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Nonlinear propagation of Gaussian beams in binary critical liquid mixtures

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We propose a theoretical study of the nonlinear propagation of a Gaussian laser beam in a mixture of two liquids. Retaining electrostriction (dipole radiation forces), thermal expansion, and thermodiffusion (Soret effect) as the major mechanisms in the perturbation of the medium by the field, we analyze their relative importances in the nonlinear refraction and nonlinear beam power losses. Our attention is mostly focused on the case of mixtures close to critical (consolute) points, where the nonlinearities are very large. We show that one crosses over from a situation dominated by thermal processes to a situation dominated by dipole radiation forces by diminishing the beam diameter and/or the distance to the critical point. The second case is shown to be well within the reach of realistic experiments. Finally, we discuss the potentialities of electrostriction as an alternative to sedimentation in the gravity field to study critical mixtures and the potentialities of critical mixtures as model media in nonlinear optical engineering.

I. INTRODUCTION

The optical nonlinearities of liquid suspensions of submicrometric particles have recently attracted a considerable interest. The electrostrictive forces resulting from the coupling between the wave and the dipole momentum induced on each particle by the field[1] can modulate the particle density and thus produce very large refractive index changes. For example, suspensions of latex spheres 100 nm in diameter feature nonlinear refractive indices n_2 about 10^5 times larger than that of CS_2.[2] The original behavior of such a system under various configurations of electromagnetic field has been theoretically analyzed[3] and could be used in various applications, for instance, phase conjugation[4] or harmonic generation.[5]

More generally, the electrostrictive forces can induce variations of concentration in any liquid mixture whose components have different indices of refraction. If the response of the medium is linear, the perturbation is proportional to the osmotic compressibility K_T (Ref. 6) and large optical nonlinearities are expected for highly compressible systems. The critical liquid mixtures are of particular interest in this respect, since K_T diverges near critical points.[7] The related divergence[8] of n_2 has been demonstrated for a critical microemulsion by self-focusing[8] or induced grating experiments.[9] n_2 values about 10^6 times that of CS_2 have been measured at about 0.1 K below the consolute (critical) temperature T_c of this particular system.

However, electrostriction is not the only possible source of optical nonlinearity of critical mixtures. Thermal processes can also cause large index variations. Light usually raises the temperature of the medium, due to the absorption by one (or more) component(s) of the mixture. This first causes a thermal expansion and a field-dependent variation of the index of refraction proportional to $\partial n/\partial T$ (T is the temperature).[10] Besides, temperature gradients can cause a spatial separation of the different components of the mixture by thermodiffusion, a process also known as the Soret effect.[11] The amplitude of the concentration gradient is given by the thermodiffusion ratio k_T/T, which is known to diverge near any consolute point.[12]

Temperature and concentration shifts affect both the real and imaginary parts of the index of refraction of the mixture ($n=n'+in''$). Critical mixtures generally feature large values of n'', even if the intrinsic absorbances of their components are very weak. In fact, much light from any incoming laser beam is scattered by thermally excited concentration fluctuations, a well-known phenomenon called "critical opalescence." The equilibrium value of n'' also goes to infinity near the critical point of the mixture.[13]

In this paper we analyze the behavior of a Gaussian laser beam propagating in a liquid binary mixture when thermal and electrostrictive processes are simultaneously taken into account.

In Sec. II we use the linearized hydrodynamic equations of binary mixtures to calculate the field-induced variations of the temperature T_E and of the concentration C_E in the medium. As we will see, the part of (C_E,T_E) due to electrostriction is everywhere proportional to the field intensity I (electrostriction is said to be "local"), while the part due to thermal processes depends on the whole intensity distribution around the point un-

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der consideration. In contrast to electrostriction, thermal nonlinearities are strongly nonlocal. From this we calculate the field-induced variations of the complex index of refraction (n'_E, n''_E).

Section III is devoted to solving the nonlinear equation of propagation of a Gaussian laser beam in a binary mixture. The calculations are worked out in the paraxial approximation,[14] so that the intensity profile of the beam is kept Gaussian everywhere in the sample. We arrive at a set of equations for the phase, the radius, and the intensity of the beam and make explicit the various terms corresponding to nonlinear refraction and nonlinear power losses due to electrostriction and thermal processes. As we will see, the essential difference between electrostriction and thermal processes (as for locality) is reflected in the distinct dependences of these terms as a function of the beam diameter.

In Sec. IV we analyze the specific case of a critical binary mixture. We examine the critical behaviors of the aforementioned terms and we comment on the validity of the previous approximations. In particular, we study the departures of electrostriction both from linearity and locality with respect to the field intensity close to T_c.

The final section (V) discusses the possibilities of using the nonlinear effects discussed in the former sections to improve our knowledge of critical mixtures, and, conversely, the possibilities of using critical mixtures as model systems in up-to-date nonlinear optical engineering problems.

This paper does not discuss longitudinal radiation pressure forces,[15] e.g., those involved for instance in particle levitation experiments. Within the frame of our analysis, which is essentially limited to linear responses as a function of the field intensity, longitudinal and transverse index shifts can be treated independently. Since we are mainly interested in the second ones to describe a self-focusing (or self-defocusing) situation, we will ignore the first ones. Besides, we believe that the longitudinal forces do not induce large concentration shifts C_E in critical mixtures (unless very close to the critical temperature) because the light scattering cross section in such media is, to first order, independent of C_E (see Sec. IV).

II. FIELD-INDUCED VARIATIONS OF THE INDEX OF REFRACTION AND OF THE TURBIDITY

The state of a binary liquid mixture is given by three variables, for instance, one component concentration C, temperature T, and hydrostatic pressure P. In this paper we are essentially interested in mixtures close to a *consolute critical point.* In other words, we will deal with situations where changing locally the concentration of one component of the mixture needs only a very small amount of energy. This means global density fluctuations (due to the *hydrostatic compression* of both species) are negligibly small compared to those due to concentration changes (corresponding to the *osmotic compression* of the mixture).[16] Things would of course be different if the system was close to a liquid-gas critical point. Let C_0 and T_0 be the values of C and T at equilibrium, i.e., in the absence of an electromagnetic field, and C_E and T_E the field-induced variations of the same variables:

$$T=T_0+T_E \ , \tag{1a}$$

$$C=C_0+C_E \ . \tag{1b}$$

C_E obeys a continuity equation:

$$\rho_0\frac{\partial C_E}{\partial t}=-\operatorname{div}\mathbf{i} \ , \tag{2}$$

where ρ_0 is the mass density of equilibrium and $\mathbf{i}$ is the field-induced mass flux. For weak perturbations, $\mathbf{i}$ is given by[16]

$$\mathbf{i}=-\alpha\nabla\mu-\beta\nabla T \ . \tag{3}$$

Here α and β are the mass and thermal transport coefficients, and μ is the difference of the chemical potentials of the two components of the mixture. Following the same notations as in (1):

$$\mu=\mu_0+\mu_E \ , \tag{4a}$$

where the field-induced part μ_E is given by the coupling energy:

$$\mu_E=-\frac{|E|^2}{8\pi\rho_0}\left[\frac{\partial n^2}{\partial C}\right] \ (\text{esu}) \ , \tag{4b}$$

where n is the index of refraction of the medium, and E the electric field.

To describe the coupling between C_E, T_E, and $|E|^2$ we use an approach similar to that of Lowdermilk and Bloembergen for gaseous mixtures.[17] Here the equations are simplified since we suppose a (hydrostatically) incompressible fluid. The combination of (3) and (4) gives

$$\mathbf{i}\simeq-\alpha\left[\frac{\partial\mu_0}{\partial C}\right]\nabla C_E-\alpha\nabla\mu_E-\left[\beta+\alpha\left[\frac{\partial\mu_0}{\partial T}\right]\right]\nabla T_E \ . \tag{5}$$

All the partial derivatives are taken at $C_0,T_0,E=0$. Combining (2) and (5) gives the mass diffusion equation:[18]

$$\frac{\partial C_E}{\partial t}=D\left[\nabla^2C_E+\frac{k_T}{T_0}\nabla^2T_E-\left[C^2\frac{\partial n^2}{\partial C}\Big/8\pi\rho_0\right]K_T\nabla^2|E|^2\right] \ . \tag{6}$$

$D=(\alpha/\rho_0)(\partial\mu_0/\partial C)$ is the mass diffusion coefficient and k_T is the so-called thermodiffusion ratio. k_T is related to D and to the thermal diffusion coefficient D_T through

$$k_TD=D_T=\frac{T_0}{\rho_0}\left[\beta+\alpha\left[\frac{\partial\mu_0}{\partial T}\right]\right] \ . \tag{7}$$

K_T is the osmotic compressibility of the mixture:

$$K_T=\frac{1}{C^2}\left[\frac{\partial\mu_0}{\partial C}\right]^{-1} \ . \tag{8}$$

We use the heat transfer equation to describe the variation of the temperature induced by the field:[18]

$$\rho_0 T\frac{\partial S}{\partial t}+\mathrm{div}(\mathbf{q}_t-\mu\mathbf{i})=Q\ . \tag{9}$$

$Q=(c_1 n_0/8\pi)\alpha_a|E|^2$ is the heat generated by the absorption of the wave in the medium.[17] α_a is the absorption coefficient, c_1 the velocity of light, and n_0 the linear index of refraction. $S=S_0+S_E$ is the entropy of the medium with a field-induced contribution S_E given by[19]

$$S_E=\frac{|E|^2}{8\pi\rho_0}\left[\frac{\partial n}{\partial T}\right]^2 . \tag{10}$$

q_t is the thermal flow which is given, to first order, by

$$\mathbf{q}_t=\left[k_T\left[\frac{\partial\mu_0}{\partial C}\right]-T_0\left[\frac{\partial\mu_0}{\partial T}\right]+\mu_0\right]\mathbf{i}-\Lambda_T\nabla T_E\ . \tag{11}$$

Λ_T is the thermal conductivity of the medium. Expressing the entropy evolution from first principles and keeping only the terms linear in E^2, one gets the thermal diffusion equation:

$$\frac{\partial T_E}{\partial t}-\frac{k_T}{C_p}\left[\frac{\partial\mu_0}{\partial C}\right]\frac{\partial C_E}{\partial t}+\frac{T_0}{8\pi\rho_0 C_p}\frac{\partial n^2}{\partial T}\frac{\partial|E|^2}{\partial t}=\frac{\Lambda_T}{\rho_0 C_p}\nabla^2 T_E+\frac{n_0 c_1}{8\pi\rho_0 C_p}\alpha_a|E|^2\ . \tag{12}$$

C_p is the heat capacity at constant pressure.

The solutions of the coupled equations (6) and (12) describe the evolution of $C_E(t)$ and $T_E(t)$ in a medium perturbed by a time varying field $|E|^2(t)$. In the following we will focus our attention on the static solution of (6) and (12). In fact, the nonlinearities in critical mixtures are very large and directly observable in steady-state conditions.[20] The generalization to dynamic effects is straightforward in that it essentially amounts to taking into account a relaxation of the form

$$\exp(-Dq^2 t)$$

for every q-wave vector component of the concentration. Strictly speaking one should also include thermal relaxation times, but these are usually very small compared to concentration relaxation times (the mass diffusivity gets vanishingly small close to a consolute point, a phenomenon known as the "critical slowing down of fluctuations," while the thermal diffusivity is nearly constant).

The zero-frequency limit of (6) and (12) reads

$$\nabla^2 T_E+\frac{n_0 c_1}{8}B_1|E|^2=0\ , \tag{13a}$$

$$\nabla^2 C_E-\frac{n_0 c_1}{8}B_1\frac{k_T}{T_0}|E|^2-B_2 K_T\nabla^2|E|^2=0\ , \tag{13b}$$

where

$$B_1=\frac{\alpha_a}{\pi\Lambda_T},\quad B_2=\frac{C^2(\partial n^2/\partial C)}{8\pi\rho_0}\ . \tag{13c}$$

Equation (13a) determines the spatial distribution of temperature induced by the field. In Eq. (13b), the last term corresponds to the electrostrictive contribution to C_E and is proportional to the osmotic compressibility. The second term is the Soret (or thermodiffusive) contribution proportional to k_T. Equations (13) show that the electrostriction is local, while the temperature profile and the related thermodiffusion-induced concentration profile are nonlocal functions of $|E|^2$ [see also Eq. (22)].

The field-induced shifts C_E and T_E result in a field-induced shift of the refractive index:

$$n_E\simeq\left[\frac{\partial n}{\partial C}\right]C_E+\left[\frac{\partial n}{\partial T}\right]T_E\ . \tag{14}$$

In this paper we deal with mixtures whose components absorb very weakly at the laser wavelength. The beam power losses are essentially due to light scattering by the concentration fluctuations of the mixture (the scattering gets very large close to a consolute point, whence the name "critical opalescence"). These losses are given by the so-called "turbidity" of the mixture τ, which is a function of both the osmotic compressibility and of the correlation length ξ of the concentration fluctuations. Assuming an Ornstein-Zernike[21,22] variation of the scattering cross section, τ is given by[13]

$$\tau=\frac{\pi^3}{\lambda_0^4\rho_0}\left[2n\frac{\partial n}{\partial C}\right]^2 k_B T(C^2 K_T)\Gamma(2(k\xi)^2)\ . \tag{15}$$

λ_0 is the laser wavelength in vacuum, and $k=2\pi n/\lambda_0$. k_B is the Boltzmann constant. The function Γ is defined by

$$\Gamma(x)=\frac{2x^2+2x+1}{x^3}\ln(1+2x)-\frac{2(1+x)}{x^2}\ . \tag{16}$$

When ξ is much smaller than λ_0, $\Gamma(2(k\xi)^2)\simeq\frac{8}{3}$.

Since τ depends on C and T, the turbidity takes on a nonequilibrium value given by

$$\tau=\tau_0+\tau_E\ , \tag{17a}$$

$$\tau_E\simeq\left[\frac{\partial\tau}{\partial C}\right]C_E+\left[\frac{\partial\tau}{\partial T}\right]T_E\ . \tag{17b}$$

At this stage, we have calculated the complex nonlinear index of refraction of the mixture from linear hydrodynamics. Consistency now requires us to insert the corresponding nonlinear polarization into Maxwell's equations, i.e., to solve a nonlinear propagation equation including n_E and τ_E. Even in simpler situations (for instance, with a simple local nonlinearity) the solution to this problem is known to be beyond the reach of analytical calculations. The problem is, however, greatly simplified if one assumes the shape of the laser transverse intensity profile is everywhere the same in the nonlinear medium. This condition amounts to imposing

$$|E|^2(r,z)=\frac{8}{n_0 c_1}\frac{P(z)}{a^2(z)}f\left[\frac{r^2}{a^2(z)}\right]\ , \tag{18}$$

where z is taken along the propagation axis, and r is the radial coordinate perpendicular to z. Equation (18) assumes that only the beam radius and the beam total power P can vary as a function of z. Practically, the invariance of the beam profile f along z is true whenever the effect of the nonlinear index of refraction on the propagation can be treated in a paraxial approximation,[14] in other words, when the nonlinearities are weak. We will then restrict our treatment to the weakly self-focusing (or self-defocusing) regimes, i.e., to beam powers much smaller than the self-focusing powers our calculations will ultimately lead to.

This in turn implies that the variations of E^2 with z are very slow compared to those with r. Under this condition, Eq. (13a) reduces to

$$\nabla_T^2 T_E(r,z) = -B_1 \frac{P(z)}{a^2(z)} f\left[\frac{r^2}{a^2(z)}\right], \tag{19}$$

where ∇_T^2 is the transverse Laplacian

$$\nabla_T^2 = \frac{1}{r}\frac{\partial}{\partial r} + \frac{\partial^2}{\partial r^2} .$$

Equation (19) has already been solved by various authors[23] interested in the heating of absorbing liquids or gases by Gaussian laser beams. Assuming cylindrical symmetry, one finds

$$T_E(r,z) = B_1 P(z) u\left[\frac{r}{a(z)}\right] . \tag{20}$$

u can be expanded as

$$u(x) = u_0\left[1 - \epsilon\frac{x^2}{a^2}\right] + \cdots . \tag{21}$$

The values of the coefficients in the expansion depend on the boundary conditions of the problem. For instance, one can impose the value of u for some multiple of the beam radius.[23]

Notice that the on-axis temperature $T_E(r=0,z)$ is proportional to the total beam power $P(z)$ rather than to the on-axis intensity. The concentration profile is now given by

$$C_E(r,z) = B_2 K_T |E|^2(r,z) - B_1 \frac{k_T}{T_0} P(z) u\left[\frac{r^2}{a^2(z)}\right] . \tag{22}$$

Combining (14), (17b), (20), and (22) we finally arrive at

$$n_E(r,z) = \left[\frac{\partial n}{\partial C}\right] B_2 K_T |E|^2(r,z) + \left[-\frac{k_T}{T_0}\left[\frac{\partial n}{\partial C}\right] + \left[\frac{\partial n}{\partial T}\right]\right] B_1 P(z) u\left[\frac{r^2}{a^2(z)}\right], \tag{23a}$$

$$\tau_E(r,z) = \left[\frac{\partial \tau}{\partial C}\right] B_2 K_T |E|^2(r,z) + \left[-\frac{k_T}{T_0}\left[\frac{\partial \tau}{\partial C}\right] + \left[\frac{\partial \tau}{\partial T}\right]\right] B_1 P(z) u\left[\frac{r^2}{a^2(z)}\right] . \tag{23b}$$

Since B_2 is proportional to $\partial n/\partial C$ [see Eq. (13c)], the first term in n_E is always positive, which means that electrostriction is always self-focusing. The thermal term contains a thermal expansion term $\partial n/\partial T$ and a thermodiffusion term

$$-\frac{k_T}{T_0}\left[\frac{\partial n}{\partial C}\right] .$$

Thermal expansion is most often defocusing ($\partial n/\partial T < 0$), but the sign of k_T depends on the particular system. Whether the overall thermal nonlinearity is self-focusing or self-defocusing is not known *a priori.*

The influence of the nonlinear turbidity is by no ways trivial. Predicting even the sign of the overall nonlinear power losses requires solving the nonlinear propagation equation. This is done in Sec. III.

III. NONLINEAR PROPAGATION OF A LASER BEAM IN A BINARY MIXTURE

We write the electric field in the form

$$\mathbf{E}(r,z,t) = \mathbf{e} A(r,z) \exp[i(\omega t - kz)] . \tag{24}$$

$A(r,z)$ is the steady-state complex amplitude, whose variation along z is supposed to be very slow on the scale of one wavelength. Under this assumption, the nonlinear propagation equation reduces to

$$2ik\frac{\partial A}{\partial z} = \nabla_\perp^2 A + k^2 P_{\mathrm{NL}} , \tag{25}$$

where P_{NL} is the nonlinear polarization corresponding to n_E and τ_E. Writing

$$A(r,z) = A_0(r,z) \exp[ik\Phi_0(r,z)] , \tag{26}$$

with A_0 real, Eq. (25) transforms into a set of two coupled equations for A_0 and Φ_0:

$$\frac{\partial A_0^2}{\partial z} = -\nabla_\perp \cdot (A_0^2 \nabla_\perp \Phi_0) - (\tau_0 + \tau_E) A_0^2 , \tag{27a}$$

$$2\frac{\partial \Phi_0}{\partial z} + \left[\frac{\partial \Phi_0}{\partial r}\right]^2 = \frac{\nabla_\perp^2 A_0}{k^2 A_0} + 2\frac{n_E}{n_0} . \tag{27b}$$

We now assume that the nonlinear medium fills the region $z > 0$ and that the beam is Gaussian. Besides, we will solve (27) in the paraxial approximation, a simplification which allows us to keep a Gaussian shape for the beam everywhere in the nonlinear medium. Then[24]

$$A_0^2(r,z)=\frac{8}{n_0c_1}\frac{P(z)}{a^2(z)}\exp\left[-\frac{r^2}{a^2(z)}\right], \tag{28a}$$

$$\Phi_0(r,z)=\psi(z)\frac{r^2}{2}+\Phi(z) . \tag{28b}$$

Let us, for instance, locate the beam waist in the $z=0$ plane. Then the initial conditions are

$$\begin{gathered}P(0)=P_0,\quad a(0)=a_0,\quad \left[\frac{\partial a}{\partial z}\right]_{z=0}=0 ,\\ \psi(0)=0,\quad \Phi(0)=0 .\end{gathered} \tag{29}$$

Inserting the paraxial developments of n_E and τ_E [Eq. (23)] into (27) and combining with (28), one gets a final set of coupled differential equations relating the power, the radius, and the phase of the beam along z:

$$\frac{P'}{P}=-\tau_0-Pb_4(1-\epsilon) , \tag{30a}$$

$$d''-d'Pb_4\epsilon=\frac{1}{d^3}\left[\frac{1}{k^2a_0^4}-P\left[\frac{2b_1}{n_0a_0^4}+\frac{2b_2\epsilon}{n_0a_0^2}d^2+\frac{\tau_0b_3}{2a_0^2}d^2+\frac{\tau_0b_4\epsilon}{2}d^4\right]\right] , \tag{30b}$$

$$\psi=\frac{d'}{d}-\frac{P}{2a_0^2d^2}(b_3+b_4\epsilon a_0^2d^2) , \tag{30c}$$

$$\phi'=-\frac{1}{k^2a_0^2d^2}+\frac{P}{n_0}\left[\frac{b_1}{a_0^2d^2}+b_2\right] . \tag{30d}$$

Here the primes denote derivations with respect to z, and $d(z)=a(z)/a_0$ is the normalized beam radius. The coupling constants b_1 to b_4 are given by

$$b_1=\frac{2}{\pi\rho_0c_1}\left[C\frac{\partial n}{\partial C}\right]^2K_T \quad \text{(electrostrictive part of } n_E) , \tag{31a}$$

$$b_2=\frac{\alpha_au_0}{\pi\Lambda_T}\left[\left[-\frac{k_T}{T_0}\right]\frac{\partial n}{\partial C}+\frac{\partial n}{\partial T}\right] \quad \text{(thermal part of } n_E) , \tag{31b}$$

$$b_3=\frac{2}{\pi\rho_0c_1}C^2\left[\frac{\partial n}{\partial C}\frac{\partial \tau}{\partial C}\right]K_T \quad \text{(electrostrictive part of } \tau_E) , \tag{31c}$$

$$b_4=\frac{\alpha_au_0}{\pi\Lambda_T}\left[\left[-\frac{k_T}{T_0}\right]\frac{\partial \tau}{\partial C}+\frac{\partial \tau}{\partial T}\right] \quad \text{(thermal part of } \tau_E) . \tag{31d}$$

From Eq. (30a) we notice that the power losses in the nonlinear mixture do deviate from the linear losses $[P(z)=P_0\exp(-\tau z)]$, but that the nonlinear losses are contributed to only by the thermal terms, not by electrostriction. The solution of Eq. (30a) reads

$$P(z)=\frac{P_0e^{-\tau_0z}}{1+\frac{P_0b_4}{\tau_0}(1-\epsilon)(1-e^{-\tau_0z})} . \tag{32}$$

The relative deviation of the power losses from the linear propagation limit is shown in Fig. 1 for various negative values of the parameter $P_0\tau_0^{-1}b_4(1-\epsilon)$. When $b_4<0$, thermal effects result in a self-induced transparency, which, for small values of b_4, is maximum at $z=(\ln 2)/\tau_0$. A positive b_4 results in a self-induced attenuation. As far as we know, such nonlinear losses have not been described before and have not been experimentally observed.

Equation (30b) describes the self-focusing or self-

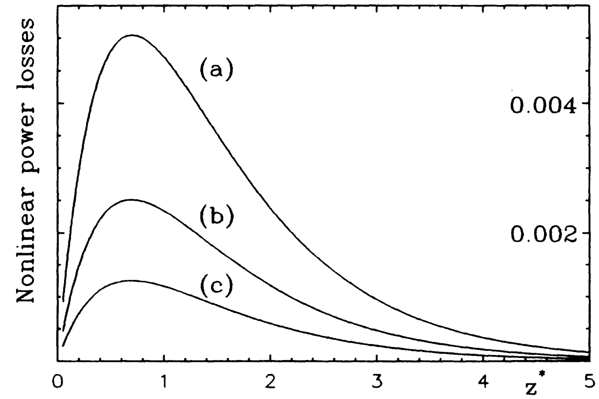

FIG. 1. Relative deviation $P(z^*)/P_0(z^*)-1$ of the beam power from its linear limit P_0 as a function of the reduced distance $z^*=\tau_0z$. τ_0 is the turbidity in the linear regime. The three curves correspond to three values of the parameter $K=P_0b_4\tau_0^{-1}(1-\epsilon)$. (a) $K=-0.02$. (b) $K=-0.01$. (c) $K=-0.05$.

defocusing of the beam. In order to get an insight into the roles of the various terms, we will introduce them successively. The linear propagation regime of course corresponds to all b_i's$=0$. In this case, (30b) reduces to

$$d''=\frac{1}{d^3(ka_0^2)^2} . \tag{33}$$

With the initial conditions given by (29), the solution of (33) is

$$d^2=1+\zeta^2 , \tag{34a}$$

with

$$\zeta=\frac{z}{ka_0^2} . \tag{34b}$$

This well-known result shows that the Fresnel length (ka_0^2) is the natural length scale in the linear diffraction regime.

If we now take into account the nonlinear refraction due to electrostriction ($b_1\neq 0$, $b_{i\neq 1}=0$), (30b) becomes

$$d''_\zeta=\frac{1}{d^3}\left[1-\frac{P_0}{P_{\rm th}}\right] , \tag{35a}$$

with

$$P_{\rm th}=\frac{n_0}{2b_1k^2} \tag{35b}$$

and

$$d^2=1+\left[1-\frac{P_0}{P_{\rm th}}\right]\zeta^2 . \tag{36}$$

Here $d''=d^2d/d\zeta^2$. Since b_1 is always positive, so is $P_{\rm th}$, which is just the self-focusing threshold power in the paraxial approximation. As is well known, true self-focusing is far beyond the domain of validity of the paraxial approximation.[25] In other words, Eq. (35a) has to be restricted to beam powers much smaller than $P_{\rm th}$.

Let us now introduce thermal nonlinear refraction ($b_1\neq 0$, $b_2\neq 0$, $b_3=b_4=0$). Equation (30b) becomes

$$d''_\zeta=\frac{1}{d^3}\left[1-\frac{P}{P_{\rm th}}\left[1+\frac{b_2\epsilon}{b_1}a_0^2d^2\right]\right] , \tag{37}$$

and d^2 is no more a simple quadratic function of ζ as before. It is, however, possible to define a formal threshold power $\tilde{P}_{\rm th}$ as

$$\tilde{P}_{\rm th}=\frac{n_0}{k^2(b_1+b_2\epsilon a_0^2)} . \tag{38}$$

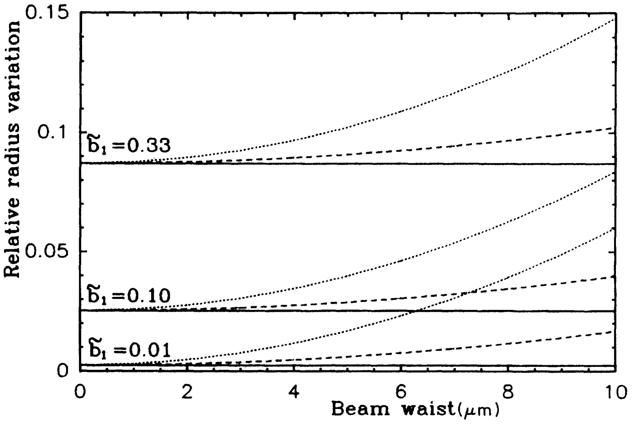

FIG. 2. Relative deviation of the beam radius $D=|d/d_0-1|$ after one Fresnel length in the nonlinear medium for different values of the electrostriction parameter b_1 and of the thermal parameter b_2. d_0 is the linear propagation limit of d and $\tilde{b}_1=(Pk^2/n_0)b_1$; $\tilde{b}_2=(P\epsilon k^2/n_0)b_2$. ——, $\tilde{b}_2=0$; — — —, $\tilde{b}_2=5\times10^{-4}$; · · · ·, $\tilde{b}_2=2\times10^{-3}\ \mu{\rm m}^{-2}$.

Notice that $\tilde{P}_{\rm th}$ now depends on a_0. This dependence is the direct consequence of the nonlocal character of the temperature profile as a function of the beam intensity. Equation (38) shows that the nonlinear refraction of large diameter beams is dominated by thermal effects. Whatever the power, increasing the beam diameter will ultimately lead to self-focusing if $b_2>0$. Figure 2 shows the results of a numerical integration of (37) in the paraxial regime ($P\ll P_{\rm th}$) at a distance equal to one Fresnel length from the beam waist ($z=0$). We have calculated the relative deviation $D(b_1,b_2)$ of the beam radius d_ζ from its value d_ζ^1 in the absence of the nonlinear effects:

$$D(b_1,b_2)=\frac{d_\zeta^1-d_\zeta}{d_\zeta^1} .$$

This deviation is plotted as a function of the beam waist, for three different values of both b_1 and b_2 (>0). Clearly $D(b_1,b_2)$ does not depend on a_0 in the electrostrictive limit ($b_2=0$) and is an increasing function of a_0 for $b_2>0$.

The value of the beam waist for which $D(b_1,b_2)=2D(b_1,b_2=0)$ can be viewed as the crossover radius $(a_0)_{\rm co}$ between the regime dominated by electrostriction $[a_0<(a_0)_{\rm co}]$ and the regime dominated by thermal effects $[a_0>(a_0)_{\rm co}]$. Clearly, $(a_0)_{\rm co}$ is an increasing function of the ratio b_1/b_2.

However, notice that a strong dependence of D on a_0 is not necessarily a definite signature of a nonlinear refraction dominated by thermal effects. In fact, nonlinear losses of electrostrictive origin (b_3) have the same form in (30b) as thermal nonlinear refraction. Separating these two effects requires additional information on the amplitude of b_3 relative to that of b_2. As we will see in Sec. IV, $b_3\simeq 0$ in critical mixtures.

Thermal nonlinear losses show up in Eq. (30b) under the form of two terms proportional to b_4. Since we essentially deal with the weakly nonlinear propagation regime, we can use the linear regime solution (34) to find out which term is the dominant one. We find that their amplitudes are similar for

$$z=2\tau^{-1} , \tag{39}$$

i.e., after two absorption lengths, which, in most practical situations, corresponds to a large number of Fresnel

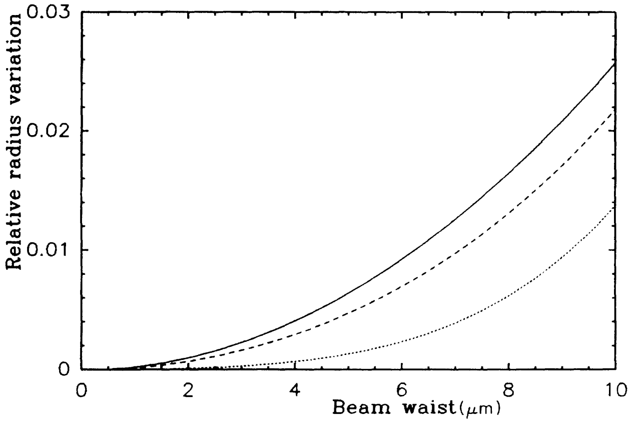

FIG. 3. Relative deviation of the beam radius after one Fresnel length with purely thermal nonlinear refraction and nonlinear power losses. The deviation is calculated for $\tilde{b}_2=9\times10^{-4}$ and for three values of $\tilde{b}_4=\frac{1}{2}P\epsilon\tau_0k^2b_4$. ——, $\tilde{b}_4=0$; — — —, $\tilde{b}_4=10^{-6}$; · · · ·, $\tilde{b}_4=3\times10^{-6}$.

lengths. In low Fresnel number geometries, the dominant nonlinear losses term in Eq. (30b) is the one in the left-hand side. One then expects a positive b_4 (self-induced attenuation) to exert a defocusing action. Figure 3 shows the beam radius deviation (b_2,b_4) (again for $\zeta=1$) due to purely thermal nonlinear refraction and losses. We have supposed both b_2 and b_4 positive. Clearly nonlinear losses tend to cancel out the self-focusing tendency of the refractive term (b_2), as we expected.

IV. CRITICAL MIXTURES

In this section we focus on the special case of critical binary mixtures. Our goal is to show that nonlinearities in these media can be made larger than in any other isotropic transparent medium, and that this performance can be achieved in domains where our linear analysis is valid and which moreover correspond to tractable experimental conditions. The complications due essentially to the saturation and to the nonlocality of electrostriction close to T_c are discussed in Sec. IV B.

A. Linear responses

In Sec. II we have used linear hydrodynamics to calculate the response of a binary mixture to the field intensity. The linear responses corresponding to the index of refraction and to the turbidity are given by the coefficients b_1 to b_4 [Eq. (31)]. Near a consolute critical point, the osmotic compressibility K_T is well known to diverge according to[22]

$$K_T\sim t^{-\gamma} . \tag{40}$$

Here t is the reduced temperature

$$t=\frac{|T-T_c|}{T_c} . \tag{41}$$

T_c is the critical temperature and γ is the so-called thermodynamic susceptibility exponent. K_T is proportional to the second moment of concentration fluctuations, which, in most practical situations, is given by the zero wave-vector light scattering cross section. Most of the experiments gave $\gamma\simeq1.24$, in agreement with modern theories of critical phenomena.[22]

The thermodiffusion constant k_T also goes to infinity near T_c. Although the critical behavior of k_T is not as well known as that of K_T, the most accurate experimental data[12] suggest that

$$k_T\underset{\sim}{\propto}\xi , \tag{42}$$

where ξ is the correlation length of the concentration fluctuations at equilibrium. ξ diverges according to

$$\xi\sim t^{-\nu} , \tag{43}$$

with $\nu\simeq0.62$.[22]

In addition to the linear responses (K_T and k_T), the other—potentially critical—quantities involved in Eq. (31) are the thermal conductivity Λ_T, the index of refraction n, and the turbidity τ. The available experimental data show that the critical variations of Λ_T (Refs. 26 and 27) and n (Ref. 28) in binary mixtures are very weak compared to those of K_T or ξ. We also expect the derivative $\partial n/\partial c$ to vary very gently close to T_c. Thus we expect the following critical behaviors:

$$b_1\propto t^{-\gamma} \tag{44}$$

and

$$b_2\underset{\sim}{\propto}t^{-\nu} . \tag{45}$$

Nonlinear losses (b_3 and b_4) are a more difficult problem because the turbidity τ depends on K_T and also on ξ. There is no general equation for the variations of ξ as a function of t and ΔC, although some tentative forms do exist.[29] For simplicity we will assume $\xi^2\propto K_T$, which is the behavior along the critical isochore. Standard scaled equations of state (see Appendix A) lead to $\partial K_T/\partial C=0$ for the critical concentration. We then expect

$$b_3\simeq0 . \tag{46}$$

We draw the same conclusion for the first term in Eq. (31d). In other words, linear electrostriction and thermodiffusion do not give rise to nonlinear power losses.

In the linear regime nonlinear power losses are entirely due to the direct heating of the sample by the laser beam. In fact,

$$b_4\propto\frac{\partial\tau}{\partial T}\propto t^{-1}\tau \tag{47a}$$

which, from Appendix A, gives

$$b_4\sim t^{-(\gamma+1)} \text{ far from } T_c(\xi\ll\lambda) , \tag{47b}$$

$$b_4\sim t^{-1}\ln t \text{ close to } T_c(\xi\gg\lambda) . \tag{47c}$$

If the heating by the beam drives the sample closer to its critical temperature (for instance, in the case of a "lower critical point" in the one phase region of the phase diagram), nonlinear losses consist in a self-induced attenuation. In the case of an "upper critical point" (again in the one phase region), we expect a self-induced transparency.

Equations (44), (45), and (47) suggest that nonlinear refractions and nonlinear power losses can be made as large as desirable at sufficiently short distances of the critical temperature. Let us now try to calculate a few orders of magnitude and decide what "close enough to T_c" practically means. We first estimate the amplitude of the electrostrictive part from the value of the paraxial electrostrictive self-focusing power [Eq. (35b)]. From (15) and (35b) we find

$$P_{\text{th}}=\frac{2\pi^2 c_1 n_0}{3\lambda_0}\frac{kT}{\tau} \tag{48}$$

in the limit of a small correlation length ($\xi \ll a$). Notice that Eq. (48) essentially relates the noise of the system (τ) to its thermodynamic susceptibility ($\propto P_{\text{th}}^{-1}$) and is just the consequence of the fluctuation-dissipation theorem. A typical example is the cyclohexane-aniline mixture, for which $\tau \sim 0.5\ \text{cm}^{-1}$ at $|T-T_c|\sim 1$ K. This gives $P_{\text{th}}\sim 1$ W and $n_2 \sim 10^{-10}\ \text{cm}^2\,\text{W}^{-1}$. Electrostrictively driven nonlinear refraction is then easily observable with molecular critical mixtures in typical conditions, and values of n_2 of the order of those obtained with polyballs suspensions can be reached with just a 10^{-2} K temperature control of the sample.

Let us now try to estimate the importances of the different contributions to the self-focusing equation (30b). The importance of thermodiffusion relative to that of electrostriction is given by the ratio b_2a^2/b_1. Clearly large beam diameters favor thermodiffusion, while the proximity to T_c should favor electrostriction. From (15), (31a), and (31b), we find

$$\left|\frac{b_2a_0^2}{b_1}\right| \simeq \frac{16\pi^3}{3}\frac{\alpha_a u_0}{\Lambda_T}\frac{c_1}{\lambda_0^4}\left(\frac{\partial n}{\partial C}\right)\left(\frac{k_T}{T}\right)\frac{k_BT}{\tau}a_0^2 \ . \tag{49}$$

Our typical example is again the aniline-cyclohexane mixture, for which values of both τ and k_T are available.[28] For $|T-T_c|\sim 1$ K, $\tau\sim 0.5\ \text{cm}^{-1}$ and $k_T \sim 20$. Taking $\partial n/\partial C \sim 0,1$, $\alpha_a \sim 10^{-3}\ \text{cm}^{-1}$, $\mu_0 \simeq 1$, $\lambda = 0.5 \times 10^{-4}$ cm, $\Lambda_T \sim 10^3\ \text{erg}\,\text{sec}^{-1}\,\text{m}^{-1}\,\text{K}^{-1}$, and $a_0 \sim 5 \times 10^{-4}$ cm, we find $b_2a^2/b_1 \sim 10$. We thus conclude that competing electrostriction and thermodiffusion should be easily observable over short distance (~ 1 Fresnel length) with usual critical mixtures.

The way in which electrostriction and thermodiffusion contribute to nonlinear refraction is shown in Fig. 4, where the self-focusing power P_{th} has been plotted as a function of t for different values of the ratio $b_2a_0^2\epsilon/b_1$. The variation of P_{th} crosses over from a $t^{-\nu}$ behavior to a $t^{-\gamma}$ behavior in a region which gets closer to T_c when $b_2a_0^2\epsilon/b_1$ increases. However, the paraxial self-focusing power is not directly accessible to experiments. A more practical picture is the relative variation D of the reduced beam radius d at some finite reduced distance ζ again as a function of t and for different values of $b_2a_0^2/b_1$ (Fig. 5). Clearly the critical increase of D near T_c is sharper when electrostriction overcomes thermodiffusion.

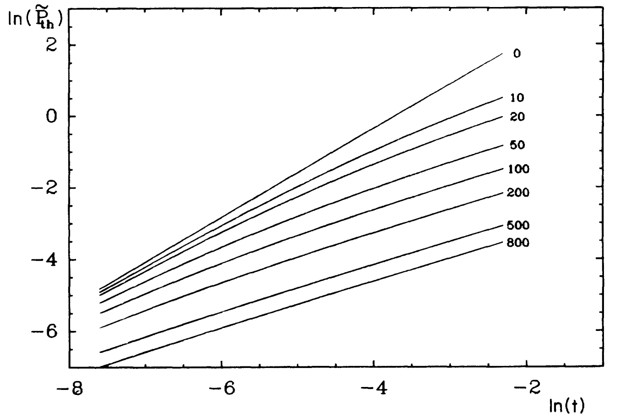

FIG. 4. ln-ln plot of the paraxial self-focusing power as a function of the distance to the critical temperature. The value of the ratio $b_2a_0^2\epsilon/b_1$ is indicated at the right of each curve.

As we discussed in Sec. III, we expect nonlinear losses to be essentially due to the term in the left-hand side of Eq. (30b) in low Fresnel number geometries. The importance of this term relative to the refractive thermodiffusion term is given by the ratio

$$dd'b_4n_0a_0^2/2b_2 \ ,$$

which is upperly bounded by

$$\frac{\dfrac{\partial\tau}{\partial T}}{\left(-\dfrac{k_T}{T}\right)\dfrac{\partial n}{\partial C}}\left(\frac{\lambda_0}{2\pi}\right)\zeta \ .$$

With the same numerical values as before, we find a ratio of the order of $6\times 10^{-4}\zeta$, suggesting that the nonlinear

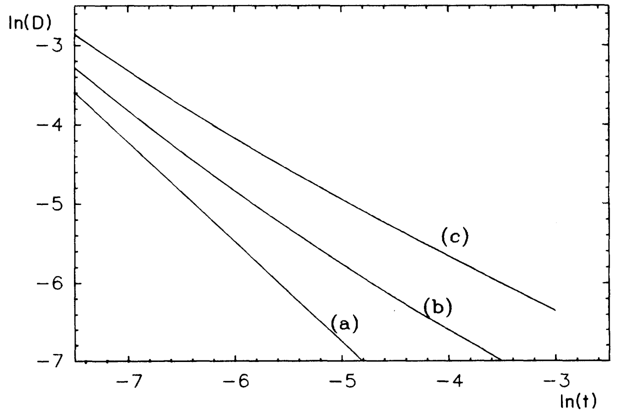

FIG. 5. ln-ln plot of the deviation of the reduced beam dimension from its linear propagation limit as a function of the distance to the critical temperature, after one Fresnel length in the nonlinear medium. The values of $b_2a_0^2\epsilon/b_1$ are 0, 30, 90 for curves (a), (b), (c), respectively.

losses do not significantly affect the beam diameter evolution in "typical" conditions. However, notice that this conclusion would not be valid very close to the critical temperatures, since

$$\frac{1}{k_T}\frac{\partial\tau}{\partial T}$$

diverges near T_c.

B. Limitations

In Sec. IV A we have seen that most of the coefficients of the linear hydrodynamic theory go to infinity at the critical point. Since infinite responses are clearly unphysical, we expect saturation effects to show up near T_c and/or for large beam intensities. Such saturation effects of course correspond to terms higher than cubic in the polarization expansion as a function of the electric field.

Electrostriction being a conservative process, one can use an equilibrium equation of state of the mixture to estimate the range of temperatures and field intensities within which the electrostrictive response is linear. One particularly simple case is the ideal solution, where

$$C\frac{\partial\mu}{\partial C}=k_B T\ . \tag{50}$$

Writing $\mu=\mu_0+\mu_E=\text{const}$, with μ_E given by (4b), one finds the result Palmer[1] worked out for an ideal suspension of polyballs. The electrostrictively induced concentration change is found to saturate for field intensities larger than a crossover value given by

$$|E^2_{\text{sat}}|=\frac{8\pi\rho_0}{\dfrac{\partial n^2}{\partial C}}k_B T\ . \tag{51}$$

The thermodynamic properties of fluids in their critical regions are well accounted for by a very general "scaled equation of state" of the form[22,30]

$$\Delta\mu=\Delta C|\Delta C|^{\delta-1}h\left[\frac{t}{|\Delta C|^{1/\beta}}\right], \tag{52}$$

$$\Delta C=C-C_0\ . \tag{53}$$

ΔC is the concentration difference from the critical concentration C_0, and

$$\Delta\mu=\mu-\mu(C_0) \tag{54}$$

is the corresponding chemical potential shift. The definitions of the β and δ exponents are given in Appendix A, together with the asymptotic behaviors of the function h. We now find (see Appendix A for the proof) a "saturating field"

$$|E^2_{\text{sat}}|\propto t^{\gamma+\beta}\ . \tag{55}$$

This relation defines the domain of validity of the linear approach we have worked out in Sec. II for electrostriction. All the proportionality constants entering Eq. (52) through (A4) are nonuniversal. Thus Eq. (55) can be quantitatively used only for comparison with one particular point (say, one temperature) where the linearity of the response as a function of the field intensity is already known. Notice that, since $\beta>0$ ($\beta\simeq0.35$), the crossover intensity $|E^2_{\text{sat}}|$ vanishes faster than K_T^{-1} when one gets close to T_c. This means that any experiment designed for testing our predictions for linear electrostriction has to be restricted to vanishingly small effects (concentration or refractive index shifts) near T_c, although the amplitude of the linear response (K_T) gets increasingly large in this region.

In contrast to electrostriction, thermodiffusion is a dissipative process and we have no equivalent to a full equation of state relating C_E (thermodiffusion) to E. Clearly, the nonlinear Soret effect is an open research field. Thus we are not able to produce a criterion such as Eq. (55) for the linearity of thermodiffusion. We are just left with the hope that absorbances of usual critical mixtures in the visible wavelengths range are small enough ($\alpha_a\sim10^{-3}$ cm^{-1} is typical) for the concentration gradients to be proportional to the temperature gradient.

Aside from this cause of saturation, a different limitation of the analysis worked out in the former sections arises when the correlation length ξ is not very small compared to the transverse dimension of the beam (a). This can be easily understood by realizing that ξ is a cutoff length in the spatial response of the fluid to a local perturbation of its concentration. When $\xi\sim a$, electrostriction itself becomes nonlocal and "saturates," although the response of the fluid remains linear as a function of $|E^2|$. Since ξ also diverges [Eq. (43)], we expect this sort of complication to show up near T_c.

The paraxial nonlocal electrostrictive self-focusing power is calculated in Appendix B. One finds

$$P_{\text{th}}=P^{\text{loc}}_{\text{th}}F\left[\frac{a}{\xi}\right], \tag{56}$$

where $P^{\text{loc}}_{\text{th}}$ is the power given by Eq. (35b). The factor $F(a/\xi)$ (see Appendix B) is larger than 1 and of course tends to 1 for $\xi\ll a$. Taking $a=5$ μm as a typical value and $\xi_0=2$ Å as an example of a molecular cutoff length (e.g., the value of ξ very far from T_c) in a molecular binary mixture, we expect severe nonlocal effects to show up within $\Delta T\sim0.7$ mK of T_c(~300 K). Macromolecular solutions[31] or microemulsions[32] feature larger cutoff lengths: $\xi_0\sim50$ Å gives $\Delta T\sim0.12$ K, a domain which is more within the reach of common experiments. Notice that nonlocality of electrostriction is not at all a marginal phenomenon in the case of polyballs suspensions,[2] since these feature cutoff lengths of the order of 0.1 μm whatever the temperature. In this case one dramatic manifestation of both nonlinear saturation and nonlocality is the irreversible clumping of polyballs in a self-focused beam.[2]

V. CONCLUSION

In this paper we have discussed various aspects of the nonlinear interaction of light with a critical mixture.[33] Among the fundamental studies of the critical properties of liquid-liquid mixtures, a few attempts have been made to use the gravity field **g** for the experimental determination of equations of state.[22,34] However, because **g** interacts with the whole volume of the sample, equilibration times are usually huge (~1 yr). Moreover, actual concentration profiles often severely deviate from expected ones because of stray temperature gradients and surface convections.[35] Electrostriction can be viewed as a sedimentation process within or outside of a laser beam. Interaction volumes can be made small, with the twofold advantage of tractable equilibration times (~ minutes) and lower sensitivity to surface-induced anomalies. The counterparts of these advantages are the complications due to laser-induced heating of the sample, to nonlinear power losses, and to the nonlocality of the concentration profiles versus the beam intensity close to the critical temperature. However, it is our hope that these different mechanisms can be separated using the conclusions drawn in Secs. III and IV, i.e., using their distinct behaviors as a function of the temperature and of the beam diameter.

On the other hand, critical mixtures are promising model systems for current nonlinear optics problems, including bistable behavior,[36] spatial soliton propagation, etc.[37] Here the specific advantage of using a critical mixture is its slowness compared to the very short response times (picoseconds) involved in systems prone to applications in nonlinear optical engineering. However, a model nonlinear medium is all the more useful as it is simple. In practice this requires the nonlinearity to reduce to a local third-order susceptibility, which in turn requires local linear electrostriction to be the leading mechanism in the nonlinear interaction of light with the mixture. As discussed in the former sections, such a condition can be met by a proper choice of the temperature and of the beam diameter, which has to be small enough for thermal effects to be negligible, but much larger than the correlation length of the concentration fluctuations.

Finally we wish to mention that convection can be a major complication of the picture drawn from our analysis. In fact, any situation where the laser beam heats the medium implies horizontal temperature gradients and consequently horizontal concentration and mass gradients. Such situations are known to trigger convection, whatever the amplitude of the gradients[38] (there is no threshold, in contrast to Rayleigh-Benard geometries). Convection can be made small if solid walls are located close to the heated volume, but not completely blocked. These convective flows can probably be detected by means of dynamic light scattering. We are currently developing an experiment using this method.

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APPENDIX A: SCALED EQUATION OF STATE AND SATURATION OF ELECTROSTRICTION

The so-called scaled equation of state reads

$$\Delta\mu=\Delta C|\Delta C|^{\delta-1}h(t|\Delta C|^{-1/\beta}) \; . \tag{A1}$$

ΔC is the shift from the critical concentration and $\Delta\mu$ is the corresponding chemical potential shift. t is the reduced temperature [Eq. (41)] and

$$\Delta\mu\propto\Delta C|\Delta C|^{\delta-1} \tag{A2}$$

is the equation of the critical isotherm ($t=0$) and

$$t|\Delta C|^{-1/\beta}=\text{const} \tag{A3}$$

is the equation of the (two phases) coexistence curve. The values of β and δ are about 0.35 and 4.6 in the Ising ($n=1$, $d=3$) universality class.[22] The function h has simple asymptotic forms:

$$h(x)\simeq\sum_{i=0}^{\infty}h_i x^i \quad \text{for } x\simeq 0 \; , \tag{A4}$$

$$h(x)\simeq\sum_{i=0}^{\infty}k_i x^{\beta(\delta+1-2i)} \quad \text{for } x\simeq\infty \; . \tag{A5}$$

The response of the fluid to an electromagnetic field through electrostriction is obtained by equating $\Delta\mu$ to μ_E [Eq. (4b)] and ΔC to C_E [Eq. (1)]. Strictly speaking, Eq. (A1) is valid only for concentration shifts which are uniform in space. Taking into account the finite size of the perturbation induced by the laser beam would need a nonlocal equation of state, which in fact is not available. Fortunately a local description should be valid in situations when the beam radius a is much larger than the correlation length ξ for the concentration fluctuations at equilibrium. Typical beam radii are of the order of a few micrometers. Reported experimental values of ξ in binary mixtures are most often much shorter,[22] so that treating electrostriction as local seems quite a realistic approximation. However, ξ diverges as $t^{-\nu}$ [with $\nu\simeq 0.62$ (Ref. 22)] so that locality will ultimately break down close to T_c. In the following we derive the conditions under which electrostriction is linear as a function of the beam intensity and ignore problems related to nonlocality.

From (A1), (A4), and (A5) we deduce the following two asymptotic regimes.

(i) $t|C_E|^{-1/\beta}\gg 1$ (far from T_c or small fields). From (A5),

$$C_E\propto E^2 t^{-\beta(\delta-1)} \; . \tag{A6}$$

With the definitions $\gamma=\beta(\delta-1)$ and

$$K_T=\frac{1}{C^2}\left[\frac{\partial\mu}{\partial C}\right]^{-1} \tag{A7}$$

for the osmotic compressibility, we obtain a linear response whose amplitude diverges as $t^{-\gamma}$ ($\gamma\simeq 1.24$).

(ii) $t|C_E|^{-1/\beta}\ll 1$ (close to T_c or large fields). From (A4),

$$C_E\propto(E^2)^{1/\delta} \; . \tag{A8}$$

In this regime, the response is strongly nonlinear. The crossover from the linear to the nonlinear regime occurs for intensities or temperatures which are related through

$$E_{\mathrm{co}}^2 \propto t^{\gamma+\beta} . \tag{A9}$$

APPENDIX B: LINEAR NONLOCAL ELECTROSTRICTION

Nonlocality arises from the fact that concentration inhomogeneities cost some amount of free energy. This is reflected in the following formulation of a generalized chemical potential:[39]

$$\Delta\mu = \Delta\mu_0(C) - \left[\frac{\partial \Delta\mu_0}{\partial \Delta C} \right]_{\Delta C=0} \xi^2(\nabla^2 C) + \cdots . \tag{B1}$$

Here $\Delta\mu_0$ is the local chemical potential. The resulting C_E is no more proportional to E^2 and is rather given by the spatial convolution product:

$$n_E(r,z) = b_1 E^2(r,z) \circ R_1(r,z) . \tag{B2}$$

The spatial response function $R_1(r,z)$ is proportional to the correlation function for the concentration fluctuations at equilibrium, as required by the fluctuation-dissipation theorem. Since

$$\left| \frac{\partial E^2}{\partial z} \right| \ll \xi^{-1} E^2 ,$$

Eq. (B2) reduces to

$$n_E(r,z) = b_1 E^2(r,z) \circ R_2(r) \tag{B3}$$

with

$$R_2(r) = \frac{1}{4\pi\xi^2} \exp(-r/\xi) . \tag{B4}$$

The paraxial development of n_E reads

$$n_E(r,z) \simeq b_1 \frac{P(z)}{a^2(z)} \left[\mu_1(x) - \mu_2(x) \frac{r^2}{a^2(z)} \right] , \tag{B5}$$

where $x = (a/2\xi)^2$ and

$$\mu_1(x) = x e^x E_i(x) , \tag{B6}$$

$$\mu_2(x) = [1 - \mu_1(x)] x . \tag{B7}$$

E_i is the exponential integral function.[40] It is easy to check that $\mu_1 = \mu_2 = 1$ when $\xi = 0$, so that one recovers the local expression (23a) for n_E.

The propagation equations for the beam diameter and for the phase are now

$$d'' = \frac{1}{d^3} \left[\frac{1}{k^2 a_0^4} - P \frac{2 b_1 \mu_2(x)}{a_0^4 n_0} \right] , \tag{B8}$$

$$\phi' = \frac{1}{k^2 a_0^2 d^2} + P \frac{b_1 \mu_1(x)}{n_0 a_0^2 d^2} . \tag{B9}$$

The paraxial self-focusing power is given by

$$P_{\mathrm{th}} = P_{\mathrm{th}}^{\mathrm{loc}} [\mu_2(x)]^{-1} . \tag{B10}$$

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