# Nonlinear spectroscopy and optical phase conjugation in cold cesium atoms

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We have employed a sample of cold cesium atoms in a magneto-optical trap to study nonlinear spectroscopy and four-wave-mixing optical phase conjugation using a noncycling transition. Reflectivities of the order of 1% have been measured for moderate levels of pump power and large angular aperture. In particular, we have performed nonlinear spectroscopy in a three-level V-type system and observed directly the ac Stark splitting of the cesium $6S_{1/2}$, $F=4$ ground state. [S1050-2947(97)08903-8]

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The ability to cool and confine neutral atoms at very low temperatures has open new avenues to perform nonlinear optical experiments in a domain where Doppler and transit broadening can be strongly reduced, thus allowing the realization of extremely high-resolution measurements [1–5]. In this domain a number of nonlinear optical effects, which are often hidden in thermal atomic vapors due to the velocity average, can be easily evidenced. Another major difference associated with the atom-laser interaction is related to the fact that with cold atoms all the atoms contribute efficiently to the signal, whereas in a Doppler-broadened medium only a small group of velocities are resonant with the laser light therefore leading to a reduction in the observed effect. Also, as we will describe later, cold atoms in a magneto-optical trap (MOT) is a very appropriate medium to perform wave-mixing spectroscopy using noncycling transitions (i.e., a transition which does not conserve the total population). It is well known that optical pumping prevents the observation of degenerate four-wave mixing (DFWM) using noncycling transitions in alkali metals [6]. Although some methods have been developed to overcome this limitation [7,8], we are able to observe DFWM signals with an efficiency several orders of magnitude higher in the MOT. This consequently increases the applicability of DFWM either as a spectroscopic tool or as a sensitive method to investigate other physical phenomena, such as optical pumping [9] and atomic diffusion [10].

In this work we have employed a sample of cold cesium atoms obtained from a MOT to investigate four-wave-mixing optical phase conjugation using a noncycling transition, and in particular to perform nonlinear spectroscopy in a three-level $V$-type system. Although other recent works have reported on the observation of DFWM and nearly DFWM in cold atoms [11–13], those experiments were done using a cycling transition, where depletion of population of the interacting ground state due to optical pumping is strongly reduced. On the other hand, since most of the trapping schemes employ the same cycling transition to cool and confine the atoms, one often has to switch off these beams while measuring the DFWM signal in order to avoid saturation effects induced by the trapping beams. The possibility of performing nonlinear spectroscopy and DFWM in cold and trapped atoms, using an atomic transition different from the trapping one will certainly be important not only for offering us a way to monitor separately the role of saturation associated with different laser beams on a specific atomic state, but also to study the dynamics of trap itself. For example, in the present work a simple pump-probe experiment performed in a MOT allows us to observe directly the ac Stark splitting of the cesium $6S_{1/2}$, $F=4$ ground state, associated with the strongly saturating trapping beams. We should mention here that the effect of the trapping beams of a MOT on the ground state of cesium was previously observed by Georgiades *et al.* [5], through the two-photon excitation of the $6S_{1/2}$-$6D_{5/2}$ cesium transition.

Our experiment was performed using cold atoms obtained from a four-beam magneto-optical trap as described previously in Ref. [14]. 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 [15]. Figure 1 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 doing the spectroscopic investigation. The trapping beams are supplied by a stabilized Ti:sapphire laser and are detuned by about 12 MHz below the resonance frequency of the cesium

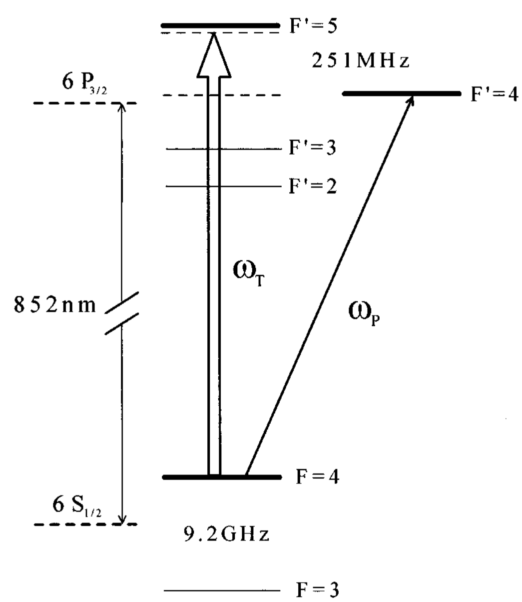


FIG. 1. Relevant energy levels of cesium ($^{133}$Cs) involved in the experiment. $\omega_T$ and $\omega_P$ indicate the frequency of the trapping and the probe beams, respectively.

 

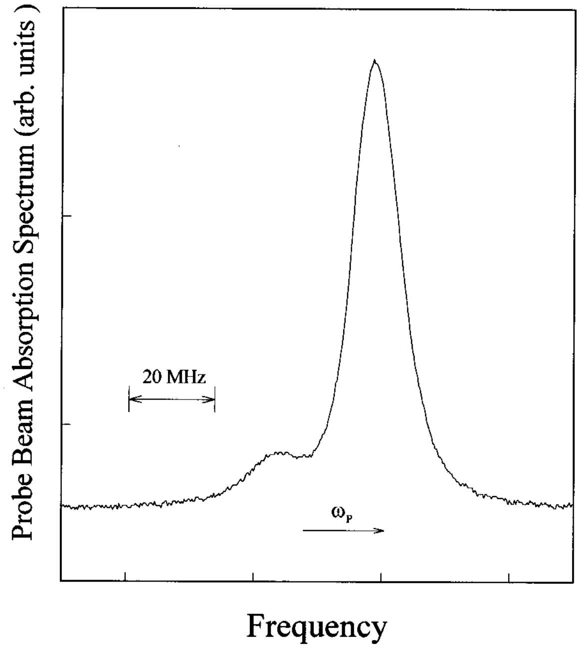


FIG. 2. Probe beam absorption spectrum around the cesium noncycling transition $6S_{1/2},F=4-6P_{3/2},F'=4$, in the presence of the strongly saturating trapping beams, detuned below the resonant frequency of the cycling transition $6S_{1/2},F=4-6P_{3/2},F'=5$.

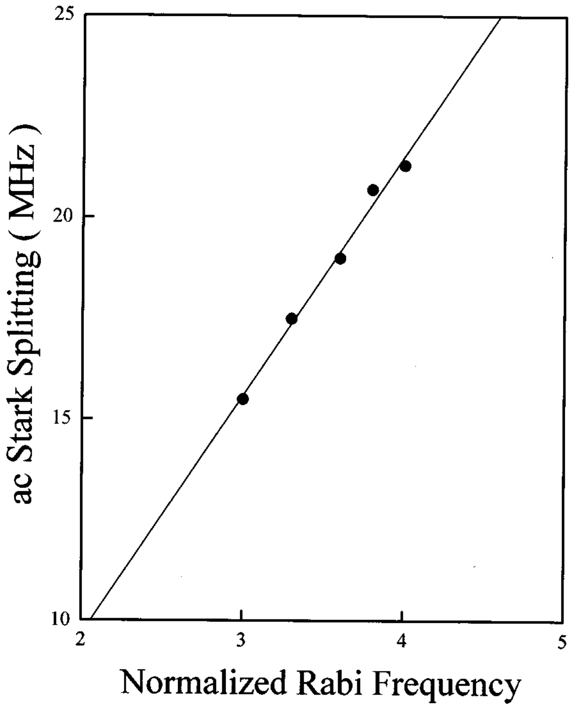


FIG. 3. Dependence of the measured ac Stark splitting as a function of the generalized Rabi frequency $\Omega/2\pi$ normalized to the cesium natural linewidth of the excited state, $\Gamma/2\pi=5.3$ MHz. The average saturation intensity is $I_S=2.5$ mW/cm$^2$ and the scaling factor $a\sim0.25$ is determined by adjusting the measured Stark splitting to the generalized Rabi frequency in just one point. The solid line corresponds to a linear best fitting.

cycling transition $6S_{1/2}$, $F=4-6P_{3/2}$, $F'=5$ at $\lambda=852$ nm. A long external cavity diode laser [16] tuned into resonance 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 [17]. Typically the number of trapped atoms, estimated by measuring the fluorescence emitted by the atomic cloud using a calibrated photodiode, is about $10^7$ atoms.

Our first set of measurements consisted in measuring the probe beam absorption spectrum of the trapped cesium atoms around the noncycling transition $6S_{1/2}$, $F=4-6P_{3/2}$, $F'=4$, which is 251 MHz from the cycling transition, as indicated schematically in Fig. 1. The probe beam is provided by a grating stabilized diode laser, with a spectral linewidth less than 1 MHz and tunable over all the cesium Doppler width of $\sim500$ MHz. The probe beam is made very weak and focused through the trap with a waist much smaller than the trap size ($\sim1$ mm). For a fixed trapping beam detuning and under the condition of strong trapping beams excitation, Fig. 2 shows a typical probe beam absorption spectrum which clearly reveals the dynamic Stark splitting of the cesium $6S_{1/2}$, $F=4$ ground state. We have measured the Stark splitting for different values of the trapping beams intensities and detunings. In Fig. 3 we have plotted this splitting as a function of the generalized Rabi frequency associated with the variation of the total intensity of the trapping beams. For a total trapping beam intensity $I_T$ and frequency detuning $\delta$, the generalized Rabi frequency can be written as $\Omega=\Gamma\sqrt{(\delta/\Gamma)^2+I_T/I_S}$, where $\Gamma/2\pi=5.3$ MHz is the cesium natural linewidth of the excited state and $I_S$ is the average saturation intensity of the cycling transition. For very high intensities the Stark splitting is known to vary linearly with the generalized Rabi frequency. By considering the large spread in the Clebsch-Gordon coefficients within the $F=4-F'=5$ manifold, we have used a value of $I_S=2.5$ mW/cm$^2$, which corresponds to an average over all the Zeeman sublevels [18]. In the plot of Fig. 3 all the measured trapping laser powers have been scaled by a factor $a\sim0.25$, which accounts for the uncertainty in determining the laser intensity in the trapping region as well as the saturation intensity. The solid line corresponds to a linear best fitting. For a fixed intensity of the trapping beams we also have measured the Stark splitting for different values of the detuning $\delta$ and observed the same type of linear dependence with the generalized Rabi frequency. Although the probe beam absorption spectrum can be obtained with a full density matrix calculation [19], we can qualitatively interpret our results in the context of the dressed-atom model [20,21]. In this approach, the intense trapping beams coupled to the atomic cycling transition $F=4-F'=5$, gives rise to successive doublets of dressed states. The doublet around the cesium ground state is coupled to the unperturbed excited state $6P_{3/2},F'=4$, by the probe beam. The frequency splitting of the dressed states in each manifold is given by the generalized Rabi frequency. Furthermore, for the trapping laser detuned below the atomic resonance, the two dressed states in the doublet have unequal populations, with the lower dressed state being more populated [21]. This fact also explains qualitatively the different peak amplitudes observed in the probe spectrum. As suggested in reference [20] this difference of population in the dressed states can be used to produce population inversion and consequently gain in the op-

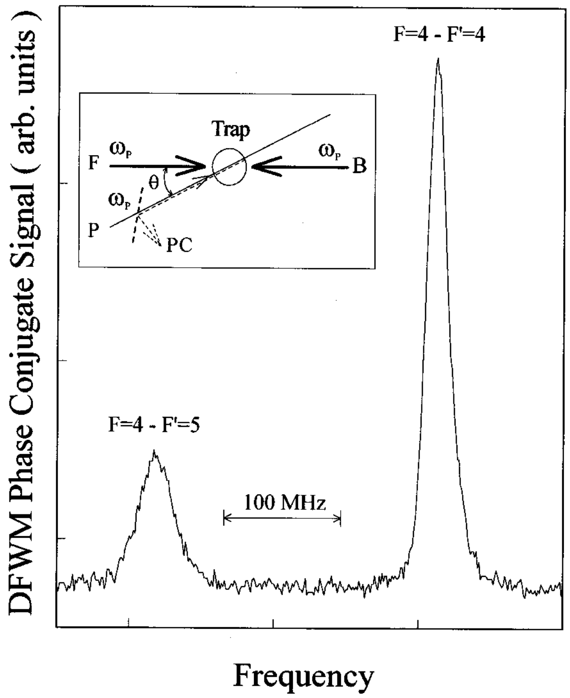


FIG. 4. Four-wave-mixing phase-conjugate spectrum, showing the peaks around the cycling $F=4-F'=5$ and the noncycling $F=4-F'=4$ cesium transitions. Inset: Experimental geometry for observing the phase-conjugate signal.

tical region. This could be achieved by the introduction of another strong pump field that is resonant with the transition between the more populated dressed state and the unperturbed excited state $F'=4$ or another higher excited state. The strong pump field will tend to equalize the population of these two states leading to a population inversion at the frequency of the other coupled transition, i.e., the transition between the involved excited state and the less populated dressed state. We are currently investigating this gain mechanism in our system. We should note that this population inversion occurs in the absence of any incoherent pumping mechanism into the excited state. Therefore, this is different from the inversion predicted in other types of three-level $V$ systems where the inversion is achieved through the presence of an incoherent spontaneous decay which transfers population between the two excited states of the three-level system [22]. It is worth mentioning that under the same experimental conditions the measured probe beam spectrum around the cycling transition also reveals the well-known Mollow's gain of the order of 10% [23]. This gain mechanism can also be interpreted as a result of stimulated emission between dressed states with inverted population [24].

We have also investigated the generation of optical phase conjugation by DFWM in the trapped cesium atoms. In order to observe the phase conjugate signal we have added another pair of counterpropagating pump beams, the forward ($F$) and the backward ($B$) beams, forming a small angle ($\theta \sim 3°$) with the direction of the probe beam ($P$), as indicated in the inset of Fig. 4. The pump beams are provided by the same grating stabilized diode laser and have the same frequency and the same linear polarization as the probe beam. To prevent optical feedback from the backward beam, we have used an optical isolator in the output of the diode laser. The forward and the retroreflected backward pump beams have approximately the same power of 24 $\mu$W and the same beam waist of $\sim 500$ $\mu$m. The probe beam has a power of 13 $\mu$W and is focused to a beam waist of $\sim 200$ $\mu$m. Under these conditions, Fig. 4 shows the DFWM spectrum versus the degenerate laser frequency $\omega_p$, for a broad frequency scan. As indicated in the figure, the peaks are associated with the cycling $F=4-F'=5$ and the noncycling $F=4-F'=4$ transitions. It is worth noticing that when the intense pump beams are resonant with the noncycling transition, a large increase in the population depletion of the absorbing ground state $6S_{1/2}$, $F=4$, occurs, due to the increasing of the optical pumping rate to the nonabsorbing ground state $6S_{1/2}, F=3$. We have verified that the presence of the DFWM beams around the noncycling transition reduces the trap absorption coefficient by approximately 30%. Nevertheless, under the above condition, we have been able to observe efficient generation of phase conjugate signal around the noncycling transition with reflectivity of order of 1%. The phase conjugate (PC) 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. By retroreflecting the probe beam we can easily align the phase-conjugate beam and calibrate the photodiode. For the spectrum shown in Fig. 4 the total trapping power used was about 80 mW.

In order to eliminate the contribution to the observed phase-conjugate signal around the cycling transition associated with the pair of counterpropagating transverse trapping beams, we slightly misalign these beams, thus introducing a mismatch that averages the corresponding signal to zero [11]. It is worth mentioning here that the phase-conjugate signal associated with the trapping beams has a complex spectral structure that reveals the presence of a narrow resonance associated to Raman processes in the ground state [1,2,11] and also broader resonances around the Mollow's spectrum. In particular, we have observed a resonant enhancement of the phase-conjugate signal, when the probe frequency is scanned around the spectral region corresponding to the Mollow's gain.

Although a density-matrix calculation for the DFWM signal around the noncycling transition, in the limit of high intensity of the trapping beams, also reveals the ac Stark splitting of the ground state, we have estimated that the ratio between the two peaks of the DFWM signal around the noncycling transition is about three orders of magnitude smaller than that predicted for the peaks in the probe absorption spectrum. This essentially explains why we have just observed a single peak in the DFWM spectrum.

As we have mentioned, DFWM around a noncycling transition have been observed previously by Knize and coworkers [7,8], in a thermal cesium vapor. In their experiments they have strongly reduced the effect of optical pumping either by introducing a repumping laser similar to the one we have in the magneto-optical trap, or by making use of the atom-wall collision process which considerably increases the relaxation rate between the two hyperfine levels of the cesium ground state. However the measured reflectivity under the condition of their experiments is very low and

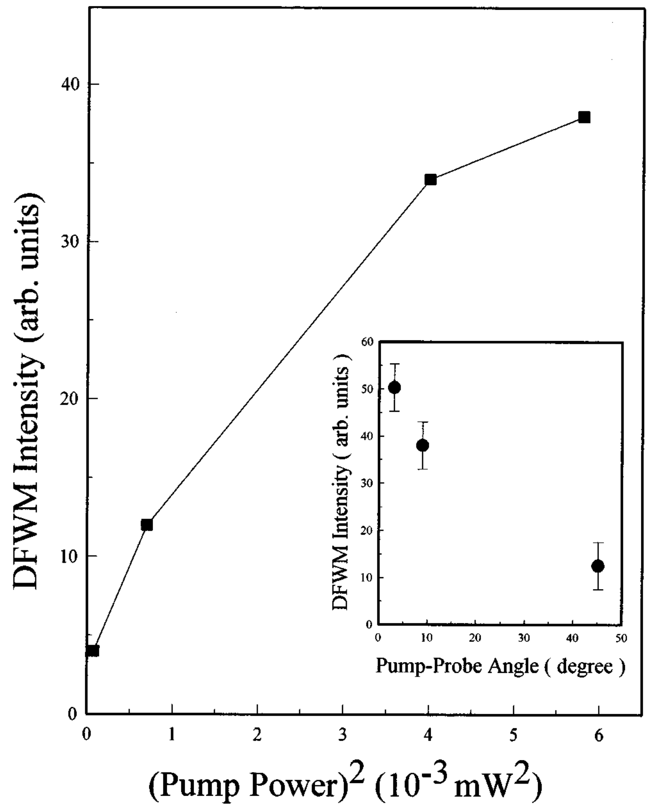


FIG. 5. Dependence of the DFWM signal with the square of the pump beams power, for the noncycling transition. Inset: Angular dependence of the DFWM signal around the noncycling transition.

ranges from $10^{-5}$ to $10^{-6}$, a value which should be compared with the $10^{-2}$ reflectivity measured in our cesium trap.

For a fixed trapping beam intensity, Fig. 5 shows the variation of the phase-conjugate signal with the square of the power of the DFWM pump beams, around the noncycling transition. As can be seen, the nonlinear behavior indicates that saturation of the third-order susceptibility $\chi^{(3)}$ is occurring. That this observed saturation is not due to a decrease in the number of atoms in the trap, was experimentally checked by measuring simultaneously the trap absorption. For the same range of pump power the DFWM signal at the cycling transition was observed to vary linearly. The presence of saturation in the four-wave-mixing process indicates that higher-order multiwave mixing processes can be observed. In principle, one can employ a spatial resolved technique to monitor separately each higher-order term of the nonlinear polarization induced in the cold atomic sample [25]. However, we have observed that for very high intensity of the DFWM beams the signal around the cycling transition is strongly enhanced while the signal at the noncycling transition drops rapidly due to optical pumping.

In the inset of Fig. 5 we show the angular dependence of the DFWM signal around the noncycling transition. Differently from the case of Doppler-broadened system where the DFWM signal decreases very rapidly with the angular aperture [26], we were able to observe efficient DFWM for pump-probe angle as large as 45 °. As it is well known [26], the DFWM signal can be interpreted as a result of the diffraction of one of the pump beam into an interference grating induced in the medium by the other incident beams. In a Doppler-broadened system the induced grating is completely washed out due to the atomic motion for short grating period, i.e., large pump-probe angle $\theta$. However, observing that in the trap the atoms are moving so slow that the time it takes for an atom to move a grating period is much longer than the excited-state lifetime, the washing out of the grating will be strongly reduced. Although there is a small decrease in the DFWM signal due to the variation in the overlapping volume, we have verified that this is not enough to explain the observed angular dependence. We also should note that our trap scheme is not isotropic so the angular dependence could also reflect the anisotropy either in its spatial or velocity distributions, as well as some dynamics aspects associated with the relaxation time of the ground state [27]. We are currently investigating the physical origin for the angular dependence of the DFWM signal both theoretically and experimentally.

In conclusion, we have observed efficient generation of optical phase-conjugate signal around a noncycling transition using cold and trapped cesium atoms in a four-beam magneto-optical trap, and measured the ac Stark splitting induced by the trapping beams in the cesium ground state. The high reflectivity and the large angular aperture of the observed DFWM signal significantly complement the possibility of its applications in spectroscopy and real-time holography.

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[1] J. W. R. Tabosa, G. Chen, Z. Hu, R. B. Lee, and H. J. Kimble, Phys. Rev. Lett. **66**, 3245 (1991).
[2] D. Grison, B. Lounis, C. Salomon, J.-Y. Courtois, and G. Grynberg, Europhys. Lett. **15**, 149 (1991).
[3] R. W. Fox, S. L. Gilbert, L. Hollberg, J. H. Marquardt, and H. G. Robinson, Opt. Lett. **18**, 1456 (1993).
[4] M. Zhu, C. W. Oates, and J. L. Hall, Opt. Lett. **18**, 1186 (1993).
[5] N. Ph. Georgiades, E. S. Polzik, and H. J. Kimble, Opt. Lett. **19**, 1474 (1994).
[6] M. Oriá, D. Bloch, M. Fichet, and M. Ducloy, Opt. Lett. **14**, 1082 (1989).
[7] R. J. Knize, J. M. C. Jonathan, B. Ai, D. S. Glassner, and J. P. Partanen, Opt. Commun. **94**, 245 (1992).
[8] B. Ai, D. S. Glassner, and R. J. Knize, Phys. Rev. A **50**, 3345 (1994).

[9] L. M. Humphrey, J. P. Gordon, and P. F. Liao, Opt. Lett. **5**, 56 (1980).
[10] D. S. Glassner and R. J. Knize, Phys. Rev. Lett. **74**, 2212 (1995).
[11] L. Hilico, P. Verkerk, and G. Grynberg, C. R. Acad. Sci. Paris **315**, 285 (1992).
[12] B. Lounis, P. Verkerk, J.-Y. Courtois, C. Salomon, and G. Grynberg, Europhys. Lett. **21**, 13 (1993).
[13] A. Hemmerich, M. Weidemüller, and T. Hänsch, Europhys. Lett. **27**, 427 (1994).
[14] C. Chesman, E. G. Lima, F. A. M. de Oliveira, S. S. Vianna, and J. W. R. Tabosa, Opt. Lett. **19**, 1237 (1994).
[15] J. W. R. Tabosa, S. S. Vianna, and C. A. Benevides, Opt. Commun. **116**, 77 (1995).
[16] L. Viana, S. S. Vianna, M. Oriá, and J. W. R. Tabosa, Appl. Opt. **35**, 368 (1996).
[17] C. Monroe, W. Swann, H. Robinson, and C. Wieman, Phys. Rev. Lett. **65**, 1571 (1990).
[18] Z. Hu and H. J. Kimble, Opt. Lett. **19**, 1888 (1994).
[19] P. Meystre and M. Sargent III, *Elements of Quantum Optics* (Springer-Verlag, Berlin, 1990).
[20] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977).
[21] K. K. Meduri, P. B. Sellin, G. A. Wilson, and T. W. Mossberg, Quantum Opt. **6**, 287 (1994).
[22] L. M. Narducci, M. O. Scully, G.-L. Oppo, P. Ru, and J. R. Tredicce, Phys. Rev. A **42**, 1630 (1990).
[23] B. R. Mollow, Phys. Rev. A **5**, 2217 (1972).
[24] T. W. Mossberg, Comments At. Mol. Phys. **32**, 75 (1995).
[25] J. W. R. Tabosa, C. L. César, M. Ducloy, and J. R. Rios Leite, Opt. Commun. **67**, 240 (1988).
[26] M. Ducloy, and D. Bloch, J. Phys. (Paris) **42**, 711 (1981).
[27] P. R. Berman, D. G. Steel, Galina Khitrova, and Jin Liu, Phys. Rev. A **38**, 252 (1988).