

Concentration mechanism for optical self-effects in stratifying liquid solutions

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A concentration mechanism is predominant in the self-focusing of light near the stratification point of liquid solutions. The threshold for the time-varying effect is inversely proportional to the kinetic mobility and is reached at moderate energy densities. Calculations are carried out for the optically induced shift of the critical temperature (a concentration self-induced transparency).

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1. When intense light beams propagate through multicomponent liquids, the thermal, electrostrictive, and orientational mechanisms for nonlinearities should be accompanied by some mechanisms which are peculiar to mixtures. These mechanisms involve (a) optically induced chemical reactions¹ and (b) optically induced redistributions of the concentration of the polarizing molecules.^{2,3} In this letter we will propose a time-varying concentration mechanism for self-focusing, and we will discuss some new physical information which can be extracted from a corresponding experiment.

The effect is of the same nature as its analog in a constant field: Molecules with a high polarizability accumulate in regions of a high electric field⁴ E , so that the concentration mechanism leads to self-focusing. The maximum change in the refractive index is $n_1 - n_2$ (n_i are the refractive indices of the components) and does not exceed values ~ 1 .

It follows from a thermodynamic analysis that $\Delta n \sim (\partial n / \partial X) \Delta X$ and $\Delta X \sim (\partial X / \partial Y) \langle E^2 \rangle$, where X and Y are the coordinate and the conjugate force. The threshold parameter for self-focusing, $\sim (\Delta n)^{-1}$, therefore decreases near the point of the phase transition, for which \pm is an order parameter: At this point, $\partial Y / \partial X \rightarrow 0$. For the concentration mechanism, X and Y are the concentration c (in a binary mixture we would have $c_1 = c$ and $c_2 = 1 - c$) and the specific chemical potential $\mu = N_A (\mu_2 / M_1 - \mu_2 / M_2)$ (μ_i and M_i are the chemical potentials and molecular masses of the components, and N_A is Avogadro's number), and the best place for observing self-focusing is near the stratification point. At this point we have $\partial \mu / \partial c \rightarrow 0$, and the concentration mechanism is predominant. In this region, a measurement of the temperature dependence or concentration dependence of the threshold or of the self-focusing length can yield the critical indices and can be used to calculate the kinetic mobility L (which has not been measured directly).

2. The concentration mechanism is described by the following dependence of μ on the intensity $|E|^2$:

$$\mu = \mu_0 - \mu_E, \quad \mu_E = (n_0 / 8\pi\rho_0) (\partial n / \partial c)_{p,T} |E|^2, \quad (1)$$

where the subscript 0 designates the parameters in a weak field. The diffusion flux is

$\mathbf{j} = -L \text{ grad} \mu$, and the continuity equation is

$$\rho_0 (\partial c / \partial t) = L \nabla^2 [(\partial \mu / \partial c) c - \mu_E |E|^2]. \quad (2)$$

The changes in the amplitude of E are described by the quasi-optical equation

$$[2ik_0 (\partial / \partial z + v_g^{-1} \partial / \partial t) - \nabla^2] E = (2k_0^2 / n_0) (\partial n / \partial c)_{p,T} c E, \quad (3)$$

where k_0 is the wave number, ∇^2 is the transverse Laplacian, and v_g is the group velocity. Near the stratification point the diffusion coefficient is $D = (L / \rho_0) (\partial \mu / \partial c) \sim \Theta^\nu$, where $\Theta = |T - \tilde{T}| / \tilde{T}$, \tilde{T} is the critical temperature, and ν is the index describing the divergence of the fluctuation correlation radius.⁵ Correspondingly, the relaxation time τ_c of the concentration distribution diverges ($\tau_c \sim a^2 / D$, where a is the beam width). This relaxation time is also long under noncritical conditions, $\tau_c \gg (1-10) \Theta^{-\nu}$ s, even if $a = 10^{-2}$ cm. At $\Theta \lesssim 10^{-2} - 10^{-4}$, we therefore have a strongly time-varying self-focusing; the pulse length is $\tau < \tau_c$.

3. An estimate based on (2) for this situation yields

$$\Delta n_c \simeq L n_0 (\partial n / \partial c)^2 |E|^2 \tau / 8 \pi (\rho_0 a)^2 \quad (4)$$

and the threshold condition⁶ $\Delta n_c / n_0 > (ka)^{-2}$, which places a lower limit on the energy density of the light beam,

$$U_t = (P \tau / \pi a^2)_t = v (\rho_0 \lambda)^2 / L (2 \pi \partial n / \partial c)^2, \quad (5)$$

where $P = v |E|^2 a^2 / 8$ is the beam power level at the entrance to the medium. In the limit $\Theta \rightarrow 0$ we have $(\partial \mu / \partial c) \sim \Theta^\gamma$, where γ is the divergence index of the thermodynamic susceptibility. The experimental values are $\nu = 0.62 - 0.68$ and $\gamma = 1.22 - 1.24$ (Ref. 5), so that we have $L \sim \Theta^{\nu-\gamma} \rightarrow \infty$, and at \tilde{T} is approached we have $U_t \rightarrow 0$.

Far from the critical point we have $D \sim (10^{-4} - 10^{-5}) \text{ cm}^2 / \text{s}$; for a large number of solutions the value of $(\partial n / \partial c)$ is $\sim 0.1 - 1$. The values of $\partial \mu / \partial c$ and L are not known as accurately; as can be seen from (2), a measurement of L would require applying an external force (electromagnetic under our assumptions). In an ideal solution we would have $\partial \mu / \partial c = RT / Mc$ (Ref. 4), and this derivative could reach $10^8 \text{ cm}^2 / \text{s}^2$ (R is the universal gas constant); i.e., $L(\Theta \approx 1) \sim 10^{-12} - 10^{-13} \text{ G/cm}^3$. With $\lambda_0 \sim 10^{-5} \text{ cm}$ and $a \sim 10^{-2} \text{ cm}$ the energies required in (5) are $(10 - 10^2) \Theta^{\nu-\gamma} \text{ J}$. The ideal-solution approximation lowers the value of L ; furthermore, values $\Theta \sim 10^{-4} - 10^{-5}$ are achievable experimentally. We may therefore conclude that the concentration mechanism for self-focusing is feasible.

4. The electrostrictive and orientational nonlinearities reach a steady state in $\tau \ll 10^{-8}$ s. In Kerr liquids we have $\Delta n_{\text{or}} \lesssim (10^{-11} - 10^{-12}) |E|^2 \text{ esu}$; electrostriction leads to similar or smaller values, $\Delta n_{\text{str}} \sim (\rho_0 / u^2) (\partial n / \partial \rho) |E|^2$, where u is the sound velocity. Using the parameter values from Sec. 3, we find $\Delta n_c / \Delta n_{\text{or}} \gg 10^6 - 10^7 \tau c$; i.e., the concentration mechanism will be predominant in the experimentally attainable neighborhood of the stratification point at $\tau \gtrsim 0.1 \mu\text{s}$.

As a rule, the thermal mechanism is a defocusing mechanism in a liquid. It follows from the heat-conduction equation that

$$\Delta n_T \simeq \delta n_0 v (\partial n / \partial T) |E|^2 \tau / 8 \pi \rho_0 c_p \quad (6)$$

under the condition $\tau < \tau_T$ ($\tau < \tau_T = a^2 \rho_0 c_p / \kappa$ is the relaxation time of the temperature

distribution; δ absorption coefficient, κ the thermal conductivity, c_p the specific heat, and v the phase velocity). Near the stratification point, only $c_p \sim \Theta^{-\alpha}$ ($\alpha \approx 0.1$) in (6) depends on Θ , so that we have $\tau_T \sim 10^{-1}$ s. There are few solutions in which the condition $\Delta n_c > |\Delta n_T|$ is satisfied. For the better values of the parameters from Sec. 3 we should meet $\delta(\partial n/\partial T) \lesssim 10^{-8} - 10^{-9} \text{ cm}^{-2} \cdot \text{K}^{-1}$.

5. The details of the wave picture of self-focusing should be included in a further study of this effect. Here the threshold parameter is the energy density, so that inhomogeneities of the original profile will grow more rapidly than by other known mechanisms; this effect may lower the actual threshold. The very concept of a threshold must be refined here: The concentration lens changes sign at the beam half-width.

At power densities corresponding to (5), some more subtle optically induced effects should also be seen. One of them is a shift of the critical point. We can estimate this shift from (1) for the model of a regular solution⁷ from the Clausius-Mosotti equation:

$$\Delta \tilde{T} = \pi M N^2 (\alpha_1 - \alpha_2)^2 (n_0^2 + 2)^3 |E|^2 / 81 R \rho_0, \quad (7)$$

where α_i are the molecular polarizabilities. Calculations from (7) for $a = 10^{-2}$ cm yield $\Delta T \sim (10^{-8} - 10^{-6}) P$ (W). We note that ΔT reaches a maximum at the beam axis; one consequence is an unusual self-induced transparency of an opalescent solution in a strong field.

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