Parametric brightening of the medium in resonant four-wave mixing

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(Submitted 23 December 1985)
Pis'ma Zh. Eksp. Teor. Fiz. **43**, No. 3, 115–118 (10 February 1986)

A parametric brightening has been observed in a two-photon-absorption medium. A suppression of the two-photon absorption by more than two orders of magnitude has been achieved. There is a good quantitative agreement between the theoretical and experimental results.

1. In this letter we report experiments which have yielded a direct observation of a parametric brightening of a medium with a two-photon absorption. To achieve the parametric brightening, we used resonant four-wave mixing, which makes it possible to not only study the dynamics of the onset of the parametric brightening but also to monitor for the absence of any significant dynamic Stark effect or saturation of the two-photon transition. The experimental results found agree within the measurement errors with theoretical predictions.

2. The parametric brightening is seen as the suppression of parametric processes, the two-photon absorption of the interacting waves, etc., as waves propagate through a medium with a two-photon (or, more generally, multiphoton) absorption. The possibility of this effect was first pointed out in Ref. 1. The physical interpretation of parametric brightening is that the two-photon excitation of the *a-b* transition by waves E_1 and E_3 is suppressed by a combinational interaction of waves E_3 and E_4, which occurs out of phase with the effect of the first two waves (Fig. 1). The parametric brightening occurs when the amplitudes and phases of the different waves satisfy certain definite relations[2]:

$$r_1E_1E_3 + r_2E_2^*E_4 = 0; \quad k_4 = k_1 + k_2 + k_3, \tag{1}$$

where r_1 and r_2 are the composite matrix elements of the *a-b* transition.

The basic experimental difficulty in attempts to observe parametric brightening is in preparing waves E_1–E_4, because it is not possible to use four beams from independent laser sources, since the phase relations in (1) must be satisfied. So far, parametric brightening has been observed experimentally in a study of multiphoton ionization.[3,4] Exactly at a three-wave resonance, parametric brightening was detected from a relative decrease in the signal corresponding to multiphoton ionization. We believe that the importance of parametric brightening as a nonlinear interference effect warrants further experimental studies under conditions such that the changes in the populations, the field-induced broadening, and the field-induced shifts of the levels are all insignificant, so that a quantitative comparison can be made between theory and experiment.

3. The method chosen by us for preparing the four waves is as follows. We assume that three waves, with frequencies ω_1, ω_2, and ω_3, are incident on the two-photon-

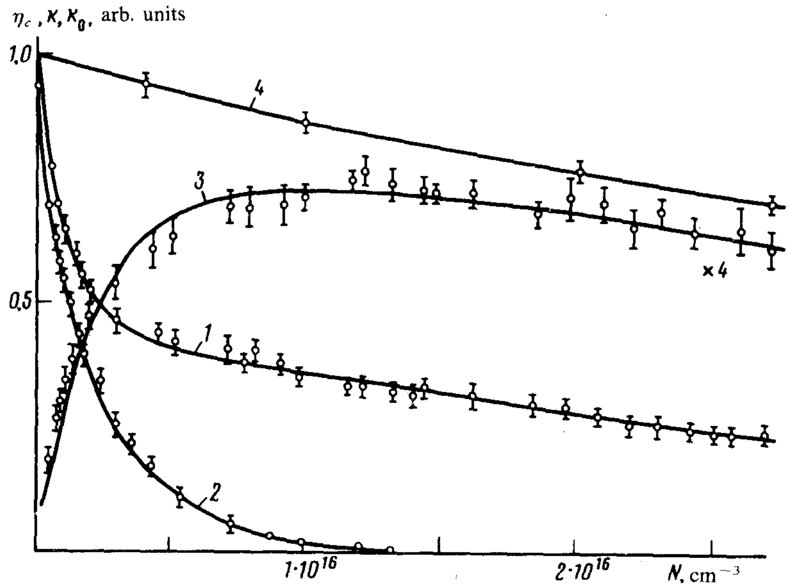

FIG. 1. Level scheme and interacting waves.

absorption medium. A fourth wave, with a frequency $\omega_4 = \omega_1 + \omega_2 + \omega_3$ is excited in the medium itself as a result of a parametric conversion under conditions of a collinear phase matching. Signal wave E_3 is much less intense than the two other waves, which are pump waves. In this case, the relative number of excited atoms is small, and the changes in the fields E_1 and E_2 in the nonlinear medium are insignificant. An upper limit is set on the pump intensities by the dynamic Stark shifts which these waves cause in the energy levels of the medium. If the conditions listed above are arranged, the amplitude and phase of wave E_4 automatically take on values such that waves E_1–E_4 begin to satisfy relation (1) over a rather large interaction distance,[5] while their further propagation no longer leads to the excitation of the *a-b* transition. In other words, parametric brightening is achieved.

As the two-photon-absorption medium, we selected atomic sodium vapor. This choice is based on the large nonlinear susceptibilities of this vapor and the fact that it is a simple matter to change the density of the vapor or, equivalently, the interaction length. The two-photon excitation of the 3*s*–4*s* transition of the sodium atoms (*a-b* in Fig. 1) is carried out by the first pump wave, a single-frequency Nd:YAG laser with a wavelength $\lambda_1 = 1.064$ μm and an organic dye laser with $\lambda_3 = 0.612$ μm and with an output line width $\Delta\nu_3 \simeq 0.05$ cm^{-1}. The source of wave E_2 is a parametric light source with $\lambda_2 = 2.2$ μm and $\Delta\nu_2 \simeq 0.5$ cm^{-1}, whose output frequency is tuned near the frequency of the 4*s*–4*p* transition in order to achieve phase matching. In an effort to suppress the effect of the transverse distribution of the intensities of the pump waves, the crossover diameter of the signal wave is chosen one-third the diameters of the crossovers of the two pump waves. Preliminary experiments on the technique of counterpropagating beams showed that at the intensities used the dynamic Stark effect has essentially no influence on the two-photon absorption.

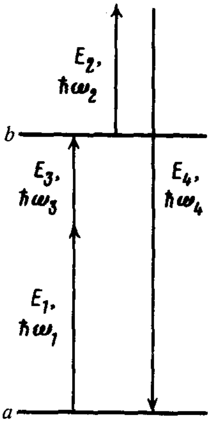

FIG. 2. Experimental and theoretical results on the transmission coefficient for the signal wave (1,2,4) and the conversion efficiency (3) versus the sodium vapor density. 1,3—$I_1 = 7$ MW/cm, $I_2 = 50$ KW/cm; 2—$I_1 = 7$ MW/cm, $I_2 = 0$; 4—$I_1 = I_2 = 0$. The effective length of the vapor is $l = 7$ cm.

4. To compare the experimental results with theory, we calculated the quantum efficiency of the conversion, η_c (the ratio of the number of photons in the excited wave to the initial number of photons in the signal wave), and the transmission coefficient (k) for the signal wave (the ratio of the energies of the signal wave before and after the two-photon-absorption medium). We also calculated the transmission coefficient (k_0) for the two-photon absorption of the signal wave in the field of the first pump wave. In this calculation, we used a linear absorption of the excited and signal waves, and we allowed for Doppler broadening of the levels and the temporal structure of the waves. We assumed that the interaction with the medium was a steady-state interaction. This is a legitimate assumption when the spectral line widths of waves E_1 and E_3 are smaller than the homogeneous broadening (Γ) of the 3s–4s transition. The maximum spectral width of the E_3 line may exceed Γ, since in our model this width is determined by the width of the phase matching.

Figure 2 shows experimental and theoretical results on the conversion efficiency η_c and the transmission coefficients k and k_0 versus the sodium vapor density. The suppression of the two-photon absorption of the signal wave by the combinational interaction of the emitted wave with the second pump wave is significant even at relatively low densities ($N \simeq 10^{15}$ cm^{-3}; curves 1 and 2). Despite the weakening of the signal wave due to the effective pumping of its energy into the excited light, the transmission coefficient k exceeds k_0, and their ratio continues to increase with increasing density. In these experiments, a suppression by more than two orders of magnitude of the two-photon absorption was achieved.

The fact that the quantum efficiency η_c reaches saturation (curve 3) at $N \simeq 10^{16}$ cm^{-3} and the slight change in k indicate that there is no interaction among all four waves over a distance exceeding $l_{\rm cr} = lN/N_{\rm cr}$ (in our case, $N_{\rm cr} = 10^{16}$ cm^{-3}). Consequently, at $lN > (lN)_{\rm cr}$ there is a situation of parametric brightening of the two-photon-absorption medium. The slight changes in η_c and k are attributed to a linear absorption of the signal wave (curve 4) and the excited wave.

The measured conversion coefficient η_c ($N = 10^{16}$ cm^{-3}) is 14% ± 5%, in comparison with the calculated value ~18%. The deviation from the limiting possible value of 25% (Ref. 6) is attributed to linear absorption and the imperfect ratio of pump intensities. The same conclusion follows from the theoretical and experimental results on the conversion efficiency and the transmission coefficient as functions of the pump intensities. These results will be discussed in more detail in a separate paper.

5. In summary, it can be asserted on the basis of these experimental results that we have been able to observe a parametric brightening of a medium due to an interference of excitations of an atomic transition under conditions of a two-photon resonance. An interference of this sort can substantially change the characteristics (both energy and spectral characteristics) of nonlinear resonant processes, e.g., a parametric frequency mixing or a selective multiphoton excitation. In addition, there is obviously interest in studying the joint effects on these processes of the parametric brightening, the motion of populations, and the field-induced shift and broadening of levels.

We are indebted to S. A. Akhmanov for a useful discussion of this study.

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Translated by Dave Parsons