High-contrast dark resonances with linearly polarized light on the D_1 line of alkali atoms with large nuclear spin

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High-contrast coherent population trapping signals were observed on the Cs D_1 line by use of a bichromatic linear polarized light (lin||lin field). A maximum absorption contrast of about 10% was obtained. This was nearly twice as high as that measured with the standard configuration of bichromatic circularly polarized light ($\sigma - \sigma$ field). The results are compared with density matrix calculations of 4 and 5 level systems. © 2009 Optical Society of America

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1. Introduction

Coherent population trapping (CPT) [1,2] resonances have recently gained renewed interest, partly through the development of chip-scale atomic clocks, such as microfabricated atomic clocks [3,4]. High contrasts and narrow resonance linewidths are important factors for the applications of CPT-based atomic frequency references [5,6]. For conventional CPT interrogation with bichromatic circularly polarized light ($\sigma - \sigma$ field), the signal amplitudes can be limited by the loss of atoms toward the extreme Zeeman sublevels $|F, m_F = \pm F\rangle$ through optical pumping. The atoms that accumulate in these states contribute to neither the CPT signal nor the background absorption. To reach the same net absorption, the vapor temperature could be increased. In many cases, however, this is not desirable, because of increased resonance broadening through spin-exchange collisions and higher power consumption.

Much effort has been dedicated to maximizing the resonance contrast without significant line broadening. It has been shown that excitation of the D_1 transition instead of the D_2 transition results in both a higher resonance contrast and a narrower resonance width [7,8]. Other approaches to increase the contrast include push–pull optical pumping with alternating circular polarizations [9], counterpropagating waves with orthogonal circular polarizations [10], interrogation with crossed linear polarizations [11], and four-wave mixing [12]. These methods excite a two-photon Λ resonance on the ground-state hyperfine sublevels with $|F, m_F = 0\rangle$, $|F+1, m_F = 0\rangle$ to minimize the frequency shift due to the magnetic field. However, most of these configurations are complex to implement.

Recently, the observation of high-contrast dark resonances was reported in the most simple setup possible: Taichenachev *et al.* [13] and Kazakov *et al.* [14] proposed to measure CPT resonances with a bichromatic linearly polarized light (lin||lin field) on the D_1 line of ^{87}Rb (with nuclear spin $I = 3/2$). They demonstrated that two Λ schemes can be formed

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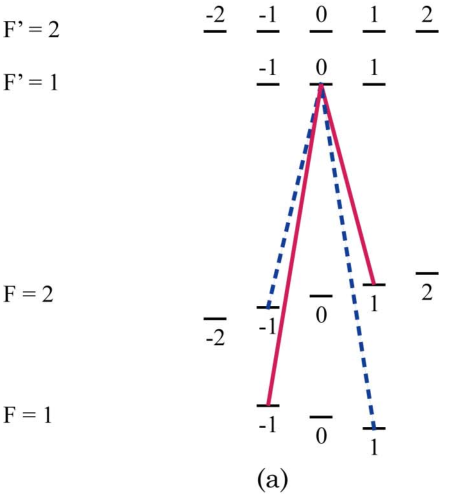
with two pairs of ground-state hyperfine sublevels simultaneously: $|F=1, m_F=-1\rangle$, $|F=2, m_F=1\rangle$ and $|F=1, m_F=1\rangle$, $|F=2, m_F=-1\rangle$ coupled with the common excited states $|F'=1, m_F=0\rangle$ [see Fig. 1(a)]. Very high CPT resonance contrasts were measured in this case, because the ground-state Zeeman components involved in the Λ system cannot couple to any other excited states by single-photon transitions. If the contribution of the nuclear spin to the Zeeman splitting is neglected, the two-photon resonance frequencies (both $|F=1, m_F=-1\rangle$, $|F=2, m_F=1\rangle$ and $|F=1, m_F=1\rangle$, $|F=2, m_F=-1\rangle$) are equal to the frequency of the 0–0 resonance formed on $|F=1, m_F=0\rangle$, $|F=2, m_F=0\rangle$. This means that at least two superposition dark states exist in the case of exact two-photon resonance for fields with linear polarizations. A CPT resonance on the ground-state hyperfine sublevels with $|F, m_F=0\rangle$, $|F+1, m_F=0\rangle$ is not excited in the case of the lin||lin configuration, because the different components of the circularly polarized light interfere destructively [15].



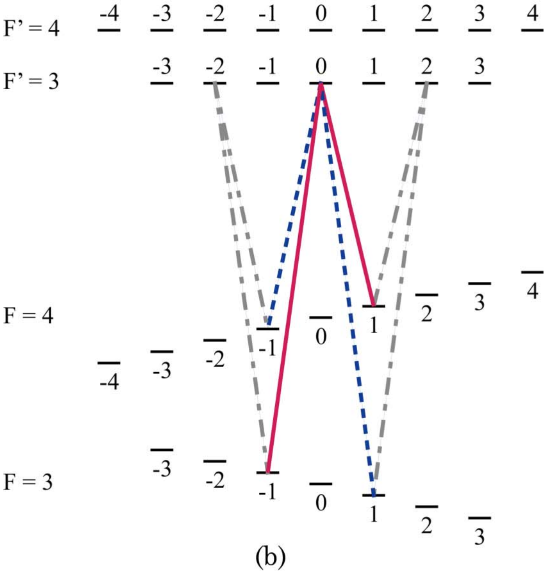


Fig. 1. (Color online) Excitation scheme with a lin||lin field on the D_1 line of (a) ^{87}Rb and (b) Cs. (a) CPT resonances in ^{87}Rb involve two pairs of ground-state hyperfine sublevels with $|F=1, m_F=-1\rangle$, $|F=2, m_F=1\rangle$ (solid lines) and $|F=1, m_F=1\rangle$, $|F=2, m_F=-1\rangle$ (dashed lines) coupled with the common excited states $|F'=1, m_F=0\rangle$. (b) CPT resonances in Cs involve two pairs of ground-state hyperfine sublevels with $|F=3, m_F=-1\rangle$, $|F=4, m_F=1\rangle$ (solid lines) and $|F=3, m_F=1\rangle$, $|F=4, m_F=-1\rangle$ (dashed lines) coupled with the common excited states $|F'=3, m_F=0\rangle$. Additional single-photon resonances are possible in the case of Cs, indicated by the alternate long and short dashed lines.

Unfortunately as pointed out in [13], this is possible only for atoms with nuclear spin $I=3/2$ on the $F'=1$ excited state. For higher angular momenta (i.e., $F'>1$) the ground-state levels $|F, m_F=-1\rangle$, $|F+1, m_F=-1\rangle$ and $|F, m_F=1\rangle$, $|F+1, m_F=1\rangle$ can also couple to the excited states $|F', m_F=-2\rangle$ and $|F', m_F=2\rangle$ and form W and M systems [see Fig. 1(b)]. This usually reduces the contrast of the CPT resonances. It was therefore expected that the CPT resonance contrast on the Cs D_1 line ($I=7/2$) is very small, due to the high nuclear spin of the Cs. Nevertheless, the CPT signal with absorption contrasts around 10% could be observed on the D_1 line of Cs atoms by use of the lin||lin configuration, even higher than the ones measured for the classic case with circularly polarized light fields. In Sections 2 and 3 the details of these CPT signals using lin||lin field excitation are described.

2. Experiments

The excitation scheme is shown in Fig. 1(b). A laser beam with a linear polarization was transmitted through a vapor of cesium atoms in a magnetic field parallel to the beam direction. Therefore, σ^+ and σ^- transitions could be excited. Two two-photon resonances of a Λ type are formed involving two pairs of ground-state hyperfine sublevels: $|F=3, m_F=-1\rangle$, $|F=4, m_F=1\rangle$ and $|F=3, m_F=1\rangle$, $|F=4, m_F=-1\rangle$. Both Λ schemes are excited through the common excited state $|F'=3, m_F=0\rangle$.

Figure 2 shows the experimental setup. The two optical fields needed for the CPT signals were derived by phase modulating the output of a single extended cavity diode laser (ECDL) in the vicinity of the D_1 line of Cs ($6^2S_{1/2} \rightarrow 6^2P_{1/2}$, $\lambda \approx 894.6\,\mathrm{nm}$). A waveguide-type electro-optic modulator (EOM) modulated the phase of the optical field with a frequency around 4.6 GHz, half of the ground-state hyperfine splitting frequency of Cs. The laser frequency was tuned such that the two first-order sidebands were resonant with the transitions to the $F'=3$ excited state. The phase modulated laser beam passed a linear polarizer with an extinction ratio of 50 dB. A vapor cell 25 mm in diameter and 25 mm long was used in the measurements at room temperature containing Cs and 10 Torr of N_2 buffer gas. The cell was surrounded by a magnetic shield and placed inside a solenoid that applied a longitudinal magnetic field of the order

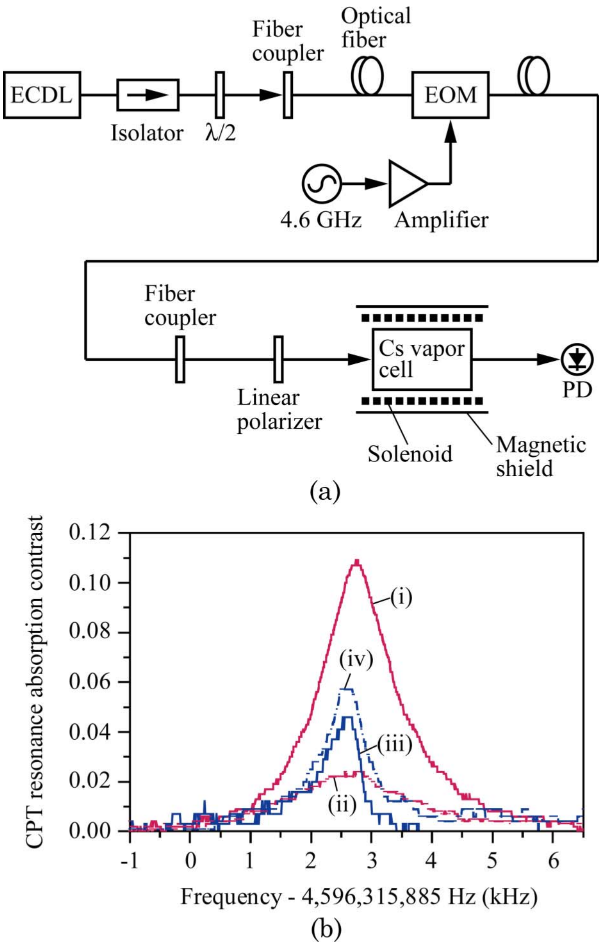


Fig. 2. (Color online) (a) Experimental arrangement used to observe the CPT phenomenon on the D_1 line of Cs atoms. ECDL, extended cavity diode laser; EOM, electro-optic modulator; PD, photo-detector. (b) CPT resonance absorption contrast with a total laser power of 2.1 mW: (i) lin||lin excitation through a common excited state $|F'=3\rangle$, (ii) lin||lin excitation through a common excited state $|F'=4\rangle$, (iii) $\sigma-\sigma$ excitation through a common excited state $|F'=3\rangle$, (iv) $\sigma-\sigma$ excitation through a common excited state $|F'=4\rangle$.

of $10\,\mu$T to lift the Zeeman energy levels' degeneracy and to separate the "clock" resonance, which has no first-order magnetic field dependence, from the field-dependent resonances. To measure the CPT resonances, the EOM frequency was scanned over a range of 10 kHz around 4.596315885 GHz. The measured beam diameter was 19 mm. The optical power transmitted through the gas cell was then detected with a Si photodiode. We performed a comparative study of four different excitation schemes of CPT resonance: (i) $|F=3\rangle\to|F'=3\rangle$, $|F=4\rangle\to|F'=3\rangle$ with lin||lin field; (ii) $|F=3\rangle\to|F'=4\rangle$, $|F=4\rangle\to|F'=4\rangle$ with lin||lin field; (iii) $|F=3\rangle\to|F'=3\rangle$, $|F=4\rangle\to|F'=3\rangle$ with $\sigma-\sigma$ field; (iv) $|F=3\rangle\to|F'=4\rangle$, $|F=4\rangle\to|F'=4\rangle$ with $\sigma-\sigma$ field. A quarter-wave plate was used in front of the Cs cell when the laser radiation was circularly polarized.

3. Results and Discussion

The CPT amplitude, the absorption contrast of the resonance, and the full width at half-maximum (FWHM) linewidth of the four different excitation schemes are shown in Fig. 3. The absorption contrast is defined as the ratio of the change in light absorption due to the CPT resonance to the absorption off CPT resonance and gives us an idea of the "darkness" of the coherent dark state, i.e., the fraction of atoms that become "dark" when the two-photon resonance condition is fulfilled. It can be seen that the CPT absorption contrast for lin||lin excitation keeps increasing with higher laser intensities, while it reaches a maximum much earlier in the $\sigma-\sigma$ schemes [16]. The highest absorption contrast for larger laser intensities was reached in the lin||lin case with the common excited state $|F'=3\rangle$. At nearly 10%, it was almost twice as high as the

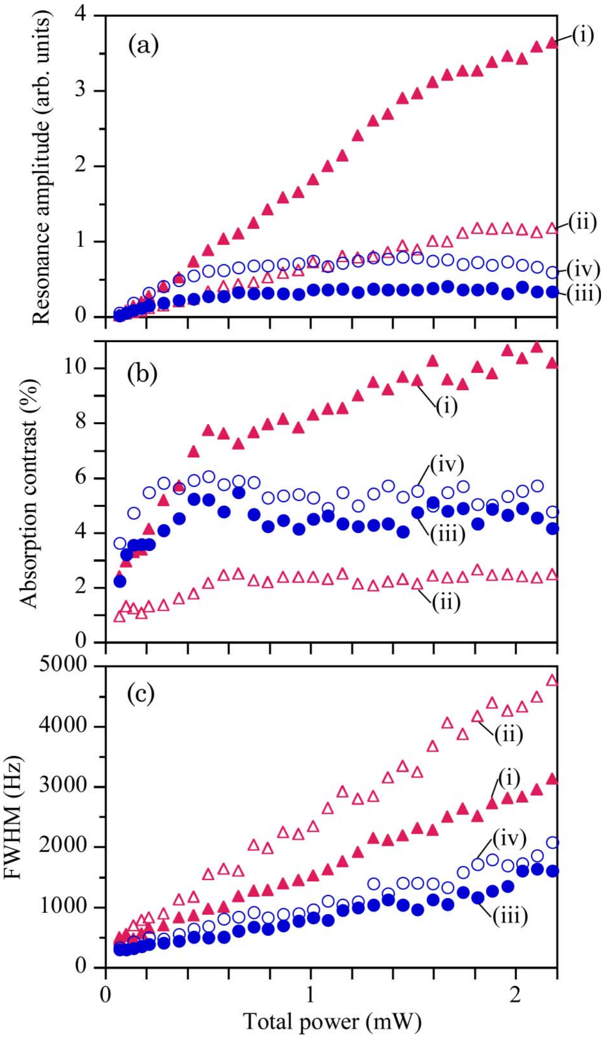


Fig. 3. (Color online) CPT (a) resonance amplitude, (b) absorption contrast, and (c) full width at half-maximum (FWHM): (i) lin||lin excitation through a common excited state $|F'=3\rangle$ (solid triangles), (ii) lin||lin excitation through a common excited state $|F'=4\rangle$ (open triangles), (iii) $\sigma-\sigma$ excitation through a common excited state $|F'=3\rangle$ (solid circles), (iv) $\sigma-\sigma$ excitation through a common excited state $|F'=4\rangle$ (open circles).

maximum contrast measured in the $\sigma-\sigma$ case. Considering the fact that Cs has 16 ground-state Zeeman levels, an absorption contrast of 10% means that a large fraction of atoms accumulates in the dark states and the destruction of the dark states due to coupling to the other excited states is not as significant, as we expected. A slight degradation of the CPT contrast and linewidth could be caused by off-resonant excitations to the excited $F'=4$ state, which is separated from the $F'=3$ state by 1.1 GHz. This splitting is considerably larger than both the Doppler width (~400 MHz) and the homogenous linewidth broadened by collisions with the buffer gas (~200 MHz), but could nevertheless contribute to some destructive interference. Furthermore, we want to note that in both excitation schemes, magnetically sensitive resonances can be observed, for example, those involving the ground states $|F=3, m_F=-1\rangle$ and $|F=4, m_F=-1\rangle$. The contrast of these resonances was almost the same as those of the clock resonances.

It can be seen from Fig. 3 that the linewidth of CPT resonance in the lin||lin case broadens roughly linearly with increased laser intensity. As expected, they are slightly broader than the widths in the $\sigma-\sigma$ case [17]. The broadening for the lin||lin fields caused by the splitting of the two CPT resonances in the magnetic field can be neglected for higher laser intensities. Through the nuclear contribution to the g factors of the hyperfine components, the two Λ resonances shift apart at a rate of 22 Hz/μT. In a 10 μT field this splitting is therefore roughly 220 Hz. Although the frequency splittings of $|F=3, m_F=-1\rangle$, $|F=4, m_F=1\rangle$ and $|F=3, m_F=1\rangle$, $|F=4, m_F=-1\rangle$ are slightly different, their position is to first order symmetrical relative to the hyperfine splitting frequency. Thus, in the case of the lin||lin excitation scheme, the presence of a small magnetic field leads only to broadening of the resonance and not to a shift [13]. Also, the neighboring CPT resonances did not overlap because the resonances split about 70 kHz in the magnetic field of 10 μT. It can be seen that in the lin||lin case, the resonance amplitude and width both increase roughly linearly for a large range of laser intensities, while the amplitude in the circular case reaches a maximum at much lower intensities. This can allow for nearly constant resonance slopes for a wide range of intensities in the lin||lin case with increased bandwidths, which could be of interest for applications such as atomic clocks less sensitive to accelerations and atomic magnetometers.

Finally, we would like to point out that the contrasts measured for the lin||lin case, when coupled to the excited $F'=4$ state, are much lower than all the other cases investigated for Cs. Taichenachev *et al.* found similar behavior in ^{87}Rb [13]. While this can be intuitively understood in the ^{87}Rb case, it is not necessarily obvious in the case of Cs. In ^{87}Rb, there is a fundamental difference between the schemes for the two excited states, because single-photon transitions couple the Λ systems to the $F'=2$ state, but not the $F'=1$ state [see Fig. 1(a)]. For Cs however, for both excited states $F'=3$ and 4, these single-photon transitions are possible, and it is more surprising that such different contrasts are measured for the two excited states.

To gain some insight into these surprising results, density matrix calculations were performed for two simplified systems. For the conventional $\sigma-\sigma$ case, a 4-level model was used similar to the one of Vanier *et al.* [16]. Two ground states $|F=3, m_F=0\rangle$, $|F=4, m_F=0\rangle$ couple to a common excited state $|F'=3,4, m_F=1\rangle$ and a fourth ground state $|F=4, m_F=4\rangle$ acts as a "trap", as shown in Fig. 4(a). The parameters were chosen to resemble the experimental conditions: the population and coherence decay rates are $\gamma=5$ MHz and $\Gamma=200$ MHz, respectively, for the excited state and $\gamma_{12}=5$ MHz and $\Gamma_{12}=200$ MHz for the ground states. The vapor is assumed to be optically thin and the atom at rest. One laser is tuned on resonance with the transition state $|F=3\rangle \rightarrow |F'\rangle$, while the second laser frequency is scanned through the Raman resonance. The ratios of Rabi frequencies are kept constant proportional to the dipole matrix elements. The density matrix was solved numerically as a function of two-photon Raman detuning to produce a spectrum of the CPT resonance. These resonances have been fitted with Lorentzian line shapes similar to the fits performed on the experimental data.

Even though this is a very simplified model of the complicated Cs atom, it has been shown to predict

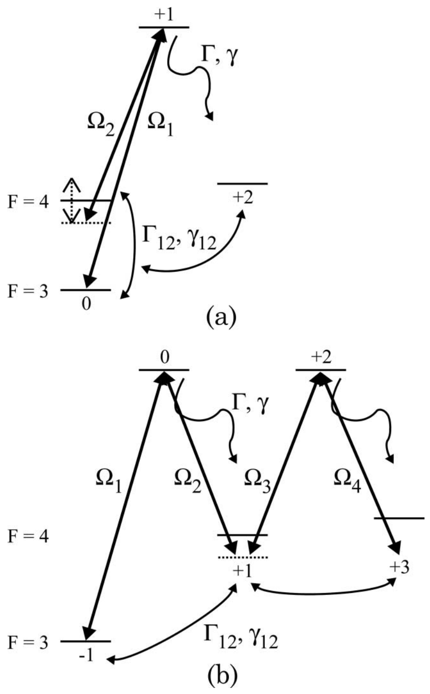


Fig. 4. Energy level diagrams used for modeling: (a) the 4-level system was used for the $\sigma-\sigma$ excitation scheme, where the $|F=4, m_F=4\rangle$ serves as a "trap" state; (b) the 5-level system was used for the lin||lin excitation scheme.

the dependence of the dark line amplitude and width well [16]. The results shown in Figs. 5(a) and 5(b) (filled circles for the $|F' = 3\rangle$ excited state and open circles for $|F' = 4\rangle$). Both excited states give very similar results although the $F' = 4$ state gives slightly higher resonance amplitudes and linewidths. For both cases, the amplitudes saturate, as more atoms are pumped into the "trap" state.

The lin||lin case was modeled as a 5-level M system as shown in Fig. 4(b). The decay rates were identical to the ones in the case with circularly polarized light. Figure 5 shows the results of the numerical solutions (filled triangles for $|F' = 3\rangle$ and open triangles for $|F' = 4\rangle$). It can be seen, that the model predicts no saturation of the amplitudes in this case. Furthermore, the results for both excited states in this case are not equal, but the $F' = 3$ states yields much higher amplitudes and narrower linewidths than the $F' = 4$ case. This is in agreement with the experimental results. Since two different very simplified models were used for the two systems, the absolute amplitudes are not necessarily comparable and the models can only give qualitative results. To make more quantitative predictions, a calculation of the full 32-level system would be useful, which is beyond the scope of this paper. Nevertheless, we can get some hints why the lin||lin case results in high resonance amplitudes by varying the coupling strengths $\Omega_3 \rightarrow \mathrm{Rp} \times \Omega_3$ and $\Omega_4 \rightarrow \mathrm{Rp} \times \Omega_4$ of the transitions to the additional excited state $|F' = 3, 4, m_F = 2\rangle$ through a common scaling factor Rp. In the limit $\mathrm{Rp} \rightarrow 0$, the system collapses to a 4-level system with a trap and an extra uncoupled excited state. For $\mathrm{Rp} \rightarrow 1$, it is the 5-level system used previously. For very low Rabi frequencies Ω_1 and Ω_2, the resonance amplitudes stay constant for low Rp and drop, as Rp increases, since more atoms are pumped out of the dark state into the additional states. For high Rabi frequencies Ω_1 and Ω_2 however, the resonance amplitudes can dramatically increase with increasing Rp. They reach a maximum around $\mathrm{Rp} = 0.8$ and decrease afterward. This suggests that, in the case of strong pumping, a large fraction of atoms can get trapped in the noncoupled "trap" state and the additional light fields can repump these atoms back into the states that can contribute to a CPT resonance. This repumping could outweigh the additional decoherence caused by the fields for moderate rates of Rp. This could be one possible explanation for the higher resonance amplitudes in the lin||lin configuration compared to the conventional sigma one.

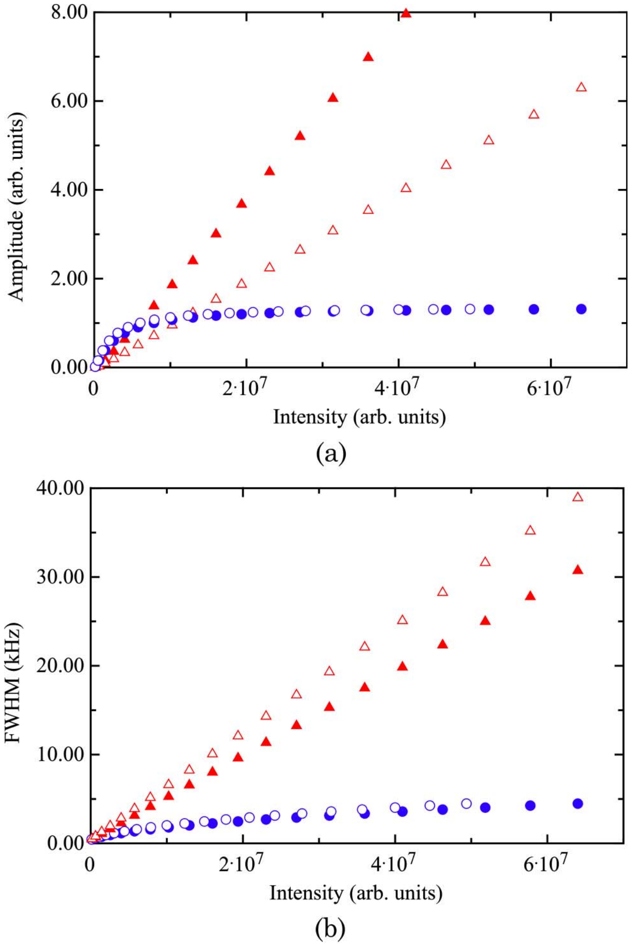


Fig. 5. (Color online) Theoretical results from the numerical density matrix calculations: (a) resonance amplitude and (b) resonance linewidth as a function of laser intensity: lin||lin excitation through a common excited states $|F' = 3\rangle$ (solid triangles) and $|F' = 4\rangle$ (open triangles). The amplitudes for the $\sigma - \sigma$ excitation through a common excited states $|F' = 3\rangle$ (solid circles) and $|F' = 4\rangle$ (open circles) are scaled by a factor of 10 for better visibility.

4. Conclusion

We have experimentally realized a high-contrast dark resonance using a scheme of excitation of the D_1 line of Cs with nuclear spin $I = 7/2$ for lin||lin light fields. A maximum absorption contrast of 10% was obtained, which was higher than that observed in the conventional $\sigma - \sigma$ case.

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