## Light-scattering fluctuations and thermal noise in photorefractive media

Robert McGraw

Rockwell International Science Center, 1049 Camino Dos Rios, Thousand Oaks, California 91360 (Received 23 December 1991)

A thermodynamic description of the photorefractive effect is developed and used to evaluate thermal fluctuations in the space-charge field of a photorefractive medium responsible for light-scattering noise. Equilibrium grating properties are obtained through minimization of the Helmholtz free energy and are found to be in agreement with results obtained by other methods. Thermal fluctuations in the space-charge field are obtained from the curvature of the free-energy surface. These fluctuations give rise to light-scattering fluctuations in the medium refractive index through the electro-optic effect and are the fundamental source of light-scattering noise. Light-scattering fluctuations associated with the optical Kerr effect are also examined. The signal-to-noise ratio and dynamic range of a photorefractive medium are determined. Stochastic noise model calculations are presented for  $BaTiO_3$  under conditions for which the dominant noise contribution is associated with the photorefractive effect. Our results suggest a very large dynamic range for photorefractive materials (120–140 dB) that should prove useful for optical signal processing applications.

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## I. INTRODUCTION

Photorefractive media constitute a large and versatile class of nonlinear optical materials with applications to holographic storage as well as to real-time optical information processing. Two closely related theoretical models of the photorefractive effect have been put forth [1,2] and successfully utilized over the past decade by a number of investigators for the description of nonlinear optical processes, including optical phase conjugation [3] and coherent beam combination via two-wave mixing [4]. Nevertheless, it appears that few studies have been directed at quantifying the fundamental limits to the photorefractive effect set by noise. In particular, questions related to the noise limits on photorefractive sensitivity to weak incident signal powers (dynamic range) have apparently not been previously addressed. In the present study it is shown that there are noise limits to the dynamic range of a photorefractive medium that are inherent in the photorefractive effect itself. The quantitative determination of these limits is the foremost objective of this paper.

During the past several years we have undertaken the investigation of light-scattering noise associated with nonlinear optical processes utilizing Kerr and artificial Kerr media [5-7]. These studies were based on a novel application of statistical thermodynamic methods to quantitatively relate the response of the medium (e.g., the strength of a grating formed by a pair of writing laser beams) to the root-mean-square (rms) amplitude of the spontaneous dielectric fluctuations that give rise to light-scattering noise. Time-averaged noise powers for Kerr media at room temperature and visible wavelengths were found to be in the microwatt range [5,6]. Moreover, stochastic simulations of nonlinear optical processes revealed significant instantaneous fluctuations in the ampli-

tude and phase of the output fields even for noise-free incident signals at power levels well into the milliwatt range [7]. The present paper extends these results to materials exhibiting the photorefractive effect. In the following sections it will be shown that the photorefractive response is also fundamentally related to light-scattering noise; however, the physics underlying this relation will be shown to differ considerably from the Kerr media case. In particular, it will be shown for photorefractive materials that much lower signal power levels can be accommodated before the effects of noise become significant.

The present paper is divided into two main parts with the first part (Sec. II) serving as the foundation for the results to follow. In Sec. II we present a new thermodynamic approach to the photorefractive effect with the objective of obtaining the Helmholtz free-energy surface associated with grating formation in the presence of a spatially modulated light intensity pattern [8]. The thermodynamic approach is described here within the framework of the Kukhtarev [1] and hopping [2] models of the photorefractive effect and is shown to give identical results for the equilibrium grating response. New expressions for the energy, entropy, and donor site chemical potentials associated with the formation of an index grating via the photorefractive effect are also derived. While these results are significant, the main reason for introducing the thermodynamic approach is that it leads naturally to a derivation of the free energy associated with nonequilibrium states for small departures from equilibrium. The occupation of these nonequilibrium states through thermal fluctuations in the medium will be shown to underlie the presence of light-scattering noise.

The second part of the paper is directed at the study of thermal fluctuations and light-scattering noise. Fluctuations in the grating amplitude are obtained in Sec. III

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from the curvature of the free-energy surface. The approach is similar to that developed in Ref. [7] for Kerr media and, where possible, we compare results for the two models. Index fluctuations that give rise to the lightscattering-noise associated with the photorefractive effect are obtained in Sec. IV. The thermal fluctuations previously described for Kerr media [5-7] are also considered here as a source of light-scattering noise for photorefractive materials that exhibit both optical Kerr and photorefractive effects. An analysis is presented for the dynamic range of a photorefractive medium with noise gratings associated with both the Kerr and photorefractive effects. The results of a computer-simulated noise calculation for the dynamic range of the photorefractive material BaTiO<sub>3</sub> are presented in Sec. V. Section VI concludes with a summary and discussion of results.

## II. THERMODYNAMIC ANALYSIS OF THE PHOTOREFRACTIVE EFFECT

This section presents an overview of the origins of the photorefractive effect followed by an analysis of the energy, entropy, and intensity contributions to the grating free energy within the framework of the Kukhtarev [1] and hopping [2] models. The first-order photorefractive grating response is then obtained through free-energy minimization.

#### A. Origins of the photorefractive effect

Initiation of the photorefractive effect occurs when a spatial variation of light intensity causes a redistribution of charge density and buildup of a space-charge field. For example, let the spatial light intensity distribution follow the simple sinusoidal grating form

$$I(x) = I_0[1 + m \cos(qx)], \qquad (2.1)$$

characterized by average intensity  $I_0$ , grating wave vector **q** with  $q = |\mathbf{q}|$ , and intensity modulation ratio *m*. The intensity grating induces a corresponding density

$$n(x) = n_0 [1 + w \cos(qx)], \qquad (2.2)$$

where  $n_0$  is the average density. In the hopping model, n refers to the number density of hopping carriers of charge  $q_e$ . In the Kukhtarev model, n refers to the number density of unionized donor sites.

A comparison of the Kukhtarev and hopping models is presented in Fig 1. Each model assumes a uniform population of fixed sites as well as a uniform distribution of fixed countercharges to maintain overall charge neutrality. In the hopping model the fixed sites may be occupied by a smaller number of mobile carriers of charge  $q_e$ . The latter are assumed to hop from site to site with a hopping rate that is proportional to the local light intensity. The net effect is to cause the carriers to diffuse from regions of higher to regions of lower light intensity. As a result, the density modulation ratio w in Eq. (2.2) is opposite in sign to the intensity modulation ratio m In the Kukhtarev model the fixed sites are occupied with a smaller number of valence-band carriers whose charge may be either posi-



FIG. 1. Comparison of the Kukhtarev and hopping models.

tive or negative. For definiteness we assume the carriers in both models to be electrons of charge  $q_e = e = -|e|$ . Mobility of carriers in the Kukhtarev model requires excitation from the valence band into the conduction band, which occurs at a rate that is proportional to the light intensity. Generation and recombination of carriers is a fast process compared to the overall redistribution of charge, and the number of carriers in the conduction band at any given time is negligible compared to the number of valence-band carriers (donors). Redistribution of charge in the Kukhtarev model may therefore be thought of as due to the redistribution of the donor population through accumulated generation, recombination, and transport of carriers via the conduction band. Consideration of the index change induced by the space-charge field through the Pockels effect is reserved for Sec. IV.

To simplify the present analysis we will neglect contributions to grating formation from the photocurrent, dark current, and externally applied electric fields. We will also neglect contributions from higher-order gratings since our primarily interest is in the small modulation ratio limit. The various components of the grating free energy will now be derived. These include contributions from energy storage in the space-charge field, configurational entropy, and the free energy associated with exposure of the donor or hopping sites, depending on which model is used, to a spatially varying light intensity.

#### B. Energy stored in the space-charge field

The energy required to build up the space-charge field  $E_{\rm SC}$  is [9]

$$U_{\rm SC} = (\epsilon_0 / 8\pi) \int E_{\rm SC}^2(x) dx , \qquad (2.3)$$

where  $\epsilon_0$  is the static dielectric constant of the medium and the integration is over the beam interaction volume  $V_S$ . The space-charge field is related to the grating modulation ratio w through Poisson's equation

$$\nabla E_{\rm SC} = -\nabla^2 \Phi = 4\pi \bar{\rho} / \epsilon_0 , \qquad (2.4)$$

where  $\Phi$  is the electrostatic potential and  $\overline{\rho}$  is the net charge density. The latter quantity follows from Eq. (2.2) and is given by

$$\bar{\rho} = n_0 e w \cos(q x) . \tag{2.5}$$

The solution to Poisson's equation is

$$E_{\rm SC}(x) = (4\pi n_0 e / \epsilon_0 q) w \sin(qx) . \qquad (2.6)$$

Substitution into Eq. (2.3) gives

$$U_{\rm SC} = (\pi/\epsilon_0)(n_0 e/q)^2 V_S w^2 = \frac{1}{4} n_0 V_S k T (k_D/q)^2 w^2 .$$
(2.7)

The last equality relates  $U_{SC}$  to the reciprocal Debye screening length  $(k_D)$  defined through the relation

$$(k_D)^2 = 4\pi n_0 e^2 / (\epsilon_0 kT)$$
 (2.8)

#### C. Configurational entropy

The entropic contribution to the free energy arises from the spatial distribution of the charge carriers in the hopping model (donors in the Kukhtarev model). This configurational entropy is also a function of the density modulation ratio w

$$S(w) = -k \int n(x) \ln n(x) dx + C$$
  
=  $-n_0 k V_S w^2 / 4$  (for  $w \ll 1$ ). (2.9)

Integration is over the beam interaction volume  $V_S$ . The last equality follows from inserting n(x) from Eq. (2.2) into the integrand and expanding the logarithm in the small w limit. Note that the constant C has been chosen so that S equals zero, its maximum value, for the uniform charge distribution (w = 0).

#### D. Chemical potential

Considering only the contributions from  $U_{\rm SC}$  and -TS to the grating free energy, one obtains from Eqs. (2.7) and (2.9) a parabolic free-energy function that is minimized for w = 0 (see Fig. 2), suggesting that the uniform charge distribution is most stable and that no grating should form. A third contribution to the free energy needs to be taken into account, which directly incorporates the effects of illumination with nonuniform light intensity and serves as the driving term for grating formation. A derivation of this new contribution to the free-energy function is provided here separately within the framework of the Kukhtarev and the hopping models of the photorefractive effect. Despite differences in the underlying physical mechanisms, it will be found that equivalent results are obtained for each model.

## 1. Kukhtarev model

In the Kukhtarev model (Fig. 1) donors become ionized in the illuminated region through the reaction

$$D \to D^+ + e \quad . \tag{2.10}$$



FIG. 2. Thermodynamic potentials for grating formation.

To derive a chemical potential for this process, we adopt the treatment of Reiss [10] and regard donor ionization in a semiconductor as a chemical process through which electrons are excited from fixed donor sites into the conduction band. Concentrations of the various species in Eq. (2.10) are then related through an equilibrium mass action expression, which in the present application is dependent on the light intensity

$$K_{eq}(I) = [D^+][e]/[D] = sI/\gamma_R$$
 (2.11)

 $[D] = n_D$  is the number density  $(cm^{-3})$  of unionized donor sites, etc. The last equality in Eq. (2.11) gives the ratio of rates for the forward to reverse processes in the equilibrium of Eq. (2.10) as specified in the Kukhtarev model. Here s is the cross section for photoionization of donors, I is the light intensity, and  $\gamma_R$  is the recombination rate.

The conditions for which the ionization of impurities in semiconductors follows the law of mass action are described in Ref. [10]. In essence, we require the absence of interaction. For electrons, this requires a concentration in the conduction band sufficiently low that the classical limit of Fermi-Dirac statistics is obeyed. This will be the case when

$$\Lambda^3 n_e \ll 1 , \qquad (2.12)$$

where

$$\Lambda = h / (2\pi m_e kT)^{1/2} \tag{2.13}$$

is the thermal de Broglie wavelength and  $n_e = [e]$  is the electron concentration. Setting *T* equal to room temperature and  $m_e$  equal to the free-electron mass gives the condition  $n_e \ll 1.3 \times 10^{19}$  for classical statistics to be valid. Thus the inequality condition given by Eq. (2.12) is easily satisfied for a photorefractive medium.

The chemical potential of a dilute distribution of electrons in the conduction band takes the form

$$\mu_e = \mu_e^0 + kT \ln[e] , \qquad (2.14)$$

where  $\mu_e^0$  is the chemical potential in the standard state. The standard state for each species is defined at a concentration of unit number density. We will also require the well-known thermodynamic relation between the change in standard state chemical potential and the equilibrium constant [11]

$$\Delta \mu^{0} = \mu_{e}^{0} + \mu_{D}^{0} + -\mu_{D}^{0} = -kT \ln K_{eq} . \qquad (2.15)$$

With the reasonable assumption that the chemical potential of ionized donors is independent of the light intensity, we obtain from the condition that  $\Delta \mu = 0$  at equilibrium, the equality

$$\mu_D(I) - \mu_D(I_0) = \mu_e(I) - \mu_e(I_0) . \qquad (2.16)$$

Combining this last result with Eq. (2.14) gives

$$\mu_D(I) - \mu_D(I_0) = kT \ln\{[e(I)]/[e(I_0)]\}, \qquad (2.17)$$

where [e(I)] is the number density of electrons at light intensity *I*. From the structure of Eq. (2.17), we see that [e(I)] is a measure of the fugacity or "escaping tendency" of the unionized donor species *D*.

The concentration of free electrons in a photorefractive material is small to the extent that  $[e] \ll [D^+], [D]$ . This means that for a closed volume sample of the material, the relative changes in species concentration with light intensity will be dominated by changes in the electron concentration and fractional changes in the concentration of ionized donors and unionized donors can be neglected. Then  $[e(I)]/[e(I_0)]=I/I_0$  from Eq. (2.11). This last result is nothing more than a statement of the proportionality between the concentration of free electrons in the conduction band and the light intensity [12]. Equation (2.17) may be now be written in the form

$$\mu_D(I) - \mu_D(I_0) = kT \ln(I/I_0) . \qquad (2.18)$$

Since a uniform intensity  $I_0$  results for m = 0, it is convenient to choose the energy scale so that the chemical potential vanishes for this case:  $\mu_D(I_0)=0$ . With this choice of scale, our final form for the spatial variation of the donor chemical potential with light intensity is

$$\mu_D(x) = kT \ln[I(x)/I_0] = kT \ln[1 + m \cos(qx)], \quad (2.19)$$

where the last equality follows from Eq. (2.1). Integration over the beam interaction volume gives

$$U_{I} = \int n(x)\mu_{D}(x)dx = kTn_{0}V_{S}mw/2 \quad (\text{for } m \ll 1) .$$
(2.20)

Equation (2.20) gives the direct contribution from light intensity to the free energy of grating formation. We will now show that an identical expression for  $U_I$  is obtained within the framework of the hopping model.

#### 2. Hopping model

In the original formulation of the hopping model, the intensity I(x) entered as a spatially dependent kinetic prefactor to an exponential expression for the hopping rate [2]. Alternatively, the spatially varying part of the intensity may be expressed as a thermodynamic potential

 $\phi_{h\nu}$ , still within the framework of the hopping model, by simply transferring it from the intensity prefactor to the exponent through the identity

$$I(x) = I_0[1 + m \cos(qx)] \equiv I_0 \exp[\phi_{hv}(x)/kT] . \qquad (2.21)$$

The redefined kinetic prefactor  $I_0$  is now a constant and a new contribution  $\phi_{h\nu}$  has been added to the potential. Henceforth we will refer to  $\phi_{h\nu}$  as the hopping potential. From the last equality in Eq. (2.21) we obtain

$$\phi_{hv}(x) = kT \ln[1 + m \cos(qx)] = \mu_D(x) , \qquad (2.22)$$

showing that the hopping potential is equivalent to the chemical potential  $\mu_D$ . Thus, the intensity contribution to the free energy takes the form

$$U_{I} = \int n(x)\phi_{hv}(x)dx = kTn_{0}V_{S}mw/2 \quad (\text{for } m \ll 1) ,$$
(2.23)

which is identical to the expression obtained above using the Kukhtarev model.

## E. Free energy minimization

On combining Eqs. (2.7), (2.9), and (2.20) [or (2.23)] we obtain the Helmholtz free energy

$$F(w) = U_I(w) + U_{SC}(w) - TS(w)$$
  
=  $\frac{1}{4}n_0 V_S kT[(k_D/q)^2 + 1]w^2 + \frac{1}{2}n_0 V_S kTmw$  (2.24)

for  $m, w \ll 1$ . Equation (2.24) is the main result of this section. The various contributions to F(w) are shown in Fig 2. Differentiation of Eq. (2.24) to obtain the minimum in free energy gives

$$w_{\rm eq} = -[(k_D/q)^2 + 1]^{-1}m . \qquad (2.25)$$

Substitution of this last result into Eq. (2.6) gives

$$E_{\rm SC}^{\rm eq}(x) = -(kTq/e)[1+(q/k_D)^2]^{-1}m\sin(qx) \qquad (2.26)$$

for the equilibrium space-charge field where Eq. (2.8) has been used. Note that since  $U_I$  is the only term in the free energy that varies linearly with w, it is the driving term (or symmetry-breaking term) for the formation of a finite amplitude equilibrium grating in the medium. Minimization of F(w) gives the equilibrium grating amplitude as a linear function of the intensity modulation ratio m. [Saturation effects have not been included in the calculation due to the small m and w approximations used in Eq. (2.24).]

The preceding thermodynamic description of the photorefractive effect is supported by the fact that Eqs. (2.25) and (2.26) are identical to the usual expressions for the grating amplitude obtained by more conventional analysis. Further support comes from the agreement between the derivations carried out separately for the Kukhtarev and hopping models. The principal advantage of the thermodynamic description is that fluctuations about equilibrium are also readily obtained by methods that will now be described.

## III. GRATING FLUCTUATIONS ASSOCIATED WITH THE PHOTOREFRACTIVE EFFECT

In the preceding section it was shown that the formation of a space-charge grating entails a reduction of the Helmholtz free energy relative to its value for a spatially uniform charge distribution. In the present section it is shown that at finite temperatures, the minimum freeenergy configuration is not necessarily the one characteristic of the medium at any given instant in time. Instead, the grating amplitude is free to fluctuate about its minimum free-energy value over a range determined by the curvature of the free-energy surface. Thus the freeenergy surface determines both the equilibrium grating response and the fluctuation range. Fluctuations in the grating amplitude are thus inherent in the photorefractive effect itself. It is important to note in this regard that grating fluctuations of the type described in the present section can occur only as the result of displacements of electrical charges throughout the beam interaction volume-by the same mechanisms as those underlying formation of the free energy minimizing signal grating. Furthermore, upon removal of the illumination source, the fluctuation component will be frozen in place in the same manner as the signal component to serve as a sensibly permanent record of the information present in the writing beams together with a statistical sample of the thermal noise.

A very general thermodynamic method for deriving fluctuation amplitudes about an equilibrium state is provided by the Einstein fluctuation formula [13]. This formula states that the probability of a fluctuation  $\delta w$  occurring spontaneously is proportional to  $\exp[-F(\delta w)/kT]$ :

$$P(\delta w) = A \exp[-F(\delta w)/kT], \qquad (3.1)$$

where  $\delta w = w - w_{eq}$  is the fluctuation amplitude,  $F(\delta w)$  is the reversible work required to create the fluctuation configuration deliberately through the application of external constraints, and A is a normalization constant. For constraint-type processes occurring at constant volume and constant temperature, the reversible work is given by the Helmholtz free energy. Taylor expansion about the equilibrium or minimum free-energy state gives

$$F(w) - F(w_{\rm eq}) = \frac{1}{2} \frac{\partial^2 F}{\partial w^2} (w - w_{\rm eq})^2 , \qquad (3.2)$$

where terms of higher than quadratic order have been neglected. The second derivative gives the curvature of the free-energy function near its minimum value. Inspection of Eqs. (3.1) and (3.2) shows that the fluctuations in w are Gaussian with mean

$$\langle w \rangle = w_{eq}$$
 (3.3)

and variance equal to

$$\langle |\delta w|^2 \rangle = kT \left[ \frac{\partial^2 F}{\partial w^2} \right]^{-1}$$
 (3.4)

The thermodynamic analysis of the previous section has furnished an explicit form for F(w), which is now utilized. Substitution from Eq. (2.24) into Eq. (3.4) gives

$$\frac{\partial^2 F}{\partial w^2} = \frac{1}{2} n_0 V_S k T[(k_D/q)^2 + 1]$$
(3.5)

for the curvature, and

$$\langle |\delta w|^2 \rangle = 2(n_0 V_S)^{-1} [(k_D/q)^2 + 1]^{-1}$$
 (3.6)

for the variance of fluctuations in the density modulation ratio w. The first and second terms in square brackets correspond to the separate contributions from the spacecharge field [Eq. (2.7)] and the entropy [Eq. (2.9)], respectively. The intensity-dependent term makes no contribution to curvature since it is linear in w.

Corresponding fluctuations will also occur in the space-charge field through Poisson's equation. With the aid of Eq. (2.6) we obtain

$$\langle |\delta E_{\rm SC}|^2 \rangle = (4\pi n_0 e / \epsilon_0 q)^2 \langle |\delta w|^2 \rangle / 2$$
  
=  $[16\pi^2 n_0 e^2 / (\epsilon_0^2 q^2 V_S)] [(k_D / q)^2 + 1]^{-1}, (3.7)$ 

where the factor of  $\frac{1}{2}$  results from averaging the square of the sine function over the beam interaction volume. This result can be written in a more compact form. Using Eq. (2.8) and expanding notation to include the wave-vector dependence of the fluctuation gives

$$\langle |\delta E_{\rm SC}(\mathbf{q})|^2 \rangle = (4\pi kT/\epsilon_0 V_{\rm S})[1+(q/k_D)^2]^{-1}$$
. (3.8)

The expression in square brackets reflects the nonlocal properties of the photorefractive medium and results in a reduction in fluctuation amplitude for  $k_D$  comparable to q. This reduction is due to the energy required to break correlations that would otherwise be present when the Debye screening length exceeds the grating spacing. A similar phenomenon occurs in a Kerr medium near a critical point [7] and is discussed in Sec. IV. It is useful to consider two limiting forms of Eqs. (3.6) and (3.8).

## A. Diffusion limit $(q^2/k_D^2 \ll 1)$

In crystals such as  $BaTiO_3$  and SBN, the photorefractive effect is diffusion limited. This limit is characterized by the property that the Debye screening length, or correlation length, is much shorter than the grating spacing. Equation (3.6) for the fluctuations in w reduces to

$$\langle |\delta w|^2 \rangle_{\text{diff limit}} = 2(n_0 V_S)^{-1} (q/k_D)^2$$
$$= q^2 \epsilon_0 kT / (2\pi n_0^2 e^2 V_S) . \qquad (3.9)$$

Similarly Eq. (3.8) becomes

$$\langle |\delta E_{\rm SC}(\mathbf{q})|^2 \rangle_{\rm diff\ limit} = (4\pi kT/\epsilon_0 V_S) .$$
 (3.10)

In the diffusion limit the grating fluctuations are dominated by the curvature contribution to the free-energy function from  $U_{SC}$ ; fluctuations are limited in size by buildup of the corresponding space-charge field and are independent of the number of unionized donor sites present in the beam interaction volume. For this limiting case, Eq. (3.10) may be deduced immediately from Eq. (2.3) and the equipartition theorem

$$U_{\rm SC} = (\varepsilon_0 / 8\pi) \int E_{\rm SC}^2(x) dx$$
  
=  $(\varepsilon_0 V_S / 8\pi) \langle |\delta E_{\rm SC}|^2 \rangle = kT/2$ . (3.11)

The last equality is identical to Eq. (3.10).

## B. Trap density limit $(q^2/k_D^2 >> 1)$

For small values of the trap density [14], the spacecharge field will be limited by the size of  $n_0$  appearing in Eq. (26) rather than by the diffusion field (kTq/e). In this limit, the correlation length is much greater than the grating spacing and Eq. (3.6) for the fluctuations in wreduces to a simple temperature-independent form

$$\langle |\delta w|^2 \rangle_{\text{trap limit}} = 2/(n_0 V_S)$$
 (3.12)

Corresponding fluctuations in the number density follow from Eq. (2.2):

$$\langle |\delta n(\mathbf{q})|^2 \rangle_{\text{trap limit}} = n_0 / V_S$$
 (3.13)

It is interesting that an identical expression to Eq. (3.13) is obtained for particle number density fluctuations in a Brownian suspension. In that well-known case [15],

$$\langle |\delta n(\mathbf{q})|^2 \rangle = n_0^2 k T \beta_T / V_S = n_0 / V_S$$
, (3.14)

where  $\beta_T$  is the isothermal compressibility and  $\delta n(\mathbf{q})$ here refers to a fluctuation in particle number density of wave vector  $\mathbf{q}$ . The last equality in Eq. (3.14) follows from the independent-particle assumption for which  $\beta_T = 1/(n_0 kT)$ . Equation (3.14) was used previously as a starting point for the analysis of light-scattering fluctuations and noise in an artificial Kerr suspension medium [5].

The preceding results are best understood by noting that fluctuations in the trap density limit are controlled by the entropy term in the free energy and are not limited by energy storage in the space-charge field. Noise in this limit may be thought of as due to statistical fluctuations in the trapped electron density. The corresponding fluctuations in the space-charge field are obtained from Eq. (3.8) in the limit  $q^2/k_D^2 \gg 1$ :

$$\langle |\delta E_{\rm SC}(\mathbf{q})|^2 \rangle_{\rm trap \ limit} = (4\pi kT/\epsilon_0 V_S)(k_D/q)^2$$
$$= 16\pi^2 n_0 e^2 / (\epsilon_0^2 q^2 V_S) , \qquad (3.15)$$

where the last equality, which provides a manifestly temperature-independent form for the fluctuations in this limit, follows using Eq. (2.8).

## **IV. LIGHT-SCATTERING NOISE**

Light-scattering noise arises from thermal fluctuations in the space-charge field [Eq. (3.8)], which induce corresponding fluctuations in the linear dielectric constant via the Pockels effect [Eq. (4.1) below]. The dielectric fluctuations, in turn, give rise to scattered or diffracted light as noise. The effect may be best understood by considering the processes involved during the writing and the readout of a pure sinusoidal grating component as shown in Fig. 3. During the write stage [Fig. 3(a)] the



FIG. 3. (a) Beam configuration for writing a signal grating of intensity modulation ratio m (solid curve).  $I_1$  and  $I_2$  are the intensities of the pump and signal beams, respectively. The dashed curve depicts a noise grating of the same phase and same wave vector **q**. (b) Read configuration showing diffracted signal (solid arrow) and diffracted noise (dashed arrow) from the field-induced and fluctuation grating components, respectively.

information-containing signal grating, represented by the solid curve, is formed from the equilibrium space-charge field of Eq. (2.26) via the Pockels effect. This grating may be probed during the read stage [Fig. 3(b)] by measuring light diffracted from it, preferably using a nonabsorbing frequency so as not to induce erasure. The diffracted signal is represented by the solid arrow labeled  $I_{\text{diff}}$  in the figure. Of particular relevance are those fluctuations in the dielectric constant, represented in Figs. 3(a) and 3(b) by the dashed curves, that have the same wave vector as the signal grating (q). Light scattered from such fluctuations during the read stage is indistinguishable from light diffracted from the signal grating and is therefore treated as noise. The scattered noise component is represented by the dashed arrow in Fig. 3(b). Noise due to scattered light from defects in the material is not treated in the present study, which addresses the fundamental noise limits associated with the photorefractive effect itself.

The symbols  $\Delta \epsilon$  and  $\delta \epsilon$  will be used throughout this section to designate variations in the dielectric constant of the medium due to signal-induced response and to noise, respectively. In all cases considered in this section, the presence of a signal grating  $\Delta \epsilon(\mathbf{q})$  is attributed solely to the photorefractive effect. In particular, it is assumed that the Kerr effect makes no contribution to the signal as will be the case, for example, when the incident light intensities are low. To determine noise, all fluctuations having the form  $\delta\epsilon(\mathbf{q})$  need to be considered, independent of their source, as potential contributions. There are two sources: (i) the fluctuations inherent in the photorefractive effect as describe above, and (ii) thermal fluctuations in density, which through the Clausius-Mossotti constant  $\partial\epsilon/\partial\rho$  induce corresponding fluctuations in the dielectric constant of the medium independent of the photorefractive effect. Each of these sources will now be separately considered.

# A. Light-scattering noise associated with the photorefractive effect

The first source of grating fluctuations to be considered is fundamentally connected with the photorefractive effect itself. As noted above, this source arises from thermal fluctuations in the space-charge field  $\delta E_{SC}$ , which in turn produce fluctuations in the linear dielectric constant  $\delta \epsilon$  through the Pockels effect. Following the notation of Shen [16]

$$\delta \epsilon = \epsilon^{(2)} \delta E_{\rm SC} , \qquad (4.1)$$

where  $\varepsilon^{(2)}$  is related to the electro-optic tensor r and the refractive index of the medium  $n_R$ . The standard relations  $\Delta n = -\frac{1}{2}n_R^3 rE$  for the index change and  $\Delta \varepsilon = 2n_R \Delta n$  yield

$$\varepsilon^{(2)} = -n_R^4 r \ . \tag{4.2}$$

Combining Eq. (3.8) for the mean-square fluctuations in the space-charge field with Eq. (4.1) gives

$$\langle |\delta \epsilon(\mathbf{q})|^2 \rangle_{\rm PR} = (4\pi kT / \epsilon_0 V_S) [1 + (q / k_D)^2]^{-1} [\epsilon^{(2)}]^2 .$$
  
(4.3)

A subscript PR has been added to designate those fluctuations associated with the photorefractive effect. Equation (4.3) is the central result of the present study. It provides the extension of our previous investigations of light-scattering noise in Kerr media [4,5] to materials exhibiting the photorefractive effect.

The field-induced change in the dielectric constant follows from Eqs. (2.26) and (4.1). Squaring and averaging over the beam interaction volume gives

$$\langle |\Delta \epsilon(\mathbf{q})|^2 \rangle = \frac{1}{2} (kTq/e)^2 [1 + (q/k_D)^2]^{-2} [\epsilon^{(2)}]^2 m^2 .$$
  
(4.4)

Here there is no need to attach the PR designation since it is assumed throughout that only the photorefractive effect contributes to the signal grating.

An important quantity is the time-averaged signal-tonoise ratio defined here as [8]

$$(S/N)_{\rm PR} = \frac{\langle |\Delta\epsilon(\mathbf{q})|^2 \rangle}{\langle |\delta\epsilon(\mathbf{q})|^2 \rangle_{\rm PR}} = [kTq^2\epsilon_0 V_S m^2 / (8\pi e^2)][1 + (q/k_D)^2]^{-1} = (n_0 V_S / 2)[1 + (k_D / q)^2]^{-1} m^2 .$$
(4.5)

Equation (4.5) provides a useful comparison of the meansquare amplitudes of the signal and noise gratings in a photorefractive medium. Interesting features of the signal-to-noise ratio include its lack of a dependence on the light intensity other than through the modulation ratio m. In addition, there is no contribution from the Pockels coefficient, which cancels by contributing in the same fashion to both signal and noise. Finally, we have the unusual property that the signal-to-noise ratio increases with temperature—with direct proportionality obtained in the diffusion limit. This last finding may be traced to the temperature dependence in the space-charge field responsible for the signal grating [Eq. (2.26)].

As the intensity modulation ratio is reduced, the effects of noise become more dominant. A useful measure of noise dominance is provided by the condition  $m = m_1$  for which the signal-to-noise ratio equals unity. From Eq. (4.5) we obtain

$$(m_1^2)_{\rm PR} = [8\pi e^2 / (kTq^2\epsilon_0 V_S)][1 + (q/k_D)^2]$$
$$= (2/n_0 V_S)[1 + (k_D/q)^2]. \tag{4.6}$$

It follows that the best dynamic range (i.e., the smallest value of  $m_1$ ) is predicted for photorefractive materials operating in the diffusion limit  $(q/k_D)^2 \ll 1$ .

Equations (4.5) and (4.6) take on a particularly simple

form in the trap limit:

$$(S/N)_{\rm PR} = n_0 V_S m^2/2 \quad (\text{trap limit}) \tag{4.7}$$

and

$$(m_1^2)_{\rm PR} = 2/(n_0 V_S)$$
 (trap limit). (4.8)

It is important that Eqs. (4.7) and (4.8) be used only in the trap limit  $n_0 \ll \epsilon_0 kTq^2/(4\pi e^2)$ . Thus the signal-to-noise ratio cannot be improved over its value in the diffusion limit by simply increasing  $n_0$  in Eq. (4.7) beyond the trap limit range. The diffusion limit remains the most favorable condition for maximization of signal to noise.

## B. Light-scattering noise associated with the optical Kerr effect

The second source of grating fluctuations to be considered is fundamentally associated with the Kerr effect. These fluctuations are present whether or not the Kerr effect makes any contribution to the signal grating. A full discussion of light-scattering noise associated with the Kerr effect is presented elsewhere [6,7]. Here we need only to adapt these findings, summarized in the next paragraph, to the present case that the signal grating is

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formed by the photorefractive effect.

The nonlinear dielectric constant or Kerr coefficient  $\epsilon_2$  is defined by the relation

$$\boldsymbol{\epsilon}(\mathbf{r}) = \boldsymbol{\epsilon}_b + \boldsymbol{\epsilon}_2 \mathbf{E}^2(\mathbf{r}) , \qquad (4.9)$$

where  $\epsilon_b$  is the background dielectric constant at optical frequency ( $\epsilon_b = n_R^2$ ) and the overbar signifies averaging over an optical period. The mean-square amplitude, or variance, of the spontaneous dielectric fluctuations associated with the Kerr effect is given by the following expression [6]:

$$\langle |\delta\epsilon|^2 \rangle_K = 8\pi k T \epsilon_2 / V_S , \qquad (4.10)$$

where the subscript K designates fluctuations associated with the Kerr effect.

Kerr media have correlation lengths that are much shorter than an optical grating spacing and Eqs. (4.9) and (4.10) will provide a good description. Nevertheless there remain important cases where long-range correlations are important. Examples include fluids near a critical point and liquid crystals near a second-order isotropic-nematic phase transition. Equations (4.9) and (4.10) have been generalized to describe such cases [7] and it is useful to examine the generalized version of Eq. (4.10) in order to illustrate certain similarities with the fluctuations associated with the photorefractive effect. In Ref. [7] we found that as the correlation length approaches the grating spacing, both  $\epsilon_2$  and  $\delta \epsilon$  acquire increasingly strong wave-vector dependence. Using the Debye-Fixman square gradient model to evaluate the free energy of grating formation near a critical point we obtained [7]

$$\langle |\delta\epsilon(\mathbf{q})|^2 \rangle_K = 8\pi k T \epsilon_2(\mathbf{q}) / V_S$$
$$= (8\pi k T \epsilon_2 / V_S) [1 + (q/q_0)^2]^{-1} \qquad (4.11)$$

as the generalized version of Eq. (4.10), where  $q_0$  is the reciprocal of the correlation length. The squarebracketed correction for long-range interaction becomes important when the correlation length approaches the grating spacing. Structural similarity between Eqs. (4.3) and (4.11) is striking. Apart from the physical distinction between the correlation length and the Debye screening length in the two models, Eqs. (4.3) and (4.11) differ only by the nondimensional factor  $2\epsilon_0\epsilon_2/[\epsilon^{(2)}]^2$ .

For any medium having a finite optical Kerr coefficient, it is necessary to consider the fluctuations described by Eq. (4.11) as a potential source of light-scattering noise. For the case of a photorefractive material we have the relation

$$(S/N)_{K} = \frac{\langle |\Delta\epsilon(\mathbf{q})|^{2} \rangle}{\langle |\delta\epsilon(\mathbf{q})|^{2} \rangle_{K}}$$
$$= (S/N)_{PR} \langle |\delta\epsilon(\mathbf{q})|^{2} \rangle_{PR} / \langle |\delta\epsilon(\mathbf{q})|^{2} \rangle_{K} .$$
(4.12)

Evaluating the right-hand side using Eqs (4.3), (4.5), and (4.11) for the usual situations that  $q/q_0 \ll 1$ , we obtain

$$(S/N)_{K} = [kTq^{2}\epsilon_{0}V_{S}m^{2}/(8\pi e^{2})] \{ [\varepsilon^{(2)}]^{2}/2\epsilon_{0}\epsilon_{2} \}$$
$$\times [1 + (q/k_{D})^{2}]^{-2} . \qquad (4.13)$$

By analogy with Eq. (4.5), the effects of noise become dominant when the modulation ratio is reduced to the level  $m = m_1$  for which the signal-to-noise ratio equals unity:

$$(m_1^2)_K = [8\pi e^2 / (kTq^2\epsilon_0 V_S)] \{2\epsilon_0\epsilon_2 / [\epsilon^{(2)}]^2\} \times [1 + (q/k_D)^2]^2.$$
(4.14)

From Eqs. (4.13) and (4.14) it is seen that light-scattering noise is again minimized for photorefractive materials operating within the diffusion limit. In the following section we will examine the factor in curly brackets in order to compare the relative importance of light-scattering noise originating from thermal fluctuations associated with the Kerr and photorefractive effects for typical photorefractive media.

#### **V. ESTIMATES OF DYNAMIC RANGE**

The methods developed in the previous sections are illustrated in this section for the write and readout of a simple sinusoidal grating using the configuration shown in Fig. 3. For the writing-beam configuration of Fig. 3(a), the intensity modulation ratio is

$$m = 2(I_1 I_2)^{1/2} / (I_1 + I_2) , \qquad (5.1)$$

where  $I_1$  and  $I_2$  are the intensities for the pump  $(I_1)$  and the signal  $(I_2)$  beams. For light of wave vector  $K_1 = K_2 = K$ , the resulting grating is of wave vector  $q = |\mathbf{q}|$ :

$$y = 2K\sin(\theta/2) , \qquad (5.2)$$

where  $\theta$  is the angle between the writing beams. Both K and  $\theta$  are measured in the material with refractive index  $n_R$ . The dynamic range is defined as the range of intensity ratios that two writing beams can induce a detectable photoretractive grating.

In the read stage [Fig. 3(b)], the intensity of the diffracted beam (signal plus noise) is proportional to the square of the total grating amplitude

$$I_{\rm diff} \sim (a_1 + \delta a_1)^2$$
, (5.3)

where  $a_1$  and  $\delta a_1$  are the amplitudes of the signal and noise grating components, respectively. From Eqs (2.26) and (4.1),

$$a_1 = -(kTq/e)[1 + (q/k_D)^2]^{-1}\varepsilon^{(2)}m .$$
 (5.4)

The noise grating component is Gaussian distributed with mean

$$\langle \delta a_1 \rangle = 0$$
 (5.5a)

and variance

$$\langle |\delta a_1|^2 \rangle = 2 \langle |\delta \epsilon|^2 \rangle$$
 (5.5b)

The factor of 2 in Eq. (5.5b) corrects for averaging the square of the sinusoidal spatial dependence of  $\delta\epsilon$  over the beam interaction volume. The right-hand side of Eq. (5.5b) is evaluated using Eqs. (4.3) or (4.11), depending on

whether the noise in question is associated with the Kerr or photorefractive effects.

Experimentally, it is expedient to conduct measurements under the condition that  $q = k_D$  since it follows from Eq. (5.4) that the diffraction efficiency is maximized when this condition is satisfied. Under this condition the signal-to-noise ratio from Eq. (4.5) is  $n_0 V_{\rm S} m^2/4$ , which equals unity for a writing-beam intensity ratio  $I_2/I_1 = 1/(n_0 V_S)$ . Figure 4 shows a calculation of lightscattering noise in the diffracted read beam for BaTiO<sub>3</sub> under the maximum diffraction efficiency condition. The figure shows the logarithm of the diffracted read beam power [in arbitrary units due to the unknown prefactor in Eq. (5.3)] versus the logarithm of the writing-beam ratio  $I_2/I_1$ . The beam interaction volume was set equal to the product of the beam cross-section area and the interaction length. For the present calculation, the beam diameter was set at 0.15 cm and the interaction length at 0.2 cm, corresponding to the experimental measurement conditions used in Ref. [17]. Other parameters used in the calculation, which were chosen as representative of Ba-TiO<sub>3</sub>, are listed in Table I. As described below, the light-scattering fluctuations associated with the Kerr effect are relatively less important for BaTiO<sub>3</sub> and were omitted from the calculation of Fig. 4. This will not always be the case for other photorefractive materials as shown below.

To obtain Fig. 4, the right-hand side of Eq. 5.5(b) for the noise variance was evaluated using Eq. (4.3) with  $q = k_D$ . To generate the figure, the x coordinate (log writing-beam ratio) was randomly selected, with uniform distribution along the scale of the figure. Next, the signal grating amplitude  $a_1$  was obtained from Eq. (5.4) with m from Eq. (5.1). Finally, a value for  $\delta a_1$  was sampled from the Gaussian distribution having mean and variance specified by Eqs. (5.5a) and (5.5b). Gaussian probability sampling was achieved using a standard computer program incorporating the Box-Muller transformation for the generation of normal deviates from random numbers



FIG. 4. Light-scattering noise in BaTiO<sub>3</sub> as simulated using the stochastic noise model. The figure shows scatter due to intensity fluctuations in the diffracted read beam as a function of the writing-beam ratio  $I_2/I_1$ . The intensity fluctuations arise from light-scattering fluctuations in the medium inherent in the photorefractive effect.

TABLE I. Parameters for estimation of dynamic range. Values for  $n_R$ , r, and  $\epsilon_0$  are representative of BaTiO<sub>3</sub> from Ref. [4] and  $n_0$ , also for BaTiO<sub>3</sub>, is from Ref. [17]. The assignment for  $\epsilon_2$  is based on a recent measurement, which suggests that the Kerr coefficient for BaTiO<sub>3</sub> is comparable to that for CS<sub>2</sub> [19].

$n_R$	2.4 ( $\lambda = 0.514 \ \mu m$ )
r	1640 pm/V $(4.9 \times 10^{-5} \text{ erg}^{-1/2} \text{ cm}^{3/2})$
$\epsilon_0$	1000
$\epsilon^{(2)}$	$-1.6 \times 10^{-3} \text{ erg}^{-1/2} \text{ cm}^{3/2} (= -n_R^4 r)$
$\epsilon_2$	$4.8 \times 10^{-11} \text{ erg}^{-1} \text{ cm}^{3}$
Т	300 K
$n_0$	$4.8 \times 10^{16} \text{ cm}^{-3}$

sampled uniformly on the interval  $\{0,1\}$  [18]. The above procedure was repeated 1000 times to obtain an equal number of points for good statistical sampling of the noise.

Expansion of the right-hand side of Eq. (5.3) gives three terms, each corresponding to a different region seen in Fig 4. The first term  $(a_1^2)$  dominates in the high probe power (low noise) region. In this case, the linear behavior (with unit slope) predicted from Eq. (5.1) for  $I_2 \leq 0.1I_1$  is observed. As the probe power is reduced further, the next-to-leading term in the expansion  $(2a_1\delta a_1)$  begins to play a role. This term, which vanishes on time averaging, results in a symmetric distribution of scatter about the average diffracted beam power due to noise. This behavior is seen in the mild noise region of Fig 4. With still further reduction of probe power (i.e., reduction of  $a_1$ ) the third term in the expansion  $(\delta a_1^2)$  begins to dominate. This last term, which does not vanish on time averaging, results in a diffracted power due entirely to noise. The resulting scatter is independent of the power in the probe since in this region the signal grating amplitude is much smaller than the rms fluctuation amplitude due to noise. This effect is best seen in the smallest writing-beam ratio range of Fig. 4. The probe power level for which the signal-to-noise ratio is unity may be computed analytically from Eq. (4.6). For the present values of  $V_S$  and  $n_0$  we obtain  $I_2/I_1 = 5.9 \times 10^{-15}$  for  $(S/N)_{PR} = 1$ .

We turn next to the comparison of Eqs. (4.6) and (4.14) for their relative importance in limiting the dynamic range of a photorefractive medium. Setting  $(m_1^2)_K = \alpha (m_1^2)_{PR}$  we obtain for  $\alpha$ 

$$\alpha = \{2\epsilon_0\epsilon_2 / [\epsilon^{(2)}]^2\} [1 + (q/k_D)^2] = \{4\epsilon_0n_2 / (n_R^7 r^2)\} [1 + (q/k_D)^2] .$$
(5.6)

For BaTiO<sub>3</sub>, the parameters in Table I yield a value of 0.035 for the expression in curly brackets and dynamic range is limited by thermal fluctuations in the space-charge field. For different materials and/or different writing-beam configurations, the curly bracketed expression in Eq. (5.6) may easily exceed unity, in which case light-scattering noise associated with the Kerr effect will dominate. Values of  $\alpha$  are computed for several other photorefractive materials in Ref. [17]. There we find  $\alpha$  values of 0.93 for KNbO<sub>3</sub>, 1.56 for Ba<sub>x</sub>Sr<sub>2-x</sub>K<sub>1-y</sub>Na<sub>y</sub>Nb<sub>5</sub>O<sub>15</sub> (BSKNN), and 3.75 for Sr<sub>x</sub>Ba<sub>1-x</sub>Nb<sub>2</sub>O<sub>6</sub> (SBN); all for the maximum diffraction

efficiency condition  $q/k_D = 1$ . Thus for KNbO<sub>3</sub> and BSKNN we predict that the thermal light-scattering fluctuations associated with the Kerr and photorefractive effects are of comparable size, while for SBN fluctuations associated with the Kerr effect are of greater importance to limiting dynamic range.

Finally we note that there will be a photon shot-noise contribution arising from fluctuations in the incident intensity of the weak writing beam when the latter becomes sufficiently small that few photons from this beam arrive during the time required for photorefractive grating formation [20]. The photon shot-noise limits to the minimum writing-beam ratio were evaluated for each of the various photorefractive materials listed above and found to be in the  $10^{-16}$  to  $10^{-17}$  range [17]. Thus, for these materials, the photon shot noise is predicted to be about 1-2 orders of magnitude smaller than the thermal noise associated with either the Kerr or photorefractive effects. The results presented in this section suggest a very large dynamical range for photorefractive materials that should prove useful in optical signal processing applications.

#### VI. DISCUSSION

In this paper a thermodynamic description of the photorefractive effect has been presented within the framework of the Kukhtarev and hopping models. The Helmholtz free energy for grating formation was derived as a sum of separate contributions from energy storage in the space-charge field, configurational entropy, and intensity-dependent chemical potential of the donor (or hopping) sites, and found to be equivalent for the two models. Thermal fluctuations in the space-charge field were obtained from the curvature of the free-energy surface near its minimum value and shown to give rise to light-scattering noise fluctuations in the linear dielectric constant through the electro-optic effect.

Two sources of light-scattering noise in photorefractive materials were examined in the present study and found to be associated with either the photorefractive or Kerr effects. In both cases thermal fluctuations in the linear dielectric constant give rise to scattered light noise. In the former case, the dielectric fluctuations arise from fluctuations in the space-charge field through the electro-optic effect. In the latter case the fluctuations are those previously associated with the Kerr coefficient of the medium through the fluctuation dissipation theorem [5-7]. The corresponding expressions for the dielectric fluctuations [Eqs. (4.3) and (4.11)] are remarkably similar, especially when the photorefractive medium, with its Debye screening of the space-charge field, is compared to a Kerr medium with screened long-range correlations near a critical point [7].

Calculations were presented for the dynamic range of a photorefractive medium both analytically and through computer simulation using the stochastic noise model [7]. For BaTiO<sub>3</sub>, the dominant contribution to noise was found to be from those fluctuations associated with the photorefractive effect. Elsewhere we have shown that for KNbO<sub>3</sub> and BSKNN the contributions from noise associated with the Kerr and photorefractive effects are comparable in magnitude, while for SBN the dominant contribution is from fluctuations associated with the Kerr effect [17]. These results suggest a very large dynamic range for photorefractive media, approximately 140 dB for BaTiO<sub>3</sub> (cf. Fig. 4) and 120–140 dB for the other materials mentioned above.

The preceding analysis has shown that light-scattering noise in photorefractive materials is inseparable from the photorefractive effect itself in the same manner that light-scattering noise in Kerr media was previously shown to be inseparable from the Kerr effect [5-7]. As such the present study provides the natural extension of our previous investigations for Kerr and artificial Kerr media to materials exhibiting the photorefractive effect. Within each class of materials, the physical processes underlying signal generation and noise are equivalent, since without the fluctuations that give rise to the noise, no driven response of the nonlinear medium to applied optical signal fields can occur. This manifestation of the fluctuation dissipation theorem implies certain fundamental limits on the design of devices employing nonlinear optical media for signal processing applications, particularly in applications requiring miniaturization, weak signal amplification, high signal throughput, or high densities of information storage. Future studies will utilize the signal-to-noise ratio to evaluate the channel capacity for optical information processing using a photorefractive medium.

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FIG. 1. Comparison of the Kukhtarev and hopping models.