Eur. Phys. J. D **6**, 127-131 (1999)

THE EUROPEAN PHYSICAL JOURNAL D
EDP Sciences

Experimental study of Velocity Changing Collisions on Coherent Population Trapping in sodium

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Received: 5 October 1998 / Received in final form: 3 December 1998

Abstract. In this paper a detailed experimental study of Coherent Population Trapping (CPT) effect on sodium induced by a dye-laser operating in a three-mode regime is presented and a detailed analysis of the role of velocity changing collisions is made. These collisions show a very small relaxation effect on the dark resonances which are visible even at high pressures. For the first time we demonstrate the persistence of the ground state coherence to pressures up to one atmosphere for a relatively "heavy" buffer gas like argon. The experimental results have been compared with theory and a very good agreement has been obtained. Preliminary results on the effect of Na-N_2 collisions on Coherent Population Trapping are presented.

PACS. 42.50.Gy Effects of atomic coherence on propagation, absorption and amplification of light – 42.50.-p Quantum optics

1 Introduction

In the last years much attention has been devoted to the Coherent Population Trapping (CPT) effect thanks to several applications of this effect in the fields of nonlinear optics, very high resolution spectroscopy and laser cooling [1]. Among other issues, the persistence of CPT in the presence of high collision rates with atoms or molecules represents a very interesting subject. Recently, the effect of the velocity changing collisions (VCC) with noble buffer gases, which affects the ground state coherences mainly at high pressures, has been theoretically and experimentally studied [2–5]. As the experiments made on this subject show contradictory results [2,3], we report here new measurements aimed at solving this disagreement. In fact, in the experiments made with Na and He, a fast CPT quenching as a function of He pressure is observed in reference [3], while a persistence of CPT up to high He pressures is reported in reference [2]. Even if in reference [4] this difference has been explained as due to different optical pumping mechanisms produced by different polarization geometries in the two experimental set-ups, nevertheless the need of a more detailed study is evident. In reference [2] also a systematic study with different noble gases is reported which shows that the relaxation rates increase with the atomic mass.

In the present experiment an apparatus quite similar to the one used in the pioneering experiment [6] is adopted. An atomic coherence between the lower levels of a three-level system in the Λ configuration is induced by a multimode dye laser. In the specific case of sodium atoms, two laser modes are resonant with two transitions coupling two Zeeman sublevels of the $3S_{1/2}$ ground state with a common level of the $3P_{1/2}$ excited state. We have studied the CPT relaxation rates in the presence of argon and nitrogen as buffer gases. We have investigated the pressure dependence of two CPT resonances upon the same experimental conditions to show the role of the optical pumping mechanism. A good agreement between the Na-Ar experimental data and the theoretical model proposed in reference [5] is obtained. The preliminary results with nitrogen surprisingly show a persistence of the coherence up to few tens of Torr.

2 Experimental apparatus

A sketch of the experimental apparatus is reported in Figure 1. A multimode dye laser with a cavity length of about 42.6 cm is tuned on the D_1 line of sodium. By inserting an intracavity thin etalon the number of modes reduces to three, about 1760 MHz spaced, which roughly corresponds to the hyperfine frequency separation of the ground state of sodium. The total output power is around 600 mW. The length of the cavity may be varied within 3 cm by moving the output mirror. This modifies the mode spacing that can be adjusted close to the zero magnetic field resonance frequency at 1771.6 MHz. When two of the three laser

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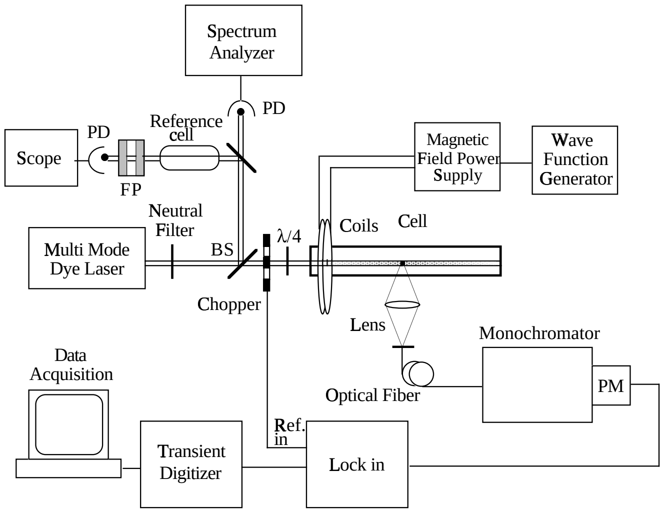

Fig. 1. Sketch of the experimental apparatus: BS = Beam Splitter; PD = Photodiode; PM = Photomultiplier.

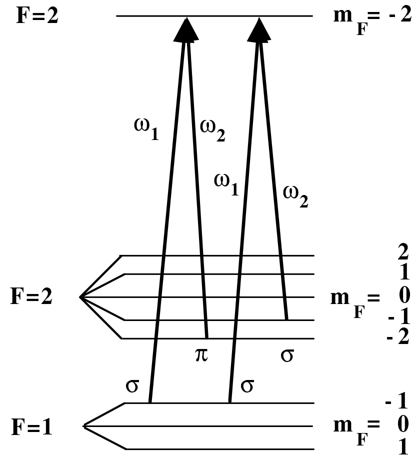

Fig. 2. Sketch of the Na ground state Zeeman sublevels. ω_1 and ω_2 represent the two laser fields. The two Λ systems are clearly visible.

modes are absorbed by the same atom, the CPT effect occurs. The main difference introduced in the present experiment in comparison to that of reference [6] is that the laser is running in a three-mode regime, where the third mode repumps the atoms staying in the wings of the velocity distribution into the Zeeman sublevels involved in the coherence. The light is circularly polarized and it is sent to a resonance cell provided with a valve which allows us to change the buffer gas pressure. The cell is immersed in a magnetic field whose axis is slightly tilted with respect to the laser beam. Because of this misalignment, the laser light polarization does not appear completely circular to the atoms, but it shows also a linear component. A small fraction of the light is sent to a radiofrequency spectrum analyzer which gives the beating signal between the modes. The monitoring of the beating is very important in order to check the coherence between the laser modes. Another fraction of the light is sent, through a reference cell, to an F.P. interferometer. In this way we may check the mode tuning with respect the Doppler broadened absorption line. The magnetic field is generated by one coil and it has a longitudinal gradient that makes the Zeeman sublevel splitting a function of the position along the laser beam. In this way the CPT resonance condition is fulfilled in a well defined region in space, where it appears as a lack of fluorescence, *i.e.* as a black line or a dark resonance crossing the fluorescence induced by the laser beam. By sweeping the magnetic field, the dark resonance moves along the cell and is detected by focusing the light emitted by the cell on an optical fiber of 1 mm diameter. The fluorescence is dispersed by a monochromator fixed on the sodium D_1 wavelength and sent to a photomultiplier connected to a transient digitizer. The signal is obtained averaging over a few magnetic field sweeps. To increase the S/N ratio the laser is modulated by a mechanical chopper and an in-phase detection is sometimes used. In order to measure precisely the magnetic field along the cell axis, we apply a radiofrequency (r.f.) field that induces a transition between the levels $(F = 2,\ m_f = -2)$, $(F = 2,\ m_f = -1)$ of the ground state; this destroys the optical pumping produced by the circularly polarized light and a bright spot appears at the spatial position of the radiofrequency resonance. By sweeping the r.f. frequency, the bright spot moves along the cell axis, so that a map of the magnetic field is obtained and the ground state level involved in the dark resonances is assigned through the Breit-Rabi formulas. As already reported in reference [6], we observed more than one dark resonance at the same time in correspondence of different values of the magnetic field, *i.e.* at different positions along the laser beam. A detailed spectroscopic analysis of all possible hyperfine coherences in sodium has been recently reported [7]. In our experiment we studied the two dark resonances shown in Figure 2: the first one corresponds to the transitions from the $(F = 1,\ m_f = -1)$ and $(F = 2,\ m_f = -2)$ levels to the $(F = 2,\ m_f = -2)$ common upper level; this resonance requires σ^- and π excitations. The second one corresponds to the transitions from the $(F = 1,\ m_f = -1)$ and $(F = 2,\ m_f = -1)$ levels to the $(F = 2,\ m_f = -2)$ common upper level and requires $\sigma^-\ \sigma^-$ excitations. In the following we will refer to them as $|A\rangle$ and $|B\rangle$ dark resonances, respectively.

3 Experimental measurements and results

Our experiment operated by fixing both the cell temperature and the laser intensity and by varying the buffer gas pressure. In order to give a quantitative description of our results and to make a comparison with the prediction of the theoretical model, we introduce the contrast C defined as

$$C = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}}\,, \tag{1}$$

where $I_{\max}$ is the sodium fluorescence level out of the dark resonance and $I_{\min}$ is the sodium fluorescence minimum in correspondence of the CPT resonance. The strength of the dark resonances, and hence the C value, depends on the relative values of the pumping and the relaxation rates. The pumping rate is primarily correlated to the intensity and tuning of the two driving e.m. fields ε_1 and ε_2. Other pumping mechanisms can be due to the presence of either other e.m. fields or collision processes which may repopulate the Zeeman sublevels of the Λ level system. In our case optical pumping is strongly enhanced by the presence of the third laser mode. As will be discussed in the next paragraph, the relaxation processes too are correlated, even if in a different way, to the presence of collisions and of e.m. fields out of the Λ scheme.

3.1 Na-Ar

In the analysis made in reference [5] the CPT effect in the presence of different relaxation processes has been considered. A model taking into account the interaction time defined by the atomic time of flight across the laser beam, the collisional damping of the atomic polarizations and of the coherences, the VCC process, has been proposed. This model has been tested *versus* the experimental results obtained in reference [2] in the case of a Na-He mixture, and we want here test it with the Na-Ar mixture results. The complete equations for the density matrix elements on the basis of coupled-noncoupled ground state linear combination are derived in reference [5] for a three-level Λ system, with the VCC process described by the velocity changing collisional relaxation rate Γ_{VCC} and the associated collisional kernel $K(v)$ for rethermalization of the atoms with velocity v. Under the assumption of strong collisions, the kernel can be approximated by

$$K(v) = \alpha \Gamma_{\mathrm{VCC}} W(v)\,, \tag{2}$$

where $W(v)$ describes the Maxwell-Boltzmann velocity distribution and $0 \le \alpha \le 1$ is an empirical constant. By integrating the evolution equations over the velocity distribution, the following analytical expression is derived for the contrast C in the steady-state regime:

$$C = \frac{1}{1 + 2(\Gamma_{\mathrm{t}} + \Gamma_{\mathrm{VCC}}(1-\alpha))/\Gamma_{\mathrm{D}}}\,. \tag{3}$$

Here Γ_{t} is the atom transit time rate across the laser beam, which assumes the form

$$\Gamma_{\mathrm{t}} = 2.405^2 \frac{D_{\mathrm{g}}}{R^2} \frac{1}{1 + 6.8\lambda/R}\,, \tag{4}$$

where R is the laser beam radius, D_{g} is the ground state atom diffusion coefficient, λ is the mean free path at the sodium temperature. The term $(1-\alpha)$ takes into account the fact that VCC may modify the atomic velocity without affecting the atomic coherence. In equation (3) Γ_{D} is the Doppler integrated pumping rate defined as

$$\Gamma_{\mathrm{D}} = \frac{\Omega_{R1}\Omega_{R2}}{2\Gamma_{\mathrm{rel}}} V(ku, \Gamma_{\mathrm{rel}})\,, \tag{5}$$

Table 1. Values of the used parameters related to our experimental conditions for the Na-Ar couple.

Parameter	Value
R laser beam radius	0.3 cm
T sodium temperature	400 °K
D_{g} diffusion coefficient	$231/p$ cm^2 s^{-1} Torr
λ mean free path	$7.05 \times 10^{-3}/p$ cm Torr
Γ_{VCC} rate of VCC	2.951×10^7 p s^{-1} Torr^{-1}
Ω_{R1} Rabi frequency	330×10^6 s^{-1}
Ω_{R2} Raby frequency	275×10^6 s^{-1}
Γ_{sp} spontaneous decay rate	62.5×10^6 s^{-1}
Γ_{dep} dephasing rate	6.15×10^7 p s^{-1} Torr^{-1}
α for σ-π	0.99994 ± 0.00002
α for σ-σ	0.9986 ± 0.0003

where Ω_{R1} and Ω_{R2} are the Rabi frequencies of the two laser driven transitions, Γ_{rel} is the relaxation rate of the optical coherences, given by

$$\Gamma_{\mathrm{rel}} = \frac{\Gamma_{\mathrm{sp}}}{2} + \Gamma_{\mathrm{dep}}\,, \tag{6}$$

where Γ_{sp} is the spontaneous decay rate and Γ_{dep} is the optical coherence dephasing rate for sodium-argon collisions. $V(ku, \Gamma_{\mathrm{rel}})$ is the Voigt function, defined as

$$V(ku, \Gamma_{\mathrm{rel}}) = \frac{1}{\sqrt{\pi} ku} \int \frac{\Gamma_{\mathrm{rel}}^2}{\Gamma_{\mathrm{rel}}^2 + z^2} e^{-(z/ku)^2} \mathrm{d}z\,, \tag{7}$$

where u is the most probable velocity and k is the wave vector of the light. The values of the collisional parameters for the mixture Na-Ar referred to our experimental conditions are reported in Table 1 with collisional values derived from reference [8]. In the Table we reported also the values for the parameter α resulting from the fit for the two different resonances. We have measured C as a function of the argon pressure p and the results are shown in Figure 3 for the $|A\rangle$ dark resonance and in Figure 4 for the $|B\rangle$ dark resonance. The C dependence on p is not the same for the two dark resonances. The $|A\rangle$ dark resonance shows a constant contrast up to about 100 Torr followed by a slow decreasing at increasing argon pressure. At a pressure larger than one atmosphere the dark resonance is still visible and its contrast is in fact dropped off by only 70%. This result is even better than that reported by [2]: in that experiment, in which the coherence is produced through the same excitation scheme as ours but with a different apparatus, the contrast goes to zero at about 650 Torr of argon. However, two main differences between the two set-ups may originate the discrepancy: the first one comes from the fact that in the experiment of reference [2] the cell is silane coated and the sodium atoms are photodesorbed from the coating at room temperature [9]. In this case, moreover, the sodium density is not exactly controllable. The second difference can be due to the stability of the laser mode structure, which cannot be directly compared. We may only recall that previous experiments [10] demonstrated for the presently used apparatus a very good coherence of the mode oscillation.

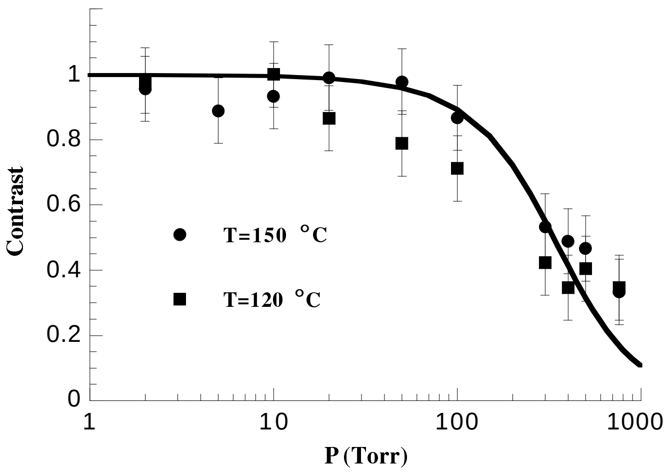

Fig. 3. Measured C values as a function of the buffer gas pressure for the $|A\rangle$ dark resonance. The solid line represents the fit according to equation (3).

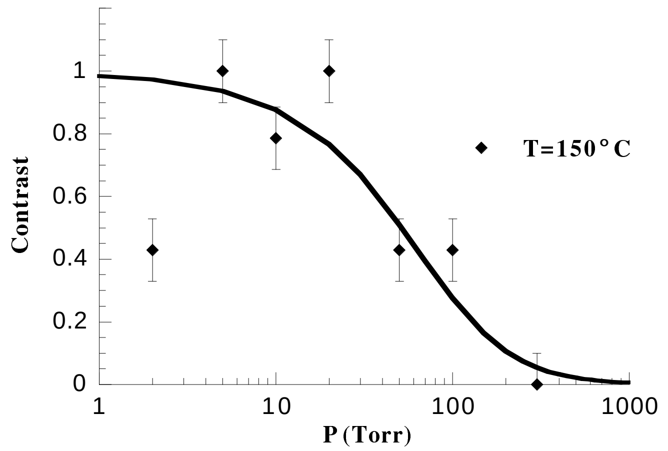

Fig. 4. Measured C values as a function of the buffer gas pressure for the $|B\rangle$ dark resonance. The solid line represents the fit according to equation (3).

The results of the fit with equation (3) are represented by the solid lines in Figures 3 and 4. In Figure 3 a remarkably good agreement is obtained, as the correlation factor $R_{corr} = 0.95$. The theoretical curve assumes Γ_{VCC} $(1 - \alpha) = 2.2 \times 10^3$ p s^{-1} $Torr^{-1}$, so that using the Γ_{VCC} value of reference [8] we found for α a value of $\alpha(|A\rangle) = 0.99994$. This extremely large value shows that the coherence is preserved also at very large pressures. In the case of the $|B\rangle$ dark resonance shown in Figure 4, the theoretical curve assumes $\Gamma_{VCC}(1 - \alpha) = 4 \times 10^4$ p s^{-1} $Torr^{-1}$, leading to an α value equal to $\alpha(|B\rangle) = 0.9986$. The value of $\Gamma_{VCC}(1-\alpha)$ is about a factor ten larger than that found for the $|A\rangle$ dark resonance and is responsible for the depeer decrease of the contrast. In Figure 4, in fact, C shows a more pronounced decrease with p and drops to zero at about 300 Torr. Also in the experiment of reference [3], in which the coherence was produced by two circularly polarized fields inducing a $\sigma^-\sigma^-$ dark resonance, C diminished quite rapidly, going to zero beyond 50 Torr of He buffer gas. This result evidences the importance of optical pumping which reduces the effect of the coherence quenching due to Na-Ar collisions.

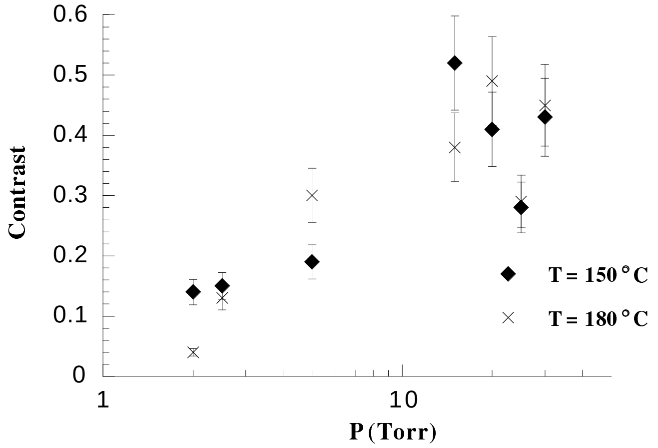

Fig. 5. Measured values for the line contrast as a function of the nitrogen buffer gas pressure for the coherence $|A\rangle$ at two different temperatures.

The different behaviour shown by the two dark resonances supports the explanation given in reference [4] to justify the difference between the results of the experiments in reference [2] and reference [4]. The VCC with the buffer gas move the atoms with zero velocity class to velocity classes out of resonance with the laser. The atoms may be re-excited by the third mode and accumulated by the optical pumping process, in the case of σ^- radiation, in the level ($F = 2$, $m_f = -2$), that is, one of the levels involved in the black line in the case of the $|A\rangle$ transition. This is not true in the case of the $|B\rangle$ transition, which involves levels depleted by the optical pumping effect. As a further support to this statement, we noticed that, when at higher temperatures the optical pumping is less effective [11], the $|B\rangle$ coherence becomes favourite in comparison to the $|A\rangle$ one [12].

3.2 Na-N_2

The collisions of Na atoms with noble gases allow a simple description as the interaction potential is quite small owing to the closed shell structure of the noble gas, which can be assumed as a spheric rigid body. Moreover, most of the interaction parameters of noble gases with alkali atoms are well known. This situation is totally different if the collisions with molecules are considered. We have made a preliminary analysis considering the Na-N_2 mixture and the experimental results are shown in Figure 5. C increases up to about 15 Torr then remains roughly constant up to about 35 Torr. It was not possible to go to higher pressures because nitrogen quenches the sodium fluorescence and the coherence cannot be monitored through the decrease of the fluorescence.

4 Conclusions

In this paper we report an experimental analysis of CPT in the presence of high pressure of argon and nitrogen. In the first case we have studied the dependence on p of two dark resonances and we have observed the persistence of one of them at pressures larger than the atmospheric one. As in the σ^{-}-π scheme the optical pumping sustains the ground state coherence, the relaxation induced by the velocity changing collisions is strongly reduced. A very good agreement has been found with theory. The preliminary results obtained with N_2 buffer gas show that the ground state coherence is particularly strong also in the presence of collisions between sodium atoms and molecules, and this new result needs a deeper experimental and theoretical analysis. It is anyway interesting to remark that with our apparatus we observe CPT at the same He pressure range as reference [3]. This confirms once more the effectiveness of the optical pumping mechanism and the importance of the third laser mode.

The authors wish to thank Prof. E. Arimondo for helpful discussions and critical reading the manuscript, Mr. M. Tagliaferri for the technical support and Mr. M. Badalassi for the vacuum techniques assistance.

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