Parametric bleaching of a two-photon absorbing medium

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A report is given of the observation of parametric bleaching of a two-photon absorbing medium by interference of two coherent channels of the excitation of a two-photon transition. Parametric bleaching caused a reduction in two-photon absorption of probe radiation by more than two orders of magnitude. A study was made of the kinetics of parametric bleaching as a result of a resonant four-wave interaction. An investigation was made of the influence of destructive interference on the spectral and energy characteristics of parametric conversion in the case of a two-photon resonance which is of practical interest. The theoretical and experimental results are compared.

INTRODUCTION

Coherence of parametric frequency mixing has the effect that the contributions made by various molecules and atoms in a medium to the intensity of the signal converted by mixing are not summed but interfere. This is manifested by the fact that the macroscopic nonlinear susceptibility of the medium is a sum of the nonlinear susceptibilities of each kind of molecule and atom. Contributions due to different energy levels of each particle in a medium interfere in a similar manner. The interference effects due to a cubic nonlinear susceptibility had been used very successfully for a long time in various spectroscopic methods, particularly in resonance active Raman spectroscopy, known usually as coherent anti-Stokes scattering (CARS).[1] Another class of interference effects observed on interaction of coherent radiation with matter is related to the interference of various excitation channels of energy levels in a medium. The most striking manifestation of such interference is parametric bleaching of a medium.

The parametric bleaching effect is manifested as follows: when waves propagate in a two-photon or, more generally, a multiphoton absorbing medium, it is found that parametric processes, two-photon absorption of interacting waves, etc. are suppressed. The possibility of this effect was first pointed out in Ref. 2. It is interpreted physically by assuming that two-photon excitation of a transition 1–2 (Fig. 1) under the action of two waves E_1 and E_3 is suppressed by an antiphase interaction with another pair of waves E_2 and E_4, i.e., parametric bleaching is due to interference of two coherent excitation channels of a two-photon transition. In a phenomenological description of this process in terms of nonlinear susceptibilities the parametric bleaching effect represents interference quenching of macroscopic nonlinear polarizations of a medium at frequencies of the interacting waves.[3–5] Generation of four such waves $E_1 - E_4$ in the optical range is possible by parametric processes in which the phase relationships between the interacting radiations are conserved.

The parametric bleaching effect alters radically the characteristics of nonlinear optical phenomena. Up to now the interference of two coherent excitation channels of an energy level had been observed experimentally (see Refs. 4–8 and the bibliography given there) in studies of resonant multiphoton ionization. A relative reduction in the multiphoton ionization signal was observed at the exact three-photon resonance. This was interpreted as the result of destructive interference between two coherent channels of excitation of an intermediate level: three-photon excitation by the pump radiation and one-photon excitation by the third harmonic generated in the investigated medium. The proof of the proposed interpretation is the absence of a resonant multiphoton ionization signal from traveling pump waves and its appearance in standing waves.

Our aim was to observe the parametric bleaching effect in the case of a two-photon resonance which is of practical interest.[9,10] We carried out experimental investigations under conditions such that a change in the populations, field broadening, and shifts of the levels were slight and, consequently, a quantitative comparison of the theoretical and experimental results was possible. The parametric bleaching effect was realized using a resonant four-wave interaction which made it possible not only to study the kinetics of the effect, but also to check the absence of a significant dynamic Stark effect and saturation of a two-photon transition, which might mask the bleaching. Moreover an investigation was made of the energy and spectral characteristics of resonant mixing of frequencies under conditions of parametric bleaching of a medium.

THEORY

It is known[2,3] that parametric bleaching of a medium (Fig. 1) occurs only for certain specific relationships between the amplitudes and phases of the interacting waves $E_1 - E_4$. These relationships can be deduced from the equations for an off-diagonal element of the density matrix[3]

$$r_1E_1E_3+r_2E_2^*E_4=0, \tag{1}$$

where r_1 and r_2 are the component matrix elements of the transition 1–2 and the electromagnetic field in the medium can be represented by

$$E=\sum_{i=1}^{4}E_i(r,t)\exp\{-i(\omega_it-k_iz)\}+\text{c.c.}$$

Suppression of two-photon excitation throughout the length of a nonlinear medium requires matching of the phase velocities of the interacting waves: $k_4=k_1+k_2+k_3$.

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Generation in the optical range of four waves satisfying the phase and amplitude relationships of Eq. (1) can be achieved most simply by parametric generation of one of the waves in a nonlinear medium. Let us assume that three waves of frequencies ω_1, ω_2, and ω_3 are incident on a two-photon absorbing medium. A fourth wave of frequency $\omega_4 = \omega_1 + \omega_2 + \omega_3$ is generated in the medium itself as a result of parametric conversion under collinear phase-matching conditions. The intensity of the signal wave E_3 is considerably less than the intensities of the other two (pump) waves E_1 and E_2. In this case we find that, firstly, the fraction of excited atoms is small and, secondly, the changes in the fields E_1 and E_2 in a nonlinear medium are slight, i.e., the conditions of a constant field are satisfied by the pump waves. The maximum intensities of the pump waves are limited by the dynamic Stark shifts of the energy levels of the medium which are induced by these waves and which give rise to frequency offsets from a two-photon resonance. The restrictions on the wave intensities are imposed because significant changes in the populations and fairly large Stark shifts of the levels reduce the excitation of a two-photon transition if it is not related to the parametric bleaching effect.

In the approximation of the interaction between plane monochromatic waves the equations of propagation appear as follows:

$$dE_3/dz+Ng_3E_3=g_0N\omega_3E_1^*E_2^*E_4,$$
$$dE_4/dz+Ng_4E_4=g_0N\omega_4E_1E_2E_3, \qquad (2)$$

where N is the concentration of the particles,

$$g_0=i\frac{\pi}{2c}\chi^{(3)}(\omega_1,\omega_3,\omega_2),$$
$$g_3=i\frac{\pi}{2c}\omega_3\chi^{(3)}(\omega_1,\omega_3,-\omega_1)|E_1|^2,$$
$$g_4=i\frac{\pi}{2c}\omega_4\chi^{(3)}(\omega_4,-\omega_2,\omega_2)|E_2|^2, \qquad (3)$$

and $\chi^{(3)}$ is a nonlinear third-order susceptibility per one atom.

We shall analyze the behavior of the other characteristics of the interaction as a function of the concentration of the particles and of the intensities of the pump waves. Firstly, we shall be interested in the quantum efficiency η of the conversion process, which is defined as the number of photons in the output wave E_4 to the initial number of photons in the signal wave E_3:

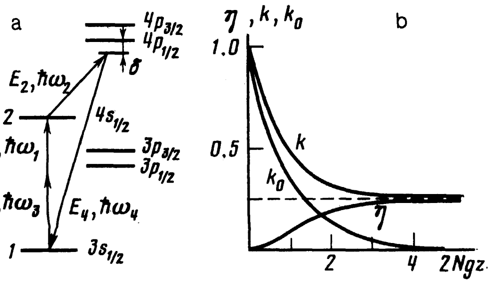

FIG. 1. (a) Schematic representation of the energy levels and of the interacting waves. (b) Dependences of the conversion efficiency η and of the transmission coefficient of the signal waves k and k_0 on the length z of the nonlinear medium in the case when $g_3 = g_4$.

$$\eta=[g_3g_4/(g_3+g_4)^2]\{1-\exp[-(g_3+g_4)Nl]\}^2, \qquad (4a)$$

where l is the length of the nonlinear medium. Secondly, we shall consider the transmission coefficient k of the signal radiation, which is equal to the ratio of the energy of the wave E_3 before and after passage through the absorbing medium:

$$k=\{[g_3\exp[-(g_3+g_4)Nl]+g_4]/(g_3+g_4)\}^2. \qquad (4b)$$

The expressions (4a) and (4b) are derived allowing for the relationship $g_3g_4 = g_0^2\omega_3\omega_4|E_1E_2|^2$, which follows from the explicit form of $\chi^{(3)}$ if we ignore the nonresonant part of the nonlinear susceptibility. We shall calculate the transmission coefficient of the signal wave under these assumptions and postulate the absence of the second pump wave, i.e., we shall find the coefficient k for $I_2 = 0$:

$$k_0=\exp(-2g_3Nl). \qquad (4c)$$

The coefficient k_0 represents two-photon absorption of the signal wave in the radiation field of the first pump wave.

We can readily show that if $N(g_3 + g_4)l \gg 1$, the waves $E_1 - E_4$ begin to satisfy the condition (1) and their subsequent propagation no longer excites the transition 1–2, i.e., we now have parametric bleaching. The conversion efficiency $\eta = g_3g_4/(g_3 + g_4)^2$ and the transmission coefficient of the signal wave $k = g_4^2/(g_3 + g_4)^2$ remain constant on increase in $N(g_3 + g_4)l$ and the two-photon absorption coefficient k_0 falls exponentially (Fig. 1b). The quantities η, k, and k_0 can be measured quite simply and the fact that the $\eta(Nl)$ and $k(Nl)$ dependences reach constant values is a demonstration of the parametric bleaching effect. Suppression of two-photon absorption is supported also by the fact that the transmission coefficient k is higher than k_0 for any value of $N(g_3 + g_4)l$.

A characteristic feature of the investigated interaction is the dependence of η and k on the intensities of the pump waves. The conversion efficiency reaches its maximum value of 25% when $g_3 = g_4$. Further increase in the pump intensities is compensated, in accordance with Eq. (1), by a reduction in the wave intensity E_4 and by an increase in the wave intensity E_3.

In contrast to the usual experimental conditions, it is not assumed in Eq. (2) that the absorption is linear and that the interacting waves have a specific time (pulsed) structure. An allowance for these aspects modifies the expressions of Eq. (4) as follows:

$$\eta=\frac{1}{\tau_3\pi^{1/2}}$$
$$\times\int dt\, g_3g_4\left\{\frac{\exp[(\gamma_1-\alpha-g_4)Nl]-\exp[(\gamma_2-\alpha-g_4)Nl]}{\gamma_1-\gamma_2}\right\}^2$$
$$\times\exp\left(-\frac{t^2}{\tau_3^2}\right),$$
$$k=\frac{1}{\tau_3\pi^{1/2}}$$
$$\times\int dt\left\{\frac{\gamma_1\exp[-(\gamma_2+g_3+\beta)Nl]-\gamma_2\exp[-(\gamma_1+g_3+\beta)Nl]}{\gamma_1-\gamma_2}\right\}^2$$
$$\times\exp\left(-\frac{t^2}{\tau_3^2}\right),$$
$$k_0=\frac{1}{\tau_3\pi^{1/2}}\int dt\exp(-2g_3l)\exp(-t^2/\tau_3^2),$$
$$2\gamma_{1,2}=\alpha+g_4-g_3-\beta\pm[(\alpha+g_4-g_3-\beta)^2+4g_3g_4]^{1/2}, \qquad (5)$$

where α and β are the linear absorption coefficients of the output and signal waves, respectively; τ_i is the duration of a pulse of the wave E_i; the nonlinear susceptibility $\chi^{(3)}$ is integrated over the Maxwellian distribution of the atomic velocities

$$\chi^{(3)} = \left(\frac{m}{2k_B T}\right)^{1/2} \int \chi^{(3)}_{\text{one}} \left(\omega + \omega \frac{v}{c}\right) \exp\left(-\frac{mv^2}{2k_B T}\right) dv, \tag{6}$$

where m is the mass of an atom and k_B is the Boltzmann constant.

In allowing for the time structure of the interacting waves we shall assume that the interaction with the medium is in a steady state. This is justified in the case when the coherence times of the waves E_1 and E_3 exceed the transverse relaxation time $1/\Gamma$ of the two-level system under discussion. In other words, we shall assume that the spectral widths of the radiation lines E_1 and E_3 are less than the homogeneous broadening of a two-photon transition. It should be pointed out that the spectral width of the E_2 line may exceed Γ, since its maximum value is governed (in our model) by the width of the phase-matching curve.

Naturally, the dependences of the conversion efficiency and of the transmission coefficients of the signal radiation on the concentration of the particles and on the pump intensities, calculated in accordance with Eq. (5), differ quantitatively from those given in Fig. 1(b), but the qualitative behavior remains as before.

APPARATUS AND MEASUREMENT METHOD

Atomic sodium vapor was used as the two-photon absorbing medium. This medium was selected because of the high values of the nonlinear susceptibilities and the ease of variation of the concentration, i.e., of the parameter Nl. Two-photon excitation of the $3s$–$4s$ transition in the sodium atom (Fig. 1a) was provided by the first pump wave from an yttrium aluminum garnet (YAG) laser and an organic dye laser (Fig. 2). The YAG laser was used as a reference source and it emitted a single longitudinal mode of the wavelength $\lambda_1 = 1.064\,\mu$ with a spectral width $\Delta\nu_1 \approx 0.01$ cm^{-1}. This width was reduced by using a pile of plates as the exit mirror and placing a low-Q Fabry-Perot étalon inside the resonator. Moreover, passive Q switching was performed by an LiF crystal with F_2^- color centers. The garnet laser emitted pulses of ~ 12 mJ energy and ~ 25 nsec duration, and with an instability less than 3%.

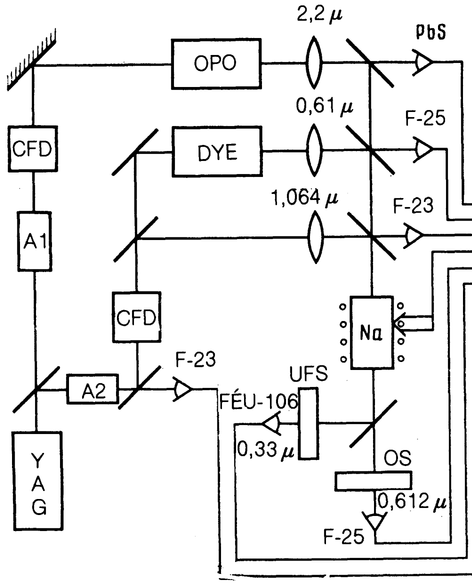

FIG. 2. Schematic diagram of the apparatus: YAG is an yttrium aluminum garnet laser; A 1 and A 2 are amplifiers; CFD are crystal frequency doublers; OPO is an optical parametric oscillator; DYE is an organic dye laser; Na is a cell with sodium vapor; UFS and OS are optical filters; F-23, F-25, FÉU-106, and PbS are photodetectors; AMCS is an automated measuring computing system.

The dye laser was excited by the second harmonic of the reference (garnet) laser and it emitted at $\lambda_3 = 0.612\,\mu$. The dispersive components in the dye laser were two diffraction gratings. The first (1200 lines/mm) operated in the near-grazing regime, whereas the second (1800 lines/mm) was operated in the autocollimation regime. Continuous frequency tuning was provided by rotation of the second grating. The width of the dye laser line was less than 0.05 cm^1 and the long-term frequency instability of this line was within 0.1 cm^{-1}.

The second pump wave was generated by an optical parametric oscillator (OPO) at $\lambda_2 = 2.2\,\mu$ and its output frequency was tunable near the frequency of the $4s$–$4p$ transition when the phase matching conditions were satisfied. The OPO was based on a lithium niobate crystal 5-cm long. The emission wavelength was tuned by altering the temperature of this crystal. The spectral width of the emission line of the OPO was measured by two methods: the method of nonlinear infrared spectroscopy (using the OPO idler wave of $2.2\,\mu$ wavelength) and with a scanning Fabry-Perot interferometer (OPO signal wave at the wavelength of $0.7\,\mu$). Both methods gave approximately the same value of $\Delta\nu_2 \approx 0.5$ cm^{-1}. The peak power of infrared radiation obtained from the OPO reached 2 kW.

The maximum intensities of the pump waves were 80 MW/cm^2 (YAG) and 150 kW/cm^2 (OPO). A weak signal wave was obtained by attenuating the dye laser radiation with optical filters. The absence of significant changes in the populations of the levels involved in the investigated two-photon transition was deduced from the spectral and concentration dependences of the two-photon absorption coefficient. The influence of the transverse distributions of the intensities in the pump waves was avoided by selecting the constriction diameter of the signal wave to be one-third of the diameters of the constrictions of both pump waves. The energies of the interacting waves before and after the nonlinear medium were determined using F-23 and F-25 photocells and a PbS photoresistor. The monitoring of the parameters, the acquisition of the data, and the on-line analysis of these data were performed by a measuring system based on an Élektronika DZ-28 microcomputer.

The tuning of the emission frequencies of the dye laser and of the OPO, needed to ensure the exact two-photon resonance and phase matching, was carried out as follows. When the concentration of the atomic vapor was $N \approx 3 \times 10^{15}$ cm^{-3} and the second pump wave was missing, we varied the dye laser frequency until the two-photon absorption was maximized ($k_0 \approx 0.3$). Then, the concentration was increased to $N \approx 3 \times 10^{16}$ cm^{-3} and the intensities of the interacting waves were attenuated by between one and a half and two orders of magnitude; the OPO frequency was then tuned to ensure the highest conversion efficiency.

The correctness of the interpretation of the results was checked by demonstrating that the two-photon absorption coefficient was not affected by the dynamic Stark effect due to the second pump wave, since this could also result in suppression of two-photon absorption. This could be done quite simply using the technique of opposite beams, i.e., by eliminating the parametric processes. Figure 3 shows the dependence of the transmission coefficient of the signal radiation on the offset δ of the OPO frequency from the one-photon resonance with the $4s_{1/2}$–$4p_{1/2}$ transition. The measurements demonstrated that for the intensities employed in our study, when the offset was $\delta \approx 7\ \mathrm{cm}^{-1}$, the dynamic Stark effect had practically no influence on the two-photon absorption coefficient. Nevertheless, when the OPO frequency was tuned to the frequencies of the $4s_{1/2}$–$4p_{1/2,\,3/2}$ transitions, a strong reduction in the two-photon absorption coefficient began to be manifested and this was due to a strong perturbation of the energy spectrum of the sodium atom. The transmission coefficient minimum lying within the doublet was related to the exact compensation of the linear susceptibility of the $4s_{1/2}$ level.

Special attention was given to the determination of the absolute value of the quantum efficiency of the conversion process, which was essential in a quantitative comparison of the experimental and theoretical results. Moreover, in some problems of practical interest the parametric bleaching effect was known to determine the maximum conversion efficiency.[11-13] All this required a careful determination of the relative spectral sensitivity of the recording system at the wavelengths of 0.612 and 0.33 μ. The calibration was carried out by two methods. The first was based on the nominal spectral characteristic of the photodetector supplied by the manufacturer and the second involved direct measurement of the quantum efficiency in the process of frequency conversion from the range 0.612 μ to 0.33 μ. Two-photon absorption of the signal wave was avoided by detuning the dye frequency from the exact two-photon resonance by 2 cm^{-1} and measuring the energy of the output wave and the change in the energy of the signal wave. The measuring system could be calibrated to within 30%.

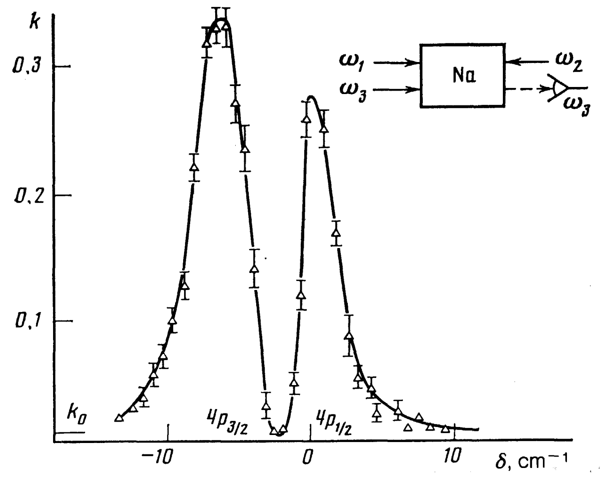

FIG. 3. Dependences of the transmission coefficient of the signal wave on the frequency of the second pump wave. The intensities of the pump waves were $I_1 = 12\ \mathrm{MW/cm}^2$ and $I_2 = 100\ \mathrm{kW/cm}^2$. The sodium vapor concentration was $7\times10^{15}\ \mathrm{cm}^{-3}$.

RESULTS AND DISCUSSION

The experimental and theoretical dependences of the conversion efficiency η, and of the transmission coefficients k and k_0 on the sodium vapor concentration are presented in Fig. 4. Suppression of two-photon absorption of the signal wave by the combination interaction of the output radiation with the second pump became observable even at relatively low ($N \approx 10^{15}\ \mathrm{cm}^{-3}$) concentrations. In spite of the weakening of the signal wave due to the effective transfer of its energy to the output radiation, the transmission coefficient k was greater than the coefficient k_0 and their ratio increased on increase in the sodium concentration. Suppression of two-photon absorption by the strong wave was by more than two orders of magnitude.

The dependence of the quantum efficiency η reached saturation at $N \approx 10^{16}\ \mathrm{cm}^{-3}$ and the transmission coefficient of the signal wave k then varied slowly (Fig. 4) indicating the absence of interaction between all four waves in a distance exceeding $l_{\mathrm{cr}} = lN/N_{\mathrm{cr}}$ (in our case, we found that $N_{cr} = 10^{16}\ \mathrm{cm}^{-3}$). Consequently, when the condition $lN > (lN)_{\mathrm{cr}}$ was satisfied, we found that two-photon absorption was reduced by parametric bleaching. The slight changes in the values of η and k were due to linear absorption of the signal (dashed curve in Fig. 4) and output radiations.

The measured conversion efficiency η ($N = 10^{16}\ \mathrm{cm}^{-3}$) was $14 \pm 5\%$, whereas the calculated value was 18%. The deviation from the maximum theoretical value of

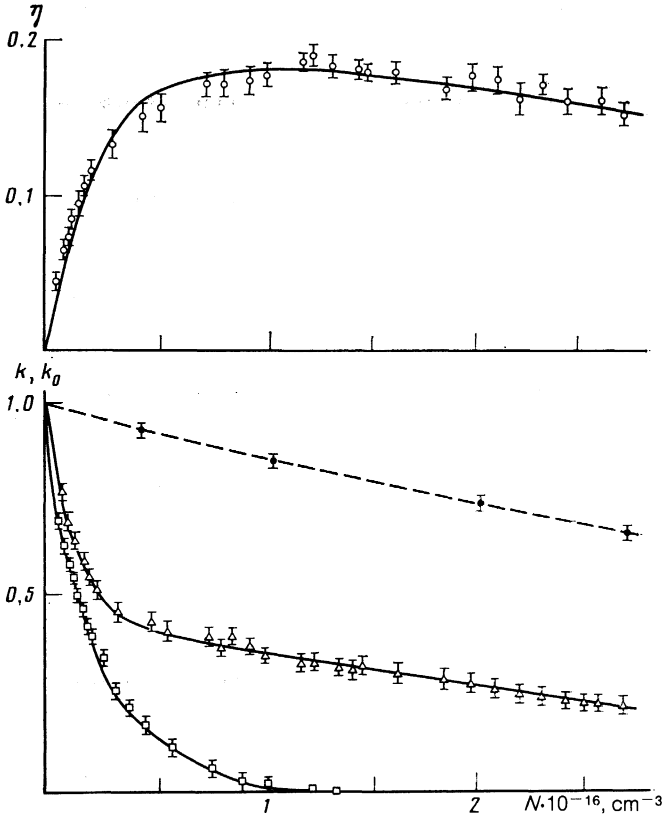

FIG. 4. Kinetics of the establishment of the parametric bleaching effect in a medium. The intensities of the pump waves were: $I_1 = 7\ \mathrm{MW/cm}^2$ and $I_2 = 50\ \mathrm{kW/cm}^2$ (the values of η are represented by circles and the values of k are represented by triangles); $I_1 = 7\ \mathrm{MW/cm}^2$, $I_2 = 0$ (the values of k_0 are represented by squares); $I_1 = I_2 = 0$ (the values of k_0 are represented by black dots). The effective length of the vapor cell was $l = 7$ cm.

25% was due to linear absorption and not quite optimal ratio of the pump wave intensities. For the optimal ratio of the intensities in question ($g_3 = g_4$) and in the presence of parametric bleaching the value of k should be approximately equal to 25%, as given by Eq. (5). In our case (Fig. 4), we found that $k \approx 35\%$, which corresponded to $g_4 > g_3$. The same conclusion was drawn from the calculated and experimental dependences of the conversion efficiency and of the transmission coefficient on the intensities of the pump waves.

The dependences of η and k on the intensities of the pump waves given in Fig. 5 also demonstrated that the parametric bleaching effect was observed. In agreement with the theory [Eq. (4a)], the dependences of the conversion efficiency on the intensities of the first or second pump waves were the same. The value of η first increased on increase in $I_{1,2}$ and then it reached its maximum value at intensities corresponding to $g_3 \approx g_4$. A further increase in I_1 or I_2 reduced the conversion efficiency η, as predicted theoretically.

On the other hand, the transmission coefficient k exhibited an asymmetric dependence on the pump wave intensities. The dependence could be studied qualitatively on the basis of Eq. (1). Assuming that the output radiation intensity was approximately constant, we found that an increase in I_2 increased the signal transmission coefficient (Figs. 5a and 5c), but an increase in I_1 reduced this transmission coefficient (Figs. 5b and 5d). The more detailed behavior of these dependences fitted Eq. (5), as demonstrated by the continuous curves in Fig. 5. The experimental points fitted well these theoretical curves.

The parametric bleaching effect was observed for quite specific ratios of the amplitudes of the interacting waves. Hence, we could carry out relative measurements of the component elements of the matrix of the investigated two-photon transition and to study the dispersion by varying the wavelengths of the interacting radiations. The absolute values of the component matrix elements could be found from the two-photon absorption data. Such spectroscopic information was carried by the dependence of the conversion efficiency on the intensity of one of the pump waves (Fig. 5). In this case there was no need to carry out absolute measurements of the intensities of the signal and output waves.

As pointed out already, in the case of the optimal ratio of the intensities of the pump wave we should have $k = 25\%$ and the value of η should pass through its maximum. This relationship between the conversion characteristics was also clearly demonstrated by the experimental dependences in Fig. 5. The slight deviation of k from 25% was due to linear absorption and it was described quantitatively by the relationships of Eq. (5). The influence of linear absorption limited the maximum attainable conversion efficiency $\eta^{\max}$: an increase in this absorption reduced $\eta^{\max}$ (we recall that $\eta^{\max} = 25\%$ should be obtained in the absence of linear absorption of waves). This was confirmed by the measured values $\eta^{\max} = 18$ and 14% obtained for sodium concentrations N of 3.5×10^{15} and 1.3×10^{16} cm^{-3} respectively.

Interference between two-photon excitations of a level affects not only the energy but also the spectral characteristics of nonlinear resonance processes. This is due to the fact that away from a two-photon resonance both two-photon absorption and inverse Raman scattering become weaker at a rate faster than the parametric coupling coefficient of the interacting radiations. The spectral characteristics of this process are described by Eq. (2) if we allow for the fact that the coefficients g_0, g_3, and g_4 are complex variables.

The dependences of the conversion efficiency and of the transmission coefficient of the signal radiation on the offset from the investigated two-photon resonance are plotted in Fig. 6. An increase in the sodium concentration increased the amplitude of the spectral curve representing the conversion efficiency, but the width of this curve remained constant (Fig. 6a). A further increase in the sodium concentration, i.e., in the parameter Nl, reduced the rate of rise of the conversion efficiency at the center because of the interference quenching of the excited two-photon transition but did not affect the wings. The curve representing the conversion effi-

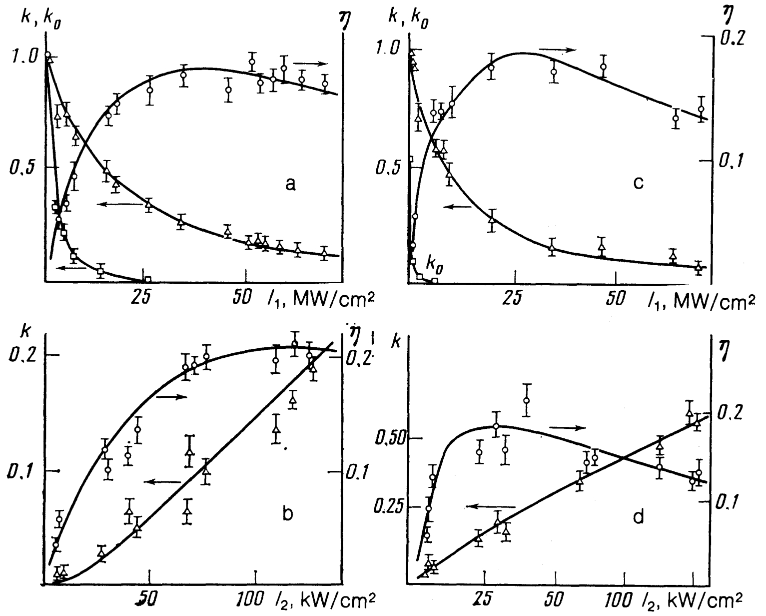

FIG. 5. Experimental and theoretical dependences of the conversion efficiency η (○) and of the signal wave transmission coefficients k_0 (□) and k (△) on the intensities of the pump waves for different sodium vapor concentrations: 3.5×10^{15} cm^{-3} (a, b) and 1.3×10^{16} cm^{-3} (c, d): (a) $I_2 = 130$ kW/cm^2; (b) $I_1 = 40$ MW/cm^2; (c) $I_2 = 100$ kW/cm^2; (d) $I_1 = 7$ MW/cm^2. The effective length of the vapor cell was $l = 10$ cm.

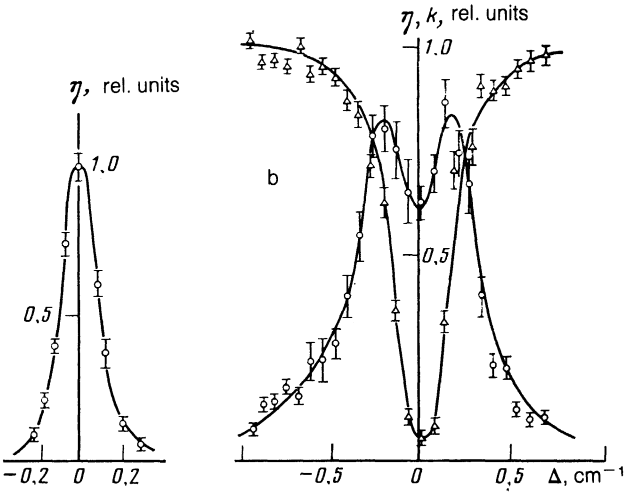

FIG. 6. Dependences of the conversion efficiency η (O) and of the signal radiation transmission coefficient k (Δ) on the detuning from a two-photon resonance. The intensities of the pump waves were $I_1 = 40$ MW/cm^2 and $I_2 = 50$ kW/cm^2. The concentration of the sodium vapor was $N = 10^{12}$ cm^{-3} (a) and 3.5×10^{15} cm^{-3} (b). The continuous curves are drawn by hand.

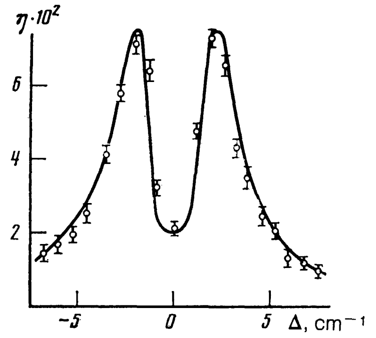

FIG. 7. Spectral dependence of the conversion efficiency Pump wave intensities $I_1 = 5$ MW/cm^2 and $I_2 = 60$ MW/cm^2. The sodium vapor concentration was $N = 4 \times 10^{16}$ cm^{-3}.

ciency became broader and the maximum value of the efficiency began to shift away from the exact two-photon resonance (Fig. 6b).

The dependences shown in Fig. 6 agreed qualitatively with those found by calculation, but we were unable to achieve a quantitative agreement within the adopted model of monochromatic waves. The differences were due to the influence of the spectral width of the line representing the second pump wave (~ 0.5 cm^{-1}), which was greater than the width of the two-photon transition. This influence could be eliminated by additional experimental investigations based on the following circumstance.

All the results obtained for the investigated sum process apply also in the case of conversion to the difference frequency $\omega_4 = \omega_1 + \omega_3 - \omega_2$ (Ref. 14). In an experimental investigation of the influence of the parametric bleaching effect on the spectral characteristics of the conversion process and the frequencies of the second pump wave ω_2 and of the signal wave ω_3 were selected to be the same for the following reasons. Firstly, in this case the phase-matching conditions were satisfied automatically and, secondly, the experimental dependences were found to be related solely to the two-photon resonance (because the offset from the intermediate resonance with the $3s$ level was sufficiently large). The first pump wave E_1 and signal wave E_3 had the same linear polarization, which was orthogonal to the polarization of the second pump wave E_2.

In these experiments the source of the first pump wave was an organic dye laser ($\lambda_1 = 0.7612\ \mu$). The signal and the second pump waves were provided by YAG laser radiation ($\lambda_{2,3} = 1.064\ \mu$). The sum frequency of these lasers was varied near the resonance with the $3s$–$4p$ transition in the sodium atom. The intensities of the pump waves were $I_1 = 5$ MW/cm^2 and $I_2 = 60$ MW/cm^2. The output radiation was selected by an optical filter and a polarizer.

A typical spectral dependence of the conversion efficiency is presented in Fig. 7. A reduction in the conversion efficiency of the exact two-photon resonance was observed because of destructive interference already for concentrations of $N = 10^{15}$ cm^{-3}. The width of the profile was in this case still slight (it was 0.4 cm^{-1} at midamplitude). When the sodium atom concentration was $N = 10^{16}$ cm^{-3}, the conversion efficiency obtained for detuning from the two-photon resonance was more than three times greater than η in the case when $\Delta = 0$. A further increase in the sodium atom concentration continued to increase the maximum efficiency and the width of the conversion efficiency curve (Fig. 7). Broadening of the spectral dependence of the conversion efficiency to 8 cm^{-1} was recorded. It is clear from Fig. 7 that theoretical and experimental results agreed quite well.

Our investigation indicated that detuning from the two-photon resonance could ensure high values of the conversion efficiency. The influence of two excitation channels of the two-photon transition, which in the case of the exact two-photon resonance resulted in parametric bleaching of the medium, broadened considerably the conversion efficiency curve. Such broadening could be several orders of magnitude greater than the spectral width of the resonantly excited transition.

CONCLUSIONS

We observed directly parametric bleaching of a two-photon absorbing medium associated with destructive interference between two coherent channels of the excitation of a two-photon transition. The observed suppression of two-photon absorption of the probe radiation by more than two orders of magnitude was due to parametric bleaching. This conclusion was drawn from the results of test experiments which excluded other possible mechanisms of the change in the two-photon absorption coefficient.

The recent interest in the various aspects of the parametric bleaching effect[5,11,13,15] is not only due to its nonlinear optical interference nature. Equally interesting is the influence of destructive interference on various characteristics of resonant nonlinear optical processes. Our experimental investigations demonstrate that the resultant destructive interference limits the degree of excitation of a multiphoton transition and thus the efficiency of resonant processes. Moreover, parametric bleaching broadens considerably and alters the profile of a resonant transition.

Direct observation of the parametric bleaching effect required that the experiments should be carried out under conditions manifesting this effect most clearly. Nevertheless, destructive interference accompanies to a greater or smaller degree all parametric resonances, because of the coherence of the parametric interaction. The multiplicity of the concomitant phenomena (shift of the populations, dynamic Stark effect, multiphoton ionization, etc.) make the influence of this interference not as obvious as in the case discussed above. However, this interference must be taken into account both in the development of methods for generation of coherent infrared and vacuum ultraviolet radiations, as well as in spectroscopic investigations by the methods of nonlinear optics.

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