

## THE OPTICS OF HETEROGENEOUS MEDIA

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### 1. INTRODUCTION

I HAD occasion to read many of Prof. C. V. Raman's papers on the optics of heterogeneous media when I had to collaborate with Prof. G. N. Ramachandran in writing the monograph on Crystal Optics for Flugge's *Handbuch der Physik*. Recently I had to go deeper into many of these because of a rather interesting problem in materials science. When a composite or a polycrystalline aggregate is elastically deformed the question arises whether the strain-continuity condition is violated at the boundary between two phases or two grains? It seemed to us that the photoelastic measurements may be more effective in deciding this issue than the usual elastic measurements. To calculate the average photoelastic tensor we had to study many of the classic papers of Prof. Raman. In this talk I hope to present some of Prof. Raman's work in this field and show how we have used them to make some new contribution to this subject.

### 2. MIRAGES

Perhaps the most famous work on the optics of heterogeneous media associated with Prof. Raman is the Raman-Nath theory (1935) where optical heterogeneity is caused by elastic wave propagation. It is not so well known that Prof. Raman wrote another classic paper along with S. Pancharatnam on the Optics of Mirages (1959) where heterogeneity arises from thermal gradients in the medium.

It is generally believed that the phenomenon of the mirage is well understood and that it arises from total internal reflection. If light passes from a medium of refractive index  $n_1$  into a medium of lower refractive index  $n_2$  then  $n_1 \sin i_1 = n_2 \sin r_1$  where  $i_1$  and  $r_1$  are the angles of incidence and refraction. As  $i_1$  increases,  $r_1$  increases and when  $n_1 \sin i_c = n_2$  the refracted ray becomes tangential to the boundary between the two media. When the angle of incidence is greater than  $i_c$  the entire energy is reflected into the first medium. Near a hot surface there is a gradient with the refractive index increasing with distance from the hot surface. Hence it is often argued that a ray of light incident on the hot surface gradually gets curved and at the layer at which the critical angle condition is satisfied it gets totally reflected. Raman and Pancharatnam showed this argument to be untenable. A ray incident at angle  $i_1$  will gradually curve as it approaches the hot surface and will travel horizontally at a layer  $L_1$  where the refractive index is  $n_{1,1} = n_1 \sin i_1$ . When the angle of incidence increases to  $i_2$  the ray does not get totally reflected at the layer  $L_1$  but travels horizontally at an earlier layer  $L_2$  where refractive index  $n_{1,2} = n_1 \sin i_2$ . Under no circumstance can the ray be totally reflected back.

From the point of view of wave optics also there is a paradox. If the paths of the rays are known, the wavefront may be obtained by drawing the surface orthogonal to the rays. Hence if a bundle of adjacent rays which are at a finite

distance apart is taken, the part of the wavefront corresponding to these rays would be finite and this would contract as it goes through the thermal gradient and would ultimately converge to a point on the limiting layer, making the energy density infinite, a result not physically admissible.

As a result of the gradient in the refractive index, the wavefront travels faster near the hot surface. This causes the lower part of the wavefront to bend more than the upper part. A plane wavefront entering the hot zone therefore gets curved and the curvature continuously changes as the wave travels onward, and it finally emerges as a plane wavefront. This description is correct only at distances far off from the limiting layer. However around the limiting layer, Raman and Pancharatnam showed that the wavefront gets distorted and finally at the limiting layer it becomes a *cusped wavefront* with two branches which at large distances become the incident and the reflected wavefronts. It is well known that such cusped wavefronts must cause caustics, and the caustic fringes.

The problem bears close resemblance to the problem of light propagation through a transparent sphere on which I worked with Prof. Raman in 1949. Two interesting features were observed. Firstly, a concentration of intensity along the periphery of the light emergent from the sphere which was evidently in the nature of a caustic. Secondly, a concentration of intensity along the axial ray which had the nature of a continuous focus. In spheres made of birefringent material two sets of caustics were observed in general. It was found that a plane wavefront entering the sphere emerged as a curved wavefront with cusps at its edges. The cusped wavefront caused the caustic and one

could observe the associated interference fringes.

Most experimenters in this field including R. W. Wood had not observed the caustics and its associated fringes in mirages. This was because of an inherent drawback in the experimental arrangement they used. The hot surface was kept horizontal and the convection currents therefore smeared out the caustic surface. Raman and Pancharatnam got over this by a rather ingenious method. To quote from the paper "the above difficulty was removed by the artifice of turning the hot plate edgewise so that the hot surface was now vertical, though its length remained horizontal. The hot air now flowed up in streamlines parallel to the surface of the plate. With the slit now vertical, it was possible to observe clearly with the aid of a pocket lens a bright caustic bordered by large number of interference fringes". They observed almost fifty fringes and established the general correctness of the theory.

Raman and Pancharatnam end their paper, by drawing attention to the fact that the theoretical technique employed by them to solve the problem was similar to that used in quantum mechanics. There is a quantum mechanical analogue to the phenomenon of mirages. When a particle of energy  $E$  meets a potential barrier of height  $V$ , there will be both reflection and transmission. The wave-vectors of the reflected and the transmitted waves are  $k_1 = \sqrt{2mE}/\hbar$  and  $k_2 = \sqrt{2m(E-V)}/\hbar$  respectively. When  $E > V$  both reflected and transmitted waves exist. However when  $E < V$ ,  $k_2$  becomes purely imaginary making the transmitted beam evanescent with no transmission of energy across the barrier. If instead of a step function we have a gradually varying potential, then total

internal reflection takes place when  $E < V$ . It can be shown that under condition of critical reflection the wave oscillates in the first medium in a manner identical to what happens when caustic fringes are formed in mirages. In this context one must mention a paper by Prof. Raman (1926) published in the *Trans. Opt. Society of America*, wherein he derived almost all the information concerning the evanescent wave of the total internal reflection using the simple Huyghen's principle. Elegant experiments were also performed by him to study these waves. One is also tempted to draw attention to an interesting paper by Prof. Raman in the *Astrophysical Journal* (1921). In 1921 Einstein suggested that the nature of light regarding its corpuscular or wave behaviour could be decided on by observing the Doppler shift of light emitted by fast moving canal rays. The arguments presented by Einstein were proved to be incorrect by young Raman who showed that Einstein had taken a wave instead of a wave packet or in other words had considered phase velocity instead of group velocity.

### 3. OPTICS OF POLYCRYSTALS

Prof. Raman was very much intrigued by the optical properties of substances like jadeite (1955) and alabaster (1954), materials which in the hands of a lapidary become objects of exquisite beauty. These materials are polycrystalline aggregates of anisotropic crystallites. In thin sections they are translucent. For example jadeite in thin sections is greenish while marble is pinkish. The brilliant whiteness of pure marble in thick sections is due to large birefringence of calcite crystallites causing intense intercrystalline reflections. Raman and Viswanathan (1955) took a

fresh look into the optics of polycrystalline aggregates. It was very clear that geometrical optics would fail to answer many of these questions. For instance as the crystallites become smaller, more and more intercrystalline reflections should occur and, decrease the transparency. This is contradictory to the experimental findings of Raman and his collaborators. Thus Raman and Viswanathan had to invoke wave optical principles to explain the problem of light transmission in polycrystals.

They used a very simple model for crystallite orientation. Each crystallite was assumed to be cubical in shape with the cube edges parallel to the principal axis of the index ellipsoid of the crystallites with  $n_1$ ,  $n_2$  and  $n_3$  as their respective refractive indices. The crystallites were assumed to be randomly packed with  $p_1$ ,  $p_2$  and  $p_3$  as probabilities of occurrence of edges with indices  $n_1$ ,  $n_2$  and  $n_3$  respectively. The incident wave was taken to be plane polarized with vibration direction parallel to the cube edge. Clearly the beam remains plane polarized throughout its travel in the medium but on emergence the phase varies randomly from point to point over the area of the polycrystal. Using these principles they obtained the following expression for the intensity of the direct beam :

$$I = I_0 P^2 \times \exp. \left\{ - \frac{4\pi^2}{\lambda^2} \bar{\tau} \sum p_1 p_2 (n_1 - n_2)^2 \right\} t \quad (1)$$

where  $P$  takes into account intercrystalline boundary reflections,  $\bar{\tau}$  is the average crystallite size,  $t$  is the thickness of the sample and  $\lambda$  the wavelength of light. They clearly showed that a randomly oriented polycrystalline aggregate of anisotropic crystallites (which are small) behaves like an isotropic solid. There is

a decrease in intensity in the exact forward direction which is a function of crystallite birefringence and varies directly as the crystallite size  $\bar{\tau}$  and inversely as  $\lambda^2$ .

We [Ranganath and Ramaseshan (1972 a), Ranganath (1972 a)] have generalized this model assuming that the index ellipsoids in various crystallites are randomly disposed in the body of the polycrystal. We have used the Poincaré sphere and the Mueller matrix method to solve this problem. When a completely polarized plane wavefront falls on the sample, different parts of this wavefront travel through different portions of the polycrystal. This results in a continuous change in polarization of the wave as it travels through the medium. This is equivalent to a random walk on the surface of the Poincaré sphere. Thus the emergent light will consist of a set of beams polarized in different states but having the same intensity. If we assume the beams to be uncorrelated we get for the direct beam

$$I = I_0 \exp. \left\{ - \frac{2\pi^2}{\lambda^2} \bar{\tau} \overline{(\delta n)^2} \right. \\ \left. \times (1 - \frac{1}{2} \cos^2 2\omega_0) \right\} t \quad (2)$$

where  $\overline{(\delta n)^2}$  is the mean crystallite birefringence,  $\omega_0$  ( $= \tan^{-1} b/a$ ) is the ellipticity of the incident wave. This equation is similar to (1) excepting for an extra factor involving  $\omega_0$ . This is a new result which shows that the intensity diminution also depends on the state of polarization of the incident beam. We also get the general result that direct beam has the same state of polarization as the incident light for optically inactive crystallites, a result amply supported by experiments of Prof. Raman. The problem of coherence between the different beams which results in diffraction

has also been considered (see Ranganath's article, p. 386).

The next question is to consider the case of birefringent and optically active crystallites. In this case the polycrystalline medium behaves like an isotropic optically active solid having a rotatory power equal to the mean rotatory power of the single crystal of which the medium is made. Optical rotation in a single crystal is described by a second rank symmetric axial tensor ( $g_{ij}$ ). Polycrystals will have a gyration  $\bar{g} = 1/3 (g_{11} + g_{22} + g_{33})$ , with  $g_{11}$ ,  $g_{22}$  and  $g_{33}$  as the principal components of ( $g_{ij}$ ). In the case of uniaxial enantiomorphic optically active crystals  $g_{11} = g_{22} \neq g_{33}$ . Only  $g_{33}$  can ordinarily be obtained by measuring rotation along the optic axis of a single crystal. The present analysis shows that by measuring rotation in a polycrystalline aggregate one can get the value of  $g_{11}$  also. In  $\alpha$ -quartz throughout the visible region it so happens that  $g_{11} = g_{22} = -1/2 g_{33}$  giving the optical rotation in the polycrystal to be zero. This is the reason why Raman and Bhat (1955) found no optical rotation in  $\alpha$ -quartz polycrystals.

Part of the optical rotation in crystals, containing of optically active molecules, is due to molecular rotation. For computing the part due to molecular rotation in the crystal one requires a complete knowledge of the rotatory anisotropy of the molecule, i.e., a knowledge of its ( $g^\circ$ ) tensor. Optical rotation in a liquid gives only  $1/3 (g_{11}^\circ + g_{22}^\circ + g_{33}^\circ)$ . To get all the principal components we must perform two more experiments in which molecules are aligned to different extents. Raman and Krishnan (1928) have studied in detail alignment of molecules of a liquid when it flows or when placed in an electric field. Their ideas have directly been used to get the principal

components of ( $g^{\circ}$ ). This change in rotation under molecular alignment would be very small and would be observable only near an absorption band.

#### 4. PIEZO-OPTICS IN POLYCRYSTALS

The general problem of light propagation in a polycrystal can now be extended to include effects of stress on the polycrystalline aggregate. We find that the polycrystal becomes linearly birefringent under a uniaxial stress. From a knowledge of piezo-optical constants of single crystals we can calculate [Ranganath and Ramaseshan (1972 b)] the average photoelastic birefringence for stress or strain continuity in the sample. Some results are shown in Table. I.

TABLE I  
Values of  $\bar{P}_{44} \times 10^{+2}$

Strain continuity		Stress continuity	
NaCl	.. -1.279		-1.647
KCl	.. -0.357		-1.536
KBr	.. -0.007		-1.228
KI	.. +0.481		-0.488
RbCl	.. +0.613		-1.655
RbBr	.. +0.858		-1.984
RbI	.. +1.029		-1.359

From Table I we conclude photoelastic studies in polycrystals can be a very sensitive tool in deciding the issue whether the strain continuity is violated or not.

#### 5. OPTICS OF PREFERENTIALLY ORIENTED POLYCRYSTALS

This problem is of relevance to a geophysicist. Tectonic movements result in preferential orientation of quartzite polycrystals present in the surface layers of the earth. Conversely systematic studies of the preferred orientation would give information about the tectonic movements. Determination of preferred orientation by X-ray methods is a laborious and time-consuming process. However optics can be used as a powerful tool in this analysis. Raman's method of studying the polarization and intensity characteristics of the diffusion haloes that surround a beam of light, emerging from the polycrystal, can be usefully employed for getting data about the preferred orientation [Ranganath (1972 b)].

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