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Speech presented at an Electrochemical Research Seminar,
Dept. of Chemistry U. of C. Berkeley, July 7, 1964.

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DIFFUSIONAL MASS TRANSFER

R. H. Muller

July 21, 1964

OPTICAL TECHNIQUES FOR THE STUDY OF
DIFFUSIONAL MASS TRANSFER

R. H. Muller

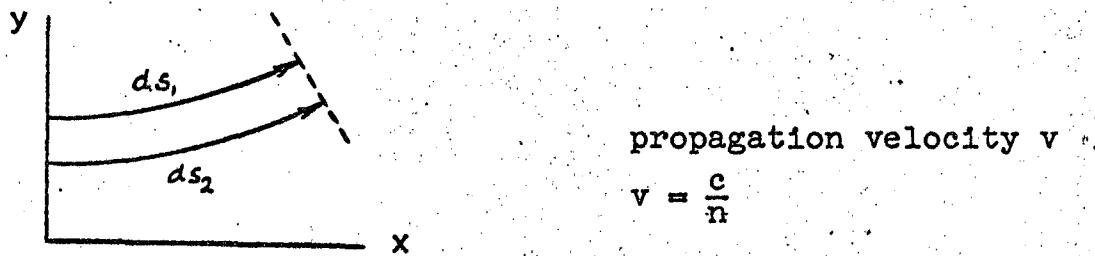
Inorganic Materials Research Division
Lawrence Radiation Laboratory
University of California
Berkeley, California

July 21, 1964

The following notes are the outline of a talk on the measurement of refractive index fields. They are, therefore, not a self-contained report, but rely on oral commentary. A review of different techniques of potential interest for the determination of diffusion constants for the study of heterogeneous mass transfer in solution is given with an extensive list of references.

Schlieren Methods

Based on curved light path due to refractive index gradient (Schliere = optical inhomogeneity). The resulting small angle light deflections can be measured in various ways, resulting in different techniques.



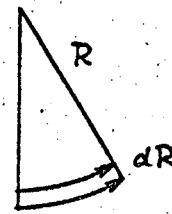
$$ds_1 = v_1 dt = \frac{c}{n} dt$$

$$ds_2 = v_2 dt = \frac{c}{n + dn} dt$$

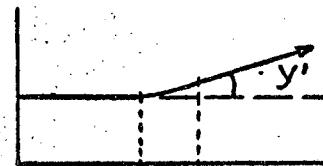
$$ds_1 n = ds_2 (n + dn)$$

Small angle deflections, radius of curvature R

$$\frac{1}{R} = \frac{1}{n} \frac{dn}{dR}$$



angle of deflection y' ($y' \ll 1$)



$$\frac{1}{R} = \frac{y'}{(1 + y'^2)^{3/2}}$$

$$y' = \int_{x_1}^{x_2} \frac{1}{n} \frac{\partial n}{\partial y} dx$$

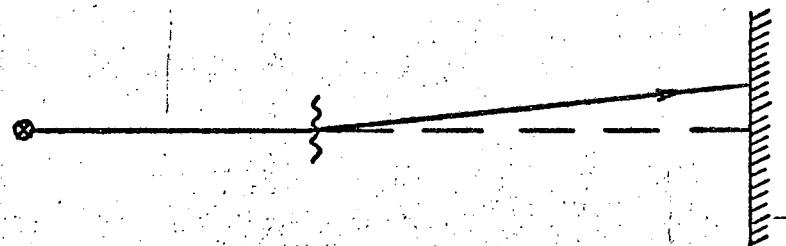
$$y' \approx \frac{1}{n} \frac{\partial n}{\partial y} (x_2 - x_1)$$

Shadowgraph

A Schlieren technique without focussing of the Schliere.

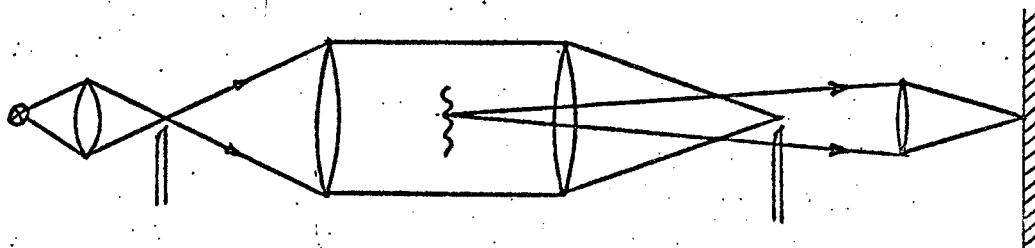
Point light source casts "shadow" of object. Deflected and undeflected light are superimposed on the screen. Only dark areas are geometrically similar to the density disturbance.

No optical system needed. Not quantitative.



Toeppler

A great variety of Schlieren methods with focussing of the Schliere are derived from Toepler's arrangement to transform the angular deflection into an intensity variation. A knife edge in the image plane of the light source prevents part or all of the undeflected light from contributing to the image of the object.



Interpretation of a Schlieren picture:

1. Extent and location of Schliere geometrically correct.
2. Brightness is a measure of refractive index gradient at any point.
3. Only the component of the light deflection normal to the knife edge is detected.

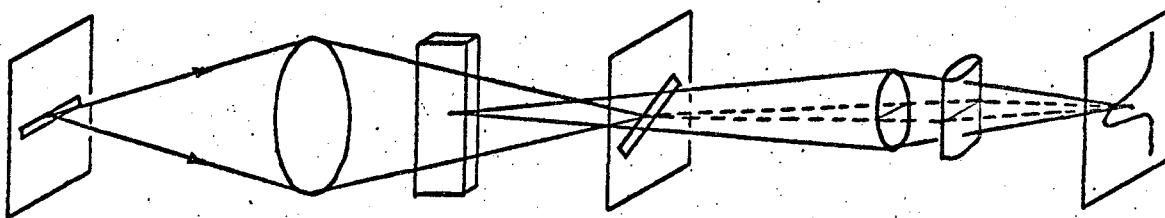
Extensions:

- a) deflection in two directions can be detected with round instead of linear sources and "edges".
- b) to avoid the necessity for photometric scanning of the picture, colored stripes can be used to differentiate between increments of deflection.

c) the principles a) and b) can be combined by use of colored segments of a circular disc.

Philpot - Svensson

Method of crossed slits for one-dimensional refractive index fields. The vertical deflection of a sheet of light is transformed into a horizontal displacement by an inclined slit. Focussing the cell in the vertical direction and the inclined slit in the horizontal direction results in a plot of distance vs. refractive index gradient.



Interferometric Methods

Based on phase difference due to different propagation rate. Phase detected by interference with another wave which may be a reference beam or another part of the same beam. Phase change ΔZ (number of wave lengths) for one-dimensional index field:

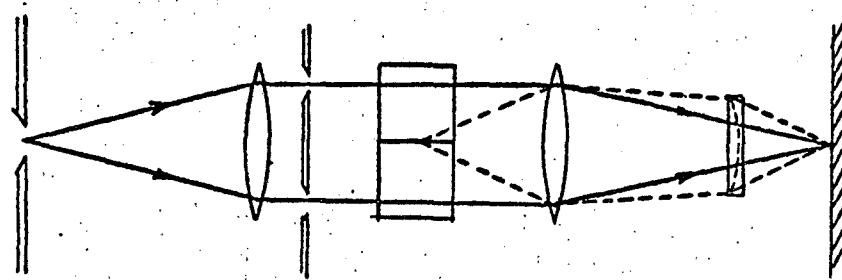
$$\Delta Z = \frac{d}{\lambda_0} \Delta n \quad d = \text{cell dimension.}$$

λ_0 = wave length in
vacuum.

Rayleigh

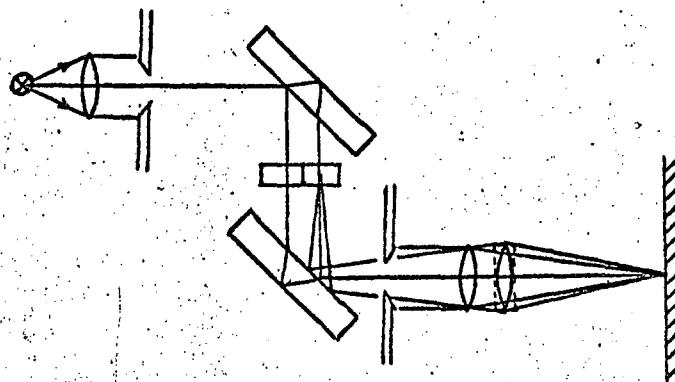
Two coherent beams formed with a double slit.* Image of slit in horizontal, of cell in vertical direction focussed.

*(division of wave front)



Jamin

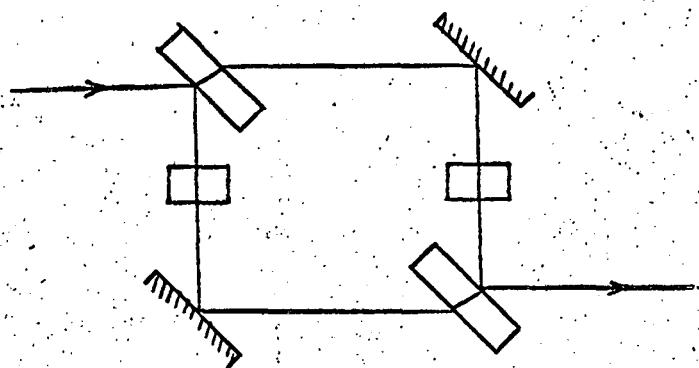
Coherent beams separated and united by reflection on front and backside of plano-parallel glass plates.* Normally used with a set of horizontal interference fringes introduced by tilting mirrors around horizontal axis. Vertical fringes can be obtained with additional optical elements according to Antweiler. Refractive index fields must be one-dimensional gradient should be parallel to fringes. Plates in parallel position result in "interferometric Schlierensystem".



*(division of amplitude)

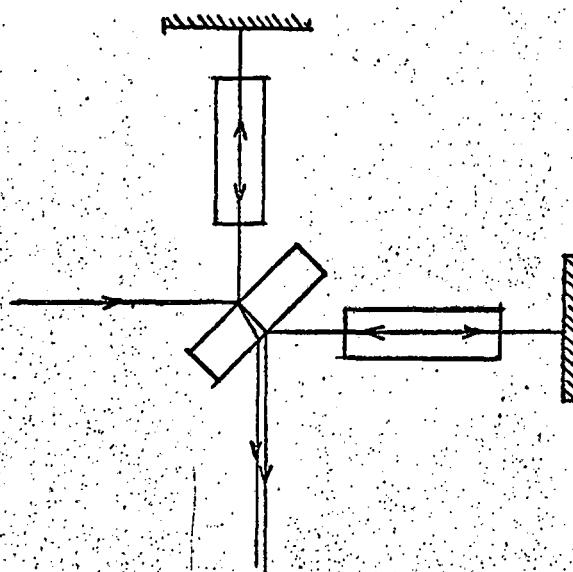
Mach-Zehnder

Four separated reflecting surfaces allow large spacing between both beams. Very expensive and difficult to align.



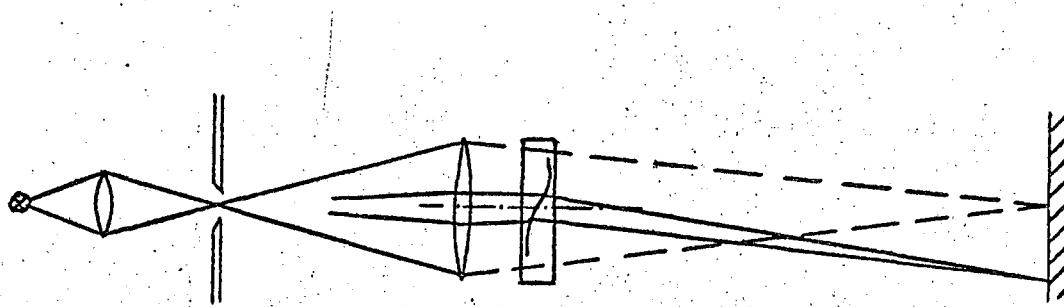
Michelson

The same plate serves to split and re-unite the beams. Cell is traversed twice in opposite directions. Has been modified by Lotmar and Guest et al..



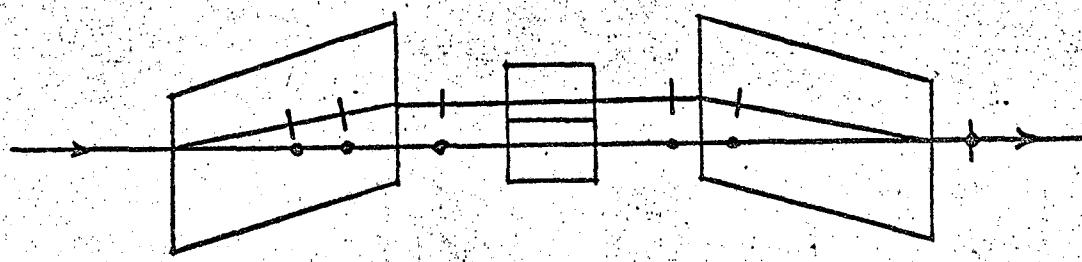
Gouy

Combines Schlieren and interference principles by bringing equally deflected rays from different parts of a diffusing boundary to interference. Index gradient curve has to be symmetrical and its shape must be known for evaluation. Used extensively for diffusion studies.



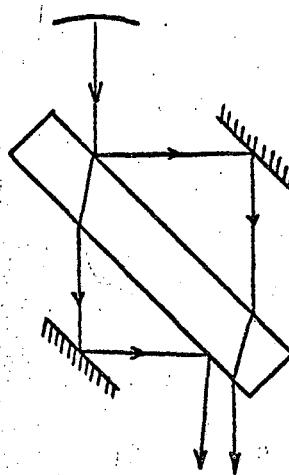
Lebedeff

Two interfering beams of small displacement are produced by a birefringent crystal plate.(division by polarization).



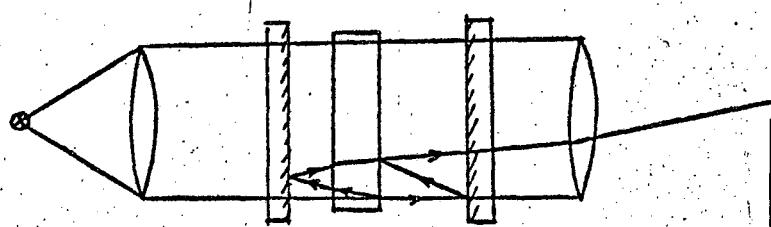
Shearing

Similar to Mach-Zehnder principle. Interference between two slightly sheared converging wave fronts. Thus, interference fringes analogous to contact fringes between spherical surfaces are obtained without a comparison piece. Several simplified versions have been proposed for testing optical components.



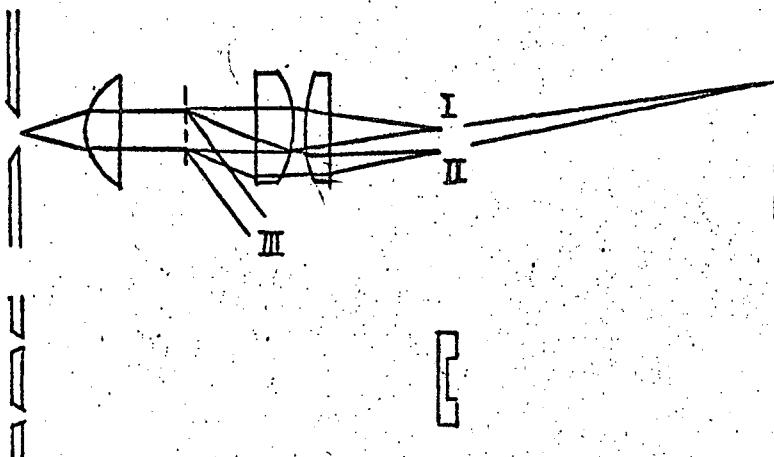
Multiple Beam

Two or more partially reflecting surfaces employed under almost normal incidence. Fringes can be made to be located in specimen. Multiple beam interference fringes can be much narrower than double beam fringes, thus allowing a higher resolution of lateral fringe displacement. Most constructions are compact and sturdy, yet little used.



Phase Contrast

A microscopic object produces a diffraction pattern of the light source in the focal plane of the objective. In the diffraction pattern of an absorbing (amplitude-) object all the maxima are in phase while in the pattern of a phase-object the zero order maximum shows a phase shift of $\lambda/4$. Correction of this phase shift produces an image like that of an amplitude object.

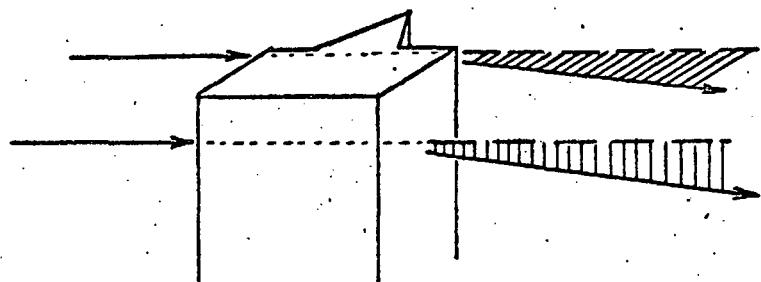


Combined Methods

Simultaneous recording of refractive index and index gradient has been achieved by combining the Toepler Schlieren system with a Mach-Zehnder Interferometer or by the combination of a Philpot - Svensson Schlieren System with a Rayleigh Interferometer.

The interferometric determination of gradients is also possible by the slight displacement of two identical cells by mechanical or optical means.

Of particular simplicity is the combination of refraction by a liquid-filled prism for refractive index with a Philpot - Svensson arrangement for the gradient.



Limitations and Errors

The geometrical resolution of optical instruments and, thus, their sensitivity is limited by light diffraction most noticeable in systems of low aperture. The curved light path due to refractive index gradients is a source of errors for the interpretation of interferometric results.

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