Phase-conjugation in the three-level resonant medium*

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The nondegenerate phase-conjugation through four-wave mixing in potassium vapour has been theoretically and experimentally considered. The investigation of spectral dependence of the phase-conjugate reflectivity shows that in the spectrum of conjugate wave there appear ranges of amplification near the characteristic seven frequencies. Two of these ranges occur because of coherent interactions between *unpopulated levels* of doublet and are shifted relatively to the pump frequency on the value of doublet splitting.

1. Introduction

Recently, the phenomenon of optical phase-conjugation different nonlinear media has drawn a considerable interest (see, e.g., [1] and the references given there). Phase-conjugation is usually investigated through degenerate four-wave mixing. The results of such experiments in resonant gases are represented, for instance, in papers [2, 3]. However, the investigation of phase-conjugation through nondegenerate four-wave mixing is of particular interest. This process has been considered theoretically in papers [4, 5], where the nonlinear medium is modelled as a two-level atomic system. Since excited state of alkali metal atoms has doublet structure in theoretical investigation of light resonant interaction the third level should be taken into account.

In this paper the nondegenerate phase-conjugation in potassium vapour has been considered both theoretically and experimentally. The theory of nondegenerate phase-conjugation in the three-level resonant medium is developed. It is shown, that taking account of the excited level doublet structure changes qualitatively the spectral composition of a conjugated wave. There occur novel spectral ranges of effective phase-conjugation, which are considerably changed by pump frequency tuning.

The paper presents experimental results obtained from the observations of nondegenerate phase-conjugation in four-wave mixing in potassium vapour. Novel spectral ranges of the signal wave conjugation have been detected.

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2. Theory

Consider the interaction of four plane linearly polarized waves — two counter-propagating pump waves E_1 and E_2 and two counter-propagating signal waves E_3 and E_4 with three-level resonance medium (1, 2 are excited levels, 3 is ground level). The angle between pump and signal directions of propagation exceeds largely the parametric interaction phase-matching angle for each wave separately. Pump waves are monochromatic with frequency ω_1 and signal waves are quasimonochromatic with frequency spectrum ω_1. By solving the equation of motion for the density matrix ϱ with phenomenological relaxation terms in a steady-state approximation, the four-wave induced polarization has been calculated. In the third order approximation, after spatial averaging over the pump wave oscillation, we get the coupled set of two following equations for slowly varying Fourier's components of weak wave amplitudes E_3 and E_4

$$\frac{\partial E_3(\omega)}{\partial z} + A_1(\omega)E_3(\omega) = A_2(\omega)E_1E_2E_4^*(2\omega_1 - \omega) - \frac{\partial E_4^*(2\omega_1 - \omega)}{\partial z}$$

$$+ A_1^*(2\omega_1 - \omega)E_4^*(2\omega_1 - \omega) = A_2^*(2\omega_1 - \omega)E_1^*E_2^*E_3(\omega) \tag{1}$$

where

$$A_i(\omega) = i\left\{ \frac{\omega_1 - \omega}{c} + q\left[\sum_{i=1}^{5} \frac{a_i}{\omega - \omega_1 + \Omega_i} - \sum_{i=1}^{2} \frac{a_i'}{(\omega - \omega_1 + \Omega_i)^2} \right] \right\}, \tag{1a}$$

$$A_2(\omega) = iq \sum_{i=1}^{7} \frac{b_i}{\omega - \omega_1 + \Omega}. \tag{1b}$$

Here $a_1 = \xi\left(\frac{2}{\Omega_1 - \Omega_3} + \frac{\eta}{\Omega_1 - \Omega_5} \right)$, $a_2' = \eta\xi\left(\frac{2\eta}{\Omega_2 - \Omega_3} + \frac{1}{\Omega_2 - \Omega_4} \right)$, the values of Ω_i, a_i and b_i are presented in Table, where the following notations are used: $q = \frac{2\pi N \omega_1 |d_{13}|^2}{c\hbar}$, $\xi = \frac{|d_{13}|^2}{\hbar^2}(|E_1|^2 + |E_2|^2)$, γ_{ik} is half-width of transition line, $(\gamma_1)^{-1}$ — atomic excited state life-time, $\varepsilon_i = \omega_1 - \omega_{i3}$ is resonance detuning between pump wave frequency ω_1 and frequency ω_{i3} of the atomic transition $3 \to i$ ($i = 1, 2$), $\Delta = \omega_{13} - \omega_{23}$ is a value of doublet splitting, d_{ik} is dipole matrix element, N — atom density, $\eta = |d_{23}|^2/|d_{13}|^2$.

From the obtained set of Eqs. (1) it is seen that the Fourier's component of the wave E_3 at frequency ω is coupled with the Fourier's component of the wave E_4^* at frequency $2\omega_1 - \omega$ according to the energy conservation law. Coefficient $A_1(\omega)$ characterizes resonant medium susceptibility at frequency ω, $A_2(\omega)$ is parametric coupling between waves. From the formula (1a) it is seen that $A_1(\omega)$ has five resonance poles corresponding to the five processes of the waves interaction with the three-level atom: 1) absorption at the frequency ω_{13} (the pole $\omega = \omega_1 - \Omega_1$), 2) absorption at the frequency ω_{23} (the pole $\omega = \omega_1 - \Omega_2$),

i	Ω_i	a_i	b_i
1	$\varepsilon_1 + i\gamma_{13}$	$1 - \xi\left\{\frac{2}{\Omega_6}\left[\frac{-2\gamma_{13}}{\gamma_1\Omega_1} + \frac{i(2\gamma_{13}-\gamma_1)}{(\Omega_1-\Omega_3)^2}\right] + \frac{\eta}{\Omega_7}\left[\frac{\Delta - i\gamma_{13} - i\gamma_{23}}{\Omega_1\Omega_5} + \frac{i(\gamma_{13}+\gamma_{23}-\gamma_{12})}{(\Omega_1-\Omega_5)^2}\right]\right\}$	$-\frac{|d_{13}|^2}{\hbar^2\Omega_1}\left[\frac{\Omega_6}{(\Omega_1-\Omega_3)(\Omega_1-i\gamma_{13})} + \frac{\eta\Omega_7}{(\Omega_1-\Omega_5)(\Omega_1-\Omega_7)}\right]$
2	$\varepsilon_2 + i\gamma_{23}$	$\eta - \eta\xi\left\{\frac{2\eta}{\Omega_7}\left[\frac{-2\gamma_{23}}{\gamma_1\Omega_2} + \frac{i(2\gamma_{23}-\gamma_1)}{(\Omega_2-\Omega_3)^2}\right] + \frac{1}{\Omega_6}\left[-\frac{\Delta + i\gamma_{13}+i\gamma_{23}}{\Omega_2\Omega_4} + \frac{i(\gamma_{13}+\gamma_{23}-\gamma_{12})}{(\Omega_2-\Omega_4)^2}\right]\right\}$	$-\frac{|d_{23}|^2}{\hbar^2\Omega_2}\left[\frac{\eta\Omega_7}{(\Omega_2-\Omega_3)(\Omega_2-i\gamma_{23})} + \frac{\Omega_6}{(\Omega_2-\Omega_4)(\Omega_2-\Omega_6)}\right]$
3	$i\gamma_1$	$-2i\xi\left[\frac{\gamma_1-2\gamma_{13}}{\Omega_6(\Omega_1-\Omega_3)^2} + \frac{\eta^2(\gamma_1-2\gamma_{23})}{\Omega_7(\Omega_2-\Omega_3)^2}\right]$	$\frac{2i|d_{13}|^2}{\hbar^2}\left[\frac{\gamma_1-2\gamma_{13}}{\Omega_1(\Omega_3-\Omega_6)(\Omega_3-\Omega_1)} + \frac{\eta^2(\gamma_1-2\gamma_{23})}{\Omega_2(\Omega_3-\Omega_7)(\Omega_3-\Omega_2)}\right]$
4	$\Delta + i\gamma_{12}$	$i\eta\xi\,\frac{\gamma_{13}+\gamma_{23}-\gamma_{12}}{\Omega_6(\Omega_2-\Omega_4)^2}$	$\frac{i|d_{23}|^2}{\hbar^2}\,\frac{\gamma_{12}-\gamma_{13}-\gamma_{23}}{\Omega_2(\Omega_4-\Omega_6)(\Omega_4-\Omega_2)}$
5	$-\Delta + i\gamma_{12}$	$i\eta\xi\,\frac{\gamma_{13}+\gamma_{23}-\gamma_{12}}{\Omega_7(\Omega_1-\Omega_5)^2}$	$i\,\frac{|d_{23}|^2}{\hbar^2}\,\frac{\gamma_{12}-\gamma_{13}-\gamma_{23}}{\Omega_1(\Omega_5-\Omega_7)(\Omega_5-\Omega_1)}$
6	$-\varepsilon_1 + i\gamma_{13}$	—	$\frac{|d_{13}|^2}{\hbar^2}\left\{\frac{1}{(\Omega_1-i\gamma_{13})(\Omega_6-\Omega_3)} + \frac{\eta}{(\Omega_6-\Omega_2)[\Omega_2+i(\gamma_{12}-\gamma_{13}-\gamma_{23})]}\right\}$
7	$-\varepsilon_2 + i\gamma_{23}$	—	$\frac{|d_{23}|^2}{\hbar^2}\left\{\frac{\eta}{(\Omega_2-i\gamma_{23})(\Omega_7-\Omega_3)} + \frac{1}{(\Omega_7-\Omega_1)[\Omega_1+i(\gamma_{12}-\gamma_{13}-\gamma_{23})]}\right\}$

3) Rayleigh light scattering at the frequency ω_1 (the pole $\omega = \omega_1 - \omega_3$), 4) stimulated light scattering at the frequency $\omega_1 - \Delta$ (the pole $\omega = \omega_1 - \Omega_4$), and 5) stimulated scattering at the frequency $\omega_1 + \Delta$ (the pole $\omega = \omega_1 - \Omega_5$).

The fourth and fifth stimulated processes of light scattering coincide by means of frequency with the Stokes and anti-Stokes frequencies of stimulated electronic Raman scattering (SERS) (between doublet levels populated by the light excitation). These resonance processes, however, differ from SERS essentially, which does not occur in the first nonlinear approximation. The main difference of these processes is that they disappear in the absence of atomic collisions. In fact, the relaxation rates (for instance, γ_{13}) can be separated into a radiative part $(1/2\gamma_1)$ and a term proportional to the pressure (γ_{13}^{pr}), so that $a_{4,5} \sim (\gamma_{12} - \gamma_{13} - \gamma_{23}) = (\gamma_{12}^{pr} - \gamma_{13}^{pr} - \gamma_{23}^{pr})$. This equality shows that the fourth and fifth resonances disappear if only radiative processes are taken into account. For the first time those new resonances have been suggested by Bloembergen in four-wave mixing processes and investigated in [6–8].

Parametric coupling coefficient $A_2(\omega)$ contains seven resonance poles $\omega = \omega_1 - \Omega_i$ $(i = 1, ..., 7)$, where sixth and seventh poles are three-photon scattering frequencies in the process of which an atom is excited absorbing two photons and eliminating one signal photon [9].

Such a complicated dispersive dependence of resonant susceptibility $A_1(\omega)$ and parametric coupling coefficient $A_2(\omega)$ essentially affects phase-conjugated wave spectral pattern. By neglecting the pump intensity depletion and solving the set of Eqs. (1) with the boundary conditions $E_3(z = 0) = E_{30}$, $E_4(z = l) = 0$, we obtain phase-conjugate reflectivity $R = |E_4(z = 0)|^2 / |E_{30}|^2$,

$$R = \left| 2A_2^*(2\omega_1 - \omega) E_1^* E_2^* \frac{\sinh ml}{2m \cosh ml + [A_1(\omega) + A_1^*(2\omega_1 - \omega)] \sinh ml} \right|^2 \quad (2)$$

where

$$m = \{1/4 [A_1(\omega) + A_1^*(2\omega_1 - \omega)]^2 - A_2(\omega) A_2^*(2\omega_1 - \omega) |E_1|^2 |E_2|^2\}^{1/2}.$$

It follows from this formula, that reflectivity sharply increases near the seven specific frequencies represented in Table. Five of them can be predicted by the linear phase-matching condition. For the illustration these frequencies are written in linear approximation (far off resonance line)

$$\omega^{(1)} = \omega_1, \ \omega^{(2)} = \omega_1 - \Omega_+, \ \omega^{(3)} = \omega_1 + \Omega_+,$$
$$\omega^{(4)} = \omega_1 - \Omega_-, \ \omega^{(5)} = \omega_1 + \Omega_- \quad (3)$$

where

$$\Omega_\pm = \frac{\text{sgn}(\varepsilon_{1,2})}{\sqrt{2}} \{qc(1 + \eta) + \varepsilon_1^2 + \varepsilon_2^2 \pm \sqrt{[\varepsilon_1^2 - \varepsilon_2^2 + qc(1 - \eta)]^2 + 4\eta q^2 c^2}\}^{1/2}.$$

One can see from (3) that the specific frequencies of phase-matching are shifted relatively to the resonance frequencies on the value proportional to the

atom density N. These seven phase-conjugate ranges correspond to four-photon processes, diagrams of which are presented in Figs. 1a–d.

The phase-conjugate reflectivity dependence on probing signal frequency is shown in Fig. 2. It is seen from the given curve that phase-conjugation reflectivity resonantly increases in the above-mentioned spectral ranges oscillating at the same time. Near $\omega^{(6)} = \omega_1 - \Omega_4$ and $\omega^{(7)} = \omega_1 - \Omega_5$ the phase-conjugation reflectivity is by several orders of magnitude smaller than that in the range
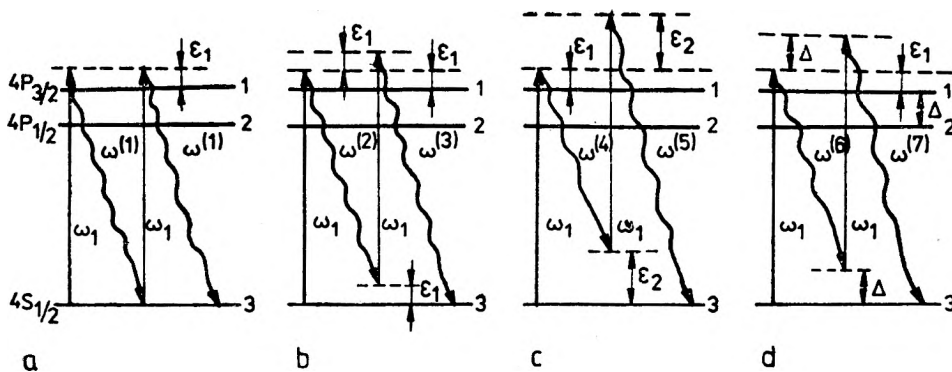

Fig. 1. The diagram of four-photon processes on a three-level atom. Two photons with frequency ω_1 (from the counter-propagating pump waves) are absorbed, and the other two are radiated with the following frequencies: (a) both photons of the same frequency $\omega^{(1)}$, (b) a photon with frequency $\omega^{(2)}$ close to the atomic absorption resonance $3 \to 1$ frequency, the second one with frequency $\omega^{(3)}$ close to the same transition three-photon scattering, (c) a photon with frequency $\omega^{(4)}$ close to the atomic transition resonance $3 \to 2$, and the second one with frequency $\omega^{(5)}$ close to the same transition three-photon scattering, (d) a photon with the fourth stimulated scattering process frequency $\omega^{(6)} = \omega_1 - \Omega_4$, the other one with the fifth stimulated scattering process frequency $\omega^{(7)} = \omega_1 - \omega_5$
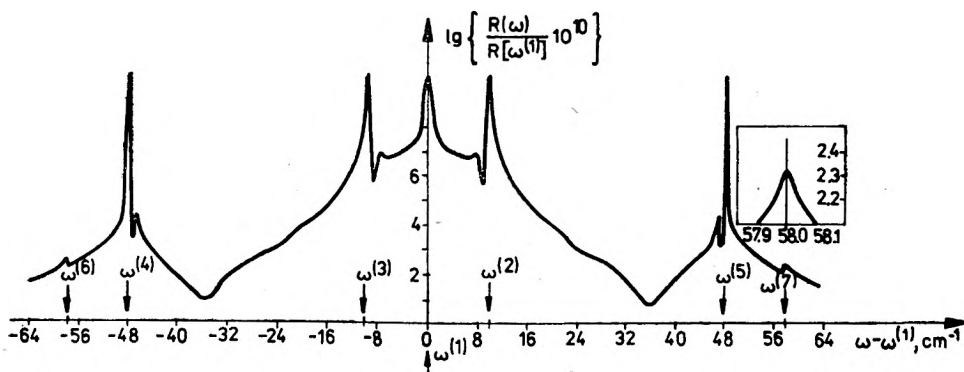

Fig. 2. The lg $R(\omega)/R(\omega^{(1)})$ vs. signal detuning at the dimensionless parameter of pump intensity $\xi/(\varepsilon_1^2 + \gamma_{13}^2) + 5 \times 10^{-2}$, $\gamma_{13} = \gamma_{23} = 1/4$ $\gamma_{12} = 10^{-2}$ cm^{-1}, $\Delta = 58$ cm^{-1}, $l = 1$ cm, $N = 10^{14}$ cm^{-3}, $\varepsilon_1 = -10$ cm^{-1}. The breaks of resonance poles are due to the fact that the pump wave saturation was not taken into account

$\omega^{(1)}-\omega^{(5)}$. It is caused by the mismatch for resonant frequencies of $\omega^{(6)}$ and $\omega^{(7)}$ at the given geometry.

The numerical estimates of R show that for all conjugation ranges, simultaneous observations of large pump wave detunings (of order of a few tens of cm^{-1}) are required, so that the pumping laser of high intensity becomes necessary.

3. Experiment

The scheme of experimental setup is shown in Figure 3. The tunable dye laser 2 with two-stage amplifier pumped by a Q-switch ruby laser 1 (with intensity ≈ 30 MW cm^{-2}) has been used as a driving oscillator. A grating (600 grooves /mm) with near grazing incident beam was used as the dye laser intracavity selector. The dye laser generation intensity was about 2 MW cm^{-2}, the pulse duration was about 30 ns. The radiation wavelength of the bandwidth of about
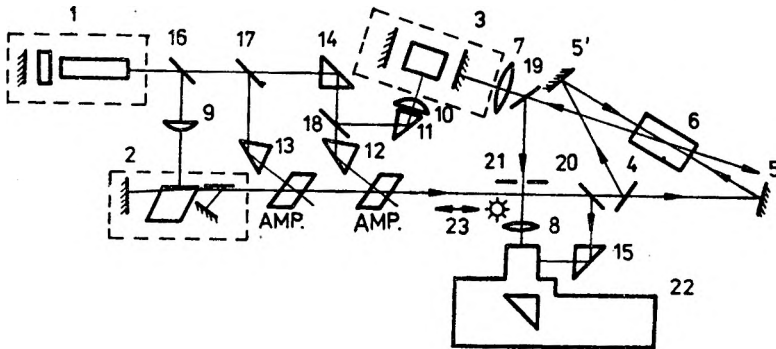

Fig. 3. The scheme of experimental setup: 1 — ruby laser, 2, 3 — dye lasers, 4 — half-transmitting mirror, 5, 5' — reflecting mirrors, potassium vapour cell, 7, 8 — spheric lenses, 9, 10 — cylindric lenses, 11–15 — prisms, 16–20 — beamsplitters, 21 — diaphragm, 22 — spectrograph, 23 — potassium lamp

0.5 cm^{-1} was tuned near both D lines of potassium doublet. To provide the counter-propagating reference pump waves the laser radiation 2 was directed into triangular interferometer, the latter consists of a half-transmitting mirror 4 and the mirrors 5, 5' with 0.98 reflectivity at the wavelength 767 nm. As a signal beam we used the radiation of the laser 3 with wide spectrum overlapping both the potassium lines focussed into 4 cm long potassium cell by means of lens 7 ($f = 120$ cm). For synchronization of lasers 2 and 3 the latter was pumped by the portion of the radiation of laser 1. The angle between reference and signal waves exceeds that of parametric scattering of each pump wave and was $\sim 4°$ [10]. During the experiment the temperature of medium was modified from 200° C to 300° C, so that potassium vapour density was $1.3 \cdot 10^{14}$ cm^{-3}–10^{16} cm^{-3}.

The spectral composition of the phase-conjugated wave was recorded by spectrograph ISP-51 (reverse linear dispersion at wavelength 767 nm is 2 nm/ /mm). The pump wave frequency was controlled during the experiment.

4. Results

The spectrograms of the conjugated wave at different temperatures of atomic potassium vapour are shown in Fig. 4. It is seen from Fig. 4a, b that at a temperature of 200 °C ($N \simeq 1.3 \cdot 10^{14}$ cm^{-3}) 3 ranges of conjugation near frequencies $\omega^{(1)}$, $\omega^{(2)}$, $\omega^{(4)}$ were detected irrespectively of detunings ε_1, ε_2. With the in-
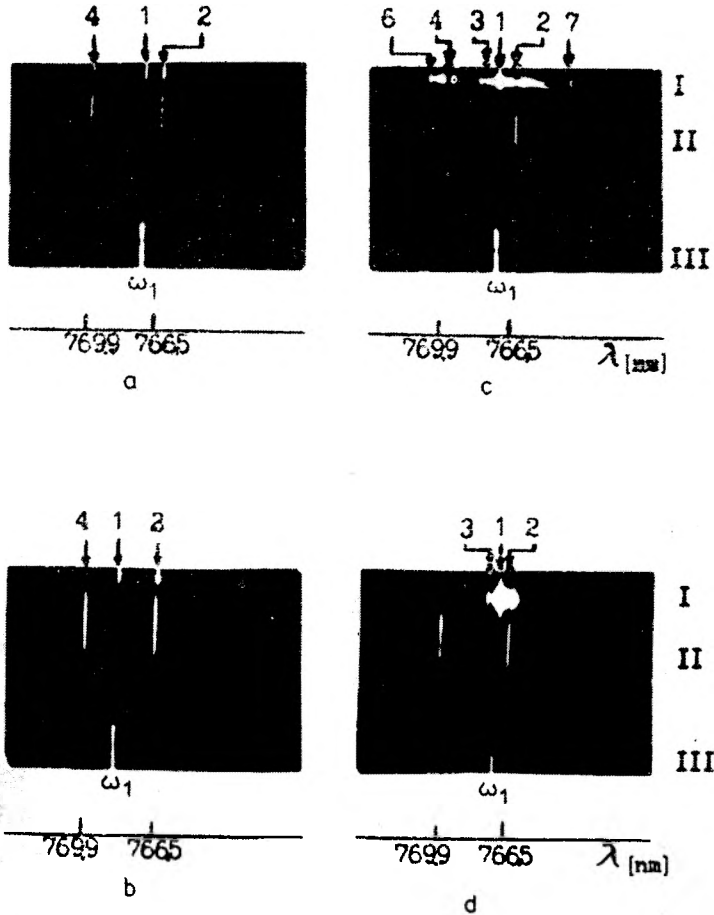

Fig. 4. Spectrograms of the conjugated wave at different temperatures: $T = 200$°C (a, b), $T = 284$°C (c), and $T = 256$°C (d). I — conjugated wave spectrum, II — potassium lamp spectrum (D_1, D_2 — lines), III — pump wave spectrum

creasing temperature there appear six ranges of conjugation near the frequencies $\omega^{(1)} - \omega^{(4)}$ and $\omega^{(6)}$, $\omega^{(7)}$ with wide conjugate background (Fig. 4c). A photometric measurement near the frequency $\omega^{(7)}$ shows that at this frequency the reflectivity exceeds about 1.5 times the nearby background. That is in good accordance with theoretical calculations (Fig. 2). Such a negligible deflection from background is due to the fact that, at given geometry of the experiment, phase matching condition is not fulfilled at frequencies $\omega^{(6)}$ and $\omega^{(7)}$, as mentioned above.

The observed temperature dependence of phase-conjugation process dynamics is caused by reflectivity dependence on N^2 in four-wave interaction. Thus, at low temperatures, i.e., at low densities, conjugation is observed only at the resonant ranges with dramatically large reflectivity. With further increase of temperature ($T = 330$ °C, $N \simeq 1.1 \cdot 10^{16}$ cm^{-3}) the efficiency decreases abruptly. It seems to be caused by broadening of atomic absorption lines and nonlinear absorption growth.

The difference in intensities of conjugate symmetric lines is represented in Fig. 4, and is possibly connected with inhomogeneous frequency distribution of incident signal wave intensity. The absence of far line $\omega^{(5)}$ near three-photon scattering frequency detection is due to the same reason. This fact is confirmed by experimental results shown in Fig. 4d. In this figure wide spectrum of incident signal wave overlaps only potassium D_2 line, moreover, three conjugation ranges were detected: one near the pump wave frequency $\omega^{(1)}$, and two symmertic ones near the frequency $\omega^{(2)}$ of resonant transition $4\,S_{1/2} \to 4P_{3/2}$ and at frequency $\omega^{(3)}$ of the same transition three-photon scattering.

Thus, the results obtained show that if the alkali metal vapour was used as a resonant medium not only phase-conjugate reflectivity can be obtained but also conjugated wave spectrum control can be made.

References

[1] DUCLOY M., Adv. in Solid State Physics **22** (1982), 35.
[2] BLOOM D. M., LIAO P. F., ECONOMOU N. P., Appl. Phys. Lett. **32** (1978), 813.
[3] TAN-NO N., HOSHIMIYA T., INABA H., IEEE J. Quant. Electron. QE-**16** (1980), 147.
[4] NILSON I., YARIV A., Appl. Opt. **18** (1979), 143.
[5] FU T., SARGENT M., III, Opt. Lett. **4** (1979), 366.
[6] BLOEMBERGEN N., *Laser spectroscopy XV*, Springer Series in Optical Sciences, Vol. 21, Springer-Verlag 1979, p. 340.
[7] PRIOR I., BOGDAN A. R., DAGENAIS M., BLOEMBERGEN N., Phys. Rev. Lett. **46** (1981), 111.
[8] GRYNBERG G., J. Phys. B: At. Mol. Phys. **14** (1981), 2089.
[9] ARUTUNYAN V. M., KANETSYAN E. G., CHALTYKYAN V. O., Zh. Eksp. Teor. Fiz. **59** (1970), 195 (Sov. Phys. JETP **32** (1970), 108).
[10] ARUTUNYAN V. M., ADONTS G. G., PAPAZYAN T. A., SARKISSYAN S. M., ARZUMANYAN G. M., MELIKSETYAN T. E., Izv. AN Arm. SSR, Fizika **12** (1974), 338.