INVESTIGATION OF INDUCED THREE-PHOTON SCATTERING OF LIGHT UPON EXCITATION NEAR RESONANT TRANSITIONS OF THE RUBIDIUM ATOM

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The spectral distribution and the intensity of induced three-photon scattering near the $5^2S_{1/2}$—$5^2P_{1/2,3/2}$ resonance transitions of the rubidium atom upon excitation of a narrow (0.2 cm^{-1}) frequency-tuned pumping line with intensities up to 30 MW/cm^2. The forced three-photon scattering was observed in the form of a wide asymmetric line. A much slower intensity falloff was always observed from the edge of the line, removed from resonance frequency ω_0 of the atomic transition. The highest-energy pulse due to induced three-photon scattering was attained upon long-wave divergence between pumping frequency ω_L and the resonance frequency, i.e., $\omega_0 - \omega_L \sim 1$-4 cm^{-1}. The dependence of the shift of the maximum of the line of induced three-photon scattering on ω_0-ω_L. It is shown that the location of the maximum and the intensity distribution of the induced three-photon scattering line are in satisfactory agreement with theoretical estimates, making allowance for the shift of atomic levels as a result of Stark's dynamic effect.

Forced three-photon scattering near the $5^2S_{1/2}$—$5^2P_{3/2}$ resonance transition of the rubidium atoms was observed previously by Badalyan et al. [1]. The present paper describes an investigation of the spectral distribution and intensity of this scattering upon excitation near the $5^2S_{1/2}$—$5^2P_{1/2,3/2}$ resonance transition employing a narrow (0.2 cm^{-1}), tuned by the pumping-line frequency (see also the paper by the present authors [2]). It was seen by comparing with analytic calculations that the observed spectral distribution of forced three-photon scattering can be attributed to the manifestation of Stark's optical effect*. The pumping pulse duration τ_L was 25 nanosec. The length of the tray with Rb vapor was l = 18 cm. A study of the interaction of the pumping ray near resonant transitions showed that there exists, near to resonance, a domain of long-wave tuning-frequency differences $\Delta_L = \omega_L - \omega_0 < 0$ (ω_L is the pumping frequency, and ω_0 is the transition frequency for an undisturbed atom), within which rather intensive radiation does not undergo self defocusing [2]. This domain is most favorable with theoretical estimates.

Retuning of ω_L in the vicinity of each of the resonance transitions of Rb

*Shifting of the forced three-photon scattering line as a result of Stark's optical effect was observed previously in potassium vapor [3, 4].

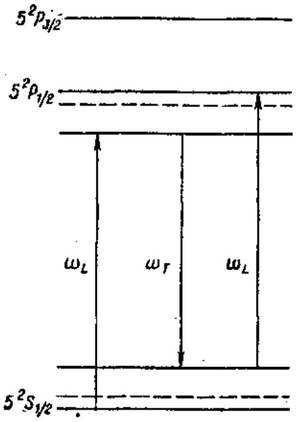

Fig. 1. Schematic of forced excitation of three-photon scattering near the $5^2S_{1/2}-5^2P_{1/2}$ transition.

there occurs an effective forced excitation of three-photon scattering (see Fig. 1). The forced three photon scattering line was observed over a wide temperature range (t from 200 to 300°C). The energy of the forced three-photon scattering W_T increased with t°. Forced three-photon scattering was observed in the direct as well as inverse direction with approximately the same effectiveness. The value of W_T attained the maximum of about $3 \cdot 10^{-4}$ J at long-wave tuning-frequency difference from 1 to 4 cm^{-1}. Forced three-photon scattering weakened upon increasing tuning-frequency difference and vanished jumpwise at the instant of appearance of self defocusing. The value of W_T dropped steeply in the immediate vicinity of resonance. At $|\Delta_L| \leqslant 0{,}5$ cm^{-1} the line of forced three-photon scattering was very weak, or was not observed at all. Forced three-photon scattering was excited in the form of a wide line, with characteristic asymmetric shape. A significantly slower intensity drop was always observed from the edge of the line, removed from resonance frequency ω_0. Figure 2a depicts the shape of a forced three-photon scattering line, excited at the inverse direction near the $5^2S_{1/2}-5^2P_{1/2}$ transition at peak pumping intensity of I_{Lm} = 30 MW/cm^2, t = 260°C (atomic population N = $6 \cdot 10^{15}$ cm^{-3}), Δ_L = -10 cm^{-1}, and its location relative to the "non-shifted" frequency of three-photon transition $\omega^0_T = 2\omega_L - \omega_0$. Here $\delta\omega_T = \omega_T - \omega^0_T < 0$, so that the line shift relative to ω^0_T agrees with the direction of the Stark shift of levels (see Fig. 1). For a fixed tuning-frequency difference the location and the shape of the forced three-photon scattering line were virtually independent of the peak pumping intensity. Forced three-photon scattering radiation in the direction of propagation of pumping had a linewidth 1.5-2 fold greater than the inverse forced three-photon scattering. Broadening occured primarily in the wing far from ω_0. The shift of the line's maximum relative to ω^0_T remained the same as upon inversed forced three-photon scattering. It can be assumed that this broadening is due to a four-photon parametric process.

The shift of the frequency of the maximum of the forced three-photon scattering line ω_{Tm} relative to ω^0_T, i.e., $\delta\omega_{Tm} = \omega_{Tm} - \omega^0_T$, as a function of Δ_L is depicted in Fig. 3 (I_{Lm} = 30 WM/cm^2, t = 260°C).

Tuning of ω_L in the vicinity of $5^2S_{1/2}-5^2P_{1/2,3/2}$ was accompanied, alongside with forced three-photon scattering, by forced VKR scattering [2]. The forced scattering line arises at higher I_L and t° than the forced three-photon scattering line. The only exception is the domain of small tuning-frequency differences $|\Delta_L| \leqslant 1$ cm^{-1}, within which the forced three-photon scattering is weak, whereas the intensity of forced VKR scattering attains a maximum.

In analytically calculating the spectral distribution of forced three-photon scattering we set $\alpha^2 = \left(\frac{\mu E_L}{\hbar \Delta_L}\right)^2 \ll 1$, where μ is the dipole matrix element of transition

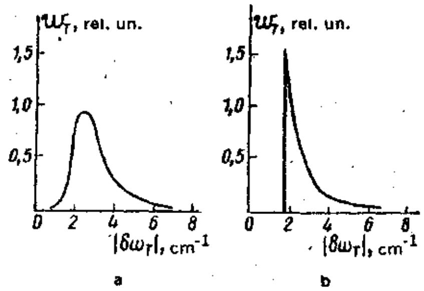

Fig. 2. Experimental (a) and analytic (b) shape of the forced three-photon scattering line.

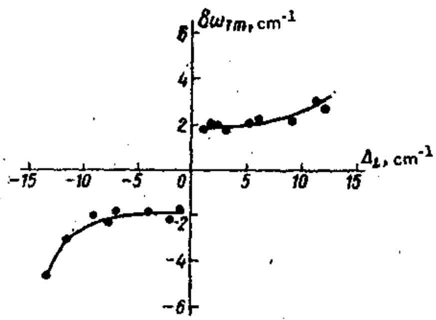

Fig. 3. Experimental shift of the maximum of the three-photon forced scattering line as a function of tuning-frequency difference Δ_L.

E_L is the amplitude of the pumping field (under experimental conditions for sufficiently high $|\Delta_L|$ forced three-photon scattering occurs at values of α^2 significantly lower than unity). The gain factor, "per passage" for "forward" forced three-photon scattering at segment 0, z is written as $G(z) = \int_0^z a(N_1 - N_2) I_L^2 dz'$, where $a \sim \mu^6/\Delta_L^4$, whereas N_1 and N_2 are the atomic populations at the initial and final levels 1 and 2. We shall assume that the energy of the forced three-photon scattering pulse is sufficiently high for saturation of the 1-2 transition to be significant. Let $\hat{z}(t)$ be a point in the tray in which the forward-scattering power $P_T(t)$ attains some level $\hat{P}_T$, which is selected to be not too small as compared with the peak power of forced three-photon scattering, and at the same time also such that there would not yet be any significant saturation of transition 1-2 in point $\hat{z}(t)$ at time t. The value of G needed for attaining power $\hat{P}_T$ is designated by $\hat{G}$. Under our conditions $\hat{P}_T \sim 0.1$ kW, $\hat{G} \sim 25$.

The threshold pumping intensity $\hat{I}_L$, determined from attainment of power $\hat{P}_T$ by forced three-photon scattering radiation, is found from the expression $aN\hat{I}_L^2 l = \hat{G}$. At $I_L > \hat{I}_L$ point $\hat{z}$ moves to the left from the end of the tray (where $z = l$) as I_L increases up to I_{Lm}. At $z > \hat{z}$, as a result of the high value of G(z), P_T increases even over the small length $\Delta z \ll l$, to values at which the populations of levels 1 and 2 become virtually equal over a time much shorter than τ_L. This makes it possible to use a simplified model, in which length Δz of the transition segment is neglected, and it is assumed that at each time t we have $N_2 = 0$, $N_1 = N$ at $z < \hat{z}(t)$ and $N_1 = N_2 = N/2$ at $\hat{z}(t) < z < l$. When both the forward and inverse scattering is taken into account, the motion of point $\hat{z}$ is determined by the condition $aNI_L^2(2\hat{z} - l) = G$. The forced three-photon scattering energy, radiated either forward or reverse toward time t is $W_T(t) = (1/2)\hbar\omega_T N\sigma(l - \hat{z})$, where σ is the cross sectional area of the beam. For $P_T(t) = \frac{dW_T}{dt}$, we obtain $P_T(t) = \frac{1}{2}\hbar\omega_T \frac{\sigma G}{aI_L^3}\frac{dI_L}{dt}$. Frequency ω_T is controlled by Stark shifting of levels, and consequently, is a function of I_L: $\omega_T = \omega_T(I_L)$. With reference to this, we obtain for the spectral density of forced three-photon scattering the expression

$$\mathcal{W}_T(\omega_T) = \frac{1}{2}\hbar\omega_T \frac{\sigma G}{aI_L^3(\omega_T)}\left|\frac{dI_L(\omega_T)}{d\omega_T}\right|,$$

where $I_L(\omega_T)$ is a function reciprocal of $\omega_T(I_L)$. This expression is valid for values of ω_T controlled by the condition $\hat{I}_L < I_L(\omega_T) < I_{Lm}$; outside the corresponding frequency interval $\mathcal{W}_T = 0$. It is seen that $\mathcal{W}_T(\omega_T)$ is independent of the pulse shape and of peak pumping intensity. Using the magnitude of the Stark shift

$$\delta\omega_T = \omega_T - \omega_T^0 = \Delta_L\,(\sqrt{1+a^2} - 1) \approx \frac{4\pi\mu^2 I_L}{ch^2\Delta_L},$$

we obtain

$$\mathcal{W}_T(\omega_T) = \frac{8\pi^2 G\sigma\omega_T\mu^4}{c^2 a\Delta_L^2 h^3 |\delta\omega_T|^3}, \quad \delta\hat{\omega}_T = \hat{\omega}_T - \omega_T^0 = \frac{4\pi\mu^2}{ch^2\Delta_L}\hat{I}_L,$$

where $\hat{\omega}_T = \omega_T(\hat{I}_L)$. The theoretical shape of $\mathcal{W}_T(\omega_T)$ for the $5^2S_{1/2}$-$5^2P_{1/2}$ at Δ_L = = -10 cm^{-1} is shown in Fig. 2b. Calculations for the general case of arbitrary α also yields $I_L \sim \Delta^2_L$, $\delta\hat{\omega}_T \sim \Delta_L$, whereas $\mathcal{W}_T(\omega_T)$ as previously, is independent of the shape and of the peak intensity of pumping.

At the assumed tuning-frequency difference the location of the maximum of the analytic line is in satisfactory agreement with experimental results (see Fig. 2). However, at low $|\Delta_L|$ the experimental shift remains constant (see Fig. 3), whereas theoretically it should decrease as $\sim|\Delta_L|$. As was shown by us previously [2], the agreement between analytic and experimental results can be significantly improved, by making allowance for depletion of pumping, which is significant at low $|\Delta_L|$.

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