

Frequency-dependent optical pumping in atomic Λ -systems

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We consider the effects of optical pumping on the conversion of laser-frequency modulation into intensity modulation by an atomic absorption line in a vapor of alkali atoms driven in a Λ -configuration. It is found that, due to optical pumping in combination with the excited-state hyperfine structure, the absorption line shape is distorted substantially as the Fourier frequency of the FM is changed. The most significant effect of the distortion is a shift of the apparent line center, which depends on how the frequency of the modulation compares with the optical pumping rate. This shift has implications for locking lasers to atomic transitions and also for FM-AM noise conversion in atomic vapors.

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Precise measurement of atomic absorption line shapes is of great importance in a variety of fields ranging from the determination of fundamental physical constants¹ to optical frequency standards.² Among the many perturbations that can affect line shape, distortion that is due to the optical field used to probe the atoms plays a unique role, as light must be present for measurement of the line shape and the light's effects can never be completely eliminated. For atoms probed on a closed transition, for example, distortion of the saturation dip line shape can arise from a redistribution of the atomic velocities as a result of spontaneous light pressure.³⁻⁵

On open transitions, where repeated scattering of photons is much less likely, optical pumping⁶ between alkali-atom ground-state hyperfine levels can also substantially distort the line shape.^{7,8} We present measurements in which line shapes are distorted by a bichromatic laser field used to probe an atomic system in a Λ -configuration. Optical pumping between the ground state's hyperfine levels changes the level populations, causing an apparent frequency shift of the peak of the optical absorption line. The magnitude of the shift depends critically on whether the laser-frequency scan rate is lower or higher than the optical pumping rate.

The experimental arrangement and laser tunings are shown in Fig. 1. A bichromatic optical field, tuned to the D_2 line of Cs at 852 nm, is incident upon a 2-cm-long cell at room temperature that contains Cs at its vapor pressure, along with a mixture of Ne and Ar buffer gases with a total pressure of 4.3 kPa. The bichromatic laser field is generated by direct modulation of the injection current of a semiconductor laser at 4.6 GHz, which results in several optical fields separated in frequency by one-half of the 9.2-GHz transition frequency between the Cs ground-state hyperfine levels. The laser is tuned such that the two first-

order sidebands form the Λ -system with the atomic transitions. However, we tune the laser fields out of Raman resonance to avoid exciting any ground-state hyperfine coherences that would unnecessarily complicate the measurement. The influence of the carrier and the other modulation sidebands can be ignored here. The light from the laser is attenuated with a neutral-density filter, and this produces a single-sideband intensity of order $50 \mu\text{W}/\text{cm}^2$ incident upon the cell's front window. The optical pumping rate corresponding to this intensity for the given buffer-gas pressure is $\sim 2\pi(120 \text{ Hz})$.

As the fields are tuned together over the two optical transitions, the transmitted light is detected with a photodiode. A roughly Lorentzian absorption profile is observed, with a FWHM of 970 MHz; the profile FWHM is dominated by collisional broadening that is due to the presence of the buffer gas.

We simultaneously modulated the frequencies of the two optical fields by combining a slow ($<100\text{-kHz}$) current modulation with the rf modulation. The change

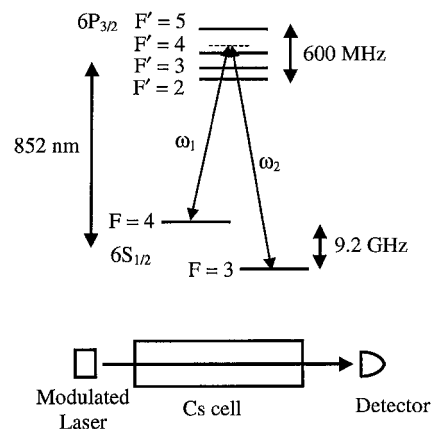


Fig. 1. Optical setup laser tunings.

in absorbance produced by this slow modulation was demodulated with a lock-in amplifier that generated a dispersive output signal. The absorption line shape and the lock-in output for a modulation frequency of 50 kHz are shown in Fig. 2. Although the lock-in output should be proportional to the derivative of the absorption line shape, it can be clearly seen that the zero of the lock-in signal does not coincide with the peak dc absorption. The shift for this particular modulation frequency is 82 MHz.

To check the dependence of the shift on modulation frequency, we carried out the following experiment. The laser's injection current was modulated with two frequencies, one, f_1 , fixed at 50 kHz, and the other, f_2 , varying from 15 Hz to 15 kHz. The modulation depth was small compared with the linewidth of the transition. The modulated absorbance caused by each frequency was demodulated with a separate lock-in, and the quadratic sum of the in- and out-of-phase components ($R - \theta$ mode) from each lock-in was monitored. As the laser was scanned across the line, the lock-in outputs traced U-shaped curves with minima near the peak absorption. The difference in optical frequency between these minima was measured as a function of modulation frequency f_2 and is plotted in Fig. 3 for three different laser intensities. When f_2 is low, the difference frequency is largest but drops to zero as f_2 is increased above a corner frequency, which depends on the optical intensity. From the data, it can be seen that this corner frequency decreases with increasing optical power.

The solid curves in Fig. 3 are fits to the data by use of the functional form⁹

$$\omega_{\text{diff}}(f) = \frac{\omega_{\text{diff}}^{(\text{max})}}{1 + (f_2/\phi_c I_{\text{opt}})^2}, \quad (1)$$

where $\omega_{\text{diff}}^{(\text{max})}/2\pi$ is the maximum frequency shift and ϕ_c is the corner frequency normalized to the average optical intensity in each of the first-order sidebands, I_{opt} . The function is fitted to all three sets of data, and a single set of fit parameters is extracted: The parameter values are $\omega_{\text{diff}}^{(\text{max})}/2\pi = 69.4$ MHz and $\phi_c = 2.6$ Hz/($\mu\text{W}/\text{cm}^2$).

We propose that the data in Figs. 2 and 3 can be explained by optical pumping combined with the atom's excited-state hyperfine structure. The absorption of the light by the atoms is the sum of the absorption of each field individually and, for an optically thin sample, can be written as $I_{\text{in}} - I_{\text{out}} = I_1\alpha(\omega_1)L + I_2\alpha(\omega_2 = \omega_1 + 9.2 \text{ GHz})L$. Here I_1 and I_2 are the intensities of the two optical fields (at optical frequencies ω_1 and ω_2 , respectively), $I_{\text{in}} = I_1 + I_2$, I_{out} is the intensity at the output from the cell, $\alpha(\omega)$ is the absorption coefficient of the atoms for an optical field of frequency ω , and L is the length of the cell. Since $\alpha(\omega_{1,2})$ depends on the number of atoms in level 1, 2, the absorption depends not only on the relative intensities of the two fields but also on the population distribution within the ground states.

The parameters $\alpha(\omega)$ depend also on the optical detuning from resonance. Since the $F = 3$ ground state

couples to the $F' = 2, 3, 4$ excited hyperfine states, and the $F = 4$ ground state couples to the $F' = 3, 4, 5$ excited states, we may account for the absorption of the two transitions by writing $\alpha(\omega_1) \approx C\alpha(\omega_2 - 2\pi\Delta)$, where C is a constant related to the different level populations and transition oscillator strengths and Δ is the separation of the center-of-mass frequency of the $F' = 2, 3, 4$ excited states from that of the $F' = 3, 4, 5$ excited states. For Cs, $\Delta \approx 270$ MHz. Because of this separation, the optical frequency at which the maximum absorption occurs depends on the population distribution of the ground states. If all the atoms are in the $F = 3$ ground state, for example, only one of the optical fields will experience absorption, and the peak absorption will occur at an optical detuning that differs by Δ from the peak that would occur if only the $F = 4$ level were populated.

As the fields are tuned slowly onto resonance, this population distribution (initially at thermal equilibrium) changes to reflect the intensity distribution between the fields. Since the absorption from one of the two lines is shifted in frequency with respect to the other, the ground-state population distribution will change as the laser tuning changes.

Suppose now that some frequency modulation is present that oscillates at a rate that is fast compared with the optical pumping time. In this case, the ground-state populations do not change with the fast modulation, and the FM-AM conversion is evaluated

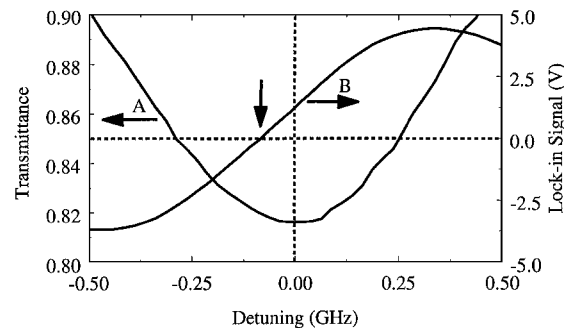


Fig. 2. (A) dc absorption line and (B) lock-in output for FM at a frequency of 50 kHz as a function of optical detuning for an average optical intensity of $50 \mu\text{W}/\text{cm}^2$ per sideband.

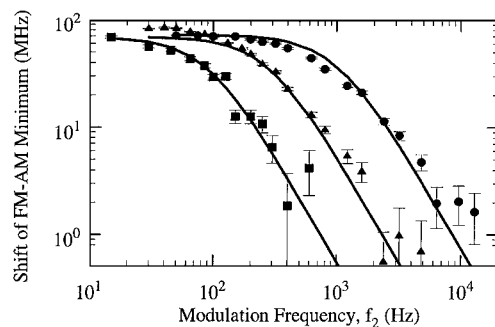


Fig. 3. Frequency difference between lock-in output zeros for modulation at a fixed frequency of 50 kHz and variable frequency f_2 . The average intensities in each of the two first-order sidebands were $170 \mu\text{W}/\text{cm}^2$ (circles), $47 \mu\text{W}/\text{cm}^2$ (triangles), and $15 \mu\text{W}/\text{cm}^2$ (squares). The solid curves are fits to Eq. (1).

with populations corresponding to the mean dc detuning from resonance. Since the dc absorption involves a population change, whereas the fast modulation does not, it is not, in general, the case that the peak of the dc absorption corresponds to the minimum of the fast FM–AM conversion. For this reason, a shift is observed in the data of Fig. 2.

Very slow modulation of the laser frequency, however, will lead to lock-in output that is exactly proportional to the derivative of the dc absorption profile. The zero of this signal then corresponds to the peak absorption. Thus, when the zeros of the lock-in outputs for fast and slow modulation are compared, a shift will be present. In Fig. 3, the first lock-in signal's zero for a frequency far above the typical optical pumping rate is compared with the second lock-in signal's zero for a frequency that varies from below to above that rate. The frequency f_2 at which the roll-off begins to occur should be roughly equal to $R_{\text{opt}}/2\pi$, where R_{opt} is the optical pumping rate. We have calculated the expected optical pumping rate by assuming a collisional broadening of 660 MHz, averaging over all the allowed transitions between Zeeman and hyperfine levels, assuming equal populations in all Zeeman levels within a hyperfine level, taking into account the unequal field intensities, and assuming equal branching ratios into the two ground states. We arrive at a rate $R_{\text{opt}}/2\pi = 3.8 \text{ Hz}/(\mu\text{W}/\text{cm}^2)$, in close agreement with the value extracted from the fit in Fig. 3. We therefore conclude that optical pumping is responsible for the shifts. A more comprehensive theoretical analysis supports this explanation.⁹

These results are important in the conversion of laser FM noise to AM noise by an atomic absorption line.^{10,11} Noise at Fourier frequencies less than the optical pumping rate will be converted to AM noise differently from noise at higher frequencies. As a result, the AM noise spectrum at the cell's output can be dramatically different in shape from the FM noise spectrum of the laser. Such considerations are important in, for example, vapor-cell frequency references, for which FM–AM conversion can be the limiting factor in determining the stability.^{10,12} In addition, some implementations of magnetometers¹³ and systems for quantum information processing¹⁴ are based on Λ -configurations and would be affected by the same processes discussed here.

Generally, however, optical pumping appears to play a role in locking lasers to the peaks of atomic absorption lines by use of FM modulation and lock-in detection. If the goal is to lock to the exact peak of the line (as is the case for optical frequency standards, for example) then in many cases optical pumping effects need to be taken into account. Although these effects are largest in a Λ -system driven by a bichromatic optical field, they can also be important for a single-frequency optical field if ground-state relaxation is present. This is because the absorption line shape, which results from transitions to the $F' = 3, 4, 5$ excited states (for the $F = 4$ ground state), is shifted in frequency with respect to the optical pumping line shape,

which results from transitions to only the $F' = 3$ and $F' = 4$ excited states. As long as the center frequency of the atomic absorption profile is different from that of the optical pumping profile, line shifts due to optical pumping that are similar to those discussed here will occur.

In conclusion, we have considered the conversion of laser-frequency modulation to AM by a sample of alkali atoms driven by a bichromatic optical field. In combination with the excited state's hyperfine structure, optical pumping results in a shift of the apparent absorption-peak frequency as measured with FM and lock-in detection. Such shifts need to be considered when one is performing precise FM or noise spectroscopy of an atomic species and may have implications for microwave frequency references, magnetometers, and quantum information processing systems based on Λ -systems.

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