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The theory of the propagation of light in polycrystalline media

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1. Introduction

Many common minerals occur in nature as polycrystalline aggregates. For example, quartz, calcite and gypsum appear in massive form respectively as quartzite, marble and alabaster, and on examining thin sections of these materials under the polarisation microscope, it is found that they consist of great numbers of crystallites variously orientated and firmly adherent to each other so as to form a coherent solid. The size, shape and manner of orientation of the crystallites may differ enormously in individual cases. Some minerals are indeed cryptocrystalline, in other words, the particles are so small that they cannot be identified by the usual polariscopic methods and require the aid of X-ray analysis to enable their true nature to be determined.

The foregoing is by way of stressing the importance alike to the mineralogist and to the physicist of a study of the physical properties of polycrystalline aggregates. It is obvious that the optical properties of the single crystal and especially its birefringence and pleochroism (if any) would play a dominant role in determining the optical characters of the polycrystalline aggregate. Considering the matter from the standpoint of geometrical optics, it is evident that when light enters a polycrystalline aggregate, it would suffer reflection at the intercrystalline boundaries. The stronger the birefringence, the greater would be the coefficient of reflection at these boundaries and hence the more quickly would the incident light be returned back towards the source. The brilliant whiteness of pure marble is thus a recognisable consequence of the strong birefringence of calcite. On the other hand, if the birefringence be feeble as in the case of quartz and gypsum, the incident light would penetrate far more deeply into the aggregate. Ultimately, all the light would necessarily be turned back provided that a sufficient thickness of the material be available and that no absorption intervenes. If the thickness of the material be insufficient, a part of the light would diffuse—a phenomenon readily observed with various materials. Light so emerging would be depolarised even if the light incident on the plate were fully polarised in the first instance.

Various considerations indicate that a purely geometric theory is inadequate to cover all the optical phenomena actually exhibited by polycrystalline media. It will suffice to mention here the question of the influence of the size of the crystallites. Geometric considerations would suggest that the smaller the crystallites, the more numerous would be the reflections and refractions at the intercrystalline boundaries, and hence the more rapidly would the incident light be diffused and extinguished in its passage through the medium. Experience however suggests that the contrary may actually be the case, and that the more fine-grained the material is, the more deeply would the light penetrate into it. Various minerals, e.g., alabaster and jade, which exhibit marked translucency, are usually fine-grained; the finer the grain, the more deeply does light penetrate into them. This suggests that the optical problems presented by polycrystalline aggregates require to be considered from the standpoint of the wave-theory of light. That indeed is the object of the present paper.

2. A simplified model

To obtain some results of physical interest and also with a view to simplify the mathematics, we shall here restrict ourselves to the case of a feebly birefringent material and consider the case in which light is incident normally on a plate with parallel faces; this is assumed to be sufficiently thick to include a great many individual crystallites but not so thick that the incident light is completely extinguished before it can emerge at the rear face. We may disregard the geometric course of the individual rays of light and view the matter purely from the wave-theoretical standpoint. Owing to the varying orientation of the individual crystallites, the waves of light entering the plate would be retarded to different extents in passing through them. To enable the resulting total retardation to be evaluated, we use a simplified model and assume the plate to be an assembly of a great number of small cubical blocks each having a common edge-length Δ and completely filling up the available space. Each block is assumed to be a single crystallite, and the three edges of each cube to be parallel to the three optic directions for which the refractive indices are μ_1, μ_2 and μ_3 respectively. To introduce the idea of varying orientation and to take account of its influence on the propagation of light through the material, we assume the incident light-beam to be plane-polarised with its vibration direction parallel to one set of edges of the cubical blocks; on the other hand, the operative refractive index of any one block may be either μ_1 or μ_2 or μ_3, the respective probabilities for these being p_1, p_2, p_3. The case where the three probabilities are equal would correspond to a random orientation of the crystallites in the present restricted sense of that term. More generally, by giving appropriate values to p_1, p_2, p_3 such that their sum remains equal to unity, we obtain a representation of a polycrystalline aggregate with any desired measure of preferred orientation along the particular direction under consideration. If, for example, we put $p_1 = 1$ while p_2 and p_3 are zero, it would mean that all the particles of the aggregate have a common refractive index for the particular direction of vibration, though the indices may be different in the perpendicular direction.

On the assumptions stated, the incident plane-polarised disturbance would remain plane-polarised in its passage through the plate, though subject to phase retardations of varying extents. The situation would no doubt be different for any actual polycrystalline material, since the incident plane-vibration would be transformed to an elliptic vibration and the parameters describing the ellipticity would alter as the disturbance passes from crystallite to crystallite. While it would no doubt be possible to deal mathematically with this general case, a very considerable simplification is effected by our present assumptions, and as we shall see, the usefulness of the results obtained is not affected thereby.

A further question needing consideration is the effect of the reflections which would occur at the boundary between every two successive blocks. This would obviously diminish the amplitude of the transmitted disturbance. As a first approximation, we may assume such diminution to be the same over all the individual elementary areas Δ^2 on the rear surface of the plate and represent it by a numerical factor of appropriate magnitude. In other words, we ignore the variation of *amplitude* over the different elementary areas Δ^2 on the rear face of the plate and consider only the variations of *phase*. The latter are in reality of much greater importance for the determination of the final observable result.

3. Mathematical formulation

Let us suppose that the wave-train before entry into the plate is represented by

$$y = \exp\left[\frac{2\pi i}{\lambda}(ct - Z)\right] \tag{1}$$

and that there are n cells along the direction of the thickness of the plate. We shall first consider a typical case in which the wave has passed through k_1 cells of refractive index μ_1, k_2 cells of refractive index μ_2 and k_3 cells of refractive index μ_3 before emerging from the plate. The numbers k_1, k_2 and k_3 can all vary from zero to n subject to the relation

$$k_1 + k_2 + k_3 = n. \tag{2}$$

The optical path retardation of the emergent wave would then be equal to $(k_1\mu_1 + k_2\mu_2 + k_3\mu_3)\Delta$.

Now the number of ways in which k_1, k_2 and k_3 cells can be orientated along a row of n cells so as to have refractive indices μ_1, μ_2 and μ_3 is obviously

$$\frac{n!}{k_1!k_2!k_3!},$$

and the probability of occurrence of each one of these cases is $p_1^{k_1}p_2^{k_2}p_3^{k_3}$. Hence the proportion of the total area of the rear surface of the plate from which a wave represented by

$$\exp\left[\frac{2\pi i}{\lambda}(ct - Z - \overline{k_1\mu_1 + k_2\mu_2 + k_3\mu_3\Delta})\right] \tag{3}$$

emerges is equal to

$$\frac{n!}{k_1!k_2!k_3!}p_1^{k_1}p_2^{k_2}p_3^{k_3}. \tag{4}$$

The emergent wave-train can now be obtained by summation of waves of the type (3) with their appropriate amplitudes and phases for all possible integral values of k_1, k_2 and k_3 satisfying the relation (2). We therefore have for the emergent wave

$$y = P\sum_{k_1+k_2+k_3=n}\frac{n!}{k_1!k_2!k_3!}p_1^{k_1}p_2^{k_2}p_3^{k_3}\exp\left[\frac{2\pi i}{\lambda}(ct - Z - \overline{k_1\mu_1 + k_2\mu_2 + k_3\mu_3\Delta})\right] \tag{5}$$

where P is a factor which is introduced to take into account the loss in intensity of the light due to reflections at the intercrystalline boundaries.

In view of the multinomial theorem, equation (5) may be rewritten as

$$y = P\exp\left[\frac{2\pi i}{\lambda}(ct - Z)\right]\left[p_1\exp\left(\frac{-2\pi i\mu_1\Delta}{\lambda}\right) + p_2\exp\left(\frac{-2\pi i\mu_2\Delta}{\lambda}\right) + p_3\exp\left(\frac{-2\pi i\mu_3\Delta}{\lambda}\right)\right]^n. \tag{6}$$

The average refractive index of the medium is clearly

$$\mu = (p_1\mu_1 + p_2\mu_2 + p_3\mu_3). \tag{7}$$

If therefore we set $\nu_1 = (\mu_2 - \mu_3)$; $\nu_2 = (\mu_3 - \mu_1)$; and $\nu_3 = (\mu_1 - \mu_2)$, we can then express μ_1, μ_2 and μ_3 as

$$\begin{aligned} \mu_1 &= \mu + (p_2\nu_3 - p_3\nu_2) \\ \mu_2 &= \mu + (p_3\nu_1 - p_1\nu_3) \\ \mu_3 &= \mu + (p_1\nu_2 - p_2\nu_1). \end{aligned} \tag{8}$$

Further, the thickness of the plate is given by $d = n\Delta$. Hence substituting the relations (8) in (6) and expanding the exponential terms in power series of their arguments, one obtains

$$y = P\exp\left[\frac{2\pi i}{\lambda}(ct - Z - \mu d)\right] \times \left\{1 - \frac{2\pi^2\Delta^2}{\lambda^2}\sum p_1(p_2\nu_3 - p_3\nu_2)^2\right\}^n. \quad (9)$$

As the birefringence is assumed to be small, we have ignored terms of third and higher powers of $(\mu_1 - \mu_2)$, $(\mu_2 - \mu_3)$ and $(\mu_3 - \mu_1)$ in (9). Also by means of a small simplification, it can be verified that

$$\sum p_1(p_2\nu_3 - p_3\nu_2)^2 = \sum p_2 p_3 \nu_1^2.$$

Hence, we can rewrite (9) as

$$y = P\exp\left[\frac{2\pi i}{\lambda}(ct - Z - \mu d)\right]\left\{1 - \frac{1}{n} \times \frac{2\pi^2\Delta d}{\lambda^2}\sum p_2 p_3 \nu_1^2\right\}^n$$

$$= PR\exp\left[\frac{2\pi i}{\lambda}(ct - Z - \mu d)\right],$$

where

$$R = \exp\left(\frac{-2\pi^2\Delta d}{\lambda^2}\sum p_2 p_3 \nu_1^2\right) \quad \text{as } n \text{ is large.}$$

The ratio of the intensity of the transmitted light to that of the incident radiation is therefore given by

$$\frac{I}{I_0} = P^2R^2 = P^2\exp\left[\frac{-4\pi^2\Delta d}{\lambda^2}\sum p_2 p_3(\mu_2 - \mu_3)^2\right]. \quad (10)$$

If the three optic axes of any cube have the same probability of being orientated in the direction of the incident light, then $p_1 = p_2 = p_3 = \frac{1}{3}$ and (10) reduces to

$$\frac{I}{I_0} = P^2\exp\left[\frac{-8\pi^2\Delta d}{9\lambda^2}\left(\sum \mu_1^2 - \sum \mu_2\mu_3\right)\right]. \quad (11)$$

4. Significance of the results

The physical meaning of the result stated in (10) is that the plane-polarised waves incident on the front of the plate emerge from the rear face of the plate also as a plane-polarised vibration but with an attenuated amplitude determined by an exponential factor involving four variables, namely the size of the particles, the thickness of the plate, the wavelength of the light and a quantity which is a measure of the birefringence of the material, since it vanishes when the three indices μ_1, μ_2, μ_3 are all equal. The appearance of λ^2 in the denominator indicates that white light entering the plate would emerge enfeebled but with the longest waves predominant, in other words, much reddened in colour. Since the wavelength λ is a small quantity, the actual intensity of the emerging light would be negligible if both Δ and d are large. The individual crystallites have, in fact, to be quite small and the total thickness traversed should be moderate if any observable fraction of the light is to emerge as a coherent optical beam. We have already assumed the birefringence to be small and the need for such assumption is reinforced by our final result which indicates that unless the three indices μ_1, μ_2, μ_3 differ from each other by quantities which are small fractions of their absolute magnitudes, no light can emerge from the rear of the plate.

We may illustrate the foregoing remarks by the case of a plate of alabaster 1 mm thick taking λ and Δ equal to 5896 A.U. and 1 μ respectively; the three indices for gypsum are $\mu_1 = 1{\cdot}520$, $\mu_2 = 1{\cdot}523$ and $\mu_3 = 1{\cdot}530$. The percentage of transmission then comes out as 13·5% but increases to 37% and 82% if $\Delta = 0{\cdot}5\,\mu$ and $0{\cdot}1\,\mu$ respectively. Thus, the formula indicates that a plate of alabaster approaches practically complete transparency as the crystallites of which it is composed approach colloidal dimensions.

The case of preferred orientations is also of interest in view of the known optical behaviour of chalcedony and of certain forms of gypsum. Taking now the general formula (10), if we put $p_1 = 1$ while p_2 and p_3 are both zero, the formula indicates that the transmission becomes complete. In other words, if the crystallites are so orientated that all of them have a common refractive index for the direction of vibration of the incident light, then we have a complete transmission of the incident light wave. But the position would be totally different for a perpendicular direction of vibration if the refractive indices for the latter direction differ from crystallite to crystallite. The general formula (10) would then show only a partial transmission depending upon the actual values of the probabilities and the refractive indices for that direction. Since the latter transmission would depend upon the thickness d, it would follow that if the light incident upon the plate be unpolarised, the state of polarisation of the emerging light would vary with the thickness of the plate. Formula (7) also indicates that the effective refractive index of the medium would be different for the two directions of vibration under consideration. Light which is plane-polarised in any arbitrary azimuth when incident on the plate, would emerge as elliptically polarised light, the parameters describing such ellipticity varying with the thickness of the plate—a phenomena readily capable of experimental verification with materials of the nature under consideration.

5. Some further remarks

The question naturally arises as to where the energy goes which disappears from the incident light according to (10). The answer to this is not far to seek. Since the reduction of amplitude is a consequence of the random variations of phase over the elementary areas of the rear face of the plate, the missing light would appear as diffracted radiation spread out in various directions surrounding the direction of the incident beam. The angular dimensions of the diffraction halo would obviously be comparable with the ratio between the wavelength λ of the light and the linear dimension Δ of the crystallites which we have assumed the material to be composed of. A diffusion halo of this type can indeed readily be observed on viewing a bright source of light through a thin plate of alabaster surrounding the sharply defined image of the source itself. According to the theory developed above, both the light emerging from the rear surface of the plate and the light appearing in the diffusion halo would be perfectly polarised if the incident light be itself plane-polarised. These results are consequences of the special assumption regarding the orientation of the crystallites which we have made. We may now ask ourselves whether they would continue to be true if the particles are orientated truly at random. The answer to this question is most readily ascertained by making a few actual observations with a plate of alabaster sufficiently thin to give a true transmission. It is then observed that the true transmission is completely polarised while the diffraction halo seen overlying it is imperfectly polarised. A more complete mathematical theory which takes account of the ellipticity resulting from the passage of light through an arbitrarily orientated crystal block would no doubt yield results in agreement with these facts of observation. It is clear, however, that the present theory suffices to indicate the state of polarisation of the transmitted light correctly and also its intensity, at least as regards the order of magnitude. But the theory fails to indicate the state of polarisation of the diffracted light accurately, since it ignores the ellipticity produced by the passage of light through a birefringent crystal in an arbitrary orientation; such ellipticity would obviously result in diverting some of the incident energy into the perpendicular component of vibration as diffracted radiation.

Summary

A formula based on wave-theoretical considerations is deduced which gives the coefficient of extinction of plane-polarised light traversing a polycrystalline aggregate in terms of the wavelength of the light, the size of the particles and their birefringence. The general formula covers the case where the particles have preferred orientation expressible by three different probability numbers for three mutually perpendicular directions, and the special case of isotropic orientation is readily derivable therefrom. The significance of the results is discussed in relation to the facts of observation.