# Dependence of molecular-laser efficiency on the emission spectrum

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It is concluded that the efficiency of a molecular laser increases when the stimulated emission is shifted from the transition characterized by the maximum gain to higher rotational states. Methods of realizing this effect are proposed.

In this paper we wish to call attention to the feasibility, in principle, of increasing the efficiency of lasers operating on vibrational-rotational transitions of molecules. This feasibility is connected with the fact that under conditions of rapid relaxation in the rotational system of the sublevels, the coefficient of conversion of the pump energy into coherent emission can depend significantly, as will be shown below, on the spectral composition of the radiation even in that spectral region where the energy of the emitted photons changes insignificantly. The decisive factor here is the rotational quantum number of the radiating states.

We consider the distribution of the molecules over the vibrational levels in the presence of a strong resonant radiation field.[1)] If the radiation saturates the transition $v,\ J-1 \to v-1,\ J$, where $v$ and $J$ are the vibrational and rotational quantum numbers, respectively, then the population densities of the vibrational states $v$ and $v-1$, in the presence of rotational-translational equilibrium, are connected by the relation

$$n_v = n_{v-1}\exp(-2J\Theta_r/T), \tag{1}$$

where $\Theta_r$ is the characteristic rotational temperature of the molecule and $T$ is the temperature of the external degrees of freedom.

The distribution (1) can be realized in the generation regime if the resonator $Q$ is high, when the threshold value of the difference between the populations of the working levels can be neglected, or else in the case of amplification under strong saturation. Formula (1) can be easily generalized to the case when several vibrational transitions, starting with $v=M$ and ending with $v=R$, take part in the generation (amplification). In this case (1) goes over into

$$n_v = n_M \exp\left\{-2\frac{\Theta_r}{T}\sum_{s=M+1}^{v} J(s)\right\}, \quad v=M+1,\ldots,R, \tag{2}$$

where $J(s)$ is the rotational quantum number of the final state of the transition in the band $v=s \to v=s-1$ and $n_M$ is the population of the $M$th vibrational state. If the threshold density of the inversion cannot be neglected, we have in lieu of (2)

$$n_v = n_M \exp\left\{-2\frac{\Theta_r}{T}\sum_{s=M+1}^{v} J(s)\right\} + \sum_{k=M+1}^{v}\Delta_{k-1,\,J(k)}^{k,\,J(k)-1} \times$$

$$\times \exp\left\{-2\frac{\Theta_r}{T}\sum_{s=k}^{v} J(s)\right\}, \tag{3}$$

where $\Delta_{v-1,J}^{v,J-1}$ is a quantity proportional to the threshold density of the inverted population for the transition $v,\ J-1 \to v-1,\ J$ and depends on the photon emission cross section $\sigma_{v-1,J}^{v,J-1}$, on the lifetime $\tau_{ph}$ of the photon in the resonator, and on the rotational quantum number $J$ in the following manner:

$$\Delta_{v-1,\,J}^{v,\,J-1} = \frac{1}{c\tau_{ph}\sigma_{v-1,\,J}^{v,\,J-1}}\,\frac{T}{\Theta_r}\,\frac{1}{2J-1}\exp\left\{\frac{\theta_r}{T}J(J+1)\right\}.$$

In molecular lasers with sufficiently short radiation pulses, when the relaxation does not play a fundamental role, the vibrational energy converted into light is

$$\mathcal{E}_L = h\nu(J)\sum_{v=M+1}^{R} v(n_v^{(0)} - n_v), \tag{4}$$

where $n_v^{(0)}$ is the population of the vibrational level $v$ produced by the pump in the absence of generation (amplification), and $h\nu(J)$ is the energy of the photon emitted in the transition $v,\ J-1 \to v-1,\ J$. For simplicity, neglect in (4) the weak dependence of the photon energy on $v$, and the number $J$ is assumed to be the same for all the considered bands. It follows from (2) that $n_v$ decreases with increasing $J$. Other conditions being equal, this leads, owing to the weak dependence of the photon energy on $J$, on an increase of the useful energy $\mathcal{E}_L$ and of the efficiency of the system. In the cw and quasi-cw regimes, the dependence of the laser energy parameters on the emission spectrum is easiest to illustrate with a two-level excitation scheme as an example. Let $W$ be the pumping rate of the level $v=1$, and let the emission be effected on the transition $v=1,\ J-1 \to v=0,\ J$. The well-known balance conditions for the working-level populations and for the number of photons in the resonator then yield readily an expression for the generation power

$$P_d = h\nu(J)\left(W - \frac{n_1}{\tau_{rel}}\right) \tag{5}$$

where $\tau_{rel}$ is the relaxation time of the level $v=1$, while $n_1$ is connected with $n_0$ by relation (1) or (3). It follows from (5) that the power loss due to relaxation should decrease with increasing rotational quantum number of the radiating states. Usually the emission spectrum of a molecular laser is automatically formed by the laser active medium itself in such a way that lasing in each band $v \to v-1$ occurs on the transition having the maximum gain. Under rotational-equilibrium conditions, the quantum number characterizing this transition lies near the value $J_m = (T/2\Theta_r)^{1/2}$. From the energy point of view, these transitions are in general not optimal. Thus, if $n_v^{(0)}$ in (4) is $\kappa$ times larger than $n_v(J_m)$, then the transfer of the generation (amplification) to higher rotational levels close to $J=\mu J_m$ increases the efficiency of the system in the ratio

$$\frac{\mathcal{E}_L(\mu J_m)}{\mathcal{E}_L(J_m)} = \frac{h\nu(\mu J_m)}{h\nu(J_m)}\,\frac{\sum\limits_{v=M+1}^{R} v\left(\kappa - \exp\left(-\frac{2\Theta_r}{T}(\mu-1)J_m\right)\right)\exp\left(-\frac{2\Theta_r}{T}J_m v\right)}{\sum\limits_{v=M+1}^{R} v(\kappa-1)\exp\left(-\frac{2\Theta_r}{T}J_m v\right)},$$

i.e., by a factor $\kappa/(\kappa-1)$ at sufficiently large $\mu$. For example, at $\kappa=1.1$ to $1.2$ this can increase the efficiency by several times. This result can be arrived at also from (5), if $\kappa$ is taken to mean the quantity $W\tau_{rel}/n_1(J_m)$. A higher efficiency can be obtained in two ways. The first calls for the use of a selective resonator that suppresses lasing on transitions with values of $J$ close to $J_m$ and ensures lasing conditions on transitions between energy levels with large values of $J$. The second is to replace the generator by a generator-amplifier system, with the spectrum of the amplified signal at resonance with the transitions corresponding to larger values of $J$, greatly exceeding $J_m$. We note that the laser efficiency can increase with increasing $J$ only if the resonator $Q$ is high enough, when the decisive contribution to the population of the vibrational levels is made by the first term in (3). When $J$ is increased, the existence of a threshold value for the population inversion comes sooner or later into play and leads, according to (4) and (5), not to an increase but to a decrease of the efficiency. It follows therefore that in a real resonator there exists an optimal value of $J$ that ensures a maximum efficiency, which does not coincide with the value of $J_m$ corresponding to the maximum gain. We emphasize that on going to large $J$ the absolute population of the rotational levels decreases, and this leads to a decrease of the gain of the active medium. Therefore, and also owing to the rapid decrease of $n_v$ with increasing $v$, it is difficult to attain the gain needed to realize the considered possibilities of increasing the efficiency in all the $v \rightarrow v-1$ bands at large values of $J$. Actually, however, to increase the laser gain it is not necessary to have $J$ large in comparison with $J_m$ in all the radiating bands. It is seen from (3) that the decisive influence is exerted on the total population of the excited states by the value of the first factor, equal to $\exp(-2J(M+1)\Theta_r/T)$. Therefore at sufficiently large $J$ in the first band (or perhaps in the first few lowest bands), it is possible to use transitions with low values of $J$ in the higher $v \rightarrow v-1$ bands. This makes it possible to attain a sufficiently high gain in all the higher bands without appreciable additional energy losses. It should be noted finally that the increase of the efficiency on going to large values of $J$ becomes more strongly pronounced the larger the parameter $\Theta_r/T$.

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[1]For simplicity, the molecule is assumed linear, Generalization of the case of an arbitrary molecule entails no principal difficulty.