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# On the relationship between artificial Kerr nonlinearities and the photorefractive effect

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## Abstract

The photorefractive effect and artificial Kerr effect are both drift and diffusion based nonlinearities. This paper treats them in a unified fashion and develops a formalism for the analysis of general nonlinearities of this type. The intensity dependence of the steady state and response time is discussed.

## 1. Introduction

The photorefractive rate equations [1], now 30 years old, have been integral in supporting the growth of the field of photorefractive nonlinear optics. They have been used to model the strength and response time of the phase gratings responsible for nonlinear effects such as optical phase conjugation, image processing and spatial solitons. Over time, several researchers in the photorefractive field have begun to display an interest in another optically mediated effect: optical tweezers. It is may not be such a coincidence that Arthur Ashkin, one of the founders of the field of photorefractive nonlinear optics [2], also helped to found the field of optical tweezers [3]. Both of these effects are mediated by photoinduced motion of small particles: the first by charge migration induced by spatial gradients in the optical intensity, the second by forces induced by spatial gradients of optical intensity. Although the optical tweezers effect is usually thought of in terms of manipulation of microscopic particles such as silica or polystyrene microspheres and biological microorganisms and cells, it can also be thought of as a nonlinear optical effect. Many of the methods of measuring forces on trapped particles rely on measurement of the displacement of the trapped particle from the axis of the trapping beam. Commonly, this is done by using a split photodiode to monitor the deviation of the trapping light after passage through the trapped microsphere acting as a ball lens. It can be thought of as a case of light interacting with light via a medium with a nonlinear optical effect. Where does this nonlinearity come from? After all, neither the trapped microsphere nor the surrounding fluid medium has any appreciable nonlinearity by themselves.

It is the combination of the two components that results in the nonlinearity. The sphere moves in response to light in the fluid medium and causes a change in the spatial distribution of refractive index, just as spatial variations in optical intensity can drive the distribution or orientation of molecules in molecular orientational or electrostrictive Kerr effects. This is the basis of the artificial Kerr medium, which was studied extensively in the early 1980s [4, 5]. These include particle orientational nonlinearities [6] and nonlinearities of optomechanical arrays realized by beads rolling on transparent substrates [7]. Similar effects have been observed and studied in critical microemulsions [8, 9], phospholipid vesicles [10] and even phototactic microorganisms [11]. It is the aim of this paper to compare the Kukhtarev and hopping models for the photorefractive effect and the theory of optical tweezers or optophoretic nonlinearities.

## 2. Artificial Kerr medium

The equations of motion in an artificial Kerr medium consisting of a suspension of transparent nanoparticles include both drift and diffusion terms. The drift term may be found from models for the viscous flow of particles under the influence of optical gradient forces. Suppose the suspension is illuminated with an optical field propagating in the  $z$  direction:

$$E(\mathbf{r}, t) = E(r, t) \cos(kz - \omega t). \quad (1)$$

Then the optical gradient force in the dipole approximation for small particles in the direction  $x$  transverse to the propagation direction  $z$  is

$$F = \alpha E \frac{\partial E}{\partial x} = \frac{\alpha}{2} \frac{\partial E^2}{\partial x} = \alpha Z \frac{\partial I}{\partial x} = -\frac{\partial \varphi_{\text{opt}}}{\partial x}, \quad (2)$$

where  $Z$  is the impedance of free space,  $\alpha$  is the particle polarizability and  $\varphi_{\text{opt}}$  is the potential energy of the particle in the optical electromagnetic field.

The polarizability  $\alpha$  of a microsphere with volume  $V_p$  and permittivity  $\varepsilon_A$  in a surrounding material with permittivity  $\varepsilon_B$  is [12]

$$\alpha = 3\varepsilon_0\varepsilon_B \frac{\varepsilon_A - \varepsilon_B}{\varepsilon_A + 2\varepsilon_B} V_p. \quad (3)$$

The velocity  $v$  of a particle with radius  $a$  moving at low Reynolds number in a Newtonian viscous fluid with viscosity  $\eta$  under the influence of a force  $F$  is given by

$$v = \frac{F}{6\pi\eta a} \quad (4)$$

so that the drift term for current density is

$$J_{\text{drift}} = \rho v = \frac{\alpha Z \rho}{6\pi\eta a} \frac{\partial I}{\partial x} = -\frac{\rho}{6\pi\eta a} \frac{\partial \varphi_{\text{opt}}}{\partial x}, \quad (5)$$

where  $\rho$  is the number density of particles. The Stokes–Einstein diffusion current density is

$$J_{\text{diffusion}} = -\frac{k_B T}{6\pi\eta a} \frac{\partial \rho}{\partial x}. \quad (6)$$

The total current density equation is thus

$$J = -\frac{\rho}{6\pi\eta a} \frac{\partial}{\partial x} (\varphi_{\text{opt}} + k_B T \ln \rho). \quad (7)$$

Current conservation implies that the resulting equation for the complete model is

$$\frac{\partial \rho}{\partial t} = -\frac{\partial J}{\partial x} = \frac{\partial}{\partial x} \frac{\rho}{6\pi\eta a} \frac{\partial}{\partial x} (\varphi_{\text{opt}} + k_B T \ln \rho). \quad (8)$$

### 3. Kukhtarev rate equations

In the photorefractive effect, the optical field of equation (1) excites charge in an electro-optic crystal such as barium titanate. These charges move under the influence of drift and diffusion to form a refractive index distribution via the electro-optic effect. The charge migration and resulting refractive index grating can be successfully modeled by the photorefractive rate equations first introduced by Kukhtarev *et al* [1]:

$$\begin{aligned} \frac{\partial N_D^+}{\partial t} &= (sI + \beta)(N_D - N_D^+) - \gamma_R n N_D^+, \\ \frac{\partial n}{\partial t} &= \frac{\partial N_D^+}{\partial t} - \frac{1}{e} \frac{\partial J}{\partial x}, \\ \frac{\partial E_{\text{sc}}}{\partial x} &= \frac{(n + N_A - N_D^+)e}{\varepsilon}, \\ J &= \mu e n \left( E_{\text{sc}} - \frac{k_B T}{e} \frac{\partial \ln n}{\partial x} \right). \end{aligned} \quad (9)$$

In these equations it is assumed that it is electrons that are photoexcited by light with intensity  $I$  from traps to the conduction band where they can move with mobility  $\mu$ . Each

photoexcited electron (number density  $n$ ) results in an empty trap (number density  $N_D^+$ ) with net charge  $e$ . The total number density of traps (both empty and occupied) is  $N_D$ , and the average number density of traps ionized in the dark is  $N_A$ . The term  $\beta$  represents the thermally excited background of electrons in the conduction band. It can be thought of as resulting from an effective dark intensity  $I_d$ . The parameter  $s$  is the photoionization cross section and  $\gamma_R$  is the recombination constant.

The first equation in (9) describes optical excitation and recombination, the second equation is for current density and number density conservation, the third equation is Gauss's law relating charge density to space charge electric field  $E_{\text{sc}}$ , and the final equation represents the current  $J$  of photoexcited particles (electrons). The first term in the current equation accounts for drift and the second term accounts for diffusion. The last equation in (9) may be written as

$$J = -\mu e n \frac{\partial}{\partial x} \left( \varphi + \frac{k_B T}{e} \ln n \right), \quad (10)$$

where  $\varphi$  is the electrostatic potential. The model can be simplified if we make the approximations

- (1)  $\frac{\partial n}{\partial t} \ll \frac{\partial N_D^+}{\partial t}$  since the number density of electrons is much less than the number density of empty traps
- (2)  $N_D^+ \ll N_D$ : the empty trap density is much less than the total trap density
- (3) The number density of electrons can be regarded as quasi-steady state

$$n = \frac{(sI + \beta)N_D}{\gamma_R N_D^+}. \quad (11)$$

Writing the combined intensity term  $sI + \beta$  as  $I^*$  we have

$$J = -\frac{\mu e I^* N_D}{\gamma_R N_D^+} \frac{\partial}{\partial x} \left( \varphi + \frac{k_B T}{e} \log \frac{I^*}{N_D^+} \right) \quad (12)$$

so that the full model, taking current conservation into account, is

$$\frac{\partial N_D^+}{\partial t} = -\frac{\partial}{\partial x} \frac{\mu I^* N_D}{\gamma_R N_D^+} \frac{\partial}{\partial x} \left( \varphi + \frac{k_B T}{e} \log \frac{I^*}{N_D^+} \right). \quad (13)$$

### 4. The hopping model

In 1980, Feinberg *et al* proposed a hopping model for the photorefractive effect in which the probability for a charge carrier to hop from one site to another is proportional to the local light intensity  $I_n$ . Suppose the total number density of sites is  $N$ . The probability  $W_n$  that a charge carrier with charge  $q$  occupies the  $n$ th site at position  $x_n$  is written as

$$\frac{dW_n}{dt} = -\sum_m D_{mn} \left[ W_n I_n \exp\left(\frac{\beta\phi_{nm}}{2}\right) - W_m I_m \exp\left(\frac{\beta\phi_{mn}}{2}\right) \right], \quad (14)$$

where  $D_{mn}$  measures the tendency for a carrier to hop from site  $m$  to site  $n$ ,  $\beta = 1/(k_B T)$  and  $\phi_{nm} = q(\varphi_n - \varphi_m)$  is the electric potential difference between sites  $n$  and  $m$  so that  $\beta\phi_{nm}$  is the ratio of the electrostatic potential energy difference between

**Table 1.** Differential equations for artificial Kerr medium, and Kukhtarev and hopping photorefractive effect.

Artificial Kerr	$\frac{\partial \rho}{\partial t} = \frac{1}{6\pi\eta a} \frac{\partial}{\partial x} \rho \frac{\partial}{\partial x} (\varphi_{\text{opt}} + k_B T \ln \rho)$	$\varphi_{\text{opt}} = -\alpha Z I$
Kukhtarev	$\frac{\partial N_D^+}{\partial t} = -\frac{\mu N_D}{\gamma_R} \frac{\partial}{\partial x} \frac{I^*}{N_D^+} \frac{\partial}{\partial x} \left( \varphi + \frac{k_B T}{e} \ln \frac{I^*}{N_D^+} \right)$	$\frac{\partial^2 \varphi}{\partial x^2} = \frac{e}{\epsilon} (N_D^+ - N_A)$
Hopping	$\frac{\partial W}{\partial t} = \frac{Dq d^2}{k_B T} \frac{\partial}{\partial x} W I \frac{\partial}{\partial x} \left( \varphi + \frac{k_B T}{q} \ln W I \right)$	$\frac{\partial^2 \varphi}{\partial x^2} = \frac{Nq}{\epsilon} (W - W_0)$

sites  $n$  and  $m$  to the thermal energy  $k_B T$ . It can be shown that this equation reduces to

$$\frac{\partial W}{\partial t} = \frac{Dq d^2}{k_B T} \frac{\partial}{\partial x} W I \frac{\partial}{\partial x} \left( \varphi + \frac{k_B T}{q} \ln W I \right), \quad (15)$$

$$\frac{\partial^2 \varphi}{\partial x^2} = \frac{Nq^2}{\epsilon} (W - W_0)$$

assuming that  $D_{nm} = D_{mn} = D$ , the trapping centres are evenly spaced by  $d$  and only nearest neighbour hopping is included. The hopping current can be read from the first equation in (15) as

$$J_{\text{hop}} = -\frac{Dq d^2}{k_B T} W I \frac{\partial}{\partial x} \left( \varphi + \frac{k_B T}{q} \frac{\partial}{\partial x} \ln W I \right). \quad (16)$$

## 5. Comparison of models

Table 1 shows the partial differential equations for the three models side by side for comparison.

The hopping model differs from the Kukhtarev model in that the particle density multiplies the intensity in the former case, but divides it in the latter case. However after application of first order perturbation theory, as described below, the two models become identical in form.

Each of these models is in the form

$$\frac{\partial \rho}{\partial t} = A \frac{\partial}{\partial x} g(\rho, F) \frac{\partial}{\partial t} (U + k_B T \ln g(\rho, F)), \quad (17)$$

where  $A$  is a constant,  $\rho$  is a generalized particle density,  $g$  is a function of the particle density and optical energy source  $F$  and  $U$  is a potential energy density. When

- (1) optical excitation by a low visibility sinusoidal fringe pattern is assumed:  $I = I_0 + m I_0 \cos(kx)$  and
- (2) the model equations are linearized by taking first order perturbations in the variables, for example  $\rho \rightarrow \rho_0 + \rho_1$  where  $\rho_1$  is small, of order  $m$ ,

they become even more similar, as shown in table 2. Table 3 shows the resulting parameters for the three linearized models: steady-state response, time constant, sensitivity and refractive index grating amplitude. The first result from this comparison that can be read from table 3 is a relation between the hopping coefficient  $D$  and the Kukhtarev parameters

$$Dd^2 = \frac{s}{\gamma_R} \frac{k_B T \mu}{e N_A} \frac{N_D}{N_A}. \quad (18)$$

The counterintuitive result that the exponential risetime for artificial Kerr media is independent of intensity is consistent

**Table 2.** Linearized differential equations for nonlinearities driven by sinusoidal fringe patterns with visibility  $m$ .

	Linearized equation	
Artificial Kerr	$\frac{d\rho_1}{dt} + \frac{k_B T k^2}{6\pi\eta a} \rho_1 = \frac{k^2 \alpha Z \rho_0 m I_0}{6\pi\eta a}$	
Kukhtarev	$\frac{dN_D^+}{dt} + \frac{(sI_0 + \beta) N_D}{N_A E_\mu} (E_p + E_d) N_D^+ = -sm I_0 N_D \frac{E_d}{E_\mu}$	$E_d = \frac{k_B T k}{e}; \quad E_p = \frac{e N_A}{\epsilon k}; \quad E_\mu = \frac{\gamma_R N_A}{\mu k}$
Hopping	$\frac{dW_1}{dt} + \frac{D I_0 (kd)^2}{E_d} (E_p + E_d) W_1 = -D (kd)^2 W_0 m I_0$	

with the observation that the initial (time zero) slope is inversely proportional to intensity and the steady-state first order particle density is proportional to intensity. The time taken to reach steady state is proportional to the product of the initial slope and the steady-state particle density is independent of intensity. On the other hand, the photorefractive risetimes are inversely proportional to intensity, while the steady-state response is intensity independent, if the dark conductivity factor  $\beta$  is neglected. In each case, the sensitivity is independent of intensity. One of the main differences between the photorefractive and artificial Kerr nonlinearities is that the artificial Kerr nonlinearity is washed out by thermal fluctuations, whereas the photorefractive nonlinearity depends on thermal fluctuations. The photorefractive effect is diffusion driven, so while the optical excitation of charge carriers depends on the intensity of the light, they will not become redistributed to form a space charge in the absence of thermally activated diffusion. The optical artificial Kerr nonlinearity is directly optically driven.

## 6. Refractive index distribution and optical nonlinearity

Once the particle distribution is known, the refractive index distribution can be determined. In the artificial Kerr medium case, we can use Bruggeman's theory when the suspended microparticles have a dimension much less than the wavelength of the light being used [13]. This theory shows that the refractive index of a suspension of microspheres with volume fraction  $f = \rho V_p$  microspheres of volume  $V_p$  is given in terms of the dielectric constants by the relation

$$f \frac{\epsilon_A - \epsilon}{\epsilon_A + 2\epsilon} + (1 - f) \frac{\epsilon_B - \epsilon}{\epsilon_B + 2\epsilon} = 0, \quad (19)$$

**Table 3.** Characteristic parameters for the three models.

	Artificial Kerr	Kukhtarev	Hopping
Steady-state particle distribution	$\frac{\rho_1}{\rho_0} = \frac{\alpha Z m I_0}{k_B T}$	$\frac{N_{D1}^+}{N_A} = -\frac{s m I_0}{s I_0 + \beta} \frac{E_d}{E_d + E_p}$	$\frac{W_1}{W_0} = -m \frac{E_d}{E_d + E_p}$
Time constant $\tau$	$\frac{6\pi\eta a}{k_B T k^2}$	$\frac{1}{s I_0 + \beta} \frac{N_A}{N_D} \frac{E_\mu}{E_d + E_p}$	$\frac{1}{D I_0 (k d)^2} \frac{E_d}{E_d + E_p}$
Sensitivity (steady state/( $m I_0 \tau$ ))	$\frac{\alpha Z}{6\pi\eta a} k^2$	$\frac{s}{\gamma_R} \frac{k_B T \mu}{e N_A} \frac{N_D}{N_A} k^2$	$D d^2 k^2$
Steady-state refractive index perturbation $n_1$	$\frac{3}{2} \frac{(n_A^2 - n_B^2)^2}{n_A^2 + 2n_B^2} \frac{m I_0 \rho_0 V_p^2}{c k_B T}$	$\frac{1}{2} n_0^3 r_{\text{eff}} \frac{s m I_0}{s I_0 + \beta} \frac{i E_p E_d}{E_p + E_d}$	$\frac{1}{2} n_0^3 r_{\text{eff}} \frac{i m E_p E_d}{E_p + E_d}$

where  $\varepsilon_A = n_A^2$  is the relative dielectric constant of the microspheres and  $\varepsilon_B = n_B^2$  is the relative dielectric constant of the fluid in which the microspheres are suspended. A Taylor expansion of equation (19) shows that the linear approximation

$$\begin{aligned} \varepsilon &= f \varepsilon_A + (1 - f) \varepsilon_B = \varepsilon_B + f(\varepsilon_A - \varepsilon_B) \\ &= \varepsilon_B + \rho_0 V_p (\varepsilon_A - \varepsilon_B) + \Delta \rho V_p (\varepsilon_A - \varepsilon_B) \end{aligned} \quad (20)$$

is in error by at most 0.2% for polystyrene beads in water. It results in the following expression for refractive index perturbation  $n_1$ :

$$n_1 = \frac{1}{2} \frac{(\varepsilon_A - \varepsilon_B)}{\varepsilon_B} \rho_1 V_p. \quad (21)$$

Combining the expression for polarizability, equation (3), the expression for the first order particle distribution (table 3) and equation (21) we find

$$n_1 = \frac{3}{2} \frac{(n_A^2 - n_B^2)^2}{n_A^2 + 2n_B^2} \frac{m I_0 \rho_0 V_p^2}{c k_B T} = \frac{3}{2} \frac{(n_A^2 - n_B^2)^2}{n_A^2 + 2n_B^2} f m \frac{U_E}{k_B T / V_p}, \quad (22)$$

where  $U_E$  is the energy density of the optical field. The magnitude of the refractive index grating is proportional to the filling factor  $f = \rho_0 V_p$ , the fringe visibility  $m$  and the ratio of the optical energy density to the thermal energy density. If we choose microspheres of a size, say 20 nm, such that the scattering coefficient  $\mu_s$  is negligible then the thermal energy density is about 1 kJ m<sup>-3</sup>. For 100  $\mu$ m diameter beams with a power of 1 W, we find  $\Delta n \sim 5 \times 10^{-5}$  resulting in a four wave mixing phase conjugate reflectivity of the order of 0.1% for coupling lengths of the order 1 cm. Use of larger microspheres, more tightly focused beams, and lower temperatures will result in higher reflectivities. However the first two methods for increasing reflectivity will also increase scattering noise.

In both the Kukhtarev and hopping models of the photorefractive effect, the refractive index perturbation  $n_1$  is derived from the space charge field  $E_{sc}$  through the effective electro-optic coefficient  $r_{\text{eff}}$ :

$$n_1 = -\frac{1}{2} n_0^3 r_{\text{eff}} E_{sc}. \quad (23)$$

The space charge field is given in terms of the first order empty trap density by

$$E_{sc} = \frac{N_{D1}^+}{N_A} E_p = -m \frac{E_p E_d}{E_p + E_d} \quad (24)$$

using the following characteristic fields: diffusion field

$$E_d = \frac{k_B T k}{e} \quad (25)$$

and limiting space charge field

$$E_p = \frac{e N_A}{\varepsilon k}. \quad (26)$$

The space charge field is 90° out of phase with respect to the optical interference pattern. The phase shift is characteristic of the diffusion dominated photorefractive effect and is responsible for two beam coupling gain, an effect that is not seen in degenerate Kerr nonlinearities. Table 3 shows the refractive index perturbation for each case.

We are now in a position to explain the intensity dependence of these nonlinearities. Why is it that the steady-state photorefractive effect is independent of average intensity (neglecting the dark conductivity term  $\beta$  in the Kukhtarev model), while the steady-state artificial Kerr nonlinearity is proportional to intensity? Consider first the Kukhtarev model. A doubling of the driving intensity does lead to a doubling of the concentration  $n$  of electrons in the conduction band. The steady-state current, equation (10) is zero. This implies that  $\varphi + (k_B T/e) \ln n$  is constant. Doubling of  $n$  leads merely to the addition of a constant to the electrostatic potential  $\varphi$ . This makes no difference to the space charge field, since it is proportional to the gradient of the potential. This result can be read right away from the Kukhtarev differential equation model in table 1. This equation also shows that the speed of response is linearly proportional to the driving intensity. The same result can also be read from the differential equations in table 1 for the hopping model. The key observation is that the intensity appears in the logarithmic terms of both the hopping and Kukhtarev equations. Since the spatial derivatives of these logarithms appear, the doubling of intensity has no effect on the steady state. In contrast, the driving intensity does not appear in the logarithmic term in the artificial Kerr model of table 1, but rather directly in the potential  $\varphi_{\text{opt}}$ . In this case, doubling of the intensity leads to a doubling of the first order density perturbation and hence a doubling of the optical nonlinearity. This equation also shows that the response time is independent of intensity.

Finally, we note that the formalism developed here can be used to quickly analyse alternative nonlinearities. For

example, consider a model for an artificial Kerr medium with charged particles. Then corresponding differential equations can be immediately written down:

$$\begin{aligned}\frac{\partial \rho}{\partial t} &= \frac{1}{6\pi\eta a} \frac{\partial}{\partial x} \rho \frac{\partial}{\partial x} (\varphi_{\text{opt}} + \varphi_{\text{el}} + k_{\text{B}} T \ln \rho), \\ \varphi_{\text{opt}} &= -\alpha Z I, \\ \frac{\partial^2 \varphi_{\text{el}}}{\partial x^2} &= -\frac{e^2}{\varepsilon} (N_{\text{D}}^+ - N_{\text{A}}).\end{aligned}\quad (27)$$

When first order perturbations are taken, we find that the steady-state response is

$$\frac{\rho_1}{\rho_0} = m \frac{\alpha Z I_0}{k_{\text{B}} T} \frac{E_{\text{d}}}{E_{\text{d}} + E_{\text{p}}}. \quad (28)$$

The nonlinearity is proportional to intensity, as could be predicted from the absence of driving intensity in the logarithmic term and the space charge saturation by  $E_{\text{p}}$  is the same as in the regular photorefractive case. The response time is

$$\tau = \frac{6\pi\eta a}{kq} \frac{1}{E_{\text{d}} + E_{\text{p}}}. \quad (29)$$

## 7. Conclusion

Drift and diffusion optical nonlinearities can be described by a unified formalism of differential equations. This formalism can be used to analyse the steady state and temporal response of such nonlinearities, including artificial Kerr media and the Kukhtarev and hopping models for the photorefractive effect. Many properties of the nonlinearities can be quickly determined by the examination of the expressions for particle current.

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