Stimulated emission in multiphoton excitation of an atom above the ionization limit

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Powerful stimulated emission in the infrared (IR) region of the spectrum was observed following three-photon excitation of a rubidium atom above the ionization limit. The phenomenon has a clearly pronounced resonant character and may be connected with the resonant ionization of the atom.

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The source of exciting radiation was a dye laser with tunable frequency. The laser radiation was focused with a telescopic system into a cell with Rb vapor. The cell was 18 cm long and had sapphire optical windows. The area of the cross section of the light beam in the cell was ~ 1 mm^2. The pump power reached $P_L \sim 300$ kW at a lasing line width 0.2 cm^{-1}. The duration of the pulse of the exciting laser was $\tau_L = 25$ nsec. The temperature of the Rb vapor was 260 °C (the atom density was $N \sim 6 \times 10^{15}$ cm^{-3}). The instrumental width of the IR monochromator in most experiments was 0.5 cm^{-1}. The recording system made it possible to record IR signals with energy up to $\sim 2 \times 10^{-7}$ J.

When the pump frequency was varied near the two-photon resonance $5^2S_{1/2}$–$5^2D_{3/2,5/2}$ (Fig. 1), a powerful directed radiation was observed on the transitions $5^2D_{5/2}$–$7^2P_{3/2}$ (4.615 μm), $5^2D_{3/2}$–$7^2P_{1/2}$ (4.683 μm), $5^2D_{3/2}$–$7^2P_{3/2}$ (4.608 μm), $4^2F_{3/2,5/2}$–$6^2D_{5/2}$ (5.268 μm), $4^2F_{3/2,5/2}$–$6^2D_{3/2}$ (5.274 μm), $6^2P_{3/2}$–$5^2D_{5/2}$ (5.231 μm), $6^2P_{1/2}$–$5^2D_{3/2}$ (5.035 μm), $6^2P_{3/2}$–$5^2D_{3/2}$ (5.239 μm), $4^2D_{5/2,3/2}$–$6^2P_{3/2}$ (2.253 μm), $4^2D_{5/2,3/2}$–$6^2P_{1/2}$ (2.293 μm), $6^2S_{1/2}$–$6^2P_{3/2}$ (2.731 μm), $6^2S_{1/2}$–$6^2P_{1/2}$ (2.790 μm). The energy of the most intense 4.61 μm line reached $\sim 10^{-4}$ J (the power averaged over the time τ_L was ~ 4 kW), and the registered width did not exceed the instrumental width of the monochromator. It could be observed also at pump powers much lower than the maximum.

We can propose the following mechanism for the onset of the stimulated emission of the IR lines. When the pump frequency approaches the frequency of the two-photon transition $5^2S_{1/2}$–$5^2D_{3/2,5/2}$[1)] the probability of the three-photon ionization in-

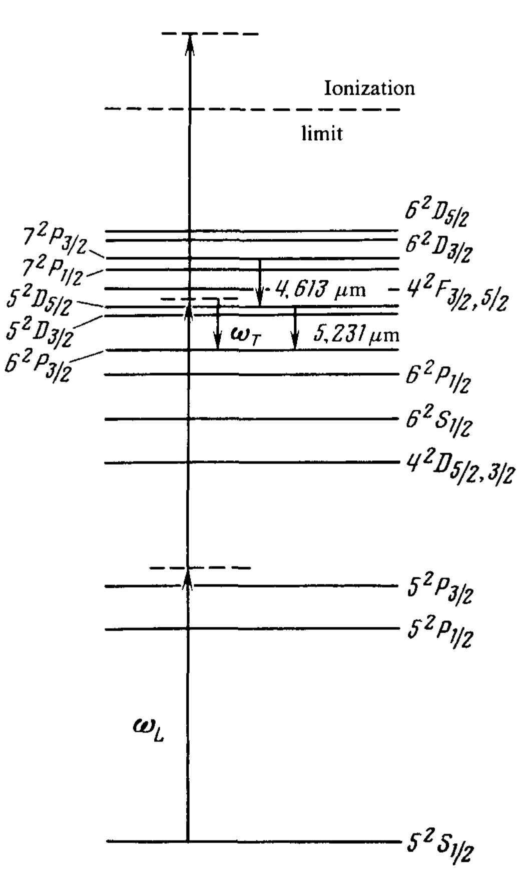

FIG. 1. Energy level scheme of the rubidium atom.

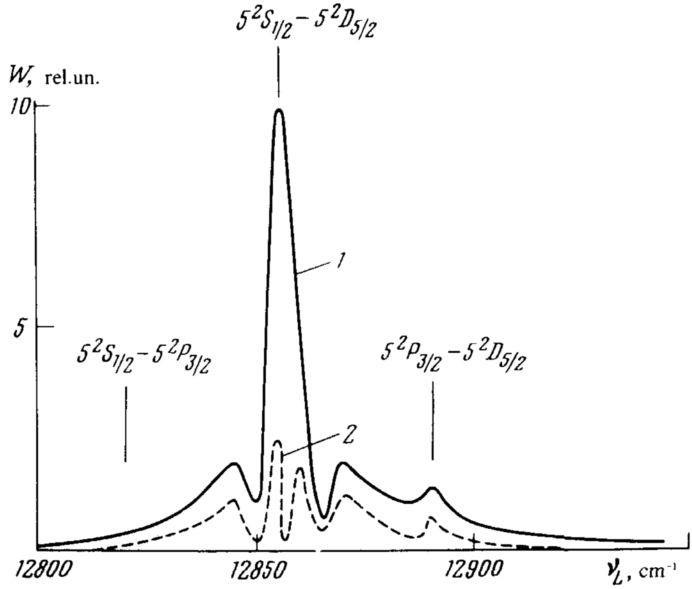

FIG. 2. Dependence of the energy of the 4.61 and 5.23 μm lines on the pump frequency: 1—4.61 μm line, 2—5.23 μm line.

creases strongly. Numerical estimates based on the results of Ref. 1 yield for the rate of the three-photon ionization in the resonance case a value $\sim 10^7$ sec^{-1}. The ionization leads to population of continuum states located near the ionization limit. When colliding with atoms, the electrons lose the excess kinetic energy and in the recombination process then accumulate on the upper levels of the atom. Cascade stimulated transi-

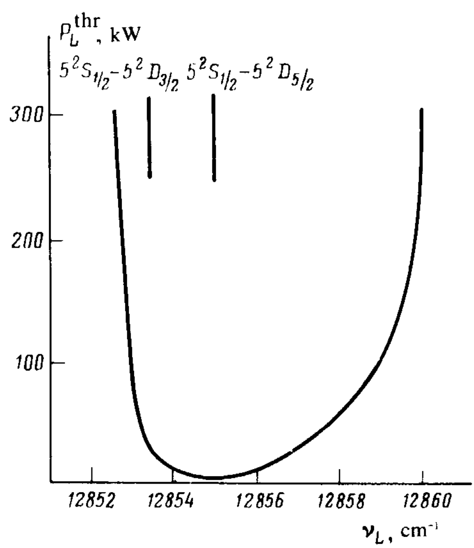

FIG. 3. Frequency dependence of the threshold of IR stimulated three-photon scattering (STS).

tions to lower levels develop from these levels and are accompanied by emission of a number of IR lines. In addition to those registered, one should expect the appearance of IR lines with larger wavelengths, but these could not be observed in our experiment because of the absorption by the sapphire windows of the cell. Numerical estimates show that the stimulated emissions of the IR lines can develop at extremely low inverted population ΔN of the levels, lower by several orders of magnitude than the concentration N of the atoms. As the cascade transitions lead to population of excited levels, single and two-photon ionization of the atom from the excited levels becomes possible in addition to the three-photon ionization from the ground level. The rapid departure of the electrons from the populated states of the continuum, due to collisions and recombination, makes it possible to attain in principle powerful continuous IR radiation.

Figure 2 shows the dependence of the IR energy of the 4.615 and 5.231 μm lines on the pump frequency, obtained at $P_L \simeq 300$ kW. It is seen that, with the exception of the frequency region located in the immediate vicinity of the two-photon resonance, the two lines correlate in behavior. This correlation must be attributed to the fact that the population of the $5^2D_{5/2}$ level, which is the initial one for the 5.231-μm line, is effected by the emission of the 4.615 μm line. The narrow 5.23 μm energy maximum at the two-photon resonance is obviously connected with the two-photon population of the $5^2D_{5/2}$ level from the ground state.[2,3] It is here that an appreciable decrease of the experimental threshold of the excitation of this line is observed.

When the laser emission frequency is varied in the immediate vicinity of the two-photon resonance $5^2S_{1/2}$–$5^2D_{5/2}$, infrared stimulated three-photon scattering of light (STS) was also observed at a frequency ω_T (Fig. 1). The threshold curve for this process is shown in Fig. 3. As seen from the figure, the threshold curve has a minimum at zero detuning from the two-photon resonance. The appreciable asymmetry in the behavior of the threshold curve is apparently connected with the fact that at long-wave detunings ~ 3 cm^{-1} a transition takes place from the level $5^2D_{3/2}$ and leads to population of the level $6^2P_{3/2}$.

When the pump frequency was varied in the vicinity of the transition $5^2P_{3/2}$–$5^2D_{5/2}$, IR STS was observed, due to the transitions $5^2P_{3/2}$–$6^2P_{3/2}$. The appearance of the IR STS line is evidence of an appreciable population of the initial level $5^2P_{3/2}$.

We note in conclusion that the stimulated emission observed in the present study under multiphoton ionization of the atom can be used to convert laser radiation into IR radiation.

[1)]In this case the detuning from the intermediate $5^2P_{3/2}$ level is also small.

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