THE ROLE OF MULTIPHOTON PROCESSES IN ESTABLISHING THE LIMITING POWER OF QUANTUM OSCILLATORS

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A multiphoton mechanism of absorption of electromagnetic radiation from an optical oscillator in the laser crystal itself is considered. An estimate is made of the limiting power of optical oscillators utilizing GaAs and ruby crystals due to the internal photoeffect in these crystals.

THE operation of the usual quantum oscillator (and also amplifier) occurs at the expense of negative absorption of electromagnetic energy in single-photon transitions between two "working" levels of the active substance. All methods of obtaining negative absorption in a medium (both those that have been practically realized already and also other conceivable ones) enable one to achieve this goal only with respect to the "working" transition and, perhaps, with respect to other transitions sufficiently close to it. The creation of negative absorption in the system with respect to all quantum transitions in it is, as is well known, impossible in principle. This circumstance leads, in particular, to the result that the multiphoton processes possible in the active substance of the oscillator in which l quanta $\hbar\omega$ participate simultaneously ($l = 2, 3, \ldots, \omega$ is the frequency of oscillation close to the frequency of the "working" transition) occur, generally speaking, accompanied by absorption of energy.[1)]

It is evident that this effect places a theoretical limit on the growth of power of a quantum oscillator. The latter follows from the fact that the probability of a single photon (working) transition increases linearly with the energy density of the radiation $\rho(\omega)$, while the probability of a multiphoton transition is $\sim\rho^l(\omega)$ ($l = 2, 3, \ldots$).

It is clear that the aforementioned effect can have practical significance only for optical quantum oscillators, and primarily in solids. The exclusion of gas lasers is due to the relatively high ionization potentials of atoms of which the generally used gas mixtures are composed (usually greater than 15 eV) so that multiphoton ionization in these mixtures becomes significant only at fields of the order of 10^8 v/cm and higher. But such radiation densities cannot be attained in gas lasers for other reasons. As regards multiphoton transitions between discrete levels of atoms which require a smaller number l of quanta, they can be realized only in specially selected mixtures containing atoms with transition frequencies ω_{kn} very close (due to the narrow line width) to the frequency $l\omega$. A special case can be presented by molecular gas mixtures with a relatively long wavelength edge of an absorption band. For example, the presence of polar molecules can lead to two- or three-photon dissociation occurring at a relatively low radiation density [1].

Crystalline solids are characterized by having many absorption bands. In future for the sake of simplicity and definiteness we shall restrict ourselves to taking into account only those absorption bands which are due to the band structure of the electronic spectrum of the crystals, i.e., to band transitions or to impurity-band transitions. It is clear that the effect under consideration will be most pronounced in lasers utilizing semiconductors for which the value of the quantum being generated $\hbar\omega$ is close to the width of the forbidden band Δ (i.e., in particular, for all the injection lasers known at present). For them there always exist (of course, only provided the width of the conduction band is not smaller than the width of the forbidden band Δ) two-photon absorption processes leading to the creation of electron-hole pairs. If the recombination time of nonequilibrium carriers is large compared to the slowing down (thermalization) time of electrons (holes) down to states close to the degenerate state, then for each elementary act of simultaneous absorption of two

[1)] At present we leave out of consideration multiphoton processes in which quanta of different frequencies participate simultaneously (in particular, Raman scattering).

quanta $\hbar\omega$ only one quantum of energy will be transferred to the lattice; the other will be expended in increasing the inverted population of the "working" transition (cf., the review article of Basov et al.[2]).

On the basis of the paper by Keldysh[3] it is not difficult to make an estimate of the limiting intensity of the field E_{lim} in a p-n junction in a semiconductor laser. In order to do this we calculate the coefficient of absorption per unit length $\alpha^{(2)}$ due to two-photon absorption. We have:

$$\alpha^{(2)} = 4\pi\sigma^{(2)}/nc, \tag{1}$$

where c is the velocity of light, n is the index of refraction inside a p-n junction, $\sigma^{(2)}$ is the effective additional conductivity due to the two-photon absorption which is determined by the condition

$$\sigma^{(2)}E^2 = 2\hbar\omega w^{(2)}. \tag{2}$$

Here $w^{(2)}$ is the number of electron-hole pairs created per unit volume per unit time as a result of the two-photon process. This quantity has been calculated by Keldysh (cf., formula (41) in [3]) and up to a factor of the order of unity has the form

$$w^{(2)} \approx \omega\left(\frac{m\omega}{\hbar}\right)^{3/2}\left(\frac{e^2E^2}{m\omega^2\Delta}\right)^2, \tag{3}$$

where m is the reduced mass of the electron and the hole ($m^{-1} = m_e^{-1} + m_h^{-1}$).

Formulas (1)—(3) lead to the following expression for the absorption coefficient $\alpha^{(2)}$:

$$\alpha^{(2)} \approx \frac{8\pi\hbar\omega^2}{nc}\left(\frac{m\omega}{\hbar}\right)^{3/2}\left(\frac{e^2}{m\omega^2\Delta}\right)^2 E^2. \tag{4}$$

The order of magnitude of the value of the limiting field intensity E_{lim} is determined by the condition $\alpha^{(2)} = \alpha$, where α is the amplification coefficient in a p-n junction taking into account all the "linear" energy losses.[2)] The value of α for a semiconductor laser is approximately equal to 200 cm^{-1}. Setting $\omega = 2 \times 10^{15}$ c/sec, $\Delta \approx 1.5$ eV (gallium arsenide), n = 3, we obtain $E_{lim} \approx 5 \times 10^5$ V/cm. From this the limiting power generated by such a laser calculated from the formula $P_{lim} \approx cE_{lim}^2 S/4\pi$ (S is the area of the ends of the p-n junction of order of magnitude 10^{-5} cm^2) is equal to 10 kW.

It is of interest to carry out an analogous estimate of the limiting power for a ruby laser, although possibly in this case the power generated is limited by an entirely different mechanism. For ruby the width of the forbidden band is equal to 5.95 eV, which corresponds to four-photon absorption. However, we shall consider the more probable process of ionization of Cr^{+++} impurities corresponding to three-photon absorption with a transition from the 2E metastable state into the conduction band.[3)] Since in this case ionization of a discrete level is taking place we shall utilize for making an estimate of the probability of simultaneous absorption of three quanta $\hbar\omega = 1.78$ eV the formula obtained for isolated atoms [cf., [3], formula (21)].

The number of electrons formed per unit volume of the conduction band per unit time is given up to a factor of the order of unity by

$$w^{(3)} \approx 70N\omega\left(\frac{I_0}{\hbar\omega}\right)^{3/2}\left(\frac{e^2E^2}{m\omega^2 I_0}\right)^3, \tag{5}$$

where $N \approx 10^{19}$ cm^{-3} is the density of chromium ions, $I_0 \approx 3.85$ eV is the ionization potential of the 2E level. On the basis of formulas (1) (with the index 2 replaced by 3), (5) and of the relation $\sigma^{(3)}E^2 = 3\hbar\omega w^{(3)}$, we obtain (n = 1.78)

$$\alpha^{(3)} \approx 10^3 N\frac{\hbar\omega^2}{c}\left(\frac{I_0}{\hbar\omega}\right)^{3/2}\left(\frac{e^2}{m\omega^2 I_0}\right)^3 E^4. \tag{6}$$

For a ruby laser the amplification coefficient is $\alpha \approx 1$ cm^{-1}. From this we obtain $E_{lim} \approx 10^7$ V/cm. The limiting power in this case turns out to be equal to ($S \approx 1$ cm^2) $P_{lim} \approx 2 \times 10^5$ MW.

Thus, the limiting power which can be removed per unit volume of a semiconductor laser (specific power) is lower by approximately a factor of one thousand compared to the limiting specific power of a ruby laser.

[1] Bunkin, Karapetyan, and Prokhorov, JETP **47**, 216 (1964), Soviet Phys. JETP **20**, 145 (1965).

[2] Basov, Krokhin, and Popov, UFN **72**, 161 (1960), Soviet Phys. Uspekhi **3**, 702 (1961).

[3] L. V. Keldysh, JETP **47**, 1945 (1964), Soviet Phys. JETP **20**, 1307 (1965).

[4] Maiman, Hoskins, D'Haenens, Asawa, and Evtuhov, Phys. Rev. **123**, 1151 (1961).

Translated by G. Volkoff
158

[2)] The value of E_{lim} obtained in this manner will always be too high due to the fact that actually the maximum of the radiation density in a laser must occur before the condition $\alpha^{(2)} = \alpha$ is satisfied.

[3)] The 2E level lies at approximately 2.1 eV above the edge of the valence band (cf.[4]).