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Optical birefringence in a fluid near its critical point with stratification under gravity

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Abstract. Electromagnetic wave propagation in an inhomogeneous medium is discussed in the context of light propagation in a fluid near its critical point, stratified under gravity. The system exhibits birefringence, where the transverse electric (TE) and the transverse magnetic (TM) waves travel with different phase-velocities, depending upon the density-gradients with height set up in the system. The possibility of experimentally detecting the predicted phenomenon is also discussed.

Keywords. Critical point phenomena; optical birefringence; stratified media.

1. Introduction

Wave propagation in inhomogeneous media has remained a field of study ever since the birth of wave optics; for instance the optics of mirage, first explained by Huygens, was subsequently reexamined on the basis of a wave optical interpretation by Raman and Pancharatnam (1959). The present day studies in the area encompass a wide range of fields like oceanography, geophysics as well as atmospheric and ionospheric studies (Berkhovskikh 1960; Tucker and Gazey 1966; Tolstoy 1973). All these studies essentially assume that the medium of wave propagation is uniform along, say, the xy plane while the nonuniformity is confined along the z-axis. In the present communication, we first point out that a one-component fluid near its critical point, in the presence of gravity, is a stratified medium of the type described above with one nonvanishing component of density gradient. We then investigate an interesting property of the system, namely optical birefringence, which appears due to the stratification of the fluid column. The possibility of experimental observation of the above phenomenon is discussed with reference to a specific system like xenon, near its critical point.

2. Theory of refractive index gradient in the critical region

Near the critical point of a one-component fluid, the isothermal compressibility ($K_T \equiv \frac{1}{\rho}\left(\frac{\partial \rho}{\partial p}\right)_T$) is known to be strongly divergent (Stanley 1971; Moldover *et al* 1979) being given by:

$$K_T^{-1} \sim (K_T^0)^{-1}\left[|\Delta T^*|^{\gamma} + a|\Delta\rho^*|^{\delta-1}\right] \tag{1}$$

to the leading orders in $|\Delta T^*|$ and $|\Delta\rho^*|$, with $|\Delta T^*| \equiv |(T-T_c)/T_c|$ and $|\Delta\rho^*| \equiv |(\rho-\rho_c)/\rho_c|$ where T_c denotes the critical temperature and ρ_c the critical density, γ and δ

being the well-known critical exponents. Due to this highly compressible nature of the fluid, significant density gradient appears in the system (Hohenberg and Barmatz 1972; Moldover *et al* 1979), under the influence of gravity. Every layer of the fluid being under a pressure, $p(z)$ due to the liquid above it, the hydrodynamic stability condition yields,

$$\mathrm{d}p/\mathrm{d}z = -\rho g \tag{2}$$

where $\rho(z)$ is the density at any height (measured from the bottom of the cell) and g is the acceleration due to gravity. Because the compressibility tends to infinity, a small pressure head $\Delta p = \rho g \Delta z$ is enough to cause a large density gradient.

Equation (2) in combination with (1) is used to eliminate $p(z)$ and give a differential equation

$$\mathrm{d}\rho/\mathrm{d}z = -g\rho^2 K_T \tag{3}$$

for the density in any layer.

Explicit solutions of (3) give the density profile $\rho(z)$ in the cell, the results of which will be presented elsewhere (Chatterjee *et al* 1983). However for the sake of completeness the final analytical expression, describing the variation of ρ with z, is quoted in Appendix I, while the graphical representation is given in figure 1. The essential point to be noted is that the density stratification of the system contributes to the stratification of the optical refractive index $n(z)$, along the z-direction. The refractive index at any point z, can be calculated from $n(z) = n(\rho)|_{\rho = \rho(z)}$ once the density profile is known. We also note that the condition of uniformity of density in the xy-plane is valid at regions not too close to the critical point, since near the critical point very large local density fluctuations occur due to critical fluctuations. This particular problem is considered in greater detail when we explore the experimental situation.

3. Wave propagation in a stratified medium

We now consider the problem of electromagnetic wave propagation in a medium, stratified in the z direction. From the Maxwell equations,

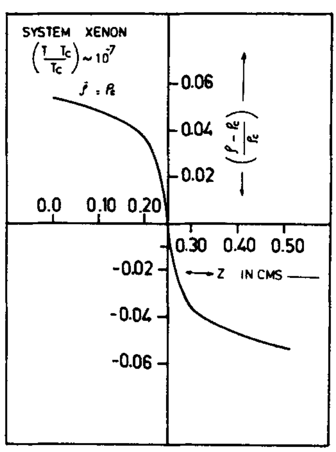

Figure 1. Density profile near the critical point.

$$\text{curl}\,\vec{H} = \frac{n^2(z)}{c}\frac{\partial \vec{E}}{\partial t}. \tag{5a}$$

$$\text{div}\, n^2(z)\vec{E} = 0. \tag{5b}$$

$$\text{curl}\,\vec{E} = -\frac{1}{c}\frac{\partial \vec{H}}{\partial t}. \tag{5c}$$

$$\text{div}\,\vec{H} = 0, \tag{5d}$$

we know that the electric and the magnetic fields can be expressed in terms of the vector and scalar potentials (for example Jackson 1962) as

$$\vec{E} = -\text{grad}\,\phi - \frac{1}{c}\frac{\partial \vec{A}}{\partial t}. \tag{6}$$

$$\vec{H} = \vec{\nabla}\times\vec{A}.$$

Invariance of $\vec{E}$ and $\vec{H}$ under gauge transformations enables one to choose the gauge in such a way that,

$$\frac{n^2}{c}\frac{\partial \phi}{\partial t} + \vec{\nabla}\cdot\vec{A} = 0. \tag{7}$$

Using equations (5)–(7) and considering a periodic variation, $A(r, t) = A(r) \exp(-iwt)$, we obtain (for example Brekhovskikh 1960) the equation

$$\nabla^2\vec{A} + k^2(z)\vec{A} + (\vec{\nabla}\text{A})\frac{\vec{\nabla}n^2}{n^2} = 0. \tag{8}$$

where

$$k^2(z) = k_0^2 n^2(z) \tag{9}$$

with

$$k_0^2 = \omega^2/C^2. \tag{10}$$

For a uniform medium, $\vec{\nabla}n^2 = 0$ and (8) reduces to that of free wave-propagation. In a stratified medium, where $\vec{\nabla}n^2$ has only the z-component $(\partial/\partial z)n^2$, we can distinguish between two directions of polarization of $\vec{A}$. In all our following discussions we shall consider $\vec{A}$ to be independent of y *i.e.* the xy plane is considered to be the plane of incidence.

Case I For $A_x = 0 = A_y$ and $A_z \neq 0$

$$E_x = \frac{i\omega}{ck^2}\frac{\partial^2 A_z}{\partial x\,\partial z}, \quad E_y = 0, \qquad E_z = \frac{i\omega}{c}\left[A_z + \frac{\partial}{\partial z}\left(\frac{1}{k^2}\frac{\partial A_z}{\partial z}\right)\right],$$

$$H_x = 0, \qquad H_y = -\frac{\partial A_z}{\partial x}, \quad H_z = 0.$$

Defining $\psi = A_z/k$ we have

$$\nabla^2\psi + k_{\text{eff}}^2(z)\psi = 0 \tag{12}$$

where

$$k_{\text{eff}}^2(z) = k^2(z) - n\frac{\partial^2}{\partial z^2}\left(\frac{1}{n}\right). \tag{13}$$

Such waves can be called transverse magnetic (TM) waves since the magnetic field is directed along y. It is seen that in wave propagation along the x-direction, $A_z \alpha \exp(ik_x x)$ so that $E_x = 0$ and hence only the z component E_z of the electric field survives.

Case II For $A_x = 0 = A_z$, $A_y \neq 0$

$$E_x = 0, \quad E_y = \frac{i\omega}{c} A_y, \quad E_z = 0$$
$$H_x = -\frac{\partial A_y}{\partial z}, \quad H_y = 0, H_z = 0 \tag{14}$$

we have $\nabla^2 A_y + k^2 A_y = 0$. (15)

These waves are referred to as transverse electric (TE) waves, the electric field being directed along y.

On examination of (9), (10), (13) and (15) we find that the dispersions of the two waves are different. Their respective phase velocities are given by

$$v_{\mathrm{TM}} = \omega / k_{\mathrm{eff}}, \tag{16a}$$

and

$$v_{\mathrm{TE}} = \omega / k. \tag{16b}$$

This implies that the effective refractive indices $\tilde{n}(z)$ of the two waves are given by,

$$\tilde{n}^2_{\mathrm{TM}}(z) = \frac{c^2}{v^2_{\mathrm{TM}}} = n^2 - \frac{n}{k_0^2} \frac{\partial^2}{\partial z^2}\left(\frac{1}{n}\right), \tag{17a}$$

$$\tilde{n}^2_{\mathrm{TE}}(z) = \frac{c^2}{v^2_{\mathrm{TE}}} = n^2. \tag{17b}$$

In other words, the system exhibits birefringence.

The birefringence arises because the E_z electric field of the TM mode can interact with the gradient of the refractive index, also along the z direction. The two dispersion relations differ by the presence of the term

$$\frac{n}{k_0^2} \frac{\partial^2}{\partial z^2}\left(\frac{1}{n}\right),$$

the origin of which can be understood as follows. From (5b), we find that the presence of any field E_z in the z-direction (*i.e.* across the layers) yields,

$$\frac{\partial E_z}{\partial z} = \frac{1}{n^2(z)} \frac{\partial n^2}{\partial z} E_z \tag{18}$$

i.e. a polarisation charge-density $(1/4\pi)(1/n^2(z))(\partial n^2/\partial z)E_z$ develops by virtue of the stratification of the dielectric constant. Any temporal fluctuation of E_z, gives rise to a displacement current, which contributes to curl $\vec{H}$. Electromagnetic waves are thus generated from all points in the medium. Since the electromagnetic potentials are all additive quantities, the effective value of $\vec{A}(r, t)$ is the sum of the $\vec{A}$'s arising out of all the displacement currents in the system. The term

$$\frac{n}{k_0^2} \frac{\partial^2}{\partial z^2}\left(\frac{1}{n}\right)$$

arises out of this phenomenon.

We next note that a fluid near its critical point exhibits significant birefringence due to very large stratification under gravity. Using (1) and (2) we find

$$\frac{\partial^2}{\partial z^2}\left(\frac{1}{n}\right) = (g\rho^2 K_T)^2\left[\frac{2}{\rho}\frac{\partial}{\partial\rho}\left(\frac{1}{n}\right)+\frac{\partial^2}{\partial\rho^2}\left(\frac{1}{n}\right)\pm\frac{1}{\rho_c}\frac{\partial}{\partial\rho}\left(\frac{1}{n}\right)\frac{a(\delta-1)|\Delta\rho^*|^{\delta-2}}{|\Delta T^*|^{\gamma}+a|\Delta\rho^*|^{\delta-1}}\right] \tag{19}$$

where the positive sign is valid for $\rho < \rho_c$ while the negative sign applies for $\rho > \rho_c$. The variation of $\partial^2/\partial z^2(1/n)$ with height is represented schematically in figure 2. Thus in the limit when $|\Delta T^*|^{\gamma} \ll a|\Delta\rho^*|^{\delta-1}$ we have, for $\rho < \rho_c$ *i.e.* above the centre of the cell,

$$\tilde{n}^2_{\mathrm{TM}}(z) = n^2(z) - \frac{n}{k_0^2}(g\rho^2 K_T)^2\frac{1}{\rho_c}\frac{\partial}{\partial\rho}\left(\frac{1}{n}\right)(\delta-1)|\Delta\rho^*|^{-1} > \tilde{n}^2_{\mathrm{TE}}(z) \tag{20}$$

and for $\rho > \rho_c$ *i.e.* below the centre of the cell,

$$\tilde{n}^2_{\mathrm{TM}}(z) = n^2(z) + \frac{n}{k_0^2}(g\rho^2 K_T)^2\frac{1}{\rho_c}\frac{\partial}{\partial\rho}\left(\frac{1}{n}\right)(\delta-1)|\Delta\rho^*|^{-1} < \tilde{n}^2_{\mathrm{TE}}(z). \tag{21}$$

where we have used the fact that $\partial/\partial\rho(1/n) < 0$.

This means that for $\rho < \rho_c$ the TE travels faster than TM while for $\rho > \rho_c$ the situation is reversed. The optics of the system can be adequately described by geometrical optics if $(\partial/\partial z)\tilde{n}(z) < k_0$.

4. Experimental observations: possible situation

The possibility of experimental verification of the above phenomenon is now discussed with reference to xenon near its critical point. The density profile of xenon close to its critical condition has been extensively studied by interferometric methods (Palmer

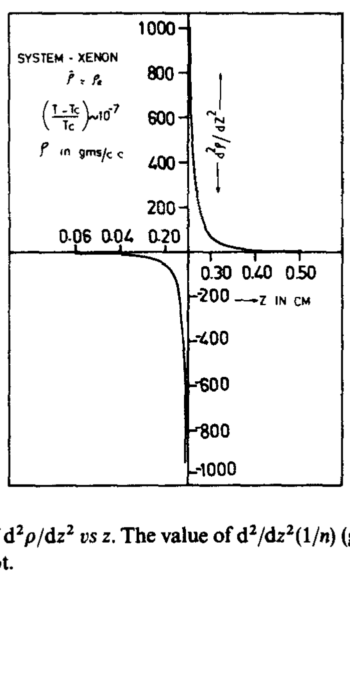

Figure 2. Plot of $d^2\rho/dz^2$ *vs* z. The value of $d^2/dz^2(1/n)$ (given in the text) can be calculated from the above plot.

1954; Wilcox and Balzarini 1968; Estler *et al* 1975; Hocken and Moldover 1976). However, these authors do not refer to the possibility of birefringence in the system. It is noted that for xenon at its critical density, the correlation length of the density fluctuations remains shorter than the wavelength of visible light for $|(T-T_c)/T_c| \gtrsim 10^{-7}$. This also eliminates the problem of multiple scattering. In fact, this is nearly the closest approach to the critical temperature that the modern experimental techniques permit. Consequently our theory is valid within this limit since the uniformity in the xy plane is then a valid assumption. For Xe, the observations (Estler *et al* 1975) give $\partial n/(\partial|\Delta\rho^*|) = 0{\cdot}13$, critical pressure = 5·7 MPa, and $\rho_c = 1110\,\mathrm{kg/m^3}$. Thus for red-light with wavelength ~ 6000 Å, for 1 % difference between $\tilde{n}^2_{\mathrm{TM}}$ and $\tilde{n}^2{}_{\mathrm{TE}}$ the necessary value of $|\Delta\rho^*|$ must be close to 0·3 %. For a 5 mm column of Xe and $|\Delta T^*| \sim 10^{-7}$ filled at an average density equal to the critical density, the above condition is attained at a height nearly 0·05 mm away from the centre. The magnitude of the birefringence varies very rapidly with height at points close to the middle of the cell. For the same system the $\Delta(n^2)$ value drops down to 10^{-4} at a point 0·1 mm away from the middle of the cell. The present-day interferometric techniques (Peterlin 1976; Tsvetkov and Frisman 1945) allow precise measurements of $\Delta(n^2)$ up to 10^{-8}. In such cases the birefringence is detectable up to $|\Delta\rho^*| \sim 3\,\%$ *i.e.* even at a height $\pm\,0{\cdot}5$ mm away from the centre of the cell, under the conditions mentioned above.

The birefringence will be more for polar type molecules having larger values of refractive index in the liquid phase. At points closer to the centre of the cell the birefringence is expected to be more pronounced. However, at a height very close to the centre the above theory breaks down if the condition $|\partial\tilde{n}/\partial z| < k_0$ is violated. Alternatively, the effect can be enhanced by using light of longer wavelength *e.g.* by working with an IR source. Several interferometric methods (for example Ramachandran and Ramaseshan 1961; Francon 1966) can be employed to study birefringence at any given layer. The advantage of this method is that the $\Delta(n^2)$ value at any particular height directly gives $\partial^2 n/\partial z^2$ whereas the conventional interferometric techniques as employed in the earlier studies require complicated calculations to extract the density gradient. The explicit calculation of the intensity pattern of light is presented in Appendix II.

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Appendix I

The explicit solution of (3) giving the density profile under gravity, can be obtained for a known value of δ whose details will be published in due course (Chatterjee *et al* 1983). For the experimentally known value $\delta \simeq 41/3$, $|\Delta\rho^*| \equiv x$ is obtained from the solution of the algebraic equation,

$$|\Delta T^*|^\gamma \ln(1+x) + a\left[\frac{x^{10/3}}{10/3} - \frac{x^{7/3}}{7/3} + \frac{x^{4/3}}{4/3} - f(x^{1/3})\right] = -gk_T^0\rho_c(z-z_0) \quad \text{(A1)}$$

for $\rho > \rho_c$

where

$$f(\phi) = 3\left[\phi - \tfrac{1}{3}\ln\frac{1+\phi}{(1-\phi+\phi^2)^{1/2}} + \tfrac{1}{3}\tan^{-1}\frac{\sqrt{3}}{2-\phi}\right]$$

while for $\rho < \rho_c$

$$|\Delta T^*|^\gamma \ln(1-x) + a\left[\frac{x^{10/3}}{10/3} + \frac{x^{7/3}}{7/3} + \frac{x^{4/3}}{4/3} + \psi(x^{1/3})\right] = gk_T^0\rho_c(z - z_0) \qquad \text{(A2)}$$

where

$$\psi(\phi) = 3\left[\tfrac{1}{3}\ln\frac{(1+\phi+\phi^2)}{1-\phi} + \frac{1}{\sqrt{3}}\tan^{-1}\frac{\phi\sqrt{3}}{2+\phi} - \phi\right]$$

Appendix II

The intensity of light at any point requires the solution of the wave-equations (12) for TE and (15) for the TM waves. The usual ray-tracing techniques are valid within the limits of geometrical optics. We consider the general situation for any oblique incidence of light. Let the light be incident at an angle θ to the plane of the layer. Since the medium is homogeneous along x, the solution of the wave equation has the formal dependence $\exp[in_i k_0 px]u(z)$ where $p = \cos\theta$ and n_i is the refractive index of the layer in which the beam originates.

The wave equation then reads

$$\frac{\partial^2 u}{\partial z^2} + \alpha^2(z)u = 0 \qquad \text{(A3)}$$

where

$$\alpha^2 = k_0^2[\bar{n}^2(z) - n_i^2 p^2]$$

with $u \equiv A_z/n$ for the TM and $u = A_y$ for the TE waves. The above equation can be solved by conventional methods of solution.

If for $z = z_1$, $\alpha = 0$, then total internal reflection occurs at this layer, which is called the limiting layer. It is obvious that the limiting layers lie at different positions for the TM and TE waves.

In the WKB approximation the solution for $u(z)$ can be written as (for example Tyres 1969)

$$u(z) \propto [1/\alpha(z)]^{1/2} \exp\left(-i\int_0^{z_1}\alpha(z)\,dz + i\pi/4\right)\left\{\exp\left(i\int_0^{z}\alpha(z)\,dz\right) - i\exp\left(2i\int_0^{z_1}\alpha(z)\,dz - i\int_0^{z}\alpha(z)\,dz\right)\right\} \qquad \text{(A4)}$$

for $z < z_1$,

$$u(z) \propto [3/\alpha(z)]^{1/2} \exp\left(-\int_{z_1}^{z}|\alpha(z)|\,dz\right) \qquad \text{(A5)}$$

for $z > z_1$.

The second term in $\{----\}$ of (A4) is absent in the absence of the total reflection and z_1 is replaced by the height of the cell.

The above formal solution enables us to calculate A_z and A_y by suitably defining the variables $u(z)$, $\alpha(z)$ and z_1 according to the conditions given in the earlier paragraph. The components of the electric, and magnetic fields and the intensity of light can be evaluated. The formal expressions are too complicated to be reproduced here. The resulting intensity pattern describes the interference phenomenon of light due to its passage through the stratified medium. It is obvious that the distribution of intensity is different for the two independent polarisations of light. In the case of total reflection, very little energy crosses the limiting layer. The reflected beam travels along the limiting layer which describes the caustic surface, in the present case (Raman and Pancharatnam 1959). Below this surface one observes interference fringes of decreasing fringe spacing and intensity as one moves away from the limiting surface. It is also clear that more detailed discussions of the caustic surfaces would require the methods of the catastrophe theory (Berry and Upstill 1980).

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