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Effects of line-narrowing of amplified spontaneous emission analyzed by a Monte Carlo model

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ABSTRACT

This work focuses on the effects caused by line-narrowing of the amplified spontaneous emission (ASE) encountered in two-photon laser-induced fluorescence (TPLIF) processes. The line-narrowing effects complicate the TPLIF problem significantly, but are important for the quantitative interpretation of the TPLIF measurements. Past models are predominately developed for one-dimensional (1D) problem under ideal conditions. Therefore, this work develops a Monte Carlo (MC) method to robustly and flexibly incorporate non-ideal conditions including arbitrary temporal, spatial, and spectral beam profiles in multi-dimension practical problems. The MC model was the applied to analyze the TPLIF processes, both in 1D and 2D. The results show that the ASE radiation is more effectively amplified when the line-narrowing effects are considered than not. Because the ASE serves as a depopulation mechanism in TPLIF, such enhanced amplification has direct implications to the quantification of signals typically measured in practice. For example, our results show that neglecting the line-narrowing effects can cause significant error in the interpretation of the LIF and ASE signals. These results are hence expected to be useful for the design and analysis of experiments.

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1. Introduction

Laser-induced Fluorescence (LIF) finds significant applications in many areas such as combustion [1] and plasma [2] diagnostics. Many light atoms of importance to combustion and plasma research (e.g., hydrogen, oxygen and carbon atom, etc.) have LIF transitions in the vacuum-ultraviolet (VUV) spectral range [3]. Two-photon LIF (TPLIF) has been developed to circumvent the experimental difficulty in the VUV range by exciting the LIF transition with the absorption of two photons [4]. Though appearing to be a natural extension of LIF motivated by a practical consideration, the TPLIF technique is fundamentally distinct from onephoton LIF in several aspects [3]. This paper addresses the aspect of ASE (Amplified Spontaneous Emission) effects in TPLIF, created by the population inversion between the excited and the de-excited states [1]. The ASE effects represent an important topic in TPLIF research and application. When the ASE effects occur, the LIF signal becomes nonlinear with respect to the number density of the target species and the laser intensity, rendering the quantitative interpretation of the signal difficult [5]. This problem becomes more acute for multi-dimensional TPLIF measurements because the ASE effects, unlike collisional quenching, are non-local in the sense that ASE effects at one location depends on the conditions at other locations [6]. Second, the ASE effects also provide a potential diagnostic approach to generate laser-like signal, with excellent directionality and spectral purity [7].

Due to such importance, a considerable amount of research efforts has been invested in the study of ASE effects and various models have been developed. Rigorous models include those based on the density matrix formation [8,9] and the Maxwell-Bloch equations [10]. Simplified models

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suitable for practical applications include those based on the rate equations (RE) [2,11,12] and Monte Carlo (MC) simulations [13].

The focal point of this current work is to examine the impact of the narrowing of the spectral lineshape of the ASE radiation. The narrowing of the spectral lineshape is caused by the fact that ASE photons near the central frequency are amplified more effectively than those off the central frequency [14], resulting in an increasingly narrower spectral lineshape of the ASE radiation as it evolves. In previous studies [2,4,13,15,16], the lineshape of the ASE radiation has been assumed to be a delta function and these studies used different methods to estimate the magnitude of the delta function. In reality, the ASE lineshape is not constant and the lineshape varies spatially as the ASE radiation propagates. Therefore, it is the goal of this current work to quantify the impact of the spectral lineshape in the modeling of ASE effects. We developed methods that can (1) solve the governing equations of ASE under arbitrary spectral lineshape, and (2) incorporate the spatial variation of the lineshape into the solution. These methods were applied to quantitatively examine the impact of ASE lineshape in TPLIF measurements. The results obtained show that (1) the consideration of lineshape narrowing led to enhanced ASE effects compared to the treatment of the lineshape as a delta function, and (2) the enhancement became more pronounced when the problem is considered in multi-dimension than in onedimension (1D). These observations are expected to be valuable for the quantitative applications of TPLIF.

The rest of the paper is organized as follows. Section 2 describes the ASE models used in this work. Section 3 discusses the narrowing of the ASE spectral lineshape and its integration in the ASE models. Sections 4 and 5 report the results obtained in 1D and 2D simulations, respectively. Finally Section 6 summarizes the paper.

2. Description of ASE models



Fig. 1. Schematic of LIF and ASE process. Panel (a). Depiction of processes involved in TPLIF and ASE. Panel (b). Schematic of ASE model in 1D. Panel(c). Illustration of the narrowing of ASE lineshape.

two-photon absorption. Atoms on the excited level can either absorb an additional photon to be ionized (level 4) or fluoresce to de-excited level (level 2) emitting a LIF photon. Then the LIF photon, as described in Section 1, triggers the ASE process when a population inversion exists. The coordinate system used in this work is also shown in Panel (a), with the positive *x*-axis defined in the propagation direction of the excitation laser pulse. These four levels are coupled via collisional quenching, stimulated emission, and spontaneous emission (not shown in Fig. 1).

With the above understanding, the following set of rate equations can be developed to describe the TPLIF and ASE process:

The notations in Eq. (1)(a-f) are defined as follows:

$$\frac{\partial}{\partial t} \begin{bmatrix} n_{1} \\ n_{2} \\ n_{3} \\ l_{L} \\ l_{ASE}^{f,0} \cdot g_{ASE}(v) \\ l_{ASE}^{b,0} \cdot g_{ASE}(v) \end{bmatrix} = \frac{\partial}{\partial x} \begin{bmatrix} 0n_{1} \\ 0n_{2} \\ 0n_{3} \\ -cl_{L} \\ -cl_{ASE}^{f,0}g_{ASE}(v) \\ cl_{ASE}^{b,0}g_{ASE}(v) \end{bmatrix} + \begin{bmatrix} -W_{13}\left(n_{1} - \frac{g_{1}}{g_{3}}n_{3}\right) + (A_{21} + Q_{21})n_{2} + Q_{31}n_{3} \\ \int_{-\infty}^{+\infty} (W_{32}^{f} + W_{32}^{b})dv\left(n_{3} - \frac{g_{3}}{g_{2}}n_{2}\right) - (A_{21} + Q_{2a})n_{2} + (A_{32} + Q_{32})n_{3} \\ W_{13}\left(n_{1} - \frac{g_{1}}{g_{3}}n_{3}\right) - \int_{-\infty}^{+\infty} (W_{32}^{f} + W_{32}^{b})dv\left(n_{3} - \frac{g_{3}}{g_{2}}n_{2}\right) - (W_{34} + A_{32} + Q_{3a})n_{3} \\ c\left(-2W_{13}\left(n_{1} - \frac{g_{1}}{g_{3}}n_{3}\right) - W_{34}n_{3}\right)hv_{L} \\ c\left(W_{32}^{f}\left(n_{3} - \frac{g_{3}}{g_{2}}n_{2}\right) + A_{32}n_{3}\frac{\Delta\Omega_{f}}{4\pi}g_{sp}(v)\right)hv \\ c\left(W_{32}^{f}\left(n_{3} - \frac{g_{3}}{g_{2}}n_{2}\right) + A_{32}n_{3}\frac{\Delta\Omega_{f}}{4\pi}g_{sp}(v)\right)hv \end{aligned}$$
(1a-f)

This section briefly summarizes the ASE models used in this work to facilitate the discussion. Panel (a) of Fig. 1 illustrates the model we used in this work to capture the major processes in TPLIF and ASE. The model involves a four-level system interacting with a laser pulse. An excitation laser pulse excites the target species from the ground level (level 1, with population denoted as n_1 and the same notation is used hereafter) to the excited level (level 3) via *x* and *t*: the space in the *x*-direction and time, respectively

c and *h*: the speed of light and the Planck constant, respectively

*I*_{*L*}: the incident laser radiation intensity

 $I_{ASE}^{0,f}$ and $I_{ASE}^{0,b}$: the peak value of irradiance of the ASE photons in the forward (i.e., positive *x*) and backward (i.e., negative *x*) directions, respectively.

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