PHYSICAL REVIEW A, VOLUME 64, 021801(R)

Temporal character of pulsed-laser cone emission

W. Gawlik,[1,2] R. Shuker,[1,3] and A. Gallagher[1]
[1]JILA, National Institute of Standards and Technology and University of Colorado, Boulder, Colorado 80309-0440
[2]M. Smoluchowski Physical Institute, Jagiellonian University, Reymonta 4, 30-059 Kraków, Poland
[3]Physics Department, Ben Gurion University of the Negev, Beer Sheva, 84105, Israel

(Received 21 March 2001; published 2 July 2001)

Temporal character of cone emission from Sr vapor excited with ∼3-ns, near-resonant laser pulses is studied with ∼0.5-ns resolution. The cone pulses appear significantly narrower than the laser pulse. Their properties (amplitude, width, energy, delay, and angular distribution) systematically depend on the laser energy and detuning in a way that contradicts present, steady-state models of cone emission.

DOI: 10.1103/PhysRevA.64.021801 PACS number(s): 42.50.Fx, 42.50.Hz, 42.65.Sf

Cone emission (CE) refers to an angularly isolated cone of light emitted in the forward direction from a medium illuminated by a laser beam. It has been detected from glass, liquids, air, and atomic vapors in dozens of experiments performed under different conditions with pulsed and steady-state, continuous wave (cw) excitation [10]. In liquids and glasses the phenomenon is reasonably well understood. In atomic vapors, however, only a cw excitation experiment [1] has yielded results that could be understood; almost all other experiments have been with pulsed lasers, yielding very different results that are not satisfactorily explained. One of the major differences in the atomic versus the liquid or glass experiments is the medium saturation, which is major for atoms, but very weak for liquids and glass. Saturation is also significantly higher for pulsed excitation than in the cw regime. In this work we concentrate on the situation where a medium consisting of "two-level" atoms is perturbed by laser pulses of some nanosecond duration and 1–10-GHz linewidth, blue detuned (∼100 GHz) from resonance, and with CE occurring on the red side of the atomic resonance in a broad frequency range typically 1–2 times the laser detuning and at 10–50-mrad angle.

Most theories of CE are based on four-wave mixing (FWM) between pump and Rabi sidebands to obtain reasonable conversion from the pump to the cone radiation [2]. FWM explains the results of cw experiments [1], yet in the pulsed experiments the fourth wave, on the high frequency side of the laser, is absent, e.g., Refs. [3,4], at least at low light intensity. References [5–7] considered Cherenkov-type emission as an alternative source of CE that does not require a fourth wave [6], however, it is yet to be determined if this type of emission could have sufficient efficiency to explain the observed intensities of the CE. Most of the existing theories of CE deal with the steady-state situation and the pulsed-laser CE is still far from even a qualitative explanation [10].

In the hopes of clarifying the causes of pulsed-laser atomic CE, we have studied the temporal character of CE. The only previous measurement [8] had insufficient resolution to see when within the laser pulse the CE was emitted. Harter and Boyd [2] measured the temporal character of the laser beam before and after the cell, finding no changes in a 3-ns pulse, but they did not study the temporal character of the CE. Yet this is an important issue for at least two reasons. First, the broad CE spectrum is often attributed to the varying laser intensity during the pulse. This predicts that the portion of the CE at small detuning arises during the small-intensity parts of the pulse, and visa versa. Next, all theories assume that only the laser beam is strong enough to severely modify the vapor dielectric constant and to produce large CE detunings. This is based on the observation that the total CE energy from one pulse is only a few percent of the laser-pulse energy. But if the CE was emitted during a very short portion of the laser pulse, the instantaneous CE power could be large enough to significantly modify the medium, and even to feed back on the laser-beam propagation. Since current theories [9] do not achieve reasonable conversion into CE, some such unexpected phenomena must be at work.

In our experiment, a 10-Hz, injection-seeded Nd:YAG beam was tripled to pump a dye laser tuned to the blue side of the Sr 5 $^1S_0$-5 $^1P_1$, 460.7-nm resonance line. The optical setup is shown schematically in Fig. 1. After spatial filtering, the spatially near-Gaussian dye-laser beam was focused onto the entrance of a 5-cm long Sr cell. Laser detuning from resonance, $\Delta_L=\nu_L-\nu_0$, was 50–100 GHz and its linewidth 1–2 GHz. The laser conditions were adjusted to yield a fairly smooth single-peaked pulse of ∼3-ns duration. To obtain the CE data, light emitted from the cell into an angular range that included the CE, but not the laser beam, was focused onto a fast Si p-i-n photodiode, whose output was recorded with a real-time, 1-GHz oscilloscope and a Polaroid camera. For some of the data, finer annular apertures (A in Fig. 1) between the cell and the focusing lens selected angular portions of CE. The diode was also illuminated by a fraction of

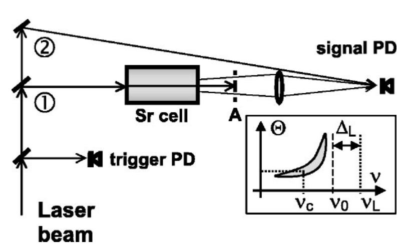

FIG. 1. The experimental setup. A pulsed dye-laser beam is split into a strong beam (1) and a weak reference (2). The inset shows schematically the range of cone angles and frequencies ν_c (shaded). When $\nu_L>\nu_0$, ν_c spans a wide range on the red side of ν_0. The characteristic dependence between θ and ν_c allows spectral resolution of CE by selection of specific angles.

the incident light (beam 2 in Fig. 1) to yield simultaneous recording of the incident and cone pulses.

To avoid errors due to temporal drifts, a measured delay of 6 ns was used between beams 1 and 2 in Fig. 1, and the cone and reference pulses were recorded on the same 10-ns oscilloscope trace. The CE pulses were recorded under well-defined conditions of Sr vapor density ($\sim 10^{14}\,\mathrm{cm}^3$ at $\sim 560\,°\mathrm{C}$), buffer gas pressure (~2 Torr of Ar), laser pulse energy and frequency, and range of cone angles transmitted to the detector. All data presented below has been taken by photographically superimposing ~100 pulses to show the effect of incident pulse fluctuations. An independent laser-pulse detector triggered the scope. Details of the beam shaping and imaging optics and the cell are described in Ref. [10] and are not shown here. As in Ref. [10], the conditions for self-focusing are determined by imaging the cell exit plane onto a charge-coupled device (CCD) camera; the intensity $I_{\mathrm{CE}}(\nu,\theta)$ is measured as a function of frequency ν and cone angle θ using a grating spectrometer and CCD camera. An example of the $I_{\mathrm{CE}}(\nu,\theta)$ spectrum is shown schematically in inset to Fig. 1.

We have confirmed a result noted in Ref. [10]: CE occurs when self-focusing takes place, either as a single self-focused filament or as a number of separated filaments within the full beam diameter. In the present experiment, we illuminated the cell with an 1–10 μJ and a 0.85-mm-diam. beam. This beam broke up into typically 5–100 separate filaments of 50–100-μm diameter, depending on the detuning. These were seen in a high-resolution image of the exit plane; the full beam size at the exit plane was still ~0.85 mm. Also in the far field, there was no visible change of the beam diameter due to the spatial beam breakup. This shows that without imaging of the exit plane, self-focusing can be unnoticed, as demonstrated in Refs. [3,11].

For the beam diameter and maximum intensity used here, CE is well resolved from the laser beam only for $\Delta_L = 30\text{–}100$ GHz. For larger detuning, cones are weak and difficult to observe, while for $\Delta_L < 30$ GHz, the cone is obscured by a diffuse, wide-angle background at the laser frequency. Figure 2 shows examples of typical incident light and CE pulses recorded with the setup of Fig. 1. Figure 2(a) depicts the narrowest observed CE pulses and defines relevant timing parameters—advancement D, delay D_0, and pulse width W full width at half maximum (FWHM)—while Fig. 2(b) shows CE pulses for various laser energies I_L. We observe that the CE pulses: (1) are much narrower and steeper than the 3-ns laser pulses; (2) generally peak on the leading edge of the incident laser pulse, rather than on its maximum; and (3) do not exhibit any component delayed from the laser pulse including later times not shown in Fig. 2.

When analyzing the pulse data taken for different laser energies I_L and detunings, we have found that most of the CE pulse parameters systematically depend on the *reduced intensity*, defined as $I_R = I_L/(\Delta_L)^2$. In particular, Fig. 3(a) presents the dependence of the peak amplitude A of the CE pulse on the incident energy and detuning, using parameter I_R. As can be seen, the data fits the linear relation $A \propto I_R$ within the experimental uncertainty. In Fig. 3(b), the dependence of the CE pulse width W is plotted versus I_R. Note that as the CE pulse amplitude shrinks, its width approaches the 0.45-ns instrumental resolution of our detection system, seen in Fig. 2(c) as the response of the diode and scope to 300-fs laser pulses. Thus, the shortest actual CE pulse must be considerably shorter than 0.45 ns. The CE energy taken as $E_{\mathrm{CE}} \propto A \cdot W$ is shown in Fig. 3(c). This is quadratic at the lower I_R, but as W in Fig. 3(b) starts to saturate at the largest I_R, so does E_{CE} in Fig. 3(c). The advancement D of the CE peak relative to the peak of the laser pulse varies from -0.2 to 0.7 ns. It is not correlated with I_R, and is not plotted here.

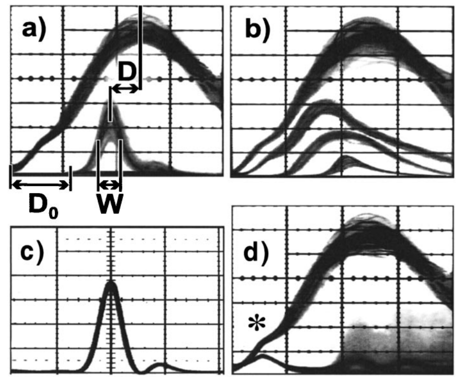

FIG. 2. Typical light pulses recorded: (a) top, incident laser; bottom, CE at $\Delta_L = 100$ GHz and $I_L = 1.9\,\mu$J. D (D_0) and W are defined as the delay between peaks (onsets) of the incident and CE pulses and the pulse width (FWHM). (b) Top, incident laser; lower, CE pulses corresponding to $\Delta_L = 80$ GHz and $I_L = 7.5$, 3.8, and 1.9 μJ (from top to bottom). (c) Response of the detection system to 300-fs light pulses. (d) Laser pulse axially transmitted through the cell (lower trace) at $\Delta_L = 0$ and $I_L = 0.7\,\mu$J. The asterisk marks the contributions due to ASE (see text). The horizontal scale is 1 ns/div. The vertical scale is arbitrarily adjusted for each trace for optimum viewing, except in (b), where all traces have the same AU.

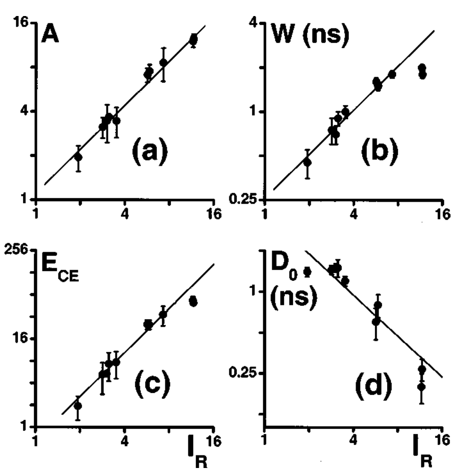

FIG. 3. Log_2-log_2 plots of dependences of the CE parameters on the reduced intensity I_R: (a) amplitude, (b) width, (c) energy=amplitude×width, (d) onset delay D_0. Straight lines represent different power dependences on I_R: linear in (a) and (b), quadratic in (c), and I_R^{-1} in (d). The vertical scale in (a) and (c) is arbitrary.

On the other hand, the delay D_0 between the onsets of the two pulses follows a $1/I_R$ dependence [Fig. 3(d)]. This shows that D_0, rather than D, is the relevant parameter for characterization of CE.

We have also analyzed the angular dependence of the CE pulses. By taking annular rings of different diameters, we were able to study temporal character of light emitted at different cone angles θ. In view of the correlation between the angle and frequency of CE, depicted in inset to Fig. 1, we were thus able to study temporal character of different spectral components ν_c of the cone emission. This method of spectral selection is not perfect: at the smaller angles it loses ν resolution and there is some leaking of the strong laser beam that is substantially spatially broadened during propagation through the cell. Nevertheless, we have found that CE pulses corresponding to angles up to 31 (21) mrad for $\Delta_L = 50(100)$ GHz have very similar temporal shape. They have $\sim$0.7-ns width and occur near the laser peak. In contrast, the pulses of cone light emitted at larger angles (42 and 31 mrad for $L=50$ and 100 GHz, respectively), i.e., corresponding to smaller detuning of c from 0, they are longer and delayed: $W=1.4\,(0.9)$ ns and $D_0=0.6\,(1)$ ns for $\Delta_L=50(100)$ GHz.

In addition to the CE pulses, we have also studied the laser pulses transmitted through the Sr vapor. The lower trace in Fig. 2(d) shows a small portion of the laser pulse transmitted through a 0.5-mm pinhole aperture $\sim$50 cm beyond the cell. The laser detuning from the resonance Δ_L is zero and the upper trace is the full incident pulse. The transmitted pulse shows a complex, rapidly varying structure after $\sim$1.5-ns delay, due to temporal pulse breakup in the cell. In addition, a short pulse of transmitted light reproducibly occurs at the leading edge of the laser pulse. This short pulse is attributed to a broadband amplified spontaneous emission (ASE) of the laser dye that occurs only in the first nanosecond, before the buildup of optical feedback within the dye-laser cavity. Since the ASE contribution is strongly collimated along the laser beam axis, there is no contribution of ASE light to the off-axis cone light. By comparison with Figs. 2(a) and 2(b) it is seen that there is no temporal coincidence between this ASE pulse and CE. This observation indicates that it is very unlikely that ASE could seed CE, as has often been speculated.

One of biggest surprises of this study of the temporal character of pulsed CE is that the CE occurs in the form of very narrow pulses that do not peak at the incident pulse intensity peak [Figs. 2(a), 2(b)], as would be expected for most nonlinear phenomena. This indicates that CE is not directly related to the time evolution of the incident pulse but represents independent, nonadiabatic buildup in a time that is much shorter than the incident pulse.

One idea, suggested by the very brief CE pulse in the near-threshold region, is that the CE may be formed during very brief periods. Then, to be consistent with the measured CE energy as a fraction of the laser-pulse energy, it must be a very significant intensity (comparable to the laser beam) during these generation periods. Most models have assumed that only the incident laser beam significantly modifies the medium, not the CE itself. But if the CE intensity is comparable to the incident intensity, this could significantly alter the propagation of the various beams. Qualitative similarity of the CE and axially transmitted laser pulses strongly supports the idea of such a feedback.

From the systematic dependences shown in Fig. 3, it follows that I_R is the relevant parameter that determines the properties of CE. Particularly interesting here are the dependences $A\propto I_R$ and $W\propto I_R$ that yield CE energy $E_{\mathrm{CE}}\propto I_R^2$. It was previously observed that the CE pulse energy is quadratic in incident pulse energy, below saturation [3,4]. It had been assumed that this resulted from a process that was a quadratic function of intensity, like four-wave-mixing or cooperative resonance fluorescence. The present data show that this relation occurs, instead, because the CE pulse amplitude and width are both linear functions of intensity below saturation. Another idea, suggested by the linearity of CE power with incident intensity, is that a large, saturated fraction of laser beam energy is converted to CE during the time when CE is generated. If approximately a constant fraction of the laser energy is converted from each saturated filament into CE, the net cone pulse amplitude should be proportional to the number of filaments, known to be proportional to the laser intensity [2]. This would explain the observed $A\propto I_R$ dependence.

The fact that cone and laser peaks do not coincide indicates that the effect is not a standard nonlinear process. In contrast to the delay D from the laser peak, which is not very meaningful, the delay of the pulse onsets D_0 exhibits a clear $1/I_R$ dependence [Fig. 3(d)]. Such dependence is characteristic for cooperative processes like superfluorescence [12] and cooperative Raman scattering [13]. In these processes, analogous dependence is observed in the delay of cooperative emission relative to the time when saturation and/or inversion is reached in an atomic system. Another well-known feature of the cooperative emission is the shortening of the cooperative emission pulses with increasing laser intensity. In our experiment, however, the opposite behavior is observed [Fig. 3(b)]. This could be due to the above mentioned increasing number of filaments at elevated I_R: even if individual CE pulses stemming from individual filaments exhibit characteristic cooperative shortening, but fluctuate temporarily, our accumulative recording of some 100 pulses with a limited time resolution would result in a wide broadened pulse.

These observations contradict earlier models based on steady-state calculations. One particularly popular model [2] assumed that CE occurs on the Rabi sideband. There was also experimental evidence showing that the maximum of CE scales like the generalized Rabi frequency Ω' [3]. On the other hand, other experimental studies [4,11,14] indicate that despite this scaling, the detuning of ν_c differs from Ω'. The present results provide additional arguments that CE *does not* occur on the Rabi sideband. If CE were generated as one of the Rabi sidebands and $\Delta_c=\nu_c-\nu_0$ were related to the incident intensity during the pulse, then as Ω' increases with intensity, the largest Δ_c would occur at the pulse peak, and the smaller Δ_c would occur on both sides of the peak, with an increasing separation as Δ_c decreases. This possibility is clearly ruled out by the measurements of CE pulses at different Θ, i.e., different ν_c, so CE cannot be directly associated with the AC-Stark enhanced emission or gain as suggested in Refs. [2,8].

This reported time-resolved study of CE has considerably expanded the available information regarding the character of pulse-excited atomic CE: it rules out the possibility that CE is seeded by laser ASE, and does not support the FWM interpretation of this phenomenon. CE may be related to temporal beam breakup found by Crenshaw and Cantrell [15] and Starostin *et al.* [16] by numerical analysis. These observations indicate that a full interpretation of CE should be based on nonperturbative and nonstationary theory including the coupled effects of both the incident and CE fields on the medium as well as atomic correlation effects.

This work was supported by the National Science Foundation. W. G. would like to thank JILA, University of Colorado, for its hospitality and support. W.G.'s work was partly supported by the Polish KBN (Grant No. 2P0301516).

[1] J. F. Valley, G. Khitrova, H. M. Gibbs, J. W. Grantham, and X. Jianin, Phys. Rev. Lett. **20**, 2362 (1990).
[2] J. Harter and R. W. Boyd, Phys. Rev. A **29**, 739 (1984).
[3] W. Chałupczak, W. Gawlik, and J. Zachorowski, Opt. Commun. **99**, 49 (1993).
[4] R. C. Hart, L. You, A. Gallagher, and J. Cooper, Opt. Commun. **111**, 331 (1994).
[5] C. H. Skinner and P. D. Kleiber, Phys. Rev. A **21**, 151 (1980).
[6] I. Golub, G. Erez, and R. Shuker, J. Phys. B **19**, L115 (1986).
[7] L. You, J. Mostowski, J. Cooper, and R. Shuker, Phys. Rev. A **44**, R6998 (1991).
[8] Y. Shevy and M. Rosenbluh, J. Opt. Soc. Am. B **5**, 116 (1988).
[9] B. D. Paul, Ph.D. thesis, University of Colorado at Boulder, 1999.
[10] B. D. Paul, M. L. Dowell, A. Gallagher, and J. Cooper, Phys. Rev. A **59**, 4784 (1999). An extensive reference list is provided, as well as a discussion of experiments and theories.
[11] W. Chałupczak, W. Gawlik, and J. Zachorowski, Phys. Rev. A **49**, 4985 (1994).
[12] A. Kumarakrishnan and X. L. Han, Phys. Rev. A **58**, 4153 (1998).
[13] V. S. Pivtosv, S. G. Rautian, V. P. Safonov, K. G. Folin, and B. M. Chernobrod, Zh. Eksp. Teor. Fiz. **81**, 468 (1981) [Sov. Phys. JETP **54**, 250 (1981)].
[14] L. Burdge and C. H. Lee, Appl. Phys. B: Photophys. Laser Chem. **28**, 197 (1982).
[15] M. E. Crenshaw and C. D. Cantrell, Opt. Lett. **13**, 386 (1988); Phys. Rev. A **39**, 126 (1989).
[16] A. N. Starostin, A. A. Pantaleev, V. I. Lebedev, S. V. Rotin, A. G. Leonov, and D. I. Chekhov, Zh. Eksp. Teor. Fiz. **108**, 1203 (1995) [JETP **81**, 660 (1995)].