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Comments on "Study of the Complex Atomic Susceptibility of Erbium-Doped Fiber Amplifiers"

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Abstract—**This comment aims to report and correct a mistake in the paper by Desurvire (*J. Lightw. Technol.*, vol. 8, no. 10, pp. 1517–1527, Oct. 1990) with regard to the expression of the complex susceptibility of quasi-three-level laser systems. The derived susceptibility formula contradicts the assumption of thermal equilibrium between sublevels within each manifold and leads to the wrong conclusion that the real and imaginary parts of the susceptibility of saturated materials do not verify the Kramers–Kronig relations (KKR). The corrected formula for the susceptibility is hereby proposed. The poles of the susceptibility obtained with the new formula are all located in the upper half plane of the complex plane, in accordance with the principle of causality. Hence, the KKR should be valid in the saturated regime.**

Index Terms—**Atomic susceptibility, Kramers–Kronig relations (KKR), saturation broadening.**

I. INTRODUCTION

In the above paper [1], an extensive study of the complex atomic susceptibility of erbium-doped fiber amplifiers was reported. Although the above paper is not recent, it is still regularly cited in the literature for the modeling of the complex susceptibility of glassy hosts doped with rare-earth ions. Hence, it appears worthwhile to point out the mistakes in the above paper, particularly when they lead to wrong conclusions about the domain of validity of Kramers–Kronig relations (KKR) in saturated media. This comment is also motivated by the fact that KKR have been recently used to relate fluctuations of the saturating pump or the signal beams in fiber lasers to that of the refractive index at the signal frequency, fluctuations that have been proposed as a possible mechanism of phase-locking of the fiber laser array at high power [2], [3] . Moreover, modification of the refractive index in the saturated regime is also important for the accurate dispersion control of chirped-pulse amplification and compression in fibers [4].

The purpose of this comment is twofold. First, we would like to expose and correct what appears to be a wrong expression of the susceptibility formula in the paper by Desurvire. Second, we would like to point out and contest the statement whereby the KKR cannot be used to calculate the real and imaginary parts of the susceptibility in saturated systems (cf. [1, p. 1522, second column]).

This comment is organized as follows. First, we recall the formula for the complex atomic susceptibility of a two-level system and discuss the physical meaning of saturation broadening. Then, the wrong formulas in the above paper of the susceptibility of a quasi-three-level system are pointed out, and the corrected formula for the complex susceptibility is provided. Finally, the poles of the susceptibility obtained with the updated formulas are shown to be all located in the upper

half plane of the complex plane, in accordance with the principle of causality. We show that this is a sufficient condition to validate the KKR, including that in the saturated regime.

II. SATURATION OF A SIMPLE TWO-LEVEL SYSTEM

Reference [1, eq. (12)] for the complex atomic susceptibility of a two-level atomic system at the frequency of the saturating field ω is given as follows:

$$\chi_E(\omega) = (\overline{N}_2 - \overline{N}_1)\frac{2\mu^2}{\varepsilon_0\hbar\Delta\omega}\frac{2\frac{\omega-\omega_{21}}{\Delta\omega} + i}{1 + 4\left(\frac{\omega-\omega_{21}}{\Delta\omega}\right)^2 + \frac{2\mu^2 E_0^2 T}{\hbar^2\Delta\omega}}$$

where $\overline{N}_2 - \overline{N}_1$ is the population difference at zero saturating field. For the complete description of the notation, please refer to [1]. Reference [1, eq. (12)] expresses the saturation broadening for a homogeneous transition in a two-level system. Although this expression is formally correct, it can be misleading. Perhaps, the following expression is a more suitable way to express [1, eq. (12)]:

$$\chi_E(\omega) = (\overline{N}_2 - \overline{N}_1)\frac{2\mu^2}{\varepsilon_0\hbar\Delta\omega}\frac{1 + 4\left(\frac{\omega-\omega_{21}}{\Delta\omega}\right)^2}{1 + 4\left(\frac{\omega-\omega_{21}}{\Delta\omega}\right)^2 + \frac{2\mu^2 E_0^2 T}{\hbar^2\Delta\omega}}g(\omega) \quad (1a)$$

where

$$g(\omega) = \frac{2\frac{\omega-\omega_{21}}{\Delta\omega} + i}{1 + 4\left(\frac{\omega-\omega_{21}}{\Delta\omega}\right)^2}$$

is the linewidth function, which is independent of the saturating field. Equation (1a) can be written as

$$\chi_E(\omega) = \left(\overline{N}_2^{\text{sat}} - \overline{N}_1^{\text{sat}}\right)_{\text{sat}}\frac{2\mu^2}{\varepsilon_0\hbar\Delta\omega}g(\omega) \quad (1b)$$

where

$$\overline{N}_2^{\text{sat}} - \overline{N}_1^{\text{sat}} = (\overline{N}_2 - \overline{N}_1)\frac{1 + 4\left(\frac{\omega-\omega_{21}}{\Delta\omega}\right)^2}{1 + 4\left(\frac{\omega-\omega_{21}}{\Delta\omega}\right)^2 + \frac{2\mu^2 E_0^2 T}{\hbar^2\Delta\omega}} \quad (2)$$

is the saturated population inversion due to the saturating field E_0 at frequency ω . Equation (1b) allows the influence of the saturating field, which acts on the population inversion, to be separated from that of the fictitious weak probe, which is used as an integrating variable in the KKR.

Remarks:

1) Equation (1b) explicitly shows that the field $E_0(\omega)$ saturates the population difference but does not actually cause any broadening of the transition. That is to say, if we probe the transition with a "weak" probe $E_p(\omega')$, then the linewidth remains unchanged, no matter how large is the value of the field E_0.

2) If the frequency of the saturating field $E_0(\omega)$ is scanned through the absorption line, then apparent broadening occurs because the saturation is more effective close to the resonance than off-resonance.

3) Actual broadening caused by the saturating field occurs if the rate of stimulated emission is fast enough to compete with the rate of

decoherence, which determines the linewidth $g(\omega)$ of the transition. The decoherence time is on the order of the picosecond range for many solid-state materials, including the erbium–glass system, while the effective lifetime on the upper manifold when stimulated emission is dominant is on the order of $h\nu/(I\sigma)$ at strong field, where σ is the effective emission cross section at frequency ω. Certainly, $h\nu/(I\sigma)$ never actually approaches the picosecond time scale; thus, the aforementioned phenomenon never happens in practice. Nonetheless, it must not happen for the hypothesis of thermal equilibrium within the manifold to be valid. This hypothesis is required in order to use the Boltzmann distribution for the population of sublevels.

We can also write (2) as

$$\overline{N}_2^{\text{sat}} - \overline{N}_1^{\text{sat}} = (\overline{N}_2 - \overline{N}_1)\frac{1}{1 + I/I_s(\omega)} \tag{3}$$

where $I = (1/2)\varepsilon_0 cn E_0^2$, and $I_s(\omega)$ is the frequency-dependent saturation parameter, which depends on the effective cross sections and can easily be measured experimentally. For the complete formulas including both the saturations of the pump and the signal, the reader is referred to [5, Sec. 3.A]. Hence, [1, eq. (12)] does not actually express the broadening of a homogeneous transition but rather the frequency-dependent saturation of the transition.

III. SATURATION OF A TWO-MANIFOLD SYSTEM

Desurvire considers a two-manifold system where thermal equilibrium is assumed to take place within the sublevels of each manifold with respective populations $\overline{N}_1$ and $\overline{N}_2$. According to the aforementioned analysis, it is easy to see that [1, eq. (18)] is wrong. Reference [1, eq. (18)] is given as

$$\chi_E(\omega) = \frac{nc}{\lambda} \sum_{k=1}^{g_2} \sum_{j=1}^{g_1} \sigma_{jk}^{\text{peak}} (p_{2k}\overline{N}_2 - p_{1j}\overline{N}_1)$$
$$\times \frac{2\frac{\omega - \omega_{jk}}{\Delta\omega} + i}{1 + 4\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)^2 + \beta_{jk}}$$

where the population of each of the sublevels p_{2k} and p_{1j} follows the Boltzmann law. We can express [1, eq. (18)] as

$$\chi_E(\omega) = \frac{nc}{\lambda} \sum_{k=1}^{g_2} \sum_{j=1}^{g_1} \sigma_{jk}^{\text{peak}} (p_{2k}\overline{N}_2 - p_{1j}\overline{N}_1)$$
$$\times \frac{1 + 4\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)^2}{1 + 4\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)^2 + \beta_{jk}} g_{jk}(\omega)$$

where

$$g_{jk}(\omega) = \frac{2\frac{\omega - \omega_{jk}}{\Delta\omega} + i}{1 + 4\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)^2} \tag{4}$$

is the linewidth function of the jk pair of sublevels, $\Delta\omega_{jk} = \Delta\omega$ is assumed to be the same as that in [1] for all transitions, and β_{jk} is the saturation parameter given by identification with [1, eq. (17)]. Reference [1, eq. (18)] contradicts the hypothesis of thermal equilibrium

inside each manifold, because the population difference for each pair of sublevels is given by

$$(p_{2k}\overline{N}_2 - p_{1j}\overline{N}_1)\frac{1 + 4\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)^2}{1 + 4\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)^2 + \beta_{jk}}$$

which implies a different saturation of each sublevel. Assuming thermal equilibrium, the correct expression instead of [1, eq. (18)] should be

$$\chi_E(\omega) = \frac{nc}{\lambda} \sum_{k=1}^{g_2} \sum_{j=1}^{g_1} \sigma_{jk}^{\text{peak}} \left(p_{2k}\overline{N}_2^{\text{sat}} - p_{1j}\overline{N}_1^{\text{sat}} \right) g_{jk}(\omega) \tag{5}$$

where $\overline{N}_2^{\text{sat}} - \overline{N}_1^{\text{sat}}$ is given by (2) and (3), and $I_s(\omega)$ is the effective saturation intensity, which can be deduced from the knowledge of the effective cross section and radiative lifetime of the medium. In fact, from the expression for $g_{jk}(\omega)$, we find that some transitions do have a stronger saturation than others as a result of interaction with the strong field at ω, i.e., $I(\omega)$, because their resonance frequency is closer to ω. However, this effect is canceled by the fast thermalization of sublevels, so that the preferred depopulation of one sublevel is rapidly spread to all other sublevels to maintain the Boltzmann distribution. The latter is so fast compared to other time scales that the relative populations p_{2k} and p_{1j} are given by the Boltzmann distribution regardless of the particular value of the frequency of the saturating field. As a consequence, [1, eqs. (34) and (35)], which are not shown here, are also wrong for the same reason that they contradict the assumption of thermalization within sublevels inside each manifold.

IV. POLES, CAUSALITY, AND KKR

The poles of (5) are that of $g_{jk}(\omega)$, which can be written as

$$g_{jk}(\omega) = \frac{\left[1 - 2i\frac{\omega - \omega_{jk}}{\Delta\omega}\right]i}{\left[1 + 2i\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)\right]\left[1 - 2i\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)\right]}$$
$$= \frac{i}{\left[1 + 2i\left(\frac{\omega - \omega_{jk}}{\Delta\omega}\right)\right]} = \frac{\Delta\omega/2}{\omega - (\omega_{jk} + i\Delta\omega/2)}.$$

The pole of g_{jk} is $\omega_p = \omega_{jk} + i(\Delta\omega/2)$, which is in the upper complex plane for any physical value of ω_{jk} and $\Delta\omega$. Hence, all the poles are located in the upper complex plane. According to Titchmarsh's theorem [6], if the function $\chi_E(z)$ is analytic in the complex $z = x + iy$ plane for $y < 0$, if $\chi_E(z)$ approaches $\chi_E(x)$ almost everywhere as y approaches 0, and if there exist a value of $K > 0$ such that $\int_0^\infty |\chi_E(x + iy)|^2 dx < K$, for $y < 0$, then two statements apply.
1) The Fourier transform of $\chi_E(\omega)$ is 0 for $t < 0$.
2) The real and imaginary parts of $\chi_E(\omega)$ are Hilbert transforms of each other.

Statement 1 is the expression for the causality principle, whereas statement 2 implies that the KKR are valid. Therefore, we conclude that KKR can be used in the case of both saturated and nonsaturated transitions. The KKR is given as

$$\chi'_E(\omega) = -\frac{2}{\pi} P \int_0^\infty \frac{\omega' \chi''_E(\omega')}{\omega'^2 - \omega^2} d\omega' \tag{6a}$$

$$\chi_E''(\omega) = \frac{2\omega}{\pi} P \int_0^\infty \frac{\chi_E'(\omega')}{\omega'^2 - \omega^2} d\omega' \qquad (6b)$$

where $\chi_E \equiv \chi_E' - i\chi_E''$, and P stands for the principal value.

In (6), the real part of the susceptibility, i.e., χ_E', is interpreted as the change of the real part of the susceptibility due to the gain profile $g(\omega)$, which is related to the imaginary part of the susceptibility χ_E'' by $g(\omega) = (\omega \chi_E''(\omega))/cn_0^2$, where n_0 is the refractive index of the material with no gain, and χ_E'' is the imaginary part of the susceptibility. The gain distribution $g(\omega)$ in a system of atoms saturated by the signal (and eventually pump field) at fixed frequency is determined with a weak probe at frequency ω, as shown in [2] and [3]. The change of refractive index resulting from the modified small-signal gain distribution was correctly computed by using the KKR.

V. CONCLUSION

In summary, we have shown that the expression for the complex susceptibility proposed by Desurvire [1, eq. (18)] is wrong because it contradicts the assumption of thermalization inside each manifold. The correct expression, i.e., (5), is proposed instead for the complex susceptibility of a two-manifold system subjected to a saturating field with fast thermalization between sublevels. Moreover, [1, eqs. (34) and (35)], which follow from [1, eq. (18)], lead to a wrong conclusion concerning the validity of KKR in the saturated regime. We showed that the KKR can be used to calculate the real part of the complex susceptibility from the imaginary part, even under saturating conditions, by using the KKR and the small-signal gain distribution. Therefore, the KKR should be valid in the saturated regime.

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