Rigorous modelling of light's intensity angular-profile in Abbe refractometers with absorbing homogeneous fluids

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Abstract. We derive an optical model for the light intensity distribution around the critical angle in a standard Abbe refractometer when used on absorbing homogenous fluids. The model is developed using rigorous electromagnetic optics. The obtained formula is very simple and can be used suitably in the analysis and design of optical sensors relying on Abbe type refractometry.

1. Introduction.

There is an increasing need for reliable small-scale refractometers for chemical and biological microfluidic devices [1-3]. However, all refractometers developed to date are assumed will be used with transparent fluids and their use with absorbing liquids is not always possible. On the other hand, one of the more robust ways to measure a refractive index of liquids is by means of an Abbe refractometer introduced many years ago [4]. Actually, most bench-top refractometers in use today are of the Abbe type. Nevertheless, to our knowledge, the Abbe refractometer has not yet been exploited in microfluidics. We believe that this is in part due to the lack of rigorous optical models for measuring with Abbe type going beyond geometrical ray optics. By Abbe type refractometers we mean refractometers in which diffuse light is refracted from a sample fluid into a prism of higher refractive index, making apparent the critical angle from which the refractive index of the fluid can be obtained. It is not difficult to realize that the basic principle of Abbe refractometers could be developed into compact optical sensors for applications in microfluidics. As most refractometers, the Abbe refractometer was thought for use with transparent materials. Nevertheless, in practice it is often used on substances with some degree of opacity without precautions. Very few works have been devoted to understand the use of an Abbe refractometer with opaque media. The use of standard Abbe refractometers with turbid colloidal fluids was investigated in Ref. [5] many years ago. But many questions regarding the correct determination of a critical angle were left open at that time and no optical model was derived then. Only recently we initiated new research on the measurement of the effective refractive index of highly turbid fluids with Abbe type refractometers [6, 7]. In the latter references we did not consider the case of samples of complex refractive index without optical scattering. Thus, the model derived in Ref. [7] is not applicable to homogeneous absorbing fluids, which are actually involved in many chemical and biological sensing applications. In standard Abbe refractometers, the sample is in the form of a fluid film at the base of an optical prism of higher refractive index. Diffuse light is transmitted through the sample to the prism. Light transmitted into the prism is confined to angles of refraction smaller than the critical angle. Thus locating de edge of the light's cone yields the sample's refractive index. However when the sample is not optically transparent, the edge becomes fuzzy and the determination of the refractive index may not be straightforward. In this work we develop a model for the light's intensity angular-profile around the critical angle in Abbe refractometers with homogeneous absorbing fluids. We suppose that light scattering within the sample fluid is negligible. Our aim is to provide a simple formula for the angular intensity profile around the critical angle in a standard Abbe refractometer that could be related to the complex refractive index of the sample.

2. Theoretical model.

In standard Abbe refractometers the sample fluid is placed between the base of a glass prism and a diffusing translucent surface. Light is incident to the diffusing surface from the side opposite to the prism and scattered within the sample fluid film as shown in Fig. 1. The light transmitted into the prism is refracted within an angular cone limited by the critical angle. In the standard methodology, the refractive index of the sample fluid is obtained from locating the critical angle from the edge of the light cone. When the sample has a complex refractive index, the angular profile around the edge of the light cone is modified with respect to that when the sample has a real refractive index. Nevertheless, from the angular intensity profile it can be possible to retrieve the real and imaginary parts of the refractive index of the sample fluid or simply sense variations of their value.

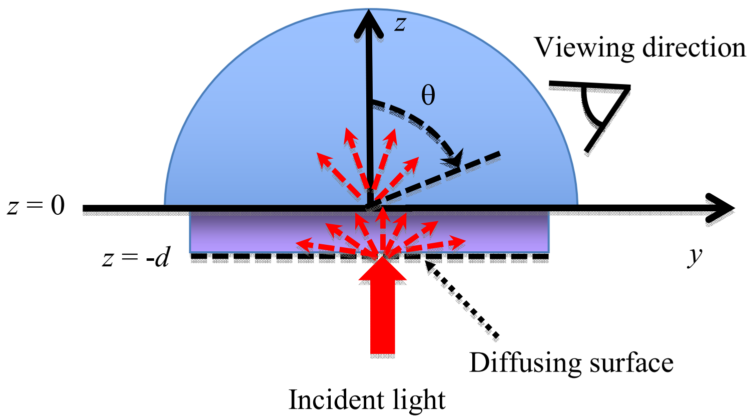

Figure 1: Schematic illustration of the basic operation of a standard Abbe refractometer.

We can follow closely the model developed in [7] for the intensity angular-profiles of light scattered within a turbid colloidal fluid and transmitted into a transparent medium of higher refractive index. The difference here is that the effective radiation sources due scattering are not distributed through the sample but all are located on a plane parallel to the interface between the absorbing liquid sample and the transparent glass of higher refractive index as illustrated in Fig. 1.

Let us start by considering an opaque rough surface, flat on the average, placed at a distance d from the bottom surface (flat) of a transparent glass prism. Let us assume that the space in-between the irregular surface and the bottom of the prism is filled with an absorbing, homogeneous liquid of complex refractive index n_m. Let us place the origin of coordinates on the interface between the prism and the liquid sample with the z-axis pointing into the prism as shown in Fig. 1. Let us assume that monochromatic light of frequency ω is incident to the rough surface from $z < -d$ and scattered in all directions. We may regard the illuminated rough surface as a random array of sources emitting incoherently in all directions. The sources can be modelled as localized electrical currents embedded in the sample liquid of refractive index n_m and distributed within a very thin film just below the liquid sample. Let us divide the radiating current into a large number of independent localized currents. The electric field radiated by the *nth* localized current can be expressed as,

$$\mathbf{E}_n^s(\mathbf{r}) = i\omega\mu_0 \int_{V_n} d^3r' \overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}')\cdot \mathbf{J}_n(\mathbf{r}'), \tag{1}$$

where V_n is the volume occupied by the *nth* current and $\overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}')$ is the Green's function dyadic of the vector Helmholtz equation in the sample medium at frequency ω. The time dependence is implicit in all equations. For observation points above all the radiating currents, we can use the following plane wave expansion of the Green function dyadic [8],

$$\overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}') = \frac{i}{8\pi^2}\int dk_x\, dk_y \frac{(\overleftrightarrow{\mathbf{I}} - \hat{\mathbf{k}}_+\hat{\mathbf{k}}_+)}{k_z}\exp[i\mathbf{k}_+\cdot(\mathbf{r}-\mathbf{r}')], \tag{2}$$

where $\mathbf{k}_\pm = k_x\hat{\mathbf{a}}_x + k_y\hat{\mathbf{a}}_y + k_z\hat{\mathbf{a}}_z$ and $k_z = (k_0^2 n_m^2 - k_x^2 - k_y^2)^{1/2}$. Making the change of variable $\mathbf{r}' = \mathbf{r}_s + \mathbf{r}_n$, where $\mathbf{r}_s$ is the position vector relative to the centre of the volume V_n containing the *nth* localized current, we get for points above the rough surface,

$$\mathbf{E}_n^s(\mathbf{r}) = \frac{-\omega\mu_0}{8\pi^2}\int dk_x\, dk_y \exp[i\mathbf{k}_+\cdot(\mathbf{r}-\mathbf{r}_n)]\,\mathbf{s}_n(k_x,k_y), \tag{3}$$

where

$$\mathbf{s}_n(k_x,k_y) = \frac{(\overleftrightarrow{\mathbf{I}} - \hat{\mathbf{k}}_+\hat{\mathbf{k}}_+)}{k_z}\cdot\int_{V_n} d^3r_s \exp(-i\mathbf{k}_+\cdot\mathbf{r}_s)\mathbf{J}_n(\mathbf{r}_s). \tag{4}$$

Since we are assuming n_m is a complex quantity, Eq. (3) is a linear superposition of inhomogeneous plane waves in the sample medium. Note that the kernel is divided by k_z which is zero when $k_x^2 + k_y^2 = k_0^2 n_m^2$. This corresponds to plane waves traveling parallel to the interface between the sample liquid and the prism (the plane $z = 0$).

Now, let us propagate these plane waves through the sample up the prism's bottom surface. For the sake of simplicity we will assume the detection plane is within the prism in the far zone. In general it is relatively simple to consider the actual case in which the detection plane is outside the prism. Let us assume that the detection plane is at a large distance from the interface and viewing towards the interface with the liquid sample. First we propagate the fields to just before the interface by evaluating Eq. (3) at $z = 0^-$. We get,

$$\mathbf{E}_n^s(\mathbf{r})\Big|_{z=0^+} = \frac{-\omega\mu_0}{8\pi^2}\int dk_x\, dk_y \exp[ik_x(x-x_n) + ik_y(y-y_n) - ik_z z_n]\mathbf{s}_n(k_x,k_y). \tag{5}$$

Then, the field just on the other side of the interface at $z = 0^+$ is obtained by splitting the fields into its transverse electric, TE, and transverse magnetic, TM, components and multiplying by the corresponding transmission coefficient. Then we change $\mathbf{s}_n(k_x,k_y)$ by,

$$\overleftrightarrow{\mathbf{t}}(k_x,k_y)\cdot\mathbf{s}_n(k_x,k_y) = t^{TE}s_n^{TE}(k_x,k_y)\hat{\mathbf{s}}_{TE} + t^{TM}s_n^{TM}(k_x,k_y)\hat{\mathbf{s}}_{TM},$$

where t^{TE} and t^{TM} are the transmission coefficients for TE and TM polarized waves respectively, $s_n^{TE}(k_x,k_y)$ and $s_n^{TM}(k_x,k_y)$ are the projections of $\mathbf{s}_n(k_1,k_2)$ on the unit vectors $\hat{\mathbf{s}}_{TE}=(\mathbf{k}_+\times\hat{\mathbf{a}}_z)/|\mathbf{k}_+\times\hat{\mathbf{a}}_z|$ and $\hat{\mathbf{s}}_{TM}=(\mathbf{k}_+\times\hat{\mathbf{s}}_{TE})/|\mathbf{k}_+\times\hat{\mathbf{s}}_{TE}|$. Since only the *z*-component of $\mathbf{k}_+$ can be complex, we can see that $\hat{\mathbf{s}}_{TE}$ is a real unit vector. However, $\hat{\mathbf{s}}_{TM}$ is in general a complex unit vector.

Now we can introduce the factor $\exp(ik_z^p z)$ with $k_z^p=(k_0^2n_p^2-k_x^2-k_y^2)^{1/2}$ in the kernel of Eq. (5) to propagate the field into the incidence medium of refractive index n_p. Then, we can propagate the transmitted field to the far zone and use the method of stationary phase to evaluate the integrals [9]. To this end, we assume that the interface transmits light through an aperture with a large but finite area *A* on the plane $z = 0$, and bring the observation point far away compared to the lateral dimensions of the aperture. We get,

$$E_n^s(\mathbf{r})=i\omega\mu_0k_p\cos\theta\frac{\exp(ik_pr)}{4\pi r}\exp(-i\mathbf{k}_+\cdot\mathbf{r}_n)\vec{\mathbf{t}}(k_1,k_2)\cdot\mathbf{s}_n(k_1,k_2) \tag{6}$$

where $k_1=k_p\sin\theta\cos\phi$ and $k_2=k_p\sin\theta\sin\phi$ are the coordinates of the stationary phase point in the $(k_x,\ k_y)$ space and $(r,\ \theta,\ \phi)$ are spherical coordinates of the observation point $\mathbf{r}$. Also, now $\mathbf{k}_+=k_1\hat{\mathbf{a}}_x+k_2\hat{\mathbf{a}}_y+k_z\hat{\mathbf{a}}_z$ with $k_z=\sqrt{k_0^2n_m^2-k_1^2-k_2^2}$, which is in general complex. That is, now we have $k_z=\mathrm{Re}\left(k_z\right)+i\,\mathrm{Im}\left(k_z\right)$. Light intensity is given by the magnitude of the time average of Poynting's vector, which in the case of self-propagating plane waves is given by $I_n=\frac{1}{2}(k_p/\omega\mu_0)\left|\mathbf{E}_n^s\right|^2$. To obtain the intensity profile in the detection plane we must add the contribution of all localized currents emitting incoherently light towards the detection plane. So we add the intensity at the detection plane due to all localized currents (we do not add the fields for incoherent sources).

For simplicity we assume that all currents are located within a very thin film of thickness *h* and lateral dimensions of area *A*. Adding up the intensity due to all localized currents and taking the limit that the volume occupied by any of the localized currents tends to zero, yields,

$$I(\mathbf{r})=\frac{1}{2}\beta D\left(\frac{\cos\theta}{r}\right)^2\iint_A dx_n\,dy_n\int_{-d-h}^{-d}dz_n\exp[2\,\mathrm{Im}(k_z^{eff})z_n]\left|\vec{\mathbf{t}}(k_1,k_2)\cdot\mathbf{s}_n(k_1,k_2)\right|^2, \tag{7}$$

where $D=\omega\mu_0k_p^3\big/16\pi^2$, β is the density of incoherently-emitting localized currents, and we assumed the area *A* is large compared to the sample's depth *d*. Since the unit vectors $\hat{\mathbf{s}}_{TE}$ and $\hat{\mathbf{s}}_{TM}$ are orthogonal to each other and we assume that light is randomly polarized we have,

$$\left|\vec{\mathbf{t}}(k_1,k_2)\cdot\mathbf{s}_n(k_1,k_2)\right|^2=\left|t^{TE}\right|^2\left|s_n^{TE}(k_1,k_2)\right|^2+\left|t^{TM}\right|^2\left|s_n^{TM}(k_1,k_2)\right|^2. \tag{8}$$

Recall that k_1 and k_2 are functions of the viewing angles θ and ϕ. The intensity profile is measured over a very small range (compared to 2π) along the polar angle θ around the direction of the edge of the light cone and about the plane of incidence, that is around $\phi = 0$. Note that $\mathbf{s}_n(k_1,k_2)$ is inversely proportional to k_z, and the magnitude of k_z is minimum when $\sqrt{k_0^2n_m^2-k_1^2-k_2^2}=\sqrt{k_0^2n_m^2-k_0^2n_p^2\sin^2\theta}$ is minimum. We may refer to the viewing angle θ that minimizes the latter expression as the critical angle. If the effective refractive index approaches a real number, the minimum of the magnitude of k_z

approaches zero and $\mathbf{s}_n(k_1,k_2)$ becomes singular. However, the transmission coefficients t^{TE} and t^{TM} are in the form of Fresnel transmission coefficients. Then, both t^{TE} and t^{TM} should be proportional to k_z. Thus, Eq. (8) does not have a singularity when $k_z = 0$.

Thus, in general $\left|\vec{\mathbf{t}}(k_1,k_2)\cdot\mathbf{s}_n(k_1,k_2)\right|^2$ should be a smooth function of k_1 and k_2, and therefore, for a fixed $\mathbf{r}_n$ we can approximate it as a constant within the small range of interest of viewing directions. Note that the magnitude of $\mathbf{s}_n$ must be proportional on the average to the intensity of the incident light I_0. The integral in Eq. (7) simplifies to,

$$I(\theta) = I_0 C \cos^2\theta \exp[-2\,\mathrm{Im}(k_z^{eff})d], \tag{9}$$

where $k_z^{eff} = k_0(n_{eff}^2 - n_p^2 \sin^2\theta)^{1/2}$ and C is an instrumental constant determined experimentally. Note that we assumed h very small. This is the main result of this paper. We must draw attention to the fact that θ in Eq. (9) is the angle of travel of light inside the prism. In a practical device the detector will be placed outside the prism and one must take into account refraction of light when exiting the prism.

3. Numerical examples

In Fig. 2a we plot the angular intensity profile of light refracted near the critical angle assuming the sample thickness is d = 100 μm and the fluid has a refractive index $n_m = 1.33 + ix$. We show plots for $x = 10^{-6}$, 10^{-5}, 10^{-4} and 10^{-3}. Note that a substance with $x = 10^{-3}$ would be completely opaque in a 1 mm thick cuvette. For the plots in Fig. 3 we assumed the refractive index of the prism is 1.515 and the wavelength in vacuum is 635 nm.

(a) (b)

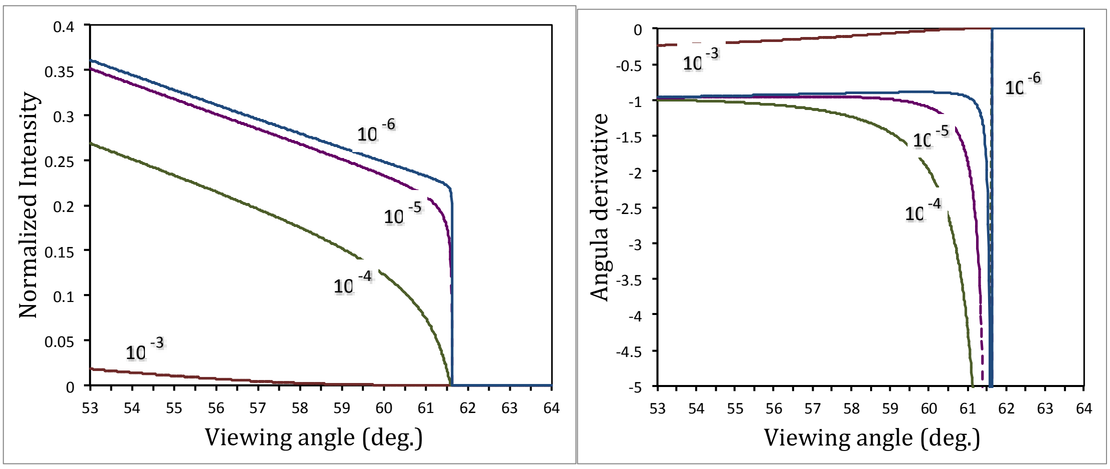

Figure 2: (a) Plots of the intensity versus the angle of refraction (inside the prism) near the critical angle for a sample fluid of refractive index $n_m = 1.33 + ix$ with $x = 10^{-6}$, 10^{-5}, 10^{-4} and 10^{-3}. The refractive index of the prism was assumed to be 1.515, the wavelength 635nm and the sample thickness = 100 μm. (b) plots of the angular derivative of the curves in (a).

We can see that for a small imaginary part of the refractive index, the intensity profile is "step-like" at the critical-angle determined by the real part of the refractive index and the refractive index of the prism. As the imaginary part of the refractive index of the sample fluid increases, the angular profile becomes smoother, but it sinks to zero at the same angle. In the plots for $x = 10^{-6}$, 10^{-5} and 10^{-4} one can se there is an inflexion point. One should take the inflexion point as the critical angle, θ_c. From it one may obtain the real part of the refractive index of the sample using the usual formula $n_s = n_p \sin^{-1}(\theta_c)$ where n_p is the refractive index of the prism and n_s would be in this case the real part of the refractive index of the sample.

In Fig. 2b we plot the angular derivative of the curves shown in Fig. 2a. Clearly the first three plots for $x = 10^{-6}$, 10^{-5} and 10^{-4} show a sharp maximum at basically the same angle. However, the curve for $x = 10^{-3}$ does not show a maximum and therefore the corresponding plot in Fig. 2a has no inflexion point. We may then conclude that standard Abbe refractometry can no longer be performed. Nevertheless, by adjusting Eq. (9) to the angular intensity profile one could retrieve the real and imaginary parts of the refractive index of the sample. We may point out here that if d is reduced the angular intensity profile for $x = 10^{-3}$ changes and an inflexion point appears. However if d is reduced too much, then the intensity profile starts to extend noticeably beyond the inflexion point. Anyhow, depending on the particular design of a refractometric device based on refraction of diffuse light near the critical angle one can use Eq. (9) as long as scattering within the sample is negligible.

4. Conclusions

We derived a simple formula for the angular intensity profile of light refracted from a homogeneous absorbing fluid into a transparent prism of higher refractive index. The formula could be used to model refractometric devices based on refraction of diffuse light from an absorbing fluid sample near the critical angle. It could also be advantageously used to retrieve the real and imaginary parts of the complex refractive index of a sample fluid from angular intensity profiles around the critical angle in Abbe type refractometers.

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