Phase conjugation by degenerate four-wave mixing in barium vapor

T. Mikropoulos, S. Cohen, M. Kompitsas, S. Goutis, and C. Baharis

Theoretical and Physical Chemistry Institute, National Hellenic Research Foundation, 48, Vassileos Constantinou Avenue, Athens 116 35, Greece

Received January 18, 1990; accepted August 24, 1990; manuscript in hand September 17, 1990

We report what is to our knowledge the first observation of phase-conjugate waves by degenerate four-wave mixing in Ba vapor near its $6s^2\ ^1S_0 \rightarrow 6s6p\ ^1P_1$ resonance transition. A maximum reflectivity of 36% at 10 mbars of Ar buffer gas pressure is observed together with a temporal pulse shortening of 50%. Conical emission in the two pump waves after their passage through the nonlinear medium is also observed. The dependence of the phase-conjugate signal on the pressure of the Ar buffer gas shows a small state-changing collisional decay rate.

Degenerate four-wave mixing is one of the most efficient methods of obtaining phase-conjugated wave fronts.[1] In atomic vapors, high reflectivities have been observed when tuning the laser frequency close to strong atomic transitions.[2,3] To our knowledge, up to now experiments have been restricted to alkali metals such as Na,[2-6] K,[7] and Rb.[8] Ba has been used only for nonlinear phenomena other than phase conjugation (PC), such as Raman conversion,[9] stimulated Stokes emission,[10] and Stokes and anti-Stokes frequency generation.[11] This Letter is the first part of an experimental study of PC by resonantly enhanced degenerate four-wave mixing in Ba vapor and is to our knowledge the first such report in an alkaline earth atom. The choice of Ba is made because of the high nonlinear efficiency of its resonance line $6s^2\ ^1S_0 \rightarrow 6s6p\ ^1P_1$ at 553.55 nm and its easy handling under heat-pipe conditions. We optimized the intensity of the phase-conjugated wave fronts for different vapor temperatures and measured it as a function of the laser intensity and the forward-beam and probe-beam intensities. We also observed the line shape of the PC spectrum under certain conditions. Moreover, we studied the PC signal at different buffer gas pressures. The collisional redistribution of the population among the upper-state m_j sublevels is not negligible during the laser pulse,[12] which in our case is ≈8 nsec. This fact probably alters the character of the four-wave mixing process because we are not dealing with a pure two-level system.[13,14] This enables one to study gas-phase dynamics, e.g., to use PC for velocity distribution measurements in the gas phase.[15]

The experimental setup is shown in Fig. 1. The light source is a XeCl excimer-laser-pumped dye laser (Lumonics TE861T-3, EPD 330), vertically polarized. The dye laser has a linewidth of ≈0.2 cm^{-1} with a pulse-to-pulse energy jitter of ≈20%. The laser beam was collimated by using a telescope and a pinhole. The pump beams were formed by the 50:50 cube beam splitter BS2 and mirrors M1 and M2. The probe beam was formed by beam splitter BS1, which has a reflectivity of ≈10%, and mirror M3. The angle between the probe and the pump beams was ≈70 mrad. The PC signal emerging from the interaction region to the opposite direction of the probe beam was detected through the 50:50 beam splitter BS3 by a 0.2-nsec rise time fast photocell (ITL-SI4H7). We verified that the signal was disappearing when each of the three input beams was blocked. The vapor cell of the conventional heat-pipe type was made of stainless steel, and the Ba vapor was confined near the center of the heat pipe by a stainless-steel mesh. The concentration of Ba atoms inside the oven was temperature controlled, and the experiments were performed in a temperature range of 620–830°C. The coherence

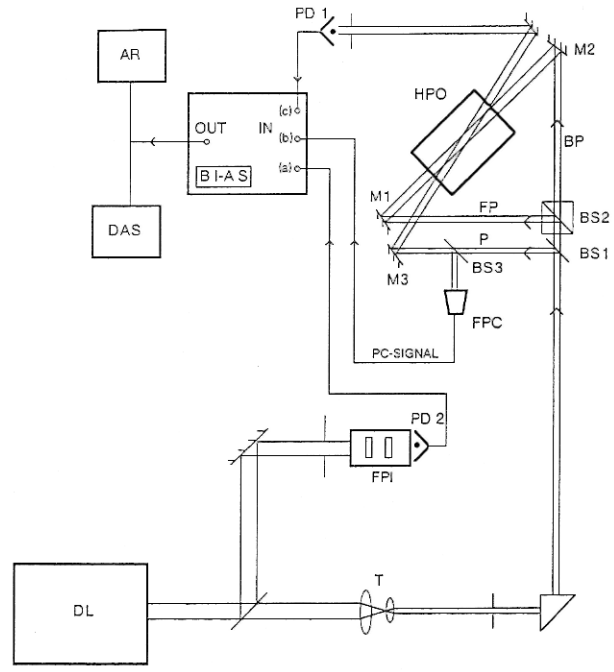

Fig. 1. Experimental setup. DL, dye-laser system; T, telescope; HPO, heat-pipe oven; FPC, fast photocell; PD's, photodiodes; FPI, Fabry–Perot interferometer; B I-A S, boxcar integrator–averager system; DAS, data-acquisition system; AR, analog recorder; M's, aluminum mirrors of ≈80% reflectivity.

0146-9592/90/221270-03$2.00/0 © 1990 Optical Society of America

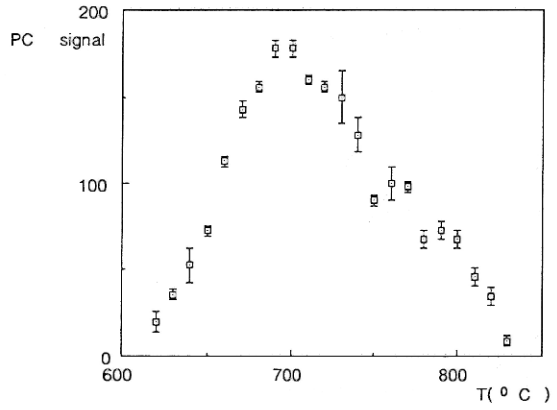

Fig. 2. PC signal as a function of the vapor temperature. The total dye-laser beam intensity is ≈1.5 MW/cm² (measured before BS1), the probe-beam intensity is ≈75 kW/cm², and detuning $\Delta\nu \approx 45$ GHz.

length of the dye laser was at least a factor of 2 larger than the interaction length, estimated to be ~1 cm.

The PC signal as a function of the vapor temperature is shown in Fig. 2. The signal increases with the temperature because of the increased susceptibility. At higher temperatures, various nonlinear effects such as self-focusing play an important role, and the signal decreases again. The maximum of the signal was found to be at 700°C, close to the melting point of Ba (≈727°C).

The maximum observed PC signal was 36% at ≈10 mbars of Ar buffer gas pressure. This relatively small value is probably due to the absorption and depletion of the strong pump waves during the interaction as well as the other competing nonlinear effects such as self-focusing, defocusing, and conical emission. The signal also shows a strong buffer gas pressure dependence. Moreover, a temporal shortening of the PC pulse by ≈50% was observed, owing to the nonlinear nature of the generation of the backward wave gain, which is proportional to the temporal product of the three input waves.[16] Another interesting observation is a conical emission in the pump beams after their passage through the nonlinear medium.[17] The shape of the transmitted pump beams when Ba atoms interact with the two counterpropagating pump beams is the same as that of Fig. 1b in Ref. 18.

In Fig. 3 the PC signal is shown as a function of the probe-beam intensity. During this measurement the dye-laser energy had a long-term stability of ≈4%. The signal seems to be approximately linear, and it should be noted that saturation does not appear up to the maximum available intensity. Figure 4 shows the dependence of the PC signal on the normalized forward pump-beam intensity. The energy of the dye laser had a long-term stability of ≈12%, and the detuning was ≈45 GHz. We observed a similar behavior as in Ref. 19, which suggests that the adiabatic following model is valid under our experimental conditions (pump intensity and detuning). The fit in Fig. 4 gives a saturation intensity of ≈136 kW/cm². This value is in good agreement with the theoretical one of 167 kW/cm², which is calculated according to $I_s = \frac{1}{2}\epsilon_0 c(h\Delta\nu/\mu)^2$, where μ is the dipole transition moment.

Figure 5 shows the dependence of the PC signal on the pressure of the Ar buffer gas. Measurements are taken in the 10–310-mbar range. We observed that the signal at the maximum pressure is not zero. Comparing these measurements with similar ones performed on Na vapor,[14] we note the same qualitative signal dependence on the pressure. However, we measure a much slower pressure-induced signal decrease since in our case the pressure values are two orders of magnitude higher than those in Ref. 14. We believe that this is due to the different energy-level structure for the Na and Ba atoms: There exist a number of lower-lying excited states in Ba ($6s5p\ ^3P_{0,1,2}$ and $6s5d\ ^1D_2$, $^3D_{1,2,3}$), while this is not the case for the

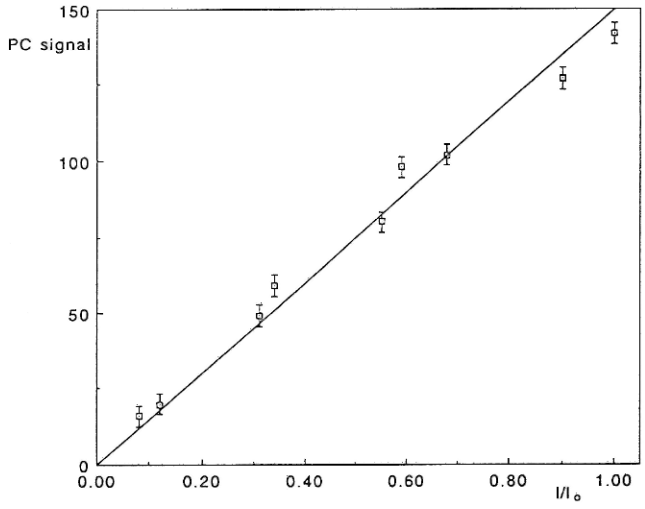

Fig. 3. PC signal as a function of the probe-beam intensity, normalized to its maximum value available, $I_0 \approx 75$ kW/cm². The experimental conditions are $I_{FP} = I_{BP}$, $I_{FP}/I_0 \approx 5$, and $\Delta\nu \approx 45$ GHz.

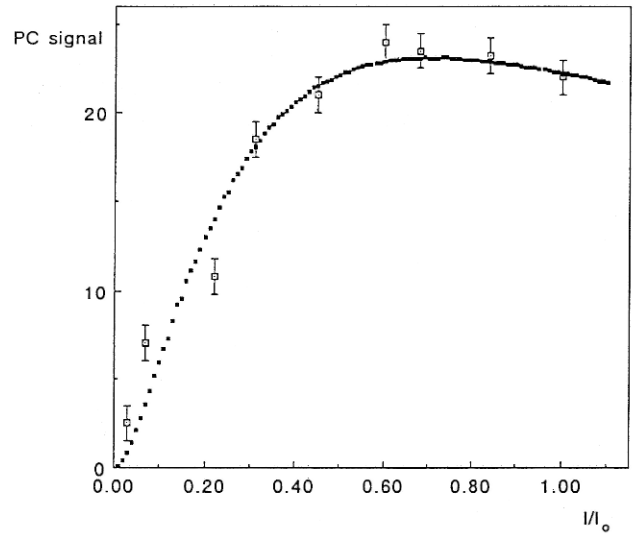

Fig. 4. PC signal as a function of the forward pump-beam intensity, normalized to its maximum value available, 386 kW/cm². $I_P \approx 75$ kW/cm², $\Delta\nu \approx$ GHz, and Ar pressure ≈8 mbars. Solid squares, fitted points; open squares, experimental data. The pump- and probe-beam intensities were always measured just before the oven windows.

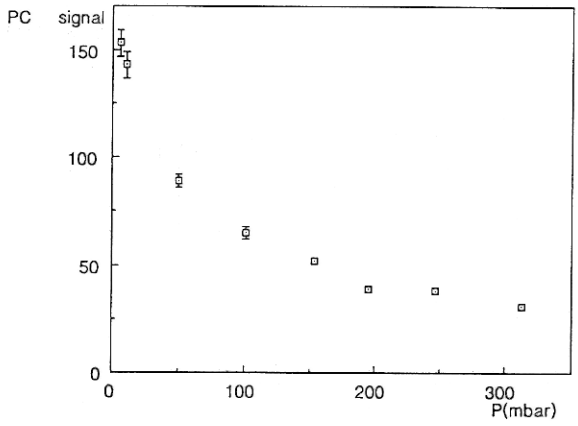

Fig. 5. PC signal as a function of the pressure of the Ar buffer gas. [Ba] = 10^{15} cm^3, and the total laser beam intensity is ≈1.5 MW/cm^2.

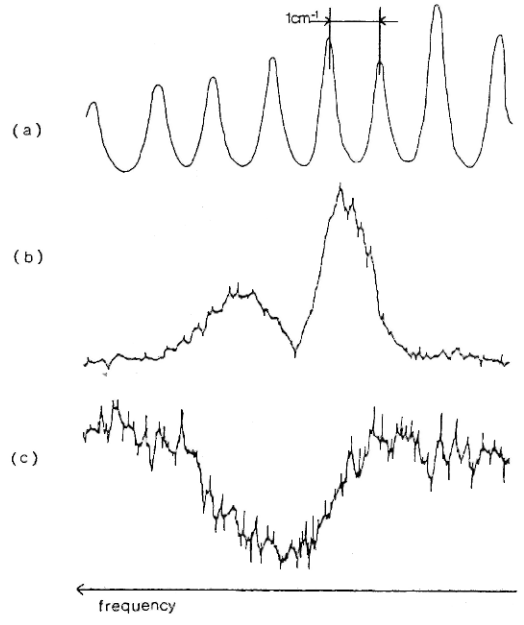

Fig. 6. (b) Typical recording of the phase-conjugate spectrum for [Ba] = 10^{15} cm^3, total beam intensity 1.5 MW/cm^2, and Ar pressure of 28 mbars. (a) Fabry–Perot transmission. (c) Absorption of the probe beam detected by PD1.

Na atom. Extended pressure-dependence measurements together with a rate-equation model are currently being studied.

Figure 6(b) shows a typical recording of the PC signal spectrum. Figure 6(c) shows the simultaneously recorded absorption spectrum (see Fig. 1), which served as the absolute reference, and Fig. 6(a) shows the transmission signal of a Fabry–Perot interferometer (F^* ≈ 15, free spectral range 1 cm^{-1}), which provides relative wavelength calibration. This method is typically accurate up to one laser linewidth. In our case we are limited by the broad absorption spectrum (probably due to power broadening) to an accuracy of ≈0.5 cm^{-1}. In Fig. 6(b) two peaks are clearly seen. We are currently conducting extensive experimental studies of the PC line shape under different conditions.

We have reported phase-conjugate waves by degenerate four-wave mixing in Ba vapor. The vapor temperature for optimum PC signal generation and its specific dependence on the buffer gas pressure observed here show that Ba is a convenient material for the study of nonlinear phenomena. In particular, the strong pulse compression by 50% of the phase-conjugated wave relative to the probe beam indicates that Ba can be used as an efficient pulse shortener.

We gratefully acknowledge the experimental assistance of Z. Pan of the Anhui Institute of Optics and Fine Mechanics, Academia Sinica, Heifei, China.

References

1. R. W. Hellwarth, J. Opt. Soc. Am. **67,** 1 (1977).
2. D. M. Bloom, P. F. Liao, and N. P. Economou, Opt. Lett. **2,** 58 (1978).
3. J. Brock, J. Fukumoto, F. Patterson, W. Garrion, G. Holleman, and L. Marabella, Proc. Soc. Photo-Opt. Instrum. Eng. **739,** 33 (1987).
4. B. Kleinmann, T. Trehin, M. Pinard, and G. Grynberg, J. Opt. Soc. Am. B **2,** 704 (1985).
5. A. C. Cefalas, T. Mikropoulos, P. Simon, J. Hebling, and C. A. Nicolaides, Appl. Phys. B **46,** 363 (1988).
6. N. Tan-no, T. Hashimiya, and H. Inaba, IEEE J. Quantum Electron. **QE-16,** 147 (1980).
7. T. Mikropoulos and Z. Pan, Appl. Phys. B **50,** 19 (1990).
8. D. Grischkowsky, N. S. Shirey, and R. J. Bennett, Appl. Phys. Lett. **33,** 805 (1978).
9. N. Djeu and R. Burnham, Appl. Phys. Lett. **30,** 473 (1977).
10. J. L. Carlsten and P. C. Dunn, Opt. Commun. **14,** 8 (1975).
11. C. H. Skinner and H. P. Palenius, in *Digest of Optical Society of America Annual Meeting* (Optical Society of America, Washington, D.C., 1975), paper Q12.
12. P. Esherick and J. J. Wynne, Comments At. Mol. Phys. **7,** 43 (1977).
13. P. F. Liao, D. M. Bloom, and N. P. Economou, Appl. Phys. Lett. **32,** 813 (1978).
14. D. G. Steel and R. A. McFarlane, Phys. Rev. A **27,** 1687 (1983).
15. T. S. Rose, W. L. Wilson, G. Wackerle, and M. D. Fayer, J. Chem. Phys. **86,** 5370 (1987).
16. A. Yariv, IEEE J. Quantum Electron. **QE-14,** 650 (1978).
17. J. Pender and L. Hesselink, IEEE J. Quantum Electron. **25,** 395 (1989).
18. G. Grynberg, E. Le Bihan, P. Verkerk, P. Simoneau, J. R. R. Leite, D. Bloch, S. Le Boiteaux, and M. Ducloy, Opt. Commun. **67,** 363 (1988).
19. P. Kumar, Opt. Lett. **10,** 74 (1985), and references therein.