Anomalous conical emission: Two-beam experiments

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We report the observation of anomalous conical emission in the first resonant transition of atomic calcium in the presence of a second probe beam. The observations do not favor the refraction-at-boundary model unless the incoherence between the beams plays a crucial role. On the other hand, an induced defocusing of a blue-shifted beam takes place in the presence of a strong red-shifted beam. This result is rather surprising, since it is observed under circumstances where self-focusing would be expected.

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Anomalous conical emission (ACE) has been observed in several metal vapors such as potassium [1], barium [2], sodium [3,4], and strontium [5] when an intense laser beam is tuned close to resonance. The cone angle and its spectrum have been reported as a function of various parameters such as laser detuning and vapor density. The main features that have been observed are (i) a strong red-shifted contribution in the cone spectrum is symmetrically but oppositely displaced with respect to the laser frequency, (ii) the cone angle increases approximately with the root of the vapor density and with the inverse root of the detuning, and (iii) the cone angle and its spectrum are insensitive to the incident laser power.

From a theoretical point of view, several explanations have been attempted with limited success. The most relevant theories that have been invoked are resonantly enhanced radiation by four-wave mixing [3], stimulated hyperfine Raman scattering with self-phase modulation [6], temporal pulse reshaping of Rabi sidebands [7,8], and Čerenkov radiation from a rapidly moving focus [9,10]. Recently, several models have been developed that include competition between different processes [11–13]. Partial agreement with some experimental results has been obtained [14]. A mechanism widely invoked to explain the angular divergence is the so-called " refraction-at-filament-boundary model" which is due to the refraction of light from a saturated refractive index within the filament and a low-intensity refractive index outside this region.

We have observed ACE in the first resonant transition of atomic calcium vapor. A detailed study of the single-beam experiments and a comparison with previous observations will be reported elsewhere. In this paper we report ACE in the presence of a second probe beam. These two-beam experiments yield unexpected results.

The experimental setup consists of a calcium oven not operating in heat-pipe mode with argon as a buffer gas. The system has been operated in a vapor density range of 3×10^{20} m^{-3} to 2×10^{21} m^{-3} corresponding to temperatures of 400 to 700 °C and pressures from 2.79×10^{-2} (2.79 Pa) to 2.69×10^{-1} mbar (26.9 Pa). The dye laser is a homemade Littman Metcalf arrangement pumped by the third harmonic of a commercial Nd: YAG laser (where YAG denotes yttrium aluminum garnet) (3550 Å). The spectral width of the laser was 5.6×10^{-2} Å and the energy per pulse 75 μJ at a repetition rate of 3 Hz with a pulse duration of 9 ns. The spectra were measured with a Czerny-Turner Spectrometer with a 0.1-Å resolution limit. The light was detected with a photomultiplier coupled to a pulse integrator and an averager system. Averages over ten pulses with a 30-ns gate window integrator were made and the analog output was sent to a plotter.

The laser beam was focused at the oven center with a 0.4-m lens achieving an energy density of 2.4×10^{-4} J/m^2 in a beam waist of 10 μm. The calcium line center $^1S_0$-$^1P_1$ was located via absorption in the linear susceptibility region. The laser wavelength as well as the spectrometer were calibrated from this line in order to have a minimum uncertainty in the detunings.

In order to test the refraction at boundary theory experimentally, we used two beams from different dye lasers tuned to different frequencies. The two beams were injected in the same direction using a beam splitter (BS2) as shown in Fig. 1. One intense laser beam was tuned on the blue side of the transition under circumstances where the generated cone was intense and clear. This corresponded to a detuning of −0.73 Å and an intensity of 55 MW cm^{-2}. The cone light was steered into a spectrometer set

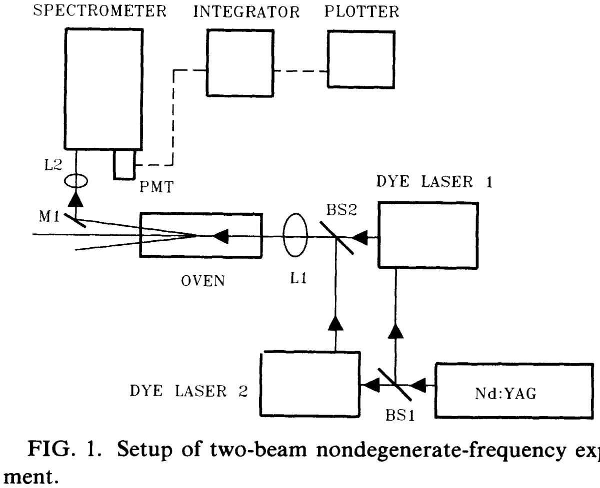

FIG. 1. Setup of two-beam nondegenerate-frequency experiment.

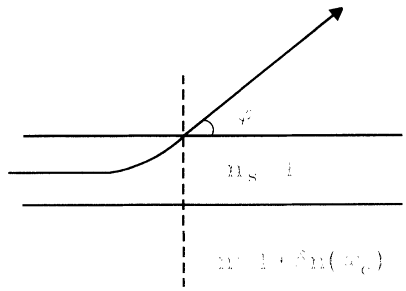

FIG. 2. Refraction-at-boundary model. An intense incident beam generates a fully saturated medium with refractive index $n_s=1$ whereas the region where the incident beam is absent exhibits a linear refractive index $n=1+\delta n$.

at the cone peak frequency. The other laser, using a probe beam with low intensity, was tuned on the red side of the transition at the center frequency of the cone generated by the first beam. This frequency corresponded to 0.73 Å and an intensity of 0.55 MW cm^{-2}.

According to the refraction-at-boundary theory, we would expect the probe beam to deviate at the cone angle as shown in Fig. 2. However, when the experiment was performed, no increase in the cone intensity was observed in the presence of the probe beam. A typical frequency resolved curve shows that the cone spectrum is identical whether the probe beam is present or not as depicted in Fig. 3 with a dotted line. The spectrum has a faint contribution of light centered at the laser frequency but with a broader linewidth and a strong red wing typically observed at the cone angle. Therefore, we conclude that the probe beam tuned at the cone center frequency does not deviate at the cone angle. Furthermore, no deviation of this beam was detected. This result does not favor the "refraction-at-boundary" proposal which is widely invoked to explain the angular dependence of the cone. An important question is whether the incoherence between

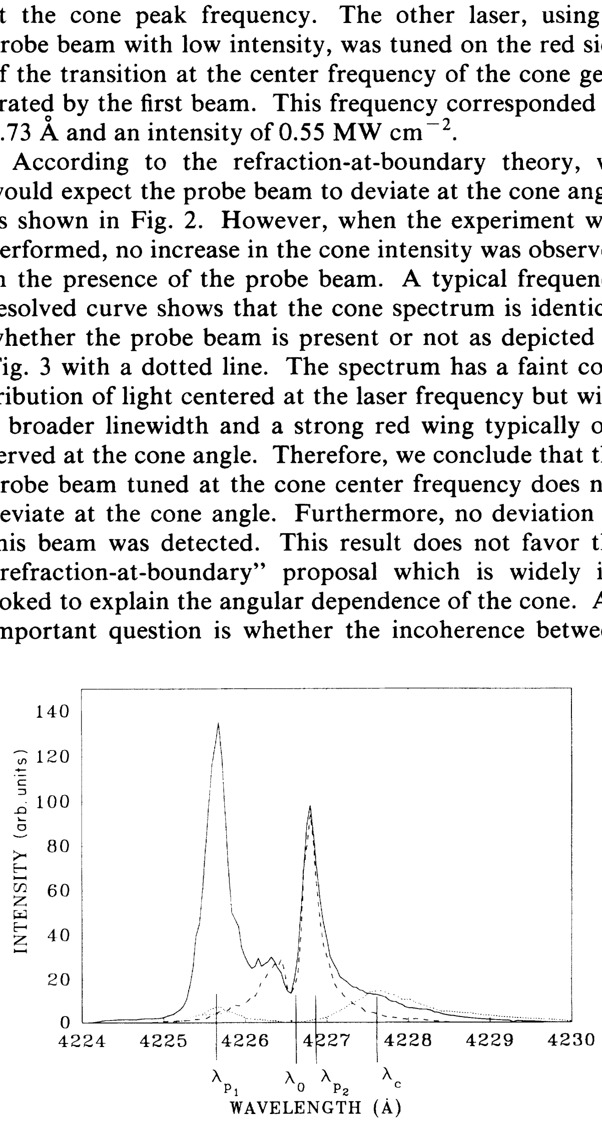

FIG. 3. Induced defocusing results. λ_p represents the pump laser wavelength, λ_0 the atomic resonance wavelength, and λ_c the cone peak wavelength as well as the second laser wavelength. The dotted line spectrum was obtained in the presence of the blue-tuned laser (1) alone. The dashed line spectrum was obtained in the presence of the red-tuned laser (2) alone. The solid line spectrum was obtained when both lasers were present.

the two laser sources is playing a relevant role in this negative result. To this end, we recall that You, Mostowksi, and Cooper [13] suggest a large correlation length of the polarization, rather than δ-correlated assumption of Valley *et al.* [12]. According to this theory [13], the correlation of the source atoms generated by the pump beam is very small as a polarization source for the probe beam since the two beams are incoherent between them.

While performing this experiment, we observed induced defocusing of the high-frequency beam in the following conditions. The blue-tuned laser labeled (1) was left as it was, that is, tuned at −0.73 Å, and the cone radiation was resolved as a function of frequency. The curve obtained is shown with a dotted line in Fig. 3. In the absence of the blue-tuned laser, the other laser labeled (2) was tuned on the red side very close to resonance at 0.20 Å and its intensity was increased to 66 MW cm^{-2}. The spectrum obtained at the cone angle is shown with a dashed line in the same figure. There is a strong contribution at the laser frequency presumably due to self-defocusing, although a significant contribution seems to come from fluorescence of the vapor. With a solid line, we depict the spectrum obtained when both beams were present. The red wing is similar to the superposition of the separate contributions. However, on the blue wing, the intensity increases considerably at the first laser frequency. Since detection is being made at the cone angle of 5 to 30 mrad, the blue-tuned laser (1) is defocused due to the presence of the red-tuned laser (2).

This result is most unforeseen since we would expect the blue laser to experience focusing rather than defocusing, even in the presence of the second beam. This prediction may be seen from the nonlinear susceptibility calculation. The steady-state refractive index for a two-level atom is given by [15]

$$n_R^2=1+\mathrm{Re}\{\chi(E)\}=1+\frac{N\mu^2}{\epsilon_0\hbar\gamma_2}\,\frac{-\dfrac{(\omega-\omega_0)}{\gamma_2}}{1+\dfrac{(\omega-\omega_0)^2}{\gamma_2^2}+\dfrac{\Omega^2}{\gamma_1\gamma_2}}\;,$$

where N is the calcium density, μ is the dipole moment, ω is the external field frequency, ω_0 is the atomic transition frequency, $\Omega=pE/\hbar$ is the Rabi frequency, and γ_1 and γ_2 are the longitudinal and transverse relaxation rates.

Under very intense fields, in the so-called saturation regime, Ω is very large and the saturated refractive index is $1(n_s=1)$. When tuned to the blue side of the transition, the detuning is positive $(\omega-\omega_0>0)$ and the refractive index is less than 1. The refractive index in the low-intensity limit is always smaller than the intensity-dependent refractive index. A beam tuned to the blue side thus exhibits focusing, since the phase of the beam with high intensity (at its center) travels slower than the phase of the beam with low intensity (far from he center). When the second beam is present, the Rabi frequency is modified by $\Omega^2=(\mu^2/\hbar^2)EE^*$, but this quantity is only amplitude dependent. Thus whether the second beam is red or blue does not change the overall sign of this term. On the red side of the transition, the detuning is negative and the situation is opposite to the above leading to defocusing. We should mention that induced focusing in a self-defocusing medium was predicted by Agrawal [16] using two coupled nonlinear equations for two beams with the same frequency. This prediction has been observed by Stentz *et al.* [17]. A similar theoretical treatment with nondegenerate frequencies has not been published to our knowledge, and we wonder whether it may account for the above observations.

The explanation of this phenomenon is not clear to us. It may be possible that the red-tuned laser (2) modifies the absorption coefficient of the medium, and the blue-tuned laser is diffracted by the spatial modulation produced by the former beam. If this were the case, the relative phase between the two lasers becomes unimportant. The spatial deviation of the blue beam (1) due to the red beam (2) may involve an energy transfer between the beams. Nonetheless, within the present experimental resolution, the peak intensity at λ_{p2} is not depleted by the presence of the first beam.

We have observed ACE in the first resonant transition of calcium vapor in the presence of a second probe beam. A two-beam nondegenerate-frequency experiment exhibited no deviation of a probe beam centered at the cone frequency, therefore not supporting the refraction at boundary theory. A similar experiment showed induced defocusing of a blue-tuned laser beam in presence of a strong red-shifted beam. To our knowledge, this induced defocusing using two beams with different frequencies in a collinear configuration has not been observed previously.

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