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Cone emission from laser-pumped two-level atoms

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In spite of the fact that cone emission was first observed over a decade ago there is still no generally accepted theory of the effect. Here we give an outline of a unified approach and point out a mechanism that supports Cherenkov-type radiation due to vacuum fluctuations as a possible source of cone emission. This can be considered as an additional mechanism to the previously discussed four-wave mixing, and initial encoding and follow-up refraction effects. We compare our treatment to previously considered models.

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Cone emission has been the subject of numerous experiments and theoretical treatments in recent years [1–8]. The effect occurs when a strong laser beam propagates in atomic vapor, for example, sodium vapor. The frequency of the laser beam should be close to, but greater than, that of the atomic resonance transition. The propagating laser beam leads to emission of other beams separated in frequency and direction from the original strong beam. Of particular interest is a beam of lower frequency (red-shifted component) which forms a cone in the far-field intensity distribution. A blue-shifted beam is also formed. The blue-shifted component is emitted in the forward direction only.

Several mechanisms leading to cone emission have been discussed but none has provided a fully satisfactory description. Probably the most complete treatment to date is the recent paper by Valley *et al.* [2]. It is believed that the process of formation and propagation of the three beams is a complicated one and involves several effects. Each effect is very simple in principle, but their combination is quite complicated. Since it is questionable whether one simple effect can adequately describe the whole process, careful theoretical analysis is needed. The paper by Valley *et al.* [2] ascribes the cone emission to an interplay between four-wave mixing (4WM) and the effects of diffractive spreading during propagation. Other authors [3], however, have invoked Cherenkov emission to explain the phenomenon.

In this paper we point out that a Cherenkov-type emission, not considered by Valley *et al.* [2], also should be taken into account when discussing cone emission. This effect relates to the initiation and generation of the frequency-shifted beams and is intrinsically quantum in nature. It comes from the spatial correlation of the medium polarization generated by spontaneous emission at the relevant frequencies, and is in many ways analogous to a spontaneous 4WM (at least in the absence of collisions) since the photons emitted in the Rabi sidebands are correlated. We show by explicit calculation that due to the interaction with the electromagnetic field, the polarization of the medium has a large correlation length [9], contrary to the δ-correlated assumption of Valley *et al.* [2]. The in-phase polarization of the medium acts as a Cherenkov-type source for the two beams (blue- and red-shifted). Our results show that the source, although quantum in nature, is closer to a coherent source than to noise. This has far-reaching consequences, especially for the angular distribution of the beams.

The system under consideration is modeled by the electromagnetic field and a medium formed by two-level atoms (Fig. 1). The electromagnetic field will be decomposed into several components. One of the fields, the pump, has large intensity and will be treated as a classical field. Under the influence of the strong pump beam the medium responds by forming two weak beams of different frequencies at the Rabi sidebands [10]. These two fields will be described by quantum-mechanical field operators, $\hat{E}_s$ and $\hat{E}_4$, and the Heisenberg equations of motion will be used to find the evolution of these fields.

It is assumed that the medium has a pencil-like shape with the Fresnel number of the order of unity. This assumption allows us to describe the propagation of waves in the framework of the paraxial approximation (PA).

We have performed careful quantum-mechanical analysis of the response of the medium to the total electromagnetic field. The system of atomic operators is solved to all orders in the strong pump field, and up to first order in the generated fields. The polarization of the medium is found by a coarse-graining procedure. Detailed analysis will be described in a future presentation [9].

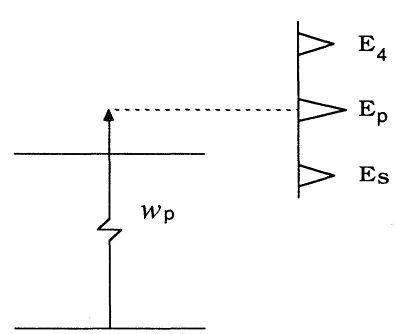

FIG. 1. The energy-level scheme of the two-level medium together with the sideband field distribution.

In the steady-state limit, under the slowly varying envelope approximation and the rotating-wave approximation, we arrive at the following equations for the propagation of the two generated sideband fields, which are similar in form to those of Valley *et al.* [2],

$$\left[\nabla_T^2 - 2ik_s\frac{\partial}{\partial z}\right]\hat{E}_s^{(-)} = -\alpha_s\hat{E}_s^{(-)} + \kappa_4\hat{E}_4^{(+)} + \hat{\beta}_s\,,$$
$$\left[\nabla_T^2 + 2ik_4\frac{\partial}{\partial z}\right]\hat{E}_4^{(+)} = -\alpha_4^*\hat{E}_4^{(+)} + \kappa_s^*\hat{E}_s^{(-)} + \hat{\beta}_4^\dagger\,, \tag{1}$$

where the $\hat{E}_j^{(-)}$ and $\hat{E}_j^{(+)}$ ($j=s$, 4) are the slowly varying envelope operators for the positive and negative frequency parts of the electric-field operator in the Heisenberg picture.

The coefficients α_s and α_4 describe the polarizability and the gain or loss of the medium subject to the strong pump. The coefficients κ_4 and κ_s describe the 4WM process. Of crucial importance are the inhomogeneous terms $\hat{\beta}_s$ and $\hat{\beta}_4$, which are the quantum noise sources that describe the spontaneous emission of photons from the pumped medium. To correctly model the propagation of the two weak beams in the medium one must solve the propagation equation for the pump beam, find the appropriate α's and κ's as well as the properties of the noise terms $\hat{\beta}$, and solve Eq. (1). This has been done [9] and some of the main conclusions are reported here.

The main results are related to the correlation of the noise terms. We have found that under the PA, the correlation functions are of the form

$$\langle\hat{\beta}_j(\mathbf{r})\hat{\beta}_{j'}^\dagger(\mathbf{r}')\rangle \sim \delta_{jj'}\frac{1}{ik_j(z-z')+k_j^2a^2} \times\exp\left[\frac{-(k_j|\delta\rho|)^2}{2[ik_j(z-z')+k_j^2a^2]}\right], \tag{2}$$

where $\mathbf{r}=\boldsymbol{\rho}+z\hat{\mathbf{z}}$, k_j ($j=s$, 4) is the center wave vector of the Rabi sideband, and a is the radius of the coarse-graining cell. Thus the noise source is not δ correlated, as assumed in Ref. [2]. On the contrary, the correlation length is long and can very easily exceed the length L of the amplifier. The reason for such a long correlation can be easily understood. The noise terms are not the vacuum electromagnetic fields themselves but rather the medium polarization induced by electromagnetic vacuum fluctuations. The electromagnetic vacuum can be viewed as a white noise, with a very short correlation time. In the steady state this very short correlation time is transformed to a very short correlation length. However, the atomic system responds to the vacuum fluctuations by essentially filtering the resonant components. Thus when driven by a white noise, the atomic system responds with a frequency bandwidth of the order of the spontaneous decay rate γ. In the steady state this bandwidth gets transformed to the correlation length, which is of the order of c/γ.

We will now describe the results of model studies which show how the long correlation length of the noise sources influences the formation of the cones. In order to gain some insights into Eqs. (1) and (2), we first study them perturbatively. Neglecting the mixing with the wave $\hat{E}_4^{(+)}$, we can write the equation for $\hat{E}_s^{(-)}$ as

$$\left[\nabla_T^2 - 2ik_s\frac{\partial}{\partial z}\right]\hat{E}_s^{(-)} = -\alpha_s\hat{E}_s^{(-)} + \hat{\beta}_s\,, \tag{3}$$

which describes the generation of $\hat{E}_s^{(-)}$ field via the noise source $\hat{\beta}_s$ and the susceptibility induced by the pump. With α_s equal to a real constant (note that the imaginary part will contribute to the gain of the field in addition to the source), this equation can be solved.

In most cases, we are not interested in the intensity at the exit window of the medium, rather we wish to calculate the intensity in the far-field limit. We will assume that the field propagates to the far field according to Huygen's principle starting from the exit window.

First we assume that $\hat{\beta}_s$ has an infinite correlation length, i.e.,

$$\langle\hat{\beta}_s(\rho,z)\hat{\beta}_s^\dagger(\rho',z')\rangle \sim e^{-\rho^2/\rho_0^2}e^{-\rho'^2/\rho_0^2}\,, \tag{4}$$

where ρ_0 is the beam waist of the pump.

We have shown [9], by following the Huygen's principle procedure for calculating the far-field intensity outlined above, that the angular distribution of the red-shifted component has the form of a cone (Fig. 2) with the cone angle

$$\theta_c = (\alpha_s)^{1/2}/k_s \approx [2(n_s-1)]^{1/2} = (2\delta n_s)^{1/2}\,, \tag{5}$$

where n_s is the phenomenological index of refraction of the s wave, and $\delta n_s = n_s - 1$. Also the index of refraction of the strongly saturated pump field has been taken to be 1. Physically we can understand this as Cherenkov emission in the following sense: The pump field propagates close to the velocity of light and via spontaneous emission leads to the creation of the source of polarization $\hat{\beta}_s$ for the field $\hat{E}_s$, whose propagation is now at c/n_s rather than c. This provides a formal background for the Cherenkov-type mechanism.

Second, we compare the previous result with that obtained in the limit of very short correlation length for the noise sources. This is, however, just a model, and it can-

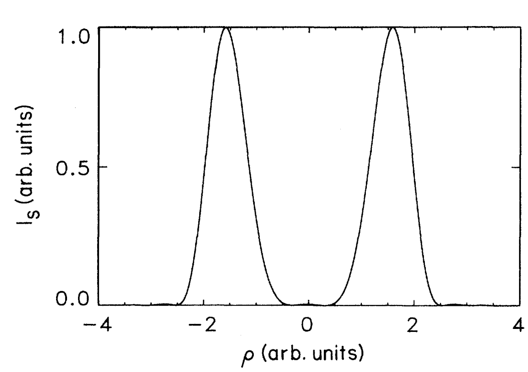

FIG. 2. Typical cross section of the cylindrical symmetric far-field intensity distribution of the red-shifted field due to noise source with long correlation length. I_s denotes the normal-ordered far-field intensity, and ρ is the transverse coordinate in the far field.

not be justified on the basis of Maxwell-Bloch equations. We assume that $\hat{\beta}_s(\rho,z)$ is a δ-correlated incoherent source (this is applicable when the coherence length is less than both the cell length L and the waist ρ_0 of the pump field). Thus

$$\langle \hat{\beta}_s(\rho,z)\hat{\beta}_s^\dagger(\rho',z')\rangle \sim e^{-\rho^2/\rho_0^2} e^{-\rho'^2/\rho_0^2} \times \delta(\rho-\rho')\delta(z-z') . \tag{6}$$

In this case, apart from an overall diffraction envelope determined by the waist ρ_0, the intensity does not depend on the transverse variables, which shows that the angular distribution is flat. Note that this is equivalent to the noise proposed by Valley *et al.* [2]. It emphasizes that the cone obtained for the long correlation length case [Eq. (4)] is due to the coherent excitation of the dipole polarization throughout the medium.

In order to study the effects of 4WM, we will neglect the source term $\hat{\beta}_s$ in Eq. (1) and then the coupled equations can be solved under similar assumption as Eq. (4) for the $\hat{\beta}_4$. Under the approximation $n_4 \approx 1$, equivalent to strong saturation of $\hat{E}_4$, the cone appears at the same position as in the pure Cherenkov case first studied without 4WM. The contrast of the cone is now reduced. Nevertheless, with the same coherent source, the angle we obtain from the 4WM is the same as from the Cherenkov radiation condition Eq. (5), while a simple analysis of the pure phase matching condition without diffraction and refraction (see below) gives a cone angle a factor of $\sqrt{2}$ smaller than Eq. (5).

Now we can also study the white-noise case equivalent to that due to Eq. (6) for the $\hat{\beta}_4$ with 4WM terms included. This gives, with $n_p \approx 1$, a minimum at $\theta_4 = (\delta n_s)^{1/2}$, again a factor of $\sqrt{2}$ smaller than Eq. (5). The first maximum is at

$$\theta_m \approx \left[\delta n_s + \frac{3\pi}{2k_pL}\right]^{1/2} , \tag{7}$$

and closely spaced subsidiary maxima occur very close to this value. Only when the overall diffraction envelope is taken into account does the resulting intensity profile appear as a well-defined cone at an angle somewhat greater than θ_4. In most cases the resulting maximum is close to θ_4, and θ_m is smaller than Eq. (5). Since Valley *et al.* [2] used δ-correlated noise in their calculations, this could be one of the reasons why the computed cone angles they obtained are always about 20% smaller than the experimental values. As we showed earlier, the δ-correlated noise in the $\hat{E}_s$ field equation will not give a cone at all. However, we see from the above calculation, that δ-correlated noise in the $\hat{E}_4^{(+)}$ field equation couples with $\hat{E}_s^{(-)}$ through 4WM. This coupling effectively filters the white noise, and makes it in a sense colored noise for the $\hat{E}_s^{(-)}$ field. The spatial phase matching then will lead to cone generation, but with a cone angle that is smaller than the θ_c of Eq. (5).

Since its experimental observation more than a decade ago, many theoretical and experimental studies of cone emission have been performed [1-8]. Some are highly numerical, and we plan to discuss them in a subsequent publication. However, many of the theories are simple enough that compact analytical formulas have been given. We will compare them with our results, and try to clarify some differences. We will write

$$n_j = n(\omega_j) = 1 + \delta n_j = 1 + \delta n(\omega_j) \quad (j = p, s, 4) .$$

Skinner and Kleiber [4] have proposed a simple parametric 4WM model, not of the same kind we are studying, but one with two additional components to 4WM with the pump and red-shifted fields. These two additional parametric components of the field at ω_1 and ω_2 mix with the pump at ω_p and the red-shifted component at ω_s, to give

$$\theta_c = 2(\delta n_s)^{1/2} \tag{8}$$

where $\delta n_s = -\delta n_p$ and $\omega_s \approx \omega_p$ is also assumed. If instead they had taken $n_p \approx 1$, they would have obtained the same result as Eq. (5).

Harter and Boyd [5] base their theory on the parametric amplification of the Rabi sidebands, which correspond to the dressed states that we have considered. Based on their observation that the pump field and the $\hat{E}_4$ field are essentially trapped in the self-focused filaments, simple Snell's law refraction of the red-shifted field $\hat{E}_s$ at the boundary of the trapped filaments gives

$$\theta_c = (\theta_0^2 + 2\delta n_s)^{1/2} \tag{9}$$

with θ_0 the internal angle of propagation within the trapped filaments due to self-focusing or diffraction. It is assumed that inside the filaments, the index of refraction is 1 because of saturation. Again for almost paraxial propagation down the filaments, i.e., putting $\theta_0 = 0$, Eq. (9) reduces to Eq. (5).

A pure 4WM of the Rabi sidebands together with both conservation of energy and all components of momentum leads to

$$\theta_c \approx (\delta n_s + \delta n_4 - 2\delta n_p)^{1/2} . \tag{10}$$

We have assumed that the pump field is propagating along the axis, and refraction due to self-focusing has been neglected. With $\delta n_4 = 0$ and $\delta n_p = 0$, Eq. (10) gives a value a factor of $\sqrt{2}$ smaller than Eq. (5). Similar formulas were obtained for the two photon pumped conical emission [11].

Golub *et al.* [3] obtain a cone angle

$$\theta_c = v_{ph}(\omega_s)/v_{gr}(\omega_p) \tag{11}$$

according to the Cherenkov radiation condition, where $v_{ph}(\omega_s)$ and $v_{gr}(\omega_p)$ are the phase velocity of the possible generated fields and the group velocity of the pump field, respectively. Then

$$\theta_c \approx [2(\delta n_s - \delta n_p)]^{1/2} . \tag{12}$$

This is consistent with our picture in which $v_{gr}(\omega_p) \sim c$ corresponds to the velocity of the pump which gives rise, via $\hat{\beta}_s$, to the source of the radiation. Essentially the same formula was obtained by Plekhanov *et al.* [7] by studying the propagation of the field, attributing it to a Cherenkov-type radiation and requiring spatial phase matching with respect to the longitudinal coordinates. They found

$$\theta_c = [2(\delta n_s + \delta n_4 - 2\delta n_p)]^{1/2} . \qquad (13)$$

Again under the same approximations, it reduces to Eq. (5).

Finally, LeBerre-Rousseau, Ressayre, and Tallet [8] obtained the same formula as Eq. (5) by a transient propagation study with effectively a long-correlation-length polarization.

Thus, although there are many different interpretations of the cone, it appears that most give the same cone angle under the approximation $n_p \approx 1$ and $n_4 \approx 1$. Only the phase matched 4WM is different, by a factor of $\sqrt{2}$.

In spite of these apparent consistencies, we agree with Valley *et al.* [2] that a completely unified theory should carefully take into account the 4WM and propagation effects, i.e., by detailed numerical simulation of Eq. (1). In this paper we have stressed, however, that the correct noise sources in these equations have a long correlation length rather than being δ correlated. Otherwise the correct cone features cannot be obtained. We emphasize that in any particular physical situation all effects (i.e., propagation, 4WM, and long-correlation-length Cherenkov-type sources) should be considered, although it is possible that according to circumstances one or the other might dominate. For example, two-photon-pumped conical emission appears to be well predicted by 4WM, whereas 4WM will be absent from cones in stimulated Raman scattering.

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