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Conical emission as cooperative fluorescence

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We have observed conical emission from a dense, strongly driven ensemble of two-level atoms without noticeable self-focusing. We present the arguments that conical emission, unaffected by autocollimation, is due to cooperative, Čerenkov-like, spontaneous emission of collisionally perturbed, optically dressed atoms rather than to parametric wave mixing. Our results indicate the importance of many-atom correlations which must be taken into account when strong light interacts with dense media.

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Conical emission (CE) is one of the most spectacular nonlinear optical phenomena associated with the propagation of an intense light beam in a dense medium. It manifests itself by the occurrence of emission in a well-defined direction at a small angle to the propagating beam, forming one (or more) characteristic light cones. CE is usually defined as a process in which the propagation of a strong, near-resonant light beam, detuned to the blue of the line center in a dense medium of two-level atoms results in the production of a light cone with the frequency shifted to the red. The phenomenon of CE very often accompanies various experiments in which strong, nearly resonant laser light interacts with an optically dense medium. It is thus necessary to study CE for a better understanding of the fundamental processes of the interaction of light with dense media, as well as for the practical reasons of maximizing the efficiency of energy deposition from a light beam into matter (e.g., in the multiphonon ionization, isotope separation, plasma research, etc.). CE has been observed in dense metal vapors (barium [1], strontium [2], and sodium [3,4]) and has been interpreted in many different ways. The apex angle of the light cone in CE strongly depends on the laser wavelength and on the density of the medium, but does not depend on the intensity of the laser. In most previous observations it has been found that the frequency of the light emitted in the cone is independent of the laser intensity but depends on its frequency [1]. The appearance of such cones has previously been observed exclusively in association with self-focusing and self-filamentation of the propagating beam. It is only in the work of Chałupczak, Gawlik, and Zachorowski [5] that CE has been studied without autocollimation and that an intensity dependence of the cone frequency has been seen.

The most popular interpretation of CE, by Harter and Boyd, is based on the four-wave mixing (FWM) in a dressed-atom model [6]. According to this model, any signal arising from the laser background or spontaneous emission at the Rabi sideband frequency, $\omega_L \pm \Omega'$, where ω_L is the laser frequency and $\Omega' = [(\omega_L - \omega_0)^2 + (ED/\hbar)^2]^{1/2}$, can be amplified by interaction with a strongly driven two-level system or by multiphoton mixing. As a result of FWM the component at $\omega_L - \Omega'$ is scattered at an angle determined by phase matching and refraction on the border of the medium perturbed and saturated by the propagating beam. According to this interpretation, the second sideband, which is amplified by the three-photon scattering (TPS) and FWM, is trapped together with the incident beam by self-focusing and filamentation. In this model the role of self-focusing is crucial. On the one hand, it is responsible for a strong refractive-index gradient on the surface of the filament, which determines the emission angle of the red sideband; on the other hand, it explains why only one cone is observed although two cones are predicted by the simple phase-matching condition. Moreover, the observed insensitivity of the cone angle and frequency to the light intensity is logically explained by a stabilization of the light intensity within a saturated filament. In a study by Shevy and Rosenbluh [4] it has been pointed out that TPS itself could produce an additional cone and that the light emitted due to TPS and FWM can interfere destructively.

Another model [7] treats CE as the result of a Čerenkov-like emission, where the laser-induced polarization moving with velocity $c/n(\omega_L)$ produces coherent radiation at $\omega_L - \Omega'$ that propagates with velocity $c/n(\omega_L - \Omega')$. This velocity difference is responsible for conical emission of the $\omega_L - \Omega'$ light. Valley *et al.* [8] interpret CE as a combination of FWM, initiated by resonance fluorescence of uncorrelated atoms, with propagation effects. You *et al.* [9] have argued recently that it is correlation of the emitters which is responsible for CE and support the model of Čerenkov radiation.

In spite of many experiments CE has eluded satisfactory explanation for a long time. In fact, it was only quite recently that a main assumption of many theories, that CE occurs on one of the Rabi sidebands, has been convincingly proved [5]. Like the well-known Mollow triplet in resonance fluorescence, CE can thus be regarded as a spectacular manifestation of the energy structure of a dressed atom.

In this Rapid Communication we present the results of measurements which clearly demonstrate that CE does not originate from FWM but is rather due to cooperative spontaneous emission. Our results are consistent with the theory of You *et al.* [9]. To perform this experiment (Fig. 1) we decided to use barium atoms, for which the $6s^2\,{}^1S_0$–$6s6p\,{}^1P_1$ (553.548 nm) transition is a

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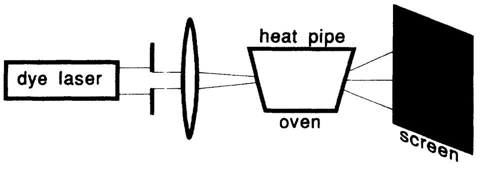

FIG. 1. Experimental setup.

good approximation to the two-level model. This eliminates many complications which may arise from additional energy levels as, e.g., in the case of fine and hyperfine structure of Na. Ba atoms, with an admixture of 5–20 Torr of argon as a buffer gas, were heated up to about 900 °C (density $N \simeq 10^{14}$ cm^{-3}) in a 5-cm-long interaction region. The vapor was illuminated by weakly focused dye-laser pulses of energy up to 30 μJ, pulse duration of 10 ns, and spectral width of ca. 5 GHz. In the measurements we report here, self-trapping was carefully avoided by keeping the light energy in the interaction region sufficiently low. Radiation at the output of the Ba oven was analyzed spatially with a linear diode array and charge-coupled-device camera, and spectrally using a 2-m grating spectrograph with a 4-GHz resolution. With the use of appropriate diaphragms, either the centrally or off-axially emitted light was selected. We have found that in contrast to earlier opinions, self-focusing and filamentation are not at all necessary for the occurrence of CE. On the contrary, the cones are even sharper and more clearly visible without self-focusing.

The absence of self-focusing is the main difference between this work and earlier experiments. Another important feature of this work is the low light intensity, which enables us to observe the primary contribution to CE unobscured by wave mixing and self-focusing which are present at higher intensities and could overwhelm the initial Čerenkov-like mechanism.

Measurements of the off-axis (cone) intensity I_C for given laser light intensity I_L give us a quadratic dependence on N, with a reabsorption correction $I_C \propto N^2 \exp(-aN)$, where a is a constant related to the absorption coefficient. This proves the coherent nature of the effect characteristic for both FWM and cooperative spontaneous emission. The I_C dependence on I_L for a given density is quadratic (Fig. 2) and saturates for higher I_L. The model of Harter and Boyd [6] predicts that both sidebands at $\omega_L \pm \Omega'$ can be parametically amplified and should be present in the spectrum behind the oven, even when seeding occurs only on one sideband. Figure 3 is a typical spectrum taken under the conditions of a clearly visible cone. Apart from the laser peak at ω_L, which is related to the central beam, and the red sideband at $\omega_L - \Omega'$, which is related to the cone [5], no other spectral feature is visible. In particular, we have not seen any component at $\omega_L + \Omega'$. We may also recall that in several earlier experiments only two spectral components have been observed [1,3,10]. The absence of the blue sideband in our spectra is most likely caused by collisions. It is known [11] that spontaneous emission from collisionally perturbed dressed atoms has an asymmetric spectrum: the amplitude of the $\omega_L + \Omega'$ sideband is strongly quenched by collisions when $\omega_L > \omega_0$ [12]. Collisions also also decrease the FWM amplitudes, but do not change their symmetry. This is also collisional broadening that determines the widths of all spectral features seen in Fig. 3. There is no evidence of any self-modulation in our experiment as neither the cone line at ω_C nor the laser line ω_L is broader than the linear absorption spectrum.

To get more insight into the role of parametric interactions in our experiment we have used a second, weaker dye laser, whose light was sent into the oven at a small angle with respect to the main beam. To see any FWM effect, the pump pulse energy had to be increased to 30–100 μJ, while the probe had 10 μJ. When the probe of frequency ω_P was tuned far from ω_0 and from the Mollow triplet (ω_L, $\omega_L \pm \Omega'$), its beam was transmitted through the vapor without any perturbation, but when ω_P was tuned to the missing sideband ($\omega_L + \Omega'$), a strong amplification of the $\omega_L + \Omega'$ and ω_L spectral components occurred. It was clearly directional, with a symmetry given by the propagation of the pump beam, and created a partial cone, giving a crescentlike shape on a screen in

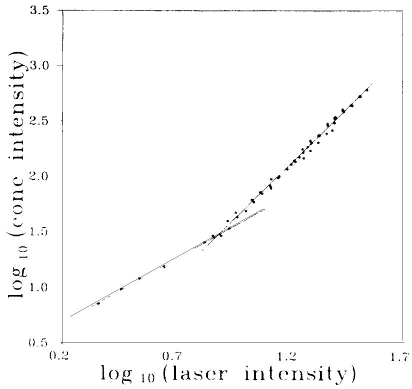

FIG. 2. Dependence of integrated cone intensity on laser light intensity in $\log_{10} \times \log_{10}$ scale (vapor density 5×10^{13} at./cm^3, $\omega_L - \omega_0 = 30$ GHz). The slopes of the dependence are about 1 for low laser intensity where essentially the scattered light background is measured and 2 for higher laser intensity.

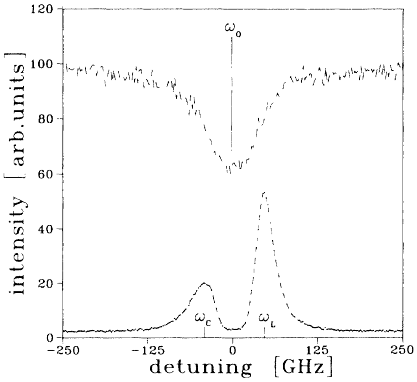

FIG. 3. Spectrum of the light observed behind the oven for vapor density 1.2×10^{14} at./cm^3, $\omega_L - \omega_0 = 47$ GHz (the lower curve). The upper curve is the linear absorption spectrum for frequency reference, taken at the same vapor density. The intensity is measured in arbitrary units.

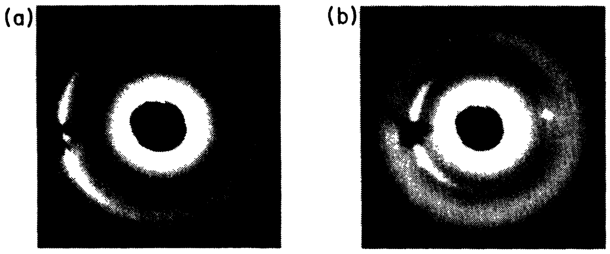

FIG. 4. Spatial light distribution behind the oven in the pump-probe beam experiments. (a) Phase-matching condition not fulfilled, the amplification of the probe beam tuned to the blue Rabi sidebands observed. (b) Proper phase matching. Parametric emission appears in the complementary angle and produces the bright spot to the right of the center. The pump and probe beams are blocked to avoid the saturation of the camera.

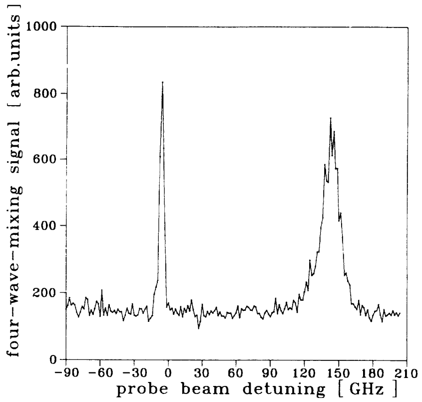

FIG. 5. Four-wave-mixing signal intensity as a function of the probe beam detuning ($\omega_P - \omega_0$) (vapor density 10^{14} at./cm^3, pump beam energy 30 μJ, probe beam energy 10 μJ, $\omega_L - \omega_0 = 63$ GHz). The intensity is measured in arbitrary units.

the far field [Fig. 4(a)]. The probe intensity behind the oven reached its maximum for $\omega_P = \omega_L$ and for the angle between the beams close to the cone angle [13]. Further studies of this phenomenon will be published elsewhere.

At the proper angle θ between the two beams and for the proper frequency ω_P, such that phase-matching and energy-conservation conditions are fulfilled, parametric emission into the complementary angle $-\theta$ appears at frequency $2\omega_L - \omega_P$ [Fig. 4(b)]. As displayed in Fig. 4(b), CE and FWM can be seen simultaneously, for the same conditions, except that FWM requires seeding by an external beam. It should be noted that for $\omega_P = \omega_L - \Omega'$ the parametric emission at $\omega_L + \Omega'$ is not trapped within the central beam, while, according to Ref. [6], this should be the case if self-focusing were not eliminated. The intensity of the FWM beam versus the detuning of the probe, i.e., the excitation spectrum of FWM (Fig. 5), shows two maxima, at $\omega_P = \omega_L + \Omega'$ and $\omega_L - \Omega'$, illustrating the generation of one sideband due to the parametric interaction of the other one with the pump (ω_L) photons. The widths of the peaks in Fig. 5 are significantly different, which we ascribe to the reabsorption at ω_0, but their amplitudes are nearly equal. This amplitude symmetry is not reproduced by the spectrum of Fig. 3, where only one of the sidebands occurs, suggesting that FWM does not play any major role in CE. Moreover, the intensity of the parametric beam follows the probe beam intensity linearly, indicating that FWM is not saturated in our conditions. One could thus reason that if the cone were due to the mixing process initiated by spontaneous emission or laser background, its intensity should also be linearly dependent on the source intensity. As the spontaneous emission into a sideband from the dressed atom is itself (for weak I_L) a quadratic function of the pump intensity while the background roughly a linear one, the function $I_C(I_L)$ should show a stronger dependence than the observed I_L^2. We have also verified that CE is not initiated by the dye fluorescence, by filtering the incoming beam using a grating monochromator. The instrumental width (15 GHz) was smaller than Ω'; thus the background at the Rabi sideband frequencies was strongly attenuated. Despite this, the cone is still clearly visible, demonstrating that CE is not caused by amplification of an external background.

In this Rapid Communication, we have presented the results of systematic studies of conical emission in a dense medium of two-level atoms, without noticeable self-focusing. Summarizing, we have found that (i) CE is a cooperative ($\propto N^2$) phenomenon, (ii) it is due to emission and not amplification of external background, (iii) it has a two-component spectrum, and (iv) it has a quadratic intensity dependence, consistent with the characteristics of dressed-atom, collisionally perturbed fluorescence. Our observations refer to relatively low light intensity. Under these conditions the effect is a volume effect not limited to a narrow light channel. All these facts support the conclusion that it is the correlated spontaneous emission (Čerenkov effect) that is responsible for the cone formation and not the process of four-wave mixing. It is possible that at higher intensities CE may be perturbed by filamentation and refraction on the boundary between the regions of different refractive indices, as described by Harter and Boyd [6], but, as shown above, this cannot constitute the primary mechanism responsible for the cone formation. The CE effect proves the existence, and spectacularly illustrates the role, of correlations in the spontaneous emission of strongly driven atoms in dense media. Such correlations may be caused by spectral filtration of vacuum fluctuations by spontaneous emission, as proposed recently by You *et al.* [9]. The possibility that these are correlation effects of the superfluorescence type cannot be excluded, because, as we have found, the cones appear only when $N \gg \lambda^{-3}$, where λ is the wavelength. These effects deserve further theoretical and experimental study because of their role in the description of quantum fluctuations and correlation effects in dense media [9,14] and in the understanding of the interaction of strong light with dense media. They also become important in the context of recent suggestions that strongly dispersive and lossless media could be obtained [15], of the influence of local fields on nonlinear processes [16], and of a better understanding of collective processes in the optical domain. In particular, it could be very interesting to observe CE from highly regular systems; e.g., in the case of crystallization in traps or storage rings [17].

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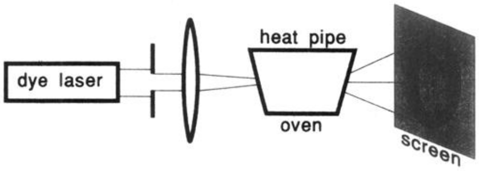
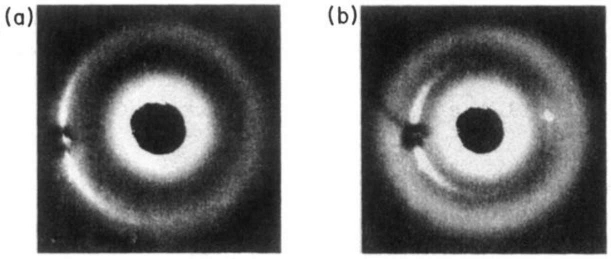


FIG. 1. Experimental setup.



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