

Comment on “Efficient diode-pumped Yb:Gd₂SiO₅ laser” [Appl. Phys. Lett. 88, 221117 (2006)]

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Recently, a new laser material, Yb:Gd₂SiO₅, was reported^{1–3} in various respected and independent journals. According to the published data, this material shows strong absorption in the vicinity of 975 nm and almost zero emission at this wavelength; the emission spectrum is interpreted as the emission cross section. In this Comment I show that such an interpretation contradicts the second law of thermodynamics. Then, I suggest an example of the emission cross section, which is consistent with the absorption cross section reported.

Various optically pumped solid-state lasers can be described considering the kinetics of excitations, taking into account only two sets of levels, the ground state and the excited state. In scientific slang, each of these two sets of levels is called a “manifold” or “level.” Each of these levels may contain several quantum states of an active center, but the interaction with phonons causes quick transfer of excitation between various sites and various sublevels of active centers. The time scale of these transitions (say, picoseconds) is small compared to the lifetime of the excitation (from several microseconds to milliseconds). In this case, the spectrum of gain does not depend on the particular way of the excitation; only the temperature and the population of the excited level are important. In this case, the temperature of the medium determines the distribution of the excitation among the sites and the sublevels; this allows to use the concept of the effective cross sections.

The spectroscopic cross sections (which are the same for absorption and emission) are not usual in the papers about solid-state lasers, and the effective cross sections are referred simply as “cross sections.” With these cross sections, the gain at frequency ω can be expressed as follows:

$$g(\omega) = n_2\sigma_{\text{emission}}(\omega) - n_1\sigma_{\text{absorption}}(\omega), \quad (1)$$

where n_1 and n_2 are the population of the ground level and that of the excited level, and σ are the effective cross sections as functions of frequency. Equation (1) can be considered as the definition of the effective cross section. To my knowledge, there is no other way to define the effective cross sections for a laser medium; we have to assume a quick transition between sublevels, and only in this case do the spectra of luminescence and gain not depend on a specific way of excitation. The effective cross sections are a convenient concept; it allows to deal with an active center of a laser medium, as if it would be a two-level system.

The cross sections σ of emission and absorption are related:

$$\sigma_{\text{emission}}(\omega) = \sigma_{\text{absorption}}(\omega) \exp\left(\frac{\hbar\omega_0}{K_B T} - \frac{\hbar\omega}{K_B T}\right), \quad (2)$$

where K_B is the Boltzmann constant and T is temperature, and ω_0 is “zero-line” frequency. Equation (2) is called “the McCumber relation.” It can be deduced for a homogeneous (single-component) medium, considering the relative populations of sublevels.^{4,5}

According to the McCumber relation, the peaks of the absorption cross section correspond to the peaks of the emission cross section, and vice versa. The McCumber relation is broken for the spectra reported by Refs. 1–3: the maximum of absorption in the vicinity of the 975 nm wavelength corresponds to the gap of the emission. By assuming that the absorption cross sections are measured correctly, the spectra of emission of Yb:Gd₂SiO₅ by Refs. 1–3 cannot be interpreted as the emission cross section. Unfortunately, all three publications specify the ordinate axis as the emission cross section. These misprints may cause the confusing conclusion that the McCumber relation does not apply to the emission cross section in widely broadened laser media such as Yb:Gd₂SiO₅.^{6,7}

The usual deduction by Refs. 4 and 5 assumes that all the active centers are equal. Therefore, we cannot apply it automatically to the new medium with huge spectral broadening. Below, the McCumber relation is deduced from the gedanken experiment with a laser medium, placed into a box with ideally reflecting walls; we have no need to assume any homogeneity or any other specific structure of sublevels of the medium.

Let us consider the medium characterized by the effective cross sections $\sigma_{\text{absorption}}$ and σ_{emission} which, at a given temperature, are functions of frequency. Let us consider a wide box with reflecting walls, filled with such a medium. Then, the time derivative of the mean number N_m of photons in the m th mode of the box can be expressed with the differential equation

$$\begin{aligned} \frac{dN_m}{dt} = & (N_m + 1)n_2v(\omega_m)\sigma_{\text{emission}}(\omega_m) \\ & - N_m n_1 v(\omega_m)\sigma_{\text{absorption}}(\omega_m), \end{aligned} \quad (3)$$

where ω_m is frequency of the m th mode and $v(\omega)$ is group velocity of light at frequency ω in the medium. This equation can be completed with the conservation of the number of excitations,

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$$Vn_2 + \sum_m N_m = \text{const}, \quad (4)$$

where V is volume of the box. The system (3) and (4) may describe the beginning of the laser action, if initially some active centers were excited and there were no photons in the box. The same system describes also the steady state, which corresponds to the thermal equilibrium. In the steady state,

$$(N_m + 1)n_2v(\omega_m)\sigma_{\text{emission}}(\omega_m) = N_m n_1 v(\omega_m)\sigma_{\text{absorption}}(\omega_m). \quad (5)$$

On the other hand, the mean number of photons in the m th mode can be expressed using the Bose distribution,

$$N_m = \frac{1}{\exp(\hbar\omega_m/K_B T) - 1}. \quad (6)$$

The combination of (5) with (6) gives the ratio of the cross sections,

$$\frac{\sigma_{\text{absorption}}(\omega)}{\sigma_{\text{emission}}(\omega)} = \frac{n_2}{n_1} \exp\left(\frac{\hbar\omega_m}{K_B T}\right), \quad (7)$$

which is equivalent to the McCumber relation (2).

The steady-state ratio n_2/n_1 determines the zero-line frequency ω_0 . Usually, the energy difference between sublevels is larger than $k_B T$, and the zero line corresponds to the energy between the lowest sublevels in the upper and lower laser manifolds.

The McCumber relation is not a specific property of the homogeneously broadened media. It holds for any medium while the effective emission cross section has sense.

The fundamental meaning of the McCumber relation can be revealed from the gedanken experiment with two idealized boxes. Let us assume that for some exotic medium the ratio of the emission and absorption cross sections deviates from relation (7), at least for some range of temperatures, at least in some narrow interval of frequencies. Let us assume that there exist media satisfying the McCumber relation. These two assumptions will lead to a contradiction.

Let us consider a box, filled with the conventional medium, for which the McCumber relation is satisfied. [For example, Ytterbium doped yttrium aluminium garnet.⁸] We let another box be filled with some medium the cross sections of which claimed to deviate from the McCumber relation, at least in some spectral range. [For example, Yb:Gd₂SiO₅ (Refs. 1–3).] For simplicity, we assume that the cross section of emission of the “exotic” medium is smaller than that we would expect from the McCumber relation, as it takes place in Fig. 1 by Ref. 1, in the vicinity of some wavelength. Let us connect these two boxes with a spectral-selective window, which reflects most of the light but is transparent in the vicinity of this wavelength. (It should be transparent at wavelength $\lambda=975$ nm in the case of the cross sections by Refs. 1–3.) The sizes of the boxes can be adjusted in such a way that the probability of absorption of a photon of this wavelength in the first box is equal to that for the second box. However, due to the break of the McCumber relation, the probability of the emission is lower in the box with exotic medium. Therefore, the box with conventional medium emits more photons than the exotic box, and the exotic box becomes warmer. Such a break of the thermal equilibrium is sufficient for a perpetual motion of the second kind.

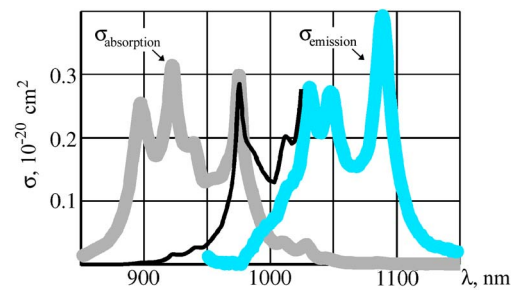


FIG. 1. (Color online) Thick curves: Cross-sections $\sigma_{\text{absorption}}$ and σ_{emission} by Ref. 1 vs wavelength λ . Thin black curve: σ_{emission} reconstructed by the McCumber relation (2) from the curve for $\sigma_{\text{absorption}}$.

The deduction of the above confirms the general statement by Ref. 4 about the McCumber relation: “Although we neglect the nonradiative transitions in our analysis, we expect our result to apply to all systems, for which the time required for thermal equilibrium within each metastable set is short compared to the lifetimes of the different sets.”

While the concept of the effective cross sections can be applied to a laser medium, the McCumber relation (2) holds. The violation of the McCumber relation would break the second law of thermodynamics. Therefore, the spectra of emission by Refs. 1–3 cannot be interpreted as emission cross section, as they are labeled in the figures.

The simplest explanation of the nonphysical result by Refs. 1–3 is reabsorption. The deviation from the McCumber relation is stronger for the highest concentration. Perhaps, the photons at $\lambda=975$ nm are emitted by Yb:Gd₂SiO₅ so often, as it is required by the McCumber relations, but absorbed in a thick sample. Such an artifact should not affect much the absorption cross section; in this case, the absorption cross section by Refs. 1–3 may be correct; then the emission cross section can be evaluated using the McCumber relation (2). In Fig. 1, the reconstructed cross section is shown with a thin black line. Values of the temperature $T=271+14$ K and the zero-line frequency $\omega_0=2\pi c/(975 \text{ nm})$ were used. For comparison, the thick lines show the cross sections of emission and absorption by Ref. 1; these data look very similar to those by Refs. 2 and 3. In Ref. 9, the reconstruction with such rough estimate of the zero-line frequency is called “Exact McCumber”; this reconstruction slightly overestimates the emission cross-section at large values of λ . Therefore I do not extend the reconstructed curve to large values of wavelength; in addition, at the right hand side of the graphic, the relative error of measurement of $\sigma_{\text{absorption}}$ is high. The adjustment of the zero-line frequency may improve the estimate, but such analysis falls outside the scope of a short comment. The authors of Refs. 1–3 have all the data to make such adjustment in their reply. The replacement of the curve for σ_{emission} by the reconstruction from the $\sigma_{\text{absorption}}$ is minimal correction of the data by Refs. 1–3, which makes them consistent with the second law of thermodynamics.

I conclude, the McCumber relation (2) applies not only to the homogeneous (uni-component) laser media but to any media with fast transitions within a metastable group of sublevels. If the quantity of the emission cross section has sense for a given medium, then, for this medium, the McCumber relation holds. By assuming the correct absorption cross section by Refs. 1–3, the emission cross section of Yb:Gd₂SiO₅ can be reconstructed (black curve in Fig. 1) with the

McCumber relation. I hope that the corrected cross sections are suitable for the design of the Yb:Gd₂SiO₅ lasers, and I should greatly appreciate an experimental confirmation of such reconstruction.

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