Broadband laser materials and the McCumber relation

Dmitrii Kouznetsov

Institute for Laser Science, University of Electro-Communications, 1-5-1 Chofu, Tokyo 182-8585, Japan

The McCumber relation can be deduced without assuming that all active centers have the same structure of sublevels. The range of validity of the McCumber relation is the same as that of the effective emission cross-section.

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The concept of effective cross-sections allow to treat the laser medium as a two-level system. Such a concept is widely used; often, these effective cross-sections are called simply "cross-sections". The McCumber relation[1,2] expresses the emission cross-seccion $\sigma_{\rm e}(\omega)$ in terms of the absorption cross-section $\sigma_{\rm a}(\omega)$:

$$\sigma_{\rm e}(\omega) \;=\; \sigma_{\rm a}(\omega)\,\exp\!\left(\hbar\frac{\omega_{\rm z}-\omega}{k_{\rm B}T}\right), \qquad (1)$$

where T is temperature, $k_{\rm B}$ is Boltzmann constant and $\omega_{\rm z}$ is zero-line frequency, at which the emission and absorption cross-sections are equal. Equation (1) is validated for various media[1−5].

The original deduction of the McCumber relation[1], as well as the adaptation in the textbook[2] assume, that all active centers are equal. It cannot be applied "as is" to the broadband laser materials with different "sites" of the active centers. This allowed the interpretation of results for $Yb : Gd_2SiO_5$ by cites[6−8] as an indication, that the effective cross-sections of broad-band composite materials have no need to satisfy the McCumber relation: the peak of $\sigma_{\rm a}$ at wavelength 950 nm corresponds to the gap of $\sigma_{\rm e}$. However, a medium with such effective cross-sections would be good not only for an efficient laser, but also for a Perpetual Motion of Second Kind[9,10]. The correction of the emission cross-section[9,10] was suggested and confirmed[11]. In order to avoid such confusions, the deduction of the McCumber relation should be generalized. After the presentation[9], I was asked for the general deduction of the McCumber relation as a substitute of the speculation[10] about the gedanken experiment with perpetual motion. Below, such a deduction is suggested.

In this paper, the generalization of the deduction of the McCumber relation is suggested. It is shown that the McCumber relation follows from the fundamental properties of the Einstein coefficients[12−15], and applies to any material with fast transitions within each of two sets of levels and relatively slow transitions between these two sets.

The sketch of sublevels of active centers is shown in Fig. 1. Consider two subsets of quantum states: level 1 and level 2. Assume slow optical transitions from level 1 to level 2. (This property makes the medium suitable for a laser action.) Assume quick transfer of energy between neighbors, which leads to the fast thermalization within each of laser levels. Then, the refractive index[16] and gain[3] are determined by the populations n_1 and n_2 of the laser levels. In this case, and only in this case, the effective cross-sections $\sigma_{\rm a}(\omega)$ and $\sigma_{\rm e}(\omega)$ of absorption and emission have sense.

Use of effective cross-sections assumes the thermalization of quantum states within each of laser levels. However, the population of the laser levels can be far from a thermal state, allowing the lasing. The gain can be expressed as

$$g(\omega) = n_2\,\sigma_{\rm e}(\omega) - n_1\,\sigma_{\rm a}(\omega), \qquad (2)$$

where n_1 and n_2 are population of lower and upper laser levels.

Keeping the consideration phenomenological, the spontaneous emission can be characterized with the Einstein coefficients[12−15]; the rate of emission of spontaneous photons at frequency ω can be expressed as $a(\omega)n_2$, where $a(\omega)$ is the probability of spontaneous emission by a random active center per time per frequency, assuming that it is excited. $a(\omega)$ is equivalent of the Einstein coefficient A_{21}. Notation a is used here to avoid confusion with the Einstein coefficient A_{21}, which has no established expression (see notes at Table 7.7 of Ref. [13]); not only value, but even dimensions of the Einstein coefficients depend on scale we use: frequencies or wavelengths.

The decay rate $1/\tau$ of the excited level can be expressed in terms of the coefficient a:

$$\frac{1}{\tau} = \int_0^\infty a(\omega)\;{\rm d}\omega. \qquad (3)$$

The cross-section $\sigma_{\rm a}(\omega)$ and $\sigma_{\rm e}(\omega)$ and the coefficient $a(\omega)$ do not depend on the populations n_1 and n_2 of the active medium and the density $D(\omega)$ of photons of frequency ω. In this approximation, the properties of the medium are determined by 3 functions $\sigma_{\rm a}(\omega)$, $\sigma_{\rm e}(\omega)$ and $a(\omega)$, and we have no need to consider nonlinear processes[17] which produce a given population. Gain and refraction index as functions of frequency are determined by the populations n_1 and n_2.

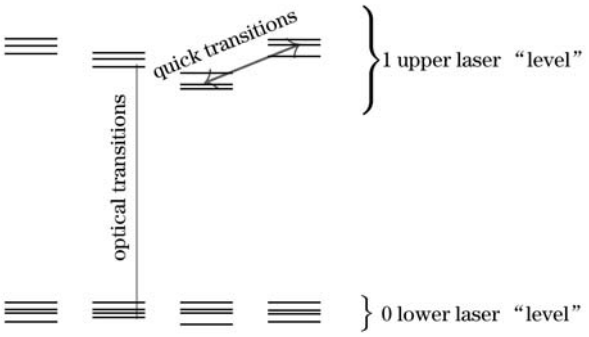

Fig. 1. Sublevels of active centers.

The functions $a(\omega)$, $\sigma_{\rm a}(\omega)$ and $\sigma_{\rm e}(\omega)$ are equivalent of the Einstein coefficients, but have an advantage: their values do not depend on system of notations. In the following, the consideration of relations between Einstein coefficients[12−15] is rewritten, taking into account sublevels (Fig. 1).

Functions $\sigma_{\rm a}(\omega)$, $\sigma_{\rm e}(\omega)$ and $a(\omega)$ of frequency ω are related, as the Einstein coefficients are. These relations can be found from the principle of detailed balance. Although the expression (2) is good for a non-equilibrium medium, it is valid also at the thermal equilibrium, when the spectral rate of emission (both spontaneous and stimulated) of photons at any frequency ω is equal to that of absorption.

Consider a thermal state. Let $v(\omega)$ be group velocity of light in the medium. The product $n_2\sigma_{\rm e}(\omega)v(\omega)D(\omega)$ is the spectral rate of stimulated emission, and $n_1\sigma_{\rm a}(\omega)v(\omega)D(\omega)$ is that of absorption; $a(\omega)n_2$ is the spectral rate of spontaneous emission. (Note that in this approximation, there is no such thing as a "spontaneous absorption".) The balance of photons gives

$$n_2\sigma_{\rm e}(\omega)v(\omega)D(\omega)+n_2a(\omega)=n_1\sigma_{\rm a}(\omega)v(\omega)D(\omega). \quad (4)$$

Rewrite it as

$$D(\omega)=\frac{\dfrac{a(\omega)}{\sigma_{\rm e}(\omega)v(\omega)}}{\dfrac{n_1}{n_2}\dfrac{\sigma_{\rm a}(\omega)}{\sigma_{\rm e}(\omega)}-1} \quad (5)$$

The thermal distribution of density of photons follows from blackbody radiation[13]:

$$D(\omega)\ =\ \frac{\dfrac{1}{\pi^2}\dfrac{\omega^2}{c^3}}{\exp\left(\dfrac{\hbar\omega}{k_{\rm B}T}\right)-1}. \quad (6)$$

Both Eqs. (5) and (6) hold for all frequencies ω. For the case of idealized two-level active centers, $\sigma_{\rm a}(\omega)=\sigma_{\rm e}(\omega)$, and $n_1/n_2=\exp\left(\frac{\hbar\omega}{k_{\rm B}T}\right)$, which leads to the relation between the probability of spontaneous emission $a(\omega)$ and the emission cross-section $\sigma_{\rm e}(\omega)$[13]. (We keep the term "probability of emission" for the quantity $a(\omega){\rm d}\omega{\rm d}t$, which is probability of emission of a photon within small spectral interval $(\omega,\omega+{\rm d}\omega)$ during a short time interval $(t,t+{\rm d}t)$, assuming that at time t the atom is excited.) The relation (6) is fundamental property of spontaneous and stimulated emission, and, perhaps, the only way to prohibit a spontaneous break of the thermal equilibrium in the thermal state of excitations and photons. For each site number s, for each sublevel number j, the partial spectral emission probability $a_{s,j}(\omega)$ can be expressed from consideration of idealized two-level atoms [12]:

$$a_{s,j}(\omega)=\sigma_{s,j}(\omega)\frac{\omega^2v(\omega)}{\pi^2c^3}. \quad (7)$$

Neglecting the cooperative coherent effects, the emission is additive: for any concentration q_s of sites and for any partial population $n_{s,j}$ of sublevels, the same proportionality between a and $\sigma_{\rm e}$ holds for the effective cross-sections:

$$\frac{a(\omega)}{\sigma_{\rm e}(\omega)}=\frac{\omega^2v(\omega)}{\pi^2c^3}. \quad (8)$$

Then, the comparison of Eqs. (5) and (6) gives

$$\frac{n_1}{n_2}\frac{\sigma_{\rm a}(\omega)}{\sigma_{\rm e}(\omega)}=\exp\left(\frac{\hbar\omega}{k_{\rm B}T}\right). \quad (9)$$

This relation is equivalent of the McCumber relation (1), if we define the zero-line frequency $\omega_{\rm z}$ as solution of equation $\left(\frac{n_2}{n_1}\right)_T=\exp\left(\frac{\hbar\omega_{\rm z}}{k_{\rm B}T}\right)$; the subscript "$T$" indicates that the ratio of populations evaluated in the thermal state with temperature T. The zero-line frequency can be expressed as

$$\omega_{\rm z}=\frac{k_{\rm B}T}{\hbar}\ln\left(\frac{n_1}{n_2}\right)_T. \quad (10)$$

We see, no specific property of sublevels of active medium is required to keep the McCumber relation. It follows from the quick transfer of energy among excited laser levels and among lower laser levels. The McCumber relation (1) has the same range of validity, as the concept of the emission cross-section itself.

The zero-line frequency is determined by Eq. (10) in terms of ratio $(n_2/n_1)_T$ of populations of levels at given thermal state with temperature T. In general, $\omega_{\rm z}$ depends on the temperature. This dependence can be expressed explicitly in terms of energies of sublevels.

Consider first the homogeneous medium, and numerate the sublevels as it is shown in Fig. 2. Let U be total number of sublevels in the system. Let the variable j numerate these sublevels. Let first L sublevels be in the lower level, they correspond to values $0\le j\le L-1$. The following $U-L$ sublevels belong to the upper level, they correspond to $L\le j\le U-1$. Let ε_j be energy of the jth sublevel. Then, the thermal-equilibrium ratio of populations

$$\left(\frac{n_2}{n_1}\right)_T=\frac{\displaystyle\sum_{j=L}^{U-1}\exp\left(-\frac{\hbar\varepsilon_j}{K_{\rm B}T}\right)}{\displaystyle\sum_{j=0}^{L-1}\exp\left(-\frac{\hbar\varepsilon_j}{K_{\rm B}T}\right)}. \quad (11)$$

For a medium with different active sites (Fig. 1), let s numerate the kinds of a site. Let q_s be the concentration of the sth site, and $\varepsilon_{q,j}$ be the energy of the jth sublevel at the sth site. Then,

$$\left(\frac{n_1}{n_0}\right)_T=\frac{\displaystyle\sum_s q_s\frac{\sum_{j=L}^{U-1}\exp\left(-\frac{\hbar\varepsilon_{s,j}}{K_{\rm B}T}\right)}{\sum_{j=0}^{U-1}\exp\left(-\frac{\hbar\varepsilon_{s,j}}{K_{\rm B}T}\right)}}{\displaystyle\sum_s q_s\frac{\sum_{j=0}^{L-1}\exp\left(-\frac{\hbar\varepsilon_{s,j}}{K_{\rm B}T}\right)}{\sum_{j=0}^{U-1}\exp\left(-\frac{\hbar\varepsilon_{s,j}}{K_{\rm B}T}\right)}}. \quad (12)$$

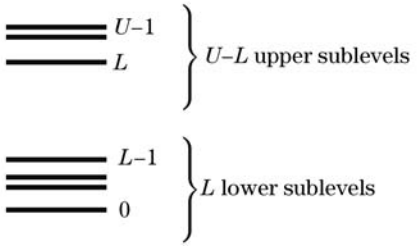

Fig. 2. Scheme of numeration of sublevels.

At small temperatures, $\frac{\hbar\varepsilon_{q,L+1} - \hbar\varepsilon_{q,L}}{k_{\rm B}T} \gg 1$, $\frac{\hbar\varepsilon_{q,1} - \hbar\varepsilon_{q,0}}{k_{\rm B}T} \gg 1$, and the only zeroth term is important in the summation. It is a typical case for the Yb-doped laser materials, when the zero-line frequency corresponds to the transition between the lowest sublevels.

The use of the formal expression (11) and, especially, (12) requires the knowledge of the energy of sublevels. It may be practical to determine the emission cross-section from the spectrum of the spontaneous emission (which is easier to measure) using Eq. (8). Then,

$$\sigma_{\rm e}(\omega) = \frac{\pi^2 c^3}{\omega^2 v(\omega)} a(\omega). \tag{13}$$

The integral of $\sigma_{\rm e}(\omega)$ can be checked using Eq. (3), while the lifetime τ is known. Then, the zero-line can be determined by comparing the ratio of the cross-sections to the exponential in the right-hand side of Eq. (9). The deviation of the right-hand side of the expression

$$\left(\frac{n_1}{n_2}\right)_T = \frac{\sigma_{\rm e}(\omega)}{\sigma_{\rm a}(\omega)} \exp\left(\frac{\hbar\omega}{k_{\rm B}T}\right) \tag{14}$$

from a constant is a measure of the error of a description of a process in terms of the effective emission cross-section. The strong deviation[6−10] may indicate that the effective emission cross-section $\sigma_{\rm e}(\omega)$ has no sense, and more detailed kinetic of excitations of various sites (or may be even subleveles) should be taken into account[18].

The McCumber relation (1) follows from the assumption of fast redistribution of energy among laser sublevels. Only in this case, the effective cross-sections can be used to characterize the laser medium. Within this concept, the inaccuracy of the McCumber relation at the tails of the spectral lines discussed by Ref. [19] should be attributed to the limitation of the effective cross-sections as physical quantities.

The deduction suggested applies to broadband materials with different sites. This approximation will be broken at low concentration of the active centers, as well as at the excitation with very strong and short pulses. In both cases, the different sites interact with electromagnetic field faster than they exchange the energy. In any of these cases, the medium cannot be characterized with the single-valued emission cross-section function $\sigma_{\rm e}(\omega)$; the effective cross-sections should be defined for each site, and the kinetic of the transfer of the excitations should be considered.

The effective emission cross-section and the McCumber relation have the same range of validity. The deviation from a constant of the right-hand side of the estimate (14) for the steady-state ratio of populations characterizes the error of measurement of the effective cross-sections.

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