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Fluctuation study of light scattered from thermal lensing media

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Statistical properties of light scattered from nonlinear thermal lensing media have been investigated for the excitation source (laser) operating below, at and above threshold of oscillation. The incident laser light undergoes thermal defocusing in such media and the scattered light exhibits completely different photon statistics as compared to that of the incident light. The statistics of the scattered light (I_s) has been characterized through its intensity distribution $P(I_s)$, first and second moments $\langle I_s \rangle$ and $\langle I_s^2 \rangle$.

1. Introduction

In conventional light scattering experiments, the scattering medium is assumed to be a linear, isotropic and homogeneous medium. This gives rise to the conclusion that the incident and scattered light exhibit linear dependence. Consequently, the scattering cross-section is completely described by the physical properties of the medium and is independent of the intensity of the excitation source [1]–[3].

From the theory of propagation of laser radiation through condensed matter it is well known that at sufficiently high values of incident intensity the susceptibility χ of the medium becomes intensity dependent. This effect is manifested in the nonlinear changes in the macroscopic parameters like dielectric constant ε, refractive index n and molecular polarizability α. To the first order in incident intensity I_0, the real part of χ gives a nonlinear contribution to the index of refraction as

$$n(I_0) = n_0 + n_2 I_0 \tag{1}$$

where n_0 is the index of refraction at low intensity limit and n_2 is the first order nonlinear coefficient. The molecular number density ϱ and average molecular polarizability $\langle \alpha \rangle$ can be related to the index of refraction through Lorenz–Lorentz expression

$$\frac{n^2-1}{n^2+1} = \frac{4\pi}{3} \varrho \langle \alpha \rangle. \tag{2}$$

It is inferred from Eq. (2) that changes in the refractive index n arise from changes in ϱ, $\langle \alpha \rangle$ or both. The physical mechanisms responsible for bringing about nonlinear changes in n are broadly classified by SVELTO [4]. From relaxation time arguments it can be shown [4] that for cw lasers (used as excitation source in light scattering experiments) thermal effects are more dominating and give larger contribution to n_2 in Eq. (1). Further this mechanism manifests itself in producing localized heating of the medium but the isotropicity of the medium is still largely maintained.

The incident laser beam has a Gaussian intensity profile (TEM_{00}). When such a beam traverses through the medium, the thermal effects cause local temperature profiles to be set up in the medium centered around the laser beam. The corresponding contribution to n_2 given as a function of time t is given by [5]

$$n_2 = \frac{\alpha_0 ct}{8\pi\varrho C_p}\left(\frac{\partial n}{\partial T}\right) \tag{3}$$

where T stands for temperature, c is the velocity of light in the medium, C_p is the specific heat at constant pressure and α_0 is spatial absorptivity. For most media $\partial n/\partial T < 0$. This causes the medium to work as a diverging lens and as a result the propagating laser beam gets defocused.

In light scattering theory both n_2 and I_0 are assumed to be small enough such that they do not make any observable contribution to n in Eq. (1). Recently, it has been proved that thermal defocusing effects are experimentally observable even when the change in the index of refraction δn from beam center to beam edge is as small as $\delta n = 10^{-5}$ [6]. This requires a corresponding temperature change of only 10^{-2}°C [7]. For example, many common organic liquids are associated with spatial absorptivities in the range from 10^{-3} to 10^{-4} cm^{-1}. Thermal defocusing phenomena can be activated in these liquids even when the incident excitation power $P \sim 1$ W. Hence, in light scattering experiments where the exciting radiation propagates a typical distance of 1 cm, thermal lensing effect will be set up in the medium and this is amenable to measurements. This has been successfully used to study the absorption properties of fluids [8]–[11].

The objective of this work is to exploit the photon statistics of the light scattered from a medium where thermal lensing effect has been set up by the incident laser light. To generalize the problem we will be investigating the statistics of the scattered light corresponding to the physical situations where the laser is operating below, at and above threshold of oscillation depending on the pump parameter.

2. Intensity of scattered light

The theory of light propagation and scattering from thermal lensing media has been discussed in details in [6], [8], [11]. The physical phenomena of thermal defocusing has been treated by Litvak [5]. We will briefly recapitulate some of the results that are relevant to our present discussion. The excitation source is a cw laser propagating in the scattering medium in the positive z-direction. The TEM_{00} mode of laser has a spatial intensity distribution given by a Gaussian function. This gives rise to a corresponding spatial temperature distribution in the scattering medium. In such a system the resulting index of refraction n exhibits a parabolic profile in space. At space-time point (r, t), it is given by [6]

$$n(r,\,t) = n(t)\left[1+\frac{p^2 r^2}{2}\left(1+\frac{1}{\left(1+\frac{2t}{t_c}\right)}\right)\right] \tag{4}$$

where

$$p^2 = -\left(\frac{\partial n}{\partial t}\right)\frac{\alpha_0 P}{\pi\lambda_T n(t)\,\omega^2}, \tag{5}$$

and $n(t)$ is given as

$$n(t) = n_0+\left(\frac{\partial n}{\partial T}\right)\frac{\alpha_0 P}{4\pi\lambda_T \ln\left(1+\frac{2t}{t_c}\right)} \tag{6}$$

where P is the power of the incident laser beam, λ_T is the thermal conductivity of the medium and ω is the Gaussian beam diameter. The parameter t_c has been used as a characteristic time scale defined as

$$t_c = \frac{\omega^2}{D_T}. \tag{7}$$

If the medium under consideration has spatial extension L, the refractive index profile $n(r, t)$ will physically transform the medium to behave as a diverging lens with focal length given by $f = -1/Lp^2$. Due to this, the physical nature of the Gaussian beam will get distorted as it propagates through the thermal lensing medium. Consequently, the beam waist size $\omega(z, t)$ will evolve with propagation distance z and time t as [6]

$$\omega(z,\,t) = \omega(0)+\frac{az^2}{4\omega_0\, t/(t+t^*)} \tag{8}$$

where $a = 2(p\omega)^2$, ω_0 is the beam radius at $z = 0$ and $t^* = t_c/2$ at $t = 0$. The parameter $\omega(0)$ is given by

$$\omega(0) = \omega_0\left[bz^2/(2\omega_0^4)+(dw/dz)_0\, z/\omega_0+1\right]. \tag{9}$$

All the parameters with suffix zero correspond to their values at $z = 0$. When light is scattered from such a system, the intensity of the scattered light I_s can be expressed as a nonlinear function of incident intensity I_0 and is given by [6]

$$I_s = \frac{I_0}{1+SI_0}. \tag{10}$$

The z and t dependence of I_s and S are taken as implicit. The parameter S is written as

$$S = -\left(\frac{\partial n}{\partial t}\right)\frac{\alpha_0 z^2}{2\pi\lambda_T n\omega_0 (G_0\, \omega_s^2 \sqrt{\pi/2}}. \tag{11}$$

The expression for I_s in Eq. (10) has been derived by taking a realistic light scattering geometry into account. In this configuration there is a pinhole that precedes the photo-multiplier tube (PMT). The solid angle over which the scattered light is collected is determined by this pinhole. The spatial aperture function of this pinhole of width ω_s is expressed as a Gaussian having the form

$$G(x) = G_0 e^{-2x^2/\omega_s^2}. \tag{12}$$

When $t \gg t^*$, we approach the steady state limit and Eq. (10) reduces to

$$I_s = I_0(1-SI_0) \quad \text{for } SI_0 \ll 1. \tag{13}$$

This expression explicitly shows the quadratic dependence of I_s on I_0 for non-vanishing SI_0 values. The physical mechanism of thermal defocusing in the scattering media will remain unaltered even when the incident laser beam is assigned a parabolic spatial intensity profile instead of a Gaussian profile discussed above [8].

Information about changes in scattering power of the incident beam by thermal lensing can be directly inferred from the differential scattering cross-section $(d\sigma/d\Omega)$ given by [12], $(d\sigma/d\Omega) = R_0^2 I_s/I_0$, where R_0 is the location of the observer related to the scattering volume and Ω defines the solid angle over which the detector collects the scattered light. Hence, from Eq. (10), the differential scattering cross-section $(d\sigma/d\Omega)$ can be explicitly determined.

3. Photon statistics of scattered light

We will be discussing the statistics of the scattered light for three explicit input conditions through the use of Eq. (10). The mode of oscillation of laser is governed by the pump parameter b [13]. Correspondingly, the laser light exhibits different photon statistics as the pump parameter b is changed [14]. We will confine ourselves to the situations where the laser is operating below ($b = -\infty$), at ($b = 0$) and above ($b = +\infty$) threshold of oscillation. For these cases, the intensity distributions of the laser light are given as:

$$P(I_0) = \frac{1}{\langle I_0 \rangle} e^{-\frac{I_0}{\langle I_0 \rangle}} \quad \text{below threshold } (b = -\infty), \tag{14}$$

$$P(I_0) = \frac{2}{\sqrt{\pi}\langle I_0 \rangle} e^{-I_0^2/\langle I_0 \rangle^2} \quad \text{at threshold } (b = 0), \tag{15}$$

$$P(I_0) = \delta(I_0 - \langle I_0 \rangle) \quad \text{well above threshold } (b = \infty). \tag{16}$$

The angular brackets represent ensemble averages. We can invoke the probability conservation relation and write [15]

$$P(I_s)\,dI_s = P(I_0)\,dI_0. \tag{17}$$

From Equations (10) and (17) it can be shown that

$$P(I_s) = \frac{P(I_0)}{(1-SI_s)^2}. \tag{18}$$

Successively, using Eqs. (14), (15) and (16) in Eq. (18) we get

$$P(I_s) = \frac{1}{\langle I_0\rangle (1-SI_s)^2} e^{-(I_s/\langle I_0\rangle(1-SI_s))} \quad \text{below threshold } (b = -\infty), \tag{19}$$

$$P(I_s) = \frac{2}{\sqrt{\pi}\langle I_0\rangle (1-SI_s)^2} e^{-(I_s^2/\langle I_0\rangle^2(1-SI_s)^2)} \quad \text{at threshold } (b = 0), \tag{20}$$

$$P(I_s) = \delta(I_0 - \langle I_0\rangle)\frac{1}{(1+S\langle I_0\rangle)^2(1-SI_s)^2} \quad \text{above threshold } (b = \infty). \tag{21}$$

Equations (19), (20) and (21) describe the probability distributions of intensity of the scattered light from the thermal lensing medium as the incident laser is taken through its threshold of oscillation. These intensity distribution functions have been normalized.

The fluctuation behaviour of the scattered intensity can be analysed through first and second order of the moments of I_s. Defining the m-th order moment as

$$\langle I_s^m\rangle = \int_0^\infty I_s^m P(I_s)\, dI_s. \tag{22}$$

Substituting expressions for appropriate $P(I_s)$ in Eq. (22) from Eqs. (19), (20) and (21) and using $m = 1$, we obtain the average intensity of the scattered light given by

$$\langle I_s\rangle = \frac{\langle I_0\rangle}{(1-e^{-n})}\left[1-(1+n)e^{-n} + \sum_{p=1}^{\infty}(-\beta)^p\left((p+1)! - e^{-n}\sum_{k=0}^{p+1}\frac{(p+1)!}{k!}n^k\right)\right] \quad \text{at } b = -\infty, \tag{23}$$

$$\langle I_s\rangle = \frac{\langle I_0\rangle}{\Phi(n)\sqrt{\pi}}\sum_{m=0}^{\infty}(-\beta)^m P(n^2/m) \quad \text{at } b = 0, \tag{24}$$

$$\langle I_s\rangle = \frac{\langle I_0\rangle}{(1+\beta)} \quad \text{at } b = \infty \tag{25}$$

where $\beta = S\langle I_0\rangle$ and $\Phi(n)$ is the error function defined as [16]

$$\Phi(n) = \frac{2}{\sqrt{\pi}}\int_0^n e^{-x^2}dx \tag{26}$$

and $P(n^2/m)$ is the probability integral of X^2-distribution [16]

$$(n^2/m) = \int_0^{n^2} x^{m/2} e^{-x}\, dx. \tag{27}$$

In the evaluations of $\langle I_s \rangle$ values given by Eq. (23)–(25) to ensure the convergence of the integrals, we have set a cutoff value for the upper limit of I_s. It is $I_s(\max) = n\langle I_0 \rangle$. In this modified formalism $P(I_s)$ describes the probability distribution of an ensemble of variables I_s bound by upper and lower limits 0 and $n\langle I_0 \rangle$ respectively instead of the ideal limits 0 and infinity. In the limit $n \to \infty$ we realize the ideal case.

Likewise, the second order moments can be calculated from Eq. (22) by substituting the appropriate expressions for $P(I_s)$ and $m = 2$. These values come out as

$$\langle I_s^2 \rangle = \frac{\langle I_0 \rangle^2}{(1-e^{-n})}\left[2-2e^{-n}\sum_{r=0}^{2}\frac{n^r}{r!}+\sum_{p=1}^{\infty}(-2\beta)^p\left((p+2)!-e^{-n}\sum_{k=0}^{p+2}\frac{(p+2)!}{k!}n^k\right)\right] \quad \text{at } b=-\infty, \tag{28}$$

$$\langle I_s^2 \rangle = \frac{\langle I_0 \rangle^2}{\Phi(n)\sqrt{\pi}}\sum_{m=0}^{\infty}(-2\beta)^m P(n^2/m+1) \quad \text{at } b=0, \tag{29}$$

$$\langle I_s^2 \rangle = \frac{\langle I_0 \rangle^2}{(1+\beta)^2} \quad \text{at } b=\infty. \tag{30}$$

The higher order moments can explicitly be evaluated following the same procedure. From the expressions for $P(I_s)$, $\langle I_s \rangle$ and $\langle I_s^2 \rangle$, it is clearly inferred that

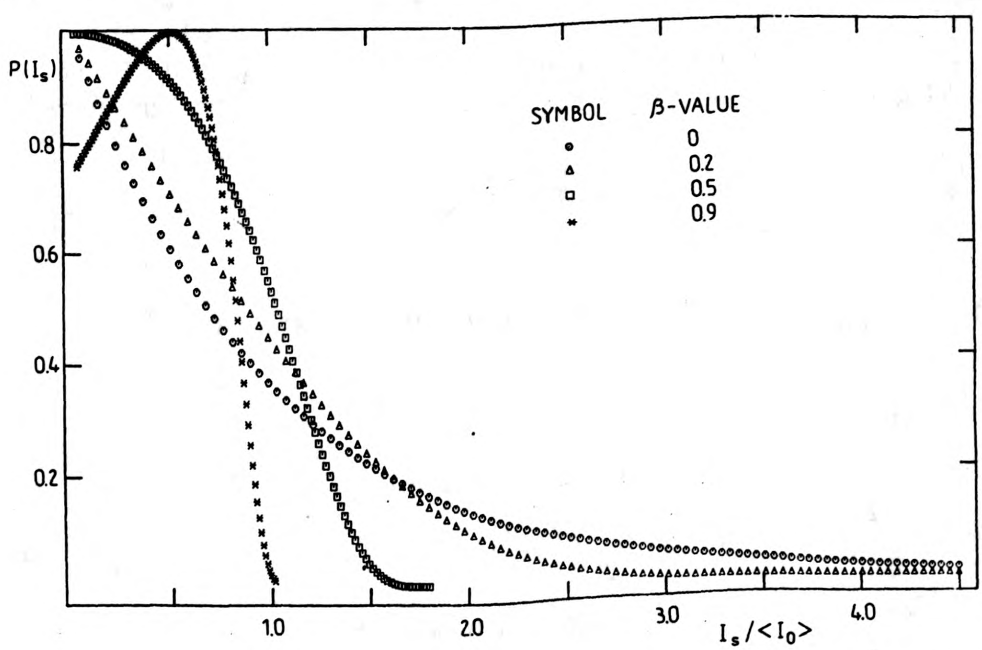

Fig. 1. Plot of intensity distribution function of the scattered light $P(I_s)$ as function of relative scattered intensity $I_s/\langle I_0 \rangle$ for the exciting laser source operating below threshold. Notice the evolution of $P(I_s)$ with increasing β

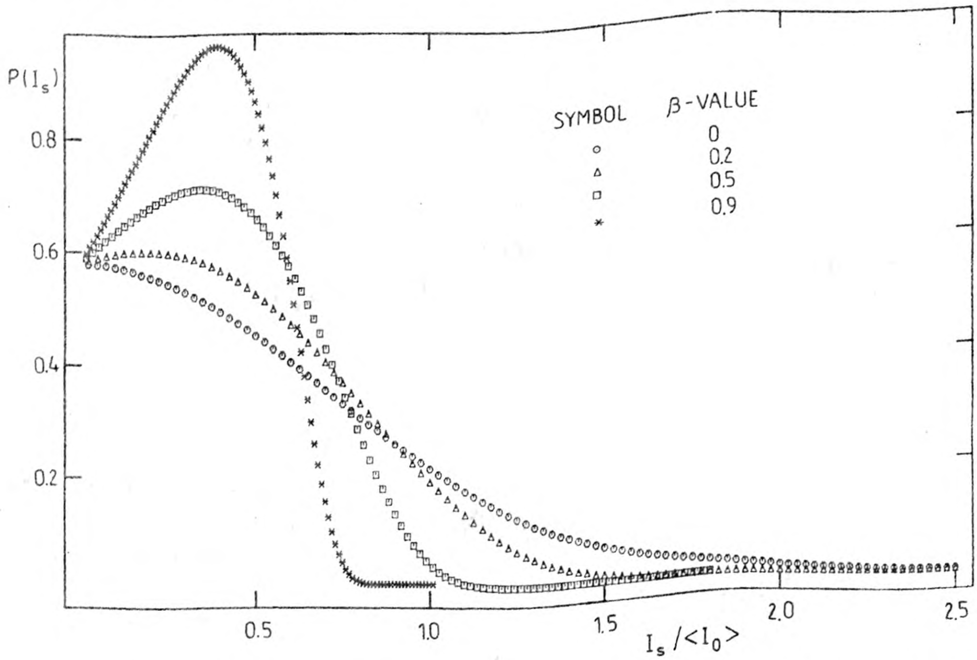

Fig. 2. Plot of intensity distribution function of the scattered light $P(I_s)$ as function of relative scattered intensity $I_s/\langle I_0\rangle$ for the exciting laser source operating at threshold. Notice the evolution of $P(I_s)$ with increasing β

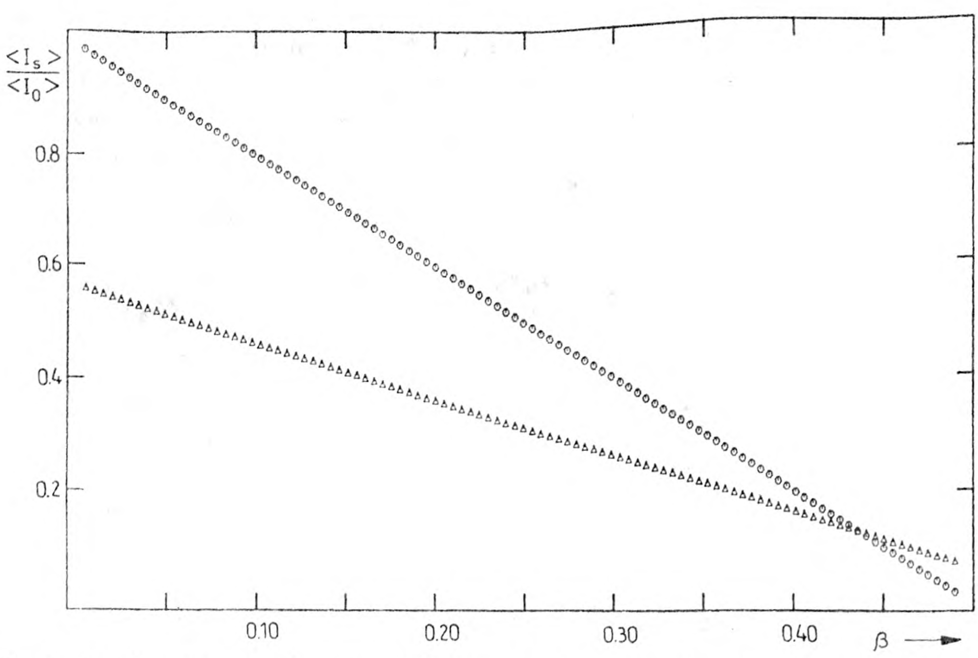

Fig. 3. Plot of average intensity relative to the average incident intensity as function of the characteristic parameter β for the exciting laser source operating below (circles) and at (triangles) threshold

most interesting change in the statistics of the scattered light arises when the incident laser is operating either below or at threshold. The situation corresponding to $b = \infty$ and the associated $P(I_0) = \delta(I_0 - \langle I_0 \rangle)$ represents an ideal condition of operation for laser that is rarely realized in practice. The $P(I_s)$ distributions are plotted as function of relative intensity $I_s/\langle I_0 \rangle$ in Figs. 1 and 2 for $b = -\infty$ and $b = 0$, respectively. The relative average scattered intensity $\langle I_s \rangle/\langle I_0 \rangle$ is plotted as function of characteristic parameter β in Fig. 3. For the numerical calculations, β values are chosen to be consistent with the realistic light scattering experiments and values for $\langle I_0 \rangle$ are taken from [13].

4. Discussion

It is clear from Figures 1 and 2 that as the characteristic parameter β is increased, the probability distribution of the scattered intensity $P(I_s)$ undergoes significant change. Particularly, when the characteristic parameter β assumes higher values $P(I_s)$ the distribution corresponding to $b = -\infty$ and $b = 0$ approaches a Gaussian like distribution function. On the other hand the evolution of the scattered intensity $\langle I_s \rangle$ with β is less dramatic. It is monotonic decreasing function of β (Fig. 3). The second moment of scattered intensity exhibits analogous behaviour.

It may sometimes be possible to minimize the effect of thermal lensing by choosing a large enough field of view of the detector. But, there are practical limitations to it. When the numerical value of $\beta \sim 10^{-2}$ or less, the thermal lensing will not have any significant contribution to the statistics of the scattered light (Eqs. (23)–(25)). This will physically mean linear dependence between I_s and I_0 in Eq. (10). This is realized experimentally by keeping the laser power very low and choosing the propagation distance z in the scattering medium to be small. These conditions are readily met in static and dynamic light scattering experiments, since the incident laser power is typically confined to ~ 100 mW. Hence, thermal lensing will not have any significant effect on these experiments as long as the laser power is limited to such low value.

An alternative approach to study the statistics of the scattered light is to use Mandel's photon counting formula [17] and evaluate the photon distribution function and its moments.

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Исследование флюктуации рассеянного света в среде вызывающей термическую расфокусировку

Исследовано статистические свойства рассеянного света в нелинейной среде термически расфокусирующей (thermal lensing) для источника возбуждения (лазер) действующего ниже, выше и на пороге осцилляции. Падающий свет подвергается термической расфокусировке в таких средах, а рассеянный свет доказывает совсем другой статистики фотонов по сравнению с падающим светом. Статистика рассеянного света (I_s) характеризована его разложением силы $P(I_s)$, первым и вторым моментом $\langle I_s \rangle$ и $\langle I_s^2 \rangle$. Охарактеризовано влияние явления термической расфокусировки на расфокусировочные опыты Райлейга и Бриллона.