kastler, "Quelques suggestions concernant la production optique et la detection optique d'une inegalite de population des niveaux de quantification spatiale des atomes. Application a l'experiance de Stern et Gerlach et a la resonance magnetique," J. Phys. Radium 11, 255–265 (1950).
- 2. W. Happer, "Optical pumping," Rev. Mod. Phys. 44, 169–249 (1972).
- 3. J. Vanier and C. Audoin, *The Quantum Physics of Atomic Frequency Standards* (Adam Hilger, Bristol, UK, 1989).
- 4. J. Kitching, L. Hollberg, S. Knappe, and R. Wynands, "Frequency-dependent optical pumping effects in driven, three-level systems," Opt. Lett. (to be published).
- A. M. Akulshin, A. A. Čelikov, and V. L. Velichansky, "Subnatural absorption resonances on the D1 line of rubidium induced by coherent population trapping," Opt. Commun. 84, 139-143 (1991).
- P. R. Hemmer, M. S. Shahriar, H. Lamela-Rivera, S. P. Smith, B. E. Bernacki, and S. Ezekiel, "Semiconductor laser excitation of Ramsey fringes by using a Raman transition in a cesium atomic beam," J. Opt. Soc. Am. B 10, 1326–1329 (1993).
- N. Cyr, M. Têtu, and M. Breton, "All-optical microwave frequency standard: a proposal," IEEE Trans. Instrum. Meas. 42, 640–649 (1993).
- 8. J. Vanier, A. Godone, and F. Levi, "Coherent population trapping in cesium: dark lines and coherent microwave emission," Phys. Rev. A 58, 2345-2358 (1998).
- G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, "An experimental method for the observation of RF transitions and laser beat resonances in oriented Na vapor," Nuovo Cimento B 36, 5–20 (1976).
- E. Arimondo and G. Orriols, "Nonabsorbing atomic coherences by coherent two-photon transitions in a three-level optical pumping," Lett. Nuovo Cimento 17, 333-338 (1976).
- E. Arimondo, "Coherent population trapping in laser spectroscopy," Prog. Opt. 35, 257 (1996).
- T. Yabuzaki, T. Mitsui, and U. Tanaka, "New type of highresolution spectroscopy with a diode laser," Phys. Rev. Lett. 67, 2453–2456 (1991).

- Th. Haslwanter, H. Ritsch, J. Cooper, and P. Zoller, "Lasernoise-induced population fluctuations in two- and threelevel systems," Phys. Rev. A 38, 5652–5659 (1988).
- R. Walser and P. Zoller, "Laser-noise-induced polarization fluctuations as a spectroscopic tool," Phys. Rev. A 49, 5067– 5077 (1994).
- B. J. Dalton and P. L. Knight, "The effects of laser field fluctuations on coherent population trapping," J. Phys. B 15, 3997-4015 (1982).
- H. G. Robinson, V. V. Vasiliev, V. L. Velichansky, L. Hollberg, and A. S. Zibrov, "Diode laser noise conversion and reduction in atomic vapor," presented at the International Conference on Atomic Physics, Boulder, Colo., July 31–Aug. 5, 1994.
- M. Bahoura, "Influence du bruit de phase d'une diode laser sur les performances ultimes de son asser vissement en frequence sur une resonance optique," Ph.D. dissertation (University of Paris XI, Paris, 1998).
- J. C. Camparo, "Conversion of laser phase noise to amplitude noise in an optically thick vapor," J. Opt. Soc. Am. B 15, 1177-1186 (1998).
- J. C. Camparo and J. G. Coffer, "Conversion of laser phase noise to amplitude noise in a resonant atomic vapor: the role of laser linewidth," Phys. Rev. A 59, 728-735 (1999).
- J. Kitching, N. Vukicevic, L. Hollberg, S. Knappe, R. Wynands, and W. Weidemann, "A microwave frequency reference based on dark-line resonances in Cs vapor," IEEE Trans. Instrum. Meas. 49, 1313–1317 (2000).
- S. Knappe, R. Wynands, J. Kitching, H. G. Robinson, and L. Hollberg, "Characterization of coherent population trapping resonances as atomic frequency references," J. Opt. Soc. Am. B (to be published).
- C. Affolderbach, A. Nagel, S. Knappe, C. Jung, D. Wiedenmann, and R. Wynands, "Non-linear spectroscopy using a vertical-cavity surface-emitting laser (VCSEL)," Appl. Phys. B 70, 407–413 (2000).
- M. Arditi and J.-L. Picque, "Precision measurement of light shifts induced by a narrow band GaAs laser in the 0-0 <sup>133</sup>Cs hyperfine transition," J. Phys. B 8, L331–L335 (1975).
- J. C. Camparo, R. P. Frueholz, and C. H. Volk, "Inhomogeneous light shift in alkali-metal atoms," Phys. Rev. A 27, 1914–1924 (1983).
- A. Nagel, S. Brandt, D. Meschede, and R. Wynands, "Light shift of coherent population trapping resonances," Europhys. Lett. 48, 385–389 (1999).