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## Degenerate Four-Wave Mixing in Cold Cesium Atoms Using a Noncycling Transition

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We present a detailed investigation on the generation of four-wave mixing optical phase conjugation using a noncyling transition of cold cesium atoms obtained from a four-beam magneto-optical trap. In particular, we study the efficiency of the four- wave mixing signal as a function of the density of trapped atoms as well as the intensity of the trapping beams. A simple theoretical model which accounts explicitly for the relaxations between the two hyperfine cesium ground states in the trap environment is in qualitative agreement with some of the observed results.

Resonant four-wave mixing (FWM) is a powerful nonlinear spectroscopic technique which has been widely employed to investigate a variety of nonlinear optical media<sup>[1]</sup>. In a low-density Doppler-broadened vapor, FWM can be used to observe sub-Doppler resonances as well as to study saturation effects associated with the third order nonlinear susceptibility  $\chi^{(3)}$ of the medium<sup>[2,3]</sup>. With the phenomenal development achieved in the field of laser cooling and trapping of neutral atoms, samples of atoms with high densities and very low temperatures can now be easily obtained. These samples of cold atoms offer the possibility to perform FWM in a completely new domain. For instance, degenerate and nearly degenerate fourwave mixing (DFWM and NDFWM) were recently employed to study the quantized atomic motion of ultracold atoms in an optical lattice<sup>[4,5]</sup>. Differently from a</sup> room temperature atomic vapor, where just a small velocity group within the sample contributes to the FWM signal, with cold atoms this contribution comes from all the atoms. Therefore the observed signal is greatly enhanced and broadening mechanism such as transit-time and second order Doppler effect are practically eliminated.

DFWM has been observed in many experiments that utilize alkali atoms, either in a thermal vapor<sup>[6]</sup> (and references therein), or with cold atoms in a magneto-optical trap (MOT)<sup>[7]</sup>. However, mainly these experiments were done using the cycling transition of the alkali atom. It is well known that optical pumping prevents the observation of DFWM in a noncycling transition, i.e., a transition which does not conserve the total population. Although some previous methods have been developed to overcome this limitation<sup>[8,9]</sup>, we have shown that the MOT is a very appropriate medium to observe DFWM optical phase conjugation using a noncycling transition<sup>[10]</sup>. In this work, we present a detailed investigation of the DFWM signal using a noncycling transition of cold cesium atoms.

Our experiment was performed using cold atoms obtained from a four-beam magneto- optical trap as described previously in reference [11]. However, in order to increase the number of trapped atoms, we have added to this trapping scheme one pair of molasses beams, with parallel linear polarization, along the transverse direction<sup>[12]</sup>. Fig. 1-(a) shows the hyperfine splitting of the cesium  $6S_{1/2}$  and  $6P_{3/2}$  states and indicates the relevant energy levels for trapping and for performing DFWM optical phase conjugation. The trapping beams are supplied by a stabilized Ti:Sapphire laser and are red-detuned by about 12 MHz from the resonance frequency of the cesium cycling transition  $6S_{1/2}$ ,  $F=4\leftrightarrow 6P_{3/2}$ , F'=5 at  $\lambda = 852$ nm. A long external cavity diode laser<sup>[13]</sup> (the repumping laser), tuned into res-

onance with the  $6S_{1/2}$ , F=3-  $6P_{3/2}$ , F'=3 or 4 transition, recycles the population lost to the hyperfine level  $6S_{1/2}$ , F=3 of the cesium ground state. The trap is loaded directly from a vapor cell at room temperature. Typically the number of trapped atoms, estimated by measuring the fluorescence emitted by the atomic cloud using a calibrate photodiode, is about  $10^7$  atoms.



Figure 1. a) Relevant energy levels of Cs involved in the trapping and in the DFWM process. b) Experimental geometry used to generate the DFWM optical phase conjugation. D is a photodiode to monitor the DFWM signal.

Due to the large hyperfine ground state splitting, the presence of the DFWM beams around the noncycling transition  $6S_{1/2}$ ,  $F=4 \leftrightarrow 6P_{3/2}$ , F'=4 will strongly increase the optical pumping rate to the non-interacting ground state  $6S_{1/2}$ , F=3, therefore leading to a reduction in the observed signal. Nevertheless, as we will show the presence of the repumping beam can accounts for the increase in the optical pumping mechanism making possible both the trapping around the cycling transition and the observation of the DFWM signal in the noncycling transition.

In Fig.1-(b) we show the experimental beam geometry for observing the DFWM signal. The forward (F) and backward (B) pumping-beams and the probe (P) beam are all provided from the same grating stabilized diode laser which has a short term frequency jitter of about 1 MHz. All the beams have the same linear polarization and the angle between the F and P beams is of order of  $\theta = 3^{\circ}$ . To prevent optical feedback from the backward beam, we used an optical isolator in the output of the diode laser. A  $\lambda/2$  plate placed before the optical isolator allowed us to vary the total power of the DFWM beams. The DFWM phase (PC) conjugate beam which propagates in the opposite direction of the probe beam is reflected by a 50-50 beam splitter and detected directly with a photodiode without the need of employing any synchronous detection system.



DFWM Laser Frequency

Figure 2. DFWM phase conjugate spectrum showing the peaks around the cycling  $F=4\leftrightarrow F'=5$  and the noncyling  $F=4\leftrightarrow F'=4$  cesium transitions.

In Fig. 2 we show the DFWM phase conjugate intensity versus the degenerate laser frequency  $\omega_L$ . For this spectrum the power of the forward, backward and probe beams are respectively  $P_F \simeq P_B \simeq 120 \mu W$  and  $P_P \simeq 40 \mu W$  and all the beams have diameters comparable with the trap size (~ 1mm). As indicated in the figure, the peaks are associated with the cycling  $F=4\leftrightarrow F'=5$  and the noncycling  $F=4\leftrightarrow F'=4$  transitions and for these values of the DFWM laser intensities the peaks have approximately the same aplitude. However, for a much higher laser intensities the spectrum changes drastically with a great enhancement of the signal around the cycling transition and with a corresponding reduction around the noncycling transition due to optical pumping. Typical value of the measured phase conjugate reflectivity in the noncycling transition is about 1%. In the spectrum presented in Fig. 2 the total trapping laser power is about 80 mW and it corresponds to an eight scan averaging. In this spectrum there is a power broadening which is mainly associated with the strong trapping beams. We have studied the dependence of the DFWM signal with the total power of the trapping beams and the observed result is shown in Fig. 3 for both types of transitions. As expected, the DFWM signal has a different behavior for the cycling and the noncycling transitions as a function of the trapping beams power. For low trapping power the signal around the noncycling transition is much smaller than the signal at the cycling transition. This can be atributed to the fact that for a fixed trapping power the DFWM beams only leads to a significant decrease in the number of trapped atoms contributing to the signal when its frequency is resonant with the noncyling transition. While the number of trapped atoms tends to saturate for high trapping powers, we see from our data that in this limit, the DFWM signal around the cycling transition saturates more rapidly as compared to the signal in the noncycling transition. In the cycling transition the DFWM signal is more strongly saturated due to the presence of the trapping beams, nearly resonant with this transition. However, a complete theoretical understanding of the observed dependence is rather complicated since we have to account for the manifold of dressed states created by the strong trapping beams.



Figure 3. Dependence of the DFWM signal as a function of the total trapping laser power: • - cycling transition;  $\blacksquare$  - noncycling transition. The trapping lasers are red-detuned by about 12 MHz from resonance.

As we have mentioned, the repumping laser plays a fundamental role for the observation of DFWM around the noncycling transition. Moreover, in the MOT the repumping laser determines the capture rate of the cooling process thereby controlling the steady state population of the MOT. Concentrating our attention on the noncycling transition  $F=4\leftrightarrow F'=4$ , where the strong trapping beams essentially only affects the population of the  $6S_{1/2}$ , F=4 ground state, we can use a simple rate equation model to calculate the populations  $N_{F=3}$  and  $N_{F=4}$  of the two hyperfine ground states  $6S_{1/2}$ , F=3 and  $6S_{1/2}$  F=4 respectively<sup>[14]</sup>. The essential feature of our treatment can be obtained considering relaxations only between these two states. Thus, if  $R_r$  and  $R_d$  represent, respectively, the repumping rate, associate with the repumping beam, and the depumping rate (or optical pumping rate) due to the DFWM beams, we obtain

$$\frac{N_{F=4}}{N_{F=3}} = \frac{R_r}{R_d} \frac{\tau_{F=4}}{\tau_{F=3}} , \qquad (1)$$

where  $\tau_{F=4}$  and  $\tau_{F=3}$  are the corresponding lifetime of each ground state level. We should note that  $\tau_{F=4}$  and  $\tau_{F=3}$  are determined by the intensity of the DFWM and repumping beams respectively. However,  $\tau_{F=4}$  has an upper limit determined by the off-resonant excitation rate associated with the trapping beams. If we assume that the total population of trapped atoms,  $N_0 =$  $N_{F=3} + N_{F=4}$  is constant, we promptly obtain that the DFWM signal around the noncycling transition, will be given by

$$I_{NC} \sim N_0^2 \left(\frac{R_r}{R_r + R_d}\right)^2 \tag{2}$$

We have used this simple model to analyze the dependence of the DFWM signal in the noncycling transition with the number of trapped atoms in the F =4 ground state, which is determined by the intensity of the repumping beam. Our experimental results are shown in Fig. 4, where we have plotted the DFWM intensity as a function of the repumping beam power. In this figure, the solid curve corresponds to the best fitting using equation (2). As we can see, our simple two-level model describes very well the observed dependence. It is worth to note that equation (2) also predicts the decreasing of the population  $N_{F=4}$  with the intensity of the DFWM beams.

5 DFWM Intensity (arb. units) 0 2 3 5 6 Repumping Laser Power (mW)

Figure 4. Dependence of the DFWM signal around the noncycling transition as a function of the power of the repumping beam. The solid curve corresponds to the best fitting using the dependence predicted by eq.(2).

In summary, we have investigated the generation of DFWM in cold cesium atoms using a noncycling transition. Although the results we have presented in this paper correspond to a small aperture angle between the forward and the probe beams, we have been able to observe efficient DFWM optical phase conjugation for aperture angle as large as 90°. This considerably increases the possibility of application of the DFWM phase conjugate signal, either in spectroscopy and in real-time holography. Another related experiment which is being pursued in our laboratory is devoted to investigate the detailed process of optical pumping induced by the DFWM beams in the MOT environment. For instance, we have observed that under appropriate condictions a single laser beam resonant with the noncycling transition can continuously lead to the production of an appreciable number of cold atoms in the lower hyperfine ground state  $6S_{1/2}$ , F=3. This could be of significant interest in the study of cold collisions involving hyperfine level changing.<sup>[15]</sup>

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