

Dressed-atom spectroscopy of cold Cs atoms

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Absorption spectra of cold Cs atoms in a magneto-optical trap under the influence of a strong external pump laser field with arbitrary detuning and intensity have been systematically investigated. The spectra featured the Rabi sidebands of each transition, and their behavior was nicely fitted by the dressed-atom theory. A pronounced dispersion-type profile with sub-natural linewidth at the pump frequency may be attributable to the stimulated Raman process by sublevels that are lightshifted and optically pumped by the pump laser. © 1996 Optical Society of America.

1. INTRODUCTION

Cold atoms trapped by laser beams, such as in a magneto-optical trap¹ (MOT), can be one of the most suitable materials for investigating the strong interaction between light and matter (dressed state). This is because such atoms are free from Doppler broadening or transit-time broadening, but the linewidths are limited to natural linewidths Γ and the density of the atoms can reach, for instance, $10^{10}/\text{cm}^3$. Besides, under ordinary experimental conditions, Rabi frequency Ω can easily be made much larger than Γ . Using these advantages, one can perform high-resolution spectroscopy of dressed atoms with cold atoms.

In fact, there have been some papers²⁻⁴ that showed that probe absorption spectra from laser-trapped cold atoms are strongly modified by the presence of intense cooling laser beams. In Fig. 1 we show our result of using such a distorted absorption spectrum for the $6^6S_{1/2}(F=4)-6^6P_{3/2}(F'=3, 4, 5)$ transition (852 nm) of cold Cs atoms in a MOT. The line shape of the $F=4-F'=5$ transition is more or less similar to the ones previously reported^{2,3} and the main spectral features, including a pronounced gain, which is fully discussed below, are explained in such a way that an atom is strongly dressed by cooling laser photons⁵ with a detuning $\Delta \equiv \omega_L - \omega_0$, where ω_L and ω_0 denote the laser frequency and the $F=4-F'=5$ transition frequency, respectively. (Of course, even before the era of laser cooling, similar spectra were observed by pump-probe spectroscopy with atomic beams.^{6,7})

Ironically enough, the very presence of the strong cooling beams has prevented a systematic investigation of pump-probe measurement (or even any other nonlinear laser spectroscopy) of cold atoms, because the cooling beams have to be present with a fixed Rabi frequency Ω and at a fixed detuning Δ to keep the atoms trapped; for example, $\Delta/2\pi = -16$ MHz in the case of Fig. 1, whereas the natural linewidth of Cs atoms is $\Gamma/2\pi = 5.3$ MHz. If Ω or Δ of the cooling laser is varied, parameters such as atomic density, size, and temperature will change because of different trapping conditions. What is desired is a dressed-atom spectroscopy with totally arbitrary Ω and Δ with all the other parameters fixed.

We can circumvent this dilemma, however, by introducing a second strong laser to dress atoms and dividing the time into a cooling-trapping period and a pump-probe period, as illustrated in Fig. 2. Most of the time (90 μs in a 100- μs repetition time) atoms are exposed to the cooling beams, as in an ordinary MOT, and this keeps them cold enough for the measurements to be made. After the cooling beams are shut off there is a pump-probe period (8 μs) when an external strong pump beam and a weak probe beam hit the cold atoms. This period should be short enough to keep the pump laser from affecting the external degrees of freedom of the atoms and to ensure that the atoms do not become diffused or fall because of gravity. On the other hand, it should be long enough to be regarded as a steady state for the atoms. With this setup, now that Ω and Δ are two free parameters, a systematic investigation of pump-probe spectroscopy should be achieved. Of course, by simply blocking the pump beam one can obtain the absorption spectrum of purely bare atoms ($\Omega = 0$).

In this paper we present the results of such a spectroscopy with cold Cs atoms, that is, cold atoms dressed by external pump photons. Rabi sidebands were clearly observed in each transition, and their behavior, including the strong gain at one sideband, was well explained within the framework of dressed-atom theory. The other feature is a pronounced dispersion structure of subnatural linewidth at the pump-laser frequency. This structure changes its symmetry when Δ changes from positive to negative. A strong gain was also observed for this dispersion structure. We discuss these rich features in what follows.

2. EXPERIMENTAL SETUP

The setup that we employed was a typical MOT.¹ During the cooling-trapping period of Fig. 2, cooling beams (typically 10 mW, with 15 mm ϕ per beam) were provided by a single-frequency Ti:sapphire laser with 500-kHz linewidth and chopped by an acousto-optic modulator as mentioned above. We fixed the detuning frequency Δ near 15 MHz below the $F=4-F'=5$ resonance by using the feedback loop with a Cs saturation spectrometer. The re-

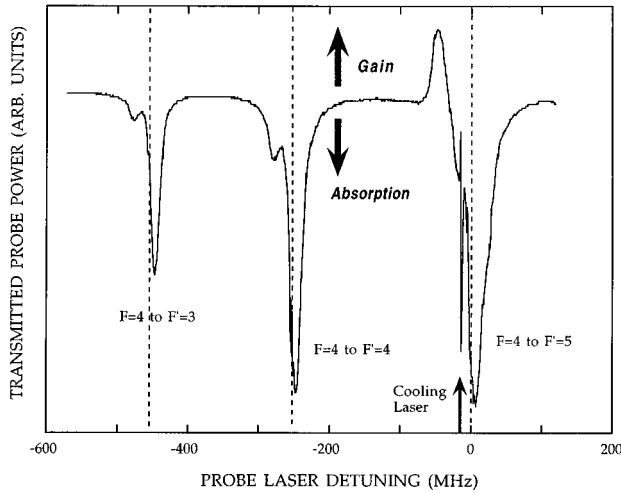


Fig. 1. Probe absorption spectra of cold Cs atoms dressed by cooling laser photons for the $F = 4 - F' = 3, 4, 5$ transitions. Cooling laser detuning Δ is indicated by the arrows at $\Delta/2\pi = -16$ MHz. The dashed lines indicate the absorption frequencies ($\omega_3/2\pi = -455.2$ MHz, $\omega_4/2\pi = -252.6$ MHz, and $\omega_5 = 0$) for the bare atoms.

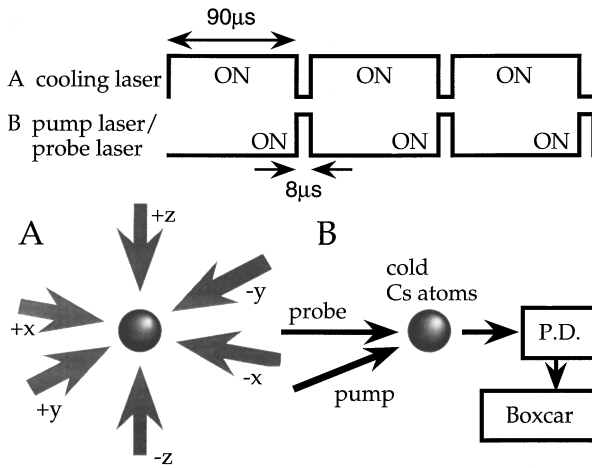


Fig. 2. Timing sequence and schematic of the experimental system; P.D., photodetector. A, Cooling-trapping period when atoms are exposed to three counterpropagating cooling beams; B, pump-probe period when dressed-atom spectroscopy is carried out.

pumping beam from a laser diode, on the other hand, was applied all the time and was resonant to the $F = 3 - F' = 4$ transition. The measured density and size of the trapped Cs atoms were $10^{10}/\text{cm}^3$ and 1 mm^3 , respectively. The temperature was determined by time-of-flight measurement to be $\sim 100 \mu\text{K}$. During the pump-probe period a strong pump beam ($\sim 5 \text{ mW}$, 500 kHz) from a second Ti:sapphire laser and a weak probe beam ($\sim 10 \mu\text{W}$, 100 kHz) from a second laser diode, both chopped by acousto-optic modulators, were focused on the trapped atoms with a lens of 30-cm focal length. Both beams were quasi-collinear, making angles with each other of ~ 20 mrad and with the horizontal cooling beams of 45 deg. The transmitted probe pulses were monitored by a photo-

detector, averaged by a boxcar, and stored in a digitizing oscilloscope as a function of probe frequency ω_p with a fixed pump frequency ω_L .

3. THEORY

Before we show the experimental results, let us apply the dressed-atom theory⁵ for the $F = 4 - F' = 3(|e3\rangle)$, $4(|e4\rangle)$, $5(|e5\rangle)$ transitions, whose frequencies are written as $\omega_3 + \omega_0$, $\omega_4 + \omega_0$, and $\omega_5 + \omega_0$ and where $\omega_3/2\pi = -455.2$ MHz, $\omega_4/2\pi = -252.6$ MHz, $\omega_5 = 0$, and $F = 4$ is denoted $|g\rangle$ hereafter. In the dressed-atom picture the level structure is illustrated in Fig. 3 for the negative detuning case $\Delta < 0$, where Δ is given by $\Delta \equiv \omega_L - \omega_5$. When $\Omega \neq 0$, strong mixing with the pump laser is possible only between $|g, n\rangle$ and $|e5, n-1\rangle$, where n is the pump photon number because it is the only closed system, and splits them by $(\Omega^2 + \Delta^2)^{1/2}$. The new eigenstates are written as $\alpha|g, n\rangle + \beta|e5, n-1\rangle$ and $\alpha|e5, n-1\rangle - \beta|g, n\rangle$, where $\alpha = \cos \theta$, $\beta = \sin \theta$, and $\theta = \tan^{-1}(-\Omega/\Delta)/2$. Now that the ground-state population is mixed up, eight transitions are possible as shown by the arrows in Fig. 3. However, because two of these transitions are those between equally populated levels, six lines should be observed. They are denoted $L3\pm$, $L4\pm$, and $L5\pm$; the peak positions, peak heights, and linewidths of these six lines can be calculated⁵ for given Ω and Δ , and the results are listed in Table 1. Once Ω and Δ are given for a given pump laser, it is easy to simulate the absorption profile numerically simply by summing these six Lorentzians.⁸

Figure 4 shows a numerically simulated three-dimensional plot of the probe absorption spectra based on Table 1 as a function of pump detuning Δ , where $\Delta/2\pi$ is varied from -60 to 60 MHz. Ω is chosen to be 50 MHz. Note that $L3+$, $L4+$, and $L5+$ are the main peaks and $L3-$, $L4-$, and $L5-$ are secondary peaks for negative detuning, $\Delta < 0$. Among these peaks only $L5-$ is a positive peak, implying amplification of the probe beam. As Δ is increased, the main peaks $L3+$ and $L4+$ become smaller while the secondary peaks $L3-$ and $L4-$ become larger, and for $\Delta > 0$ they interchange their roles. Peak $L5+$ starts from a large absorption at $\Delta < 0$, becomes

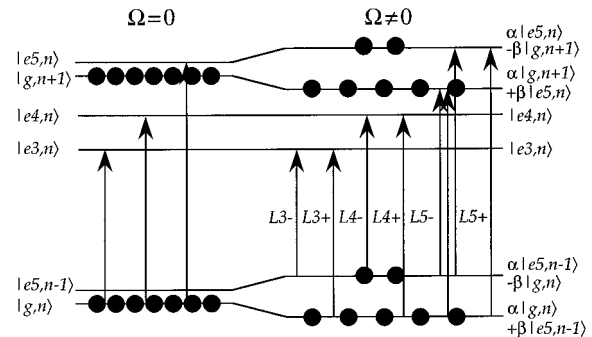


Fig. 3. Energy levels of Cs atoms dressed by the pump laser for $\Omega = 0$ and $\Omega \neq 0$ when $\Delta < 0$. When $\Delta > 0$, the $|g, n\rangle$ level and the $|e5, n-1\rangle$ level switch positions.

Table 1. Line Positions (Peak Frequencies), Peak Heights, and Line Widths of Rabi Sidebands in Cs $F = 4 - F' = 3, 4, 5$ ($L3\pm, L4\pm$ and $L5\pm$) Transitions for Negative Detuning $\Delta < 0$ Based on Dressed-Atom Theory^a

Line	Position	Height	Width
L3-	$\omega_3 + \frac{\Delta}{2} - \frac{\sqrt{\Omega^2 + \Delta^2}}{2}$	$H_3 \frac{\sin^6 \theta}{\cos^4 \theta + \sin^4 \theta}$	$\Gamma(1 + \cos^2 \theta)$
L3+	$\omega_3 + \frac{\Delta}{2} + \frac{\sqrt{\Omega^2 + \Delta^2}}{2}$	$H_3 \frac{\cos^6 \theta}{\cos^4 \theta + \sin^4 \theta}$	$\Gamma(1 + \sin^2 \theta)$
L4-	$\omega_4 + \frac{\Delta}{2} - \frac{\sqrt{\Omega^2 + \Delta^2}}{2}$	$H_4 \frac{\sin^6 \theta}{\cos^4 \theta + \sin^4 \theta}$	$\Gamma(1 + \cos^2 \theta)$
L4+	$\omega_4 + \frac{\Delta}{2} + \frac{\sqrt{\Omega^2 + \Delta^2}}{2}$	$H_4 \frac{\cos^6 \theta}{\cos^4 \theta + \sin^4 \theta}$	$\Gamma(1 + \sin^2 \theta)$
L5-	$\omega_5 + \Delta - \sqrt{\Omega^2 + \Delta^2}$	$-H_5 \frac{\sin^4 \theta (\cos^2 \theta - \sin^2 \theta)}{\cos^4 \theta + \sin^4 \theta}$	$\Gamma(1 + 2 \sin^2 \theta \cos^2 \theta)$
L5+	$\omega_5 + \Delta + \sqrt{\Omega^2 + \Delta^2}$	$H_5 \frac{\cos^4 \theta (\cos^2 \theta - \sin^2 \theta)}{\cos^4 \theta + \sin^4 \theta}$	$\Gamma(1 + 2 \sin^2 \theta \cos^2 \theta)$

^aFor positive detuning $\Delta > 0$, all sin and cos should be interchanged. $\omega_3/2\pi = -455.2$ MHz, $\omega_4/2\pi = -252.6$ MHz, and $\omega_5 = 0$, Γ is the natural linewidth, and $\theta = 1/2 \tan^{-1}[-(\Omega/\Delta)]$; $H_3:H_4:H_5 = 5.47:16.41:34.38$.

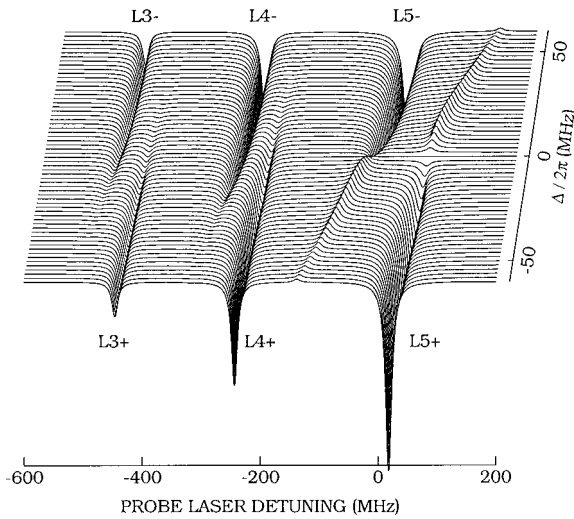


Fig. 4. Three-dimensional plot of numerical simulation of probe absorption spectra dressed by the pump laser photons based on the formula of Table 1. The Rabi frequency is $\Omega = 50$ MHz.

smaller, and finally reaches zero at $\Delta = 0$; then it experiences a maximum gain and finally becomes zero again at $\Delta = +\infty$. Peak L5- evolves exactly an opposite way. The peak positions of six lines form three hyperbolas, which form an anti-crossing at $\Delta = 0$, where separations are given by Ω for $L3\pm$ and $L4\pm$.

4. EXPERIMENTAL RESULTS

Typical absorption spectra as functions of probe laser detuning $\omega_p - \omega_0$ are shown in Fig. 5, along with the theoretical fitting (fitting parameters are $\Omega/2\pi = 55$ MHz and $\Delta/2\pi = -34.8$ MHz). Figure 5 looks very similar to Fig. 1, and this shows that, in spite of the different experimen-

tal configurations, cold Cs atoms dressed by external pump photons in the setup of Fig. 2 give spectra similar to those dressed by cooling laser photons. We can now summarize several spectral features: (1) a blue shift in the absorption peaks (L3+, L4+, and L5+) for three transitions (note that the dotted lines indicate the positions for bare atoms), (2) second absorption peaks (L3- and L4-) in the $F = 4 - F' = 3, 4$ lines (the Autler-Townes doublets^{5,11,12}), (3) strong, broad gain ($\sim 12\%$, hereafter called the dressed-atom gain) at L5- (the Mollow spectrum^{6,7}), and (4) a pronounced dispersion structure with subnatural linewidth at ω_L . The left-hand portion of this dispersion exhibits much stronger gain than the

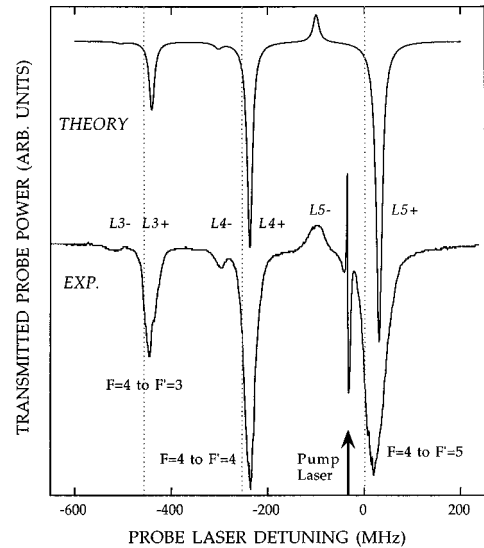


Fig. 5. Experimental (lower) and best-fit theoretical (upper) probe absorption spectra of cold Cs atoms dressed by pump-laser photons. Pump-laser detuning Δ is indicated by the arrow at $\Delta/2\pi = -34.8$ MHz. The dotted lines indicate the absorption frequencies for the bare atoms.

dressed-atom gain (dispersion gain). The agreement of all these features with the dressed-atom theory is reasonably good, except for the dispersion structure. The peak positions are almost perfectly matched with the theory, as discussed below. The linewidths are much broader in the experiment than in the theory, which could be due to a nonuniform magnetic field in the elongated atom cloud by the pump beam. With the cloud size of ~ 1 mm and a magnetic field gradient of ~ 10 G/cm, the line broadening that is due to the inhomogeneous magnetic field is estimated to be ~ 1.4 MHz. This is still a little smaller than what is observed in the experiment. If, however, we consider the pump beam blowing up the cold atoms to make the cloud size much larger than the stationary cloud, the line broadening may be explainable.

To study further the behavior of dressed atoms, we obtained probe transmission spectra for various pump detunings Δ ; we show them in Fig. 6(a). (The uppermost trace is the case of a bare atom, i.e., without a pump beam.) The pronounced dispersion structure can always be observed and, fortunately, this is a convenient and precise marker of the position of Δ . Now we can clearly see the peak shift of each line for various detuning parameters in a similar fashion to that for Fig. 4. The peak positions of the six lines as a function of Δ are plotted in Fig. 6(b), along with the theoretical fit. It is clear that each peak exhibits hyperbolic behavior, with an anticrossing at the resonance point. Moreover, the gain and the absorption of Rabi sidebands for $L5\pm$ switch their signs at resonance. Autler-Townes doublets of $L3\pm$ and $L4\pm$ are now very clear in the central spectrum of $\Delta/2\pi = -2$ MHz. These observations are evidence that the behavior of cold atoms under the influence of a strong pump field is indeed explained nicely by the dressed-atom theory.

Now we shift our attention to the pronounced dispersion structure at ω_L . The peak-to-peak separation in this structure is ~ 2 MHz, narrower than $\Gamma/2\pi (=5.3$ MHz) yet much wider than those that appear in the atomic crystal spectroscopies, which are of the order of 1–100 kHz.^{13,14} This structure is observable only when the pump beam is right or left circularly polarized and, more importantly, the symmetry of the dispersion changes according to the sign of detuning Δ [see Fig. 6(a)], that is, red gain and blue absorption for $\Delta < 0$ and red absorption and blue gain for $\Delta > 0$. (We have not observed any polarization dependence of the probe beam.) Previously, in the Mollow absorption spectrum the appearance of the dispersion structure at the pump frequency was well known and in fact could be theoretically analyzed.^{9,10} However, in those theories the dispersion gave the opposite symmetry (red absorption and blue gain for $\Delta < 0$ and red gain and blue absorption for $\Delta > 0$) and far smaller magnitude. There have also been reports of a subnatural dispersion structure in the probe spectra of trapped atoms.^{2,3,15,16} In those cases, however, the symmetry of the dispersion was the same (red gain and blue absorption), regardless of the sign of detuning Δ .

Our observations should therefore stem from a different mechanism from those mentioned above. The sub-

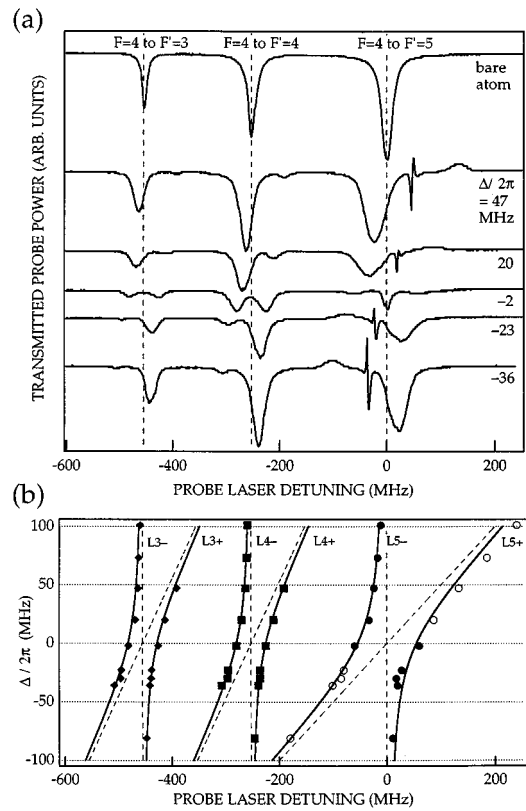


Fig. 6. (a) Probe absorption spectra of cold Cs atoms. Top trace: bare-atom case (no pump beam). Lower five traces: spectra with various detunings Δ . (b) Plot of peak positions in absorption spectra for various detuning parameters Δ . The solid curves are theoretical results with fitting parameter $\Omega/2\pi = 55$ MHz. Open circles, gain; filled diamonds, squares, and circles, absorption of $L3\pm$, $L4\pm$, and $L5\pm$ lines, respectively.

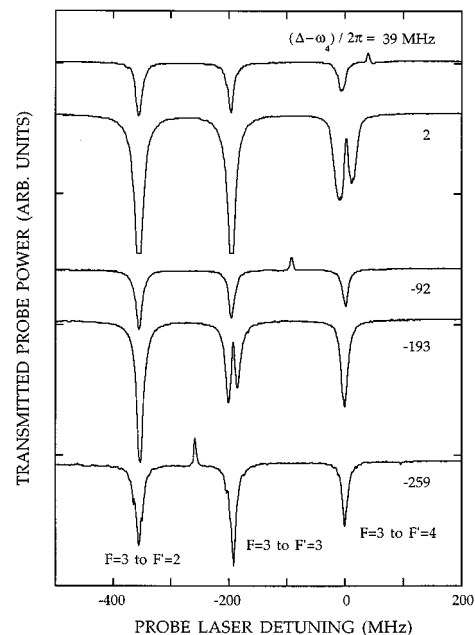


Fig. 7. Probe absorption spectra of cold Cs atoms for the $F = 3 - F = 2, 3, 4$ transitions with various pump detunings Δ .

natural width can be explainable if it is due to a Λ -type stimulated Raman process with a small splitting in the ground-state $F = 4$ level. In this model for σ_+ (σ_-) light, the most populated $m_F = 4(-4)$ level becomes the lowest level when $\Delta < 0$ and the highest when $\Delta > 0$. This qualitatively explains our observations.

A similar type of stimulated Raman gain was clearly observed in the probe absorption spectra for the $F = 3 - F' = 2, 3, 4$ transitions with the pump beam on, as shown in Fig. 7. Here the positive peak indicates the stimulated Raman gain, and it was observed when the pump frequency ω_L and the probe frequency ω_p satisfied the condition $\omega_L - \omega_p = 9.2$ GHz, i.e., the hyperfine splitting of the ground-state $F = 3$ and $F = 4$ states. This Raman gain implies that the $F = 4$ level is more populated than the $F = 3$ level because of the strong re-pumping beam. In particular, when ω_L is resonant to the $F = 4 - F' = 4$ (the second trace from the top in Fig. 7) or the $F = 4 - F' = 3$ transition (the fourth trace from the top in Fig. 7), the absorption signal becomes quite large because of the real population transfer from the $F = 4$ to the $F = 3$ level, and we have transparency within the absorption profile.

5. SUMMARY

In summary, by dividing time into a cooling-trapping period and a pump-probe period we have successfully performed pump-probe spectroscopy of the $F = 4$ transitions of cold Cs atoms in a versatile manner. The observed spectra were in reasonable agreement with the dressed-atom theory, except for the pronounced dispersion structure. Strong gains were observed at both ω_L (dispersion gain) and $\omega_L + \sqrt{\Omega^2 + \Delta^2}$ or $\omega_L - \sqrt{\Omega^2 + \Delta^2}$ (dressed-atom gain), depending on whether $\Delta > 0$ or $\Delta < 0$. The large dispersion gain can be explained in terms of a stimulated Raman process from Zeeman sublevels that are light shifted and optically pumped by the pump beam, although it should be investigated further in depth. A similar type of stimulated Raman gain was observed when pump and probe frequencies differed by the hyperfine splitting frequency of the ground state. This type of time-division spectroscopy can open the door to any type of nonlinear spectroscopy, such as phase conjugation and the transient grating method in cold atoms.

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