β factor in a random laser

Gijs van Soest and Ad Lagendijk*
Van der Waals–Zeeman Instituut, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands
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We develop a definition for the β factor, the fraction of spontaneous emission that seeds the laser process, for a random laser. With the wavelength-dependence of the gain (and potentially scattering) being the only possible criterion in the competition between gain and loss, our concept of β is based on the spectral properties of the spontaneous emission and laser light. We find $\beta \approx 0.1$. We discuss the apparent similarities and differences between the β for a cavity and a random laser.

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I. INTRODUCTION

Random lasers are strongly scattering media with optical gain. These systems have many features in common with more conventional lasers based on an optical gain medium enclosed in a cavity with two mirrors to enhance stimulated emission. For example, a threshold for lasing action and frequency narrowing has been observed in random lasers based on multiply scattering colloidal dispersions in dyes [1]. Evidently, the optical properties of random lasers are quite different from that of conventional lasers: the propagation of pump and fluorescence light is diffusive, and—in absence of well-defined cavity modes—there is no "preferred direction" in feedback and loss processes.

Spontaneous emission is usually the seed for lasing, both in cavity and in random lasers. However, not all spontaneous emission participates in the laser process. The fraction of spontaneous radiation that does contribute to lasing is called β. In the science of conventional lasers this parameter is of great interest because of the promise of a "thresholdless laser" with $\beta=1$, in which all spontaneous emission is radiated into the lasing mode [2,3].

The "sharpness" of the laser threshold is governed by the value of β. Solving the laser rate equations with $\beta=0$ yields a sharp bend in the field energy density W as a function of pump rate r, a discontinuity in the derivative at the threshold r_{th}. Below threshold $W=0$ and above $W \propto r-r_{\mathrm{th}}$. In the other limit, $\beta=1$, $W \propto r$. For $0<\beta<1$ there is a threshold, which becomes less sharp as β gets larger [4].

Random lasers have been described until now without β [5], implicitly assuming it to be unity, yet the observation of a nonzero threshold does clearly necessitate a $\beta<1$. A reliable numerical value of β is indispensable for a model describing the response of a random laser to an applied pump pulse [6].

II. SPONTANEOUS EMISSION SEEDING IN CAVITY AND RANDOM LASERS

In a cavity laser, light that is emitted outside a resonant mode of the cavity (outside being either of the wrong direction or the wrong wavelength) does not stimulate further emission and leaves the cavity without contributing to the laser field. Hence, an estimate for β involves geometric parameters such as the acceptance solid angle of the lasing modes or the mode volume, as well as frequency [7]. As is illustrated in Fig. 1, β is the overlap in wave vector $\mathbf{k}$ between spontaneous emission and laser mode.

These geometric restrictions do not apply to the random case, because of the lack of direction in the feedback mechanism, multiple scattering. The only selection criterion is the spectral dependence of the gain. Compared to the spontaneous emission the spectrum narrows above threshold around the maximum of the net gain of the medium. The exponential growth of the emitted intensity with gain coefficient $\kappa_g(\lambda)$ is responsible for the narrowing. Typical (neat dye) spontaneous emission and (high pump fluence) random laser spectra are shown in Fig. 2, normalized to their respective maxima. Since spontaneous emission of a wavelength outside the narrowed spectrum cannot contribute to the laser process, we use the overlap between below- and above-threshold emission spectra for a definition of β.

Evidently, the gain narrowing due to the wavelength dependence of $\kappa_g(\lambda)$ does not cause more light to be emitted by the system above threshold, it is just spectrally redistributed. The wavelength variation of the gain of the amplifying

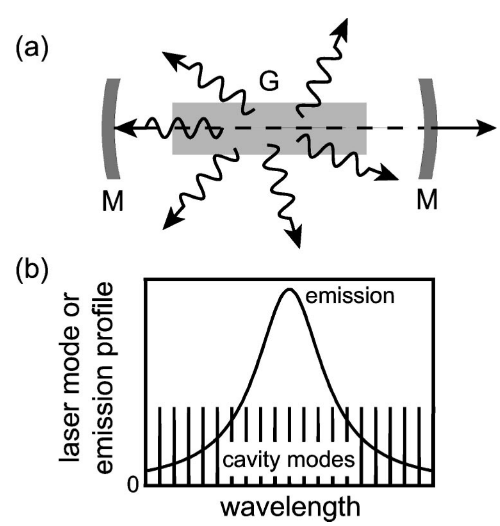

FIG. 1. Origin of β in a cavity laser. Spontaneous emission, originating from the gain medium G that is to contribute to lasing has to be in the correct mode of the cavity formed by mirrors M, i.e., there has to be overlap in direction (a) and in frequency (b).

*Corresponding author. Email address: adlag@phys.uva.nl

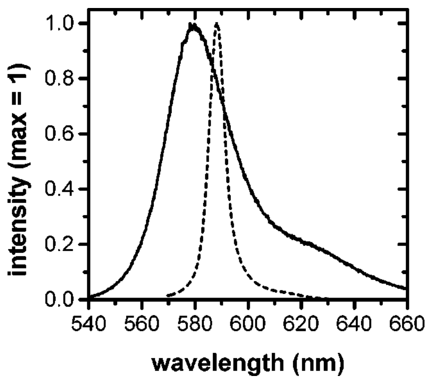

FIG. 2. Spectra of spontaneous emission [solid line; neat 0.1 mM solution of sulforhodamine B in methanol under low intensity cw excitation from an Ar^{+} laser (514.5 nm)] and of random laser emission well above threshold [dashed line; 1 mM sulforhodamine B in methanol with TiO_2 coloidal scatterers [8], the transport mean free path $\ell = 10$ μm, excited with a frequency-doubled Q-switched Nd:YAG (Nd:yttrium aluminum garnet) laser (532 nm)]. The "laser" spectrum is considerably narrower than the spontaneous emission source it originates from.

medium is the only selection mechanism in a random laser that distinguishes "laser" light from spontaneous emission. This means that the bend at the laser threshold in the curve of emitted intensity vs pump power is only observed in the frequency range near the maximum, and should not be seen in the spectrally integrated intensity.

In a cavity laser, the threshold can be observed in the total output intensity of the laser mode, because there is an additional selection mechanism for lasing that is often much more restrictive than the spectral dependence of the gain, the mode profile of the cavity. Only light radiated in the "right" solid angle, i.e., subtended by the lasing mode, contributes, and the dominance of stimulated transitions above the laser threshold causes the abrupt change in behavior. If the radiation from a cavity laser would be collected in all directions, the threshold would not be observed, because all the radiation, stimulated (laser) and spontaneous (nonlaser) light, is detected, analogous to a measurement of spectrally integrated emission from a random laser.

III. QUANTITATIVE CONSTRUCTION OF β IN A RANDOM LASER

The transport of laser light in a random laser is described in the simplest case by the following equations:

$$\frac{\partial W_\ell}{\partial t} = D\nabla_z^2 W_\ell + \sigma_e c n_1 W_\ell + \frac{\beta}{\tau} n_1, \tag{1}$$

$$\frac{\partial n_1}{\partial t} = \sigma_a c n_0 W_p - \sigma_e c n_1 W_\ell - \frac{1}{\tau} n_1, \tag{2}$$

accompanied by another diffusion equation for the pump light, see Ref. [6] for more details. $W_{\ell,p}(z,t)$ are the laser and pump light densities, $n_{0,1}(z,t)$ is the density of dye molecules in the ground and excited states, with $n = n_0 + n_1$ the total molecular density, c the speed of light in the medium, $\sigma_{a,e}$ the molecular absorption and stimulated emission cross sections, and $D = \frac{1}{3} c \ell$ is the diffusion coefficient for light. Further, ℓ is the transport mean free path and τ is the excited state lifetime. Equations (1) and (2) are the random laser analog of the well-known kinematic rate equations describing the dynamics of conventional lasers [4].

We will now explain what β means in Eq. (1) and how to obtain it. To incorporate the spectral dependence we rewrite Eq. (1) in terms of the specific energy density $W_\ell(\lambda) = W_\ell(\lambda; \mathbf{r}, t)$.

$$\frac{\partial}{\partial t} W_\ell(\lambda) d\lambda = D\nabla_z^2 W_\ell(\lambda) d\lambda + \sigma_e(\lambda) c n_1 W_\ell(\lambda) d\lambda + \frac{n_1}{\tau} L(\lambda) M(\lambda) d\lambda. \tag{3}$$

Here, $L(\lambda) d\lambda$ is the spontaneous emission spectral density function, with $\int_0^\infty L(\lambda) d\lambda = 1$. Integration over the entire spectrum yields Eq. (1) from Eq. (3). Since W_ℓ should only include the laser light (not all spontaneous emission), $L(\lambda)$ is multiplied by a "spectral participation factor" $M(\lambda)$, describing the coupling of the spontaneous emission to the laser process. $M(\lambda)$ excludes spontaneous emission outside the lasing band from W_ℓ. The exact shape of $M(\lambda)$ is immaterial for the current discussion, as long as it is peaked in a small wavelength range $\pm \delta$ around the central wavelength above threshold λ_ℓ, and $M(\lambda) \leqslant 1$. Following these arguments, we can restrict the integration domain to $\lambda_\ell \pm \delta$, where λ is the center wavelength of the emission spectrum above threshold. Outside this range $W_\ell(\lambda)$, $M(\lambda) \approx 0$. In this small wavelength domain we can take all cross sections to be constant.

$$\frac{\partial}{\partial t} \int_{\lambda_\ell - \delta}^{\lambda_\ell + \delta} W_\ell(\lambda) d\lambda = D\nabla_z^2 \int_{\lambda_\ell - \delta}^{\lambda_\ell + \delta} W_\ell(\lambda) d\lambda + \sigma_e(\lambda_\ell) c n_1 \int_{\lambda_\ell - \delta}^{\lambda_\ell + \delta} W_\ell(\lambda) d\lambda + \frac{n_1}{\tau} \int_{\lambda_\ell - \delta}^{\lambda_\ell + \delta} M(\lambda) L(\lambda) d\lambda. \tag{4}$$

Now $W_\ell = \int_{\lambda_\ell - \delta}^{\lambda_\ell + \delta} W_\ell(\lambda) d\lambda$, so to equate Eqs. (1) and (4) we define

$$\beta \equiv \int_{\lambda_\ell - \delta}^{\lambda_\ell + \delta} M(\lambda) L(\lambda) d\lambda, \tag{5}$$

and $\sigma_e = \sigma_e(\lambda_\ell)$.

For Eq. (2) the same procedure is followed, except for the multiplication by $M(\lambda)$ of the spontaneous emission term, since n_1 is not spectrally dependent. Thus, β does not appear in Eq. (2), as it should not.

IV. DISCUSSION

In the treatment of a cavity laser, β appears not only in the spontaneous emission term but also in the gain coefficient [2,4]. This is because of the fundamental requirement that, if the average occupation number of the laser mode is 1, the spontaneous and stimulated emission be equal. Via this back door, which cannot be locked in a system with discrete modes, the β that was intuitively introduced for the spontaneous emission seed, enters in the stimulated emission term. In the derivation above, we have only worked with energy densities, without having to refer to photon numbers. This is allowed by the absence of discrete modes in a random laser, due to the continuity of the space variables. If $g(\lambda,\lambda_0)$ is the lineshape function centered at λ_0, then our use of $\sigma_e(\lambda_\ell)$ instead of $\sigma_e(\lambda_0)\int_0^\infty g(\lambda,\lambda_0)d\lambda$ may be seen as the analog of the β in the gain coefficient.

If the scattering mechanism is wavelength dependent, as can occur in a system in which the scatterers are monodisperse Mie spheres, the resulting narrowed spectrum may be altered due to the improved feedback, or, equivalently, smaller transport term, near scattering resonances. Effectively, this means that $D=D(\lambda)$, where D decreases near a resonance. The definition of β is not changed by this process, because it uses the experimentally obtained laser spectrum to determine the overlap with the spontaneous emission spectrum of the active medium.

From Eq. (5), it is obvious that the spectral shape of $M(\lambda)$ will quantitatively influence β. The above-threshold spectrum is, in principle, the outcome of several wavelength dependent processes: amplification $\kappa_g(\lambda)$, seeding by spontaneous emission via $L(\lambda)$, and possibly feedback via $D(\lambda)$. The spectral participation factor $M(\lambda)$ is clearly a simplification of this complex problem, foregoing many subtleties. But the bottomline—spontaneous emission outside the laser spectrum does not contribute to W_ℓ—is encompassed in the $M(\lambda)$ construct. It therefore suffices to say that $M(\lambda)$ must be similar to the normalized above-threshold spectrum.

β reflects the narrowing of the spectrum above the threshold, which is connected to the sharpness of the laser threshold. β takes into account the spectral redistribution in a calculation that is wavelength independent; the coupling between different wavelengths [9] in Eqs. (1) and (2) would make them much less compact numerically. This use of β was shown to yield results that quantitatively agree with experiments [6].

Another matter is the recent observation of very narrow peaks in the spectra of disordered ZnO films [10]. These features are so sharp that they cannot be caused by simple gain narrowing. The peaks must originate from some resonant process, which is still debated but is possibly akin to resonant modes in a cavity. Such resonances are wave vector specific, and resulting effects cannot be explained in a purely diffusion framework. In our concept of a random laser β, we assume all wave vector information has been averaged out. The β factor for this kind of random laser should describe the coupling between the emitted field and the microscopic random electromagnetic mode structure of the material. As such, it resembles more the β known from cavity lasers.

A. Quantitative estimate

The numerical value for β is needed for the full calculation of the inversion. It typically turns out to be of the order

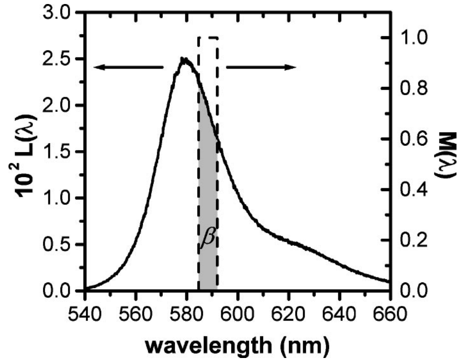

FIG. 3. Illustration of the construction of $\beta=\int_{\lambda_0-\delta}^{\lambda_0+\delta}M(\lambda)L(\lambda)d\lambda$, where $L(\lambda)d\lambda$ is the specific spontaneous emission spectral density (left axis) and $M(\lambda)$ is the coupling to the random laser process (right axis). In this example we use for $M(\lambda)$ a step function, yielding $\beta=0.14$.

of 0.1. Our method to construct β from the spectra is outlined in Fig. 3. For demonstration purposes we take $M(\lambda)$ to be 1 (perfect coupling) inside $\lambda_\ell\pm\delta$, where 2δ is the full width at half maximum of the spectrum. This yields almost certainly to an overestimation of β, but only by factors of order unity. With this $M(\lambda)$, we get $\beta=\int_{-\delta}^{\delta}L(\lambda-\lambda_\ell)d\lambda\approx0.14$ from Eq. (5). Using the spectrum normalized to the maximum as shown in Fig. 2 for $M(\lambda)$ yields $\beta\approx0.07$.

For comparison, we show a measurement of the output energy as a function of the pump pulse energy in Fig. 4, measured at the spectral peak of the emission. The solid line is a best fit of the solution of the (normal) laser rate equation to the experimental data points, with β and the threshold energy as free parameters. It shows $\beta=0.14\pm0.03$ and the threshold at $I_p=25$ μJ.

This value of β is quite sizable compared to the β factors encountered in conventional lasers, typically 10^{-8} for gas lasers, 10^{-5} for commercial semiconductor lasers, and up to 10^{-1} for (hardly "conventional") microcavity systems [2].

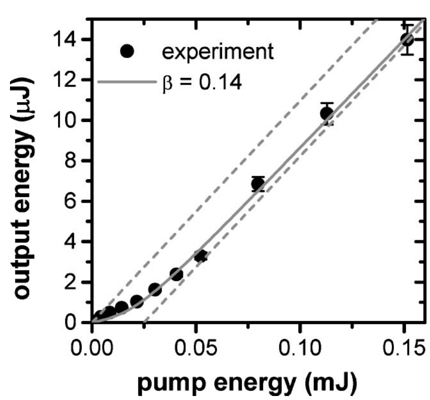

FIG. 4. Output energy of a random laser at the spectral peak as a function of pump pulse energy. Points: from experiment (parameters as in Fig. 2), solid line: fit to data points with a solution of (cavity) laser rate equations showing $\beta=0.14$. For $\beta=1$, the output energy increases linearly from zero pump energy, while for $\beta=0$ the emitted pulse energy is zero up to the threshold at $I_p=0.025$ mJ, and increase linearly from there (broken lines).

The physical background for this large magnitude is of course the ''soft'' selection mechanism solely by spectral overlap in random lasers, in contrast to the much more stringent requirements on the wave vector **k** imposed by discrete modes. However, the lack of direction in the emission (while making a large β possible) renders the random laser useless for the purposes large-β lasers are desired for, such as thresholdless directional emission and controlled quantum electrodynamics experiments.

V. CONCLUSIONS

In this paper, we have introduced a definition of the spontaneous emission factor β for a random laser, based on the spectral overlap between spontaneous and above-threshold laser emission spectra. The numeric value can easily be obtained from experimental data. An accurate value of β is indispensable for a quantitative understanding of the gain dynamics in a random laser, and for correct description of the response of a random laser to a pump pulse. The use of β following from the construction presented here reproduces experimental data very well.

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