Proc. Indian Acad. Sci. **A39** 265–271 (1954)

On the chromatic diffusion halo and other optical effects exhibited by pearls

SIR C V RAMAN and D KRISHNAMURTI
(Memoir No. 52 from the Raman Research Institute, Bangalore)

1. Introduction

The present paper concerns itself with the optical phenomena observed when a pearl is illuminated by a light-source of small angular dimensions and is viewed in the reverse direction. They were briefly reported upon in these *Proceedings*[1] but will be described and discussed rather more fully in the present communication. It appears worthwhile doing this, for, as was explained in the earlier paper, it is these phenomena that make the pearl appear so attractive an object when illuminated by extended light-sources.

As is well-known, the material of the pearl consists of an immense number of tiny crystals of aragonite held in an encompassing network of conchyolin. In the previous paper, the details of this structure and its optical behaviour have been discussed in detail and it is therefore not necessary to traverse the same ground here. The photographs reproduced as figures 21 and 22 in plate V are however of interest, as they illustrate the conclusions there stated. Figure 21 is an enlarged view of the shell of a cultured pearl as observed between crossed polaroids in parallel light. It was prepared by grinding down a fragment of the shell into a flat thin plate and mounting it in canada balsam between glass cover slips. From the birefringence pattern seen in the photograph, it is evident that the material consists of successive layers of approximately spherical shape superimposed on each other. However, the extinctions are far from being uniform, nor are the rings regularly spaced, thereby indicating that neither the optic orientation of the crystallites nor the spacing of the successive layers is completely regular. Figure 22 exhibits a small region of the same preparation as seen under a microscope between crossed nicols in parallel light; the approximately linear arrangement of the crystallites is well shown.

2. Some general observations

It is obvious that a material consisting of discrete crystallites would be optically turbid and would strongly diffuse the light traversing it. The explanation usually given of the lustre of pearls as due to internal reflection and/or diffraction at the surface ignores this feature and is totally inadequate. Actually, we observe three distinct effects: (a) the reflection and diffraction of light by the stratifications; (b) a chromatic diffusion halo which surrounds the direction of reflection; and (c) a diffusion of light through large angles consequent on a "whispering gallery effect". It is the co-operation of these three phenomena that results in the characteristic lustrous appearance of the pearl.

Figures 1, 2, 3 and 4 in plate I illustrate all the three phenomena referred to above. They are photographs of a natural pearl illuminated by a light-source of small angular dimensions and viewed in the reverse direction. The pearl was not perfectly spherical and in consequence, the four figures which were recorded in slightly different settings of the pearl show recognisable differences. In figure 1, we observe a single spot at the centre and this is surrounded by a diffuse halo. In figures 2, 3 and 4 we observe two, three and four bright spots respectively, as also a diffuse halo which has its maximum intensity in the vicinity of the last of the spots. Outside the halo, there is a general luminosity which has its maximum intensity at or near the spherical periphery, thereby making the latter appear very clearly visible.

Before we proceed to describe and discuss these effects in detail, it should be remarked that they have their analogues in the phenomena observed and described by one of us many years ago[2,3] in the case of the iridescent molluscan shells. But there are differences consequent on the fact that we are now concerned with strongly curved layers instead of plane or nearly plane stratifications.

3. The reflection-diffraction spectra

The optical effects arising at the surface of a stratified medium depend on the angle at which its layers meet the external surface, as also upon the actual configuration of that surface. In the particular case when the stratifications are perfectly parallel to the external face, the incident light-waves would be reflected at that face and also at each of the boundaries between the successive strata. Since these surfaces are curved in the present case, such reflections would result in a well-defined image of the source being visible when the pearl is viewed through a suitably held lens. The image would exhibit colour consequent on the mutual interference of the reflections from the successive boundaries.

If the stratifications of the material meet its surface at an angle, the external boundary would present the aspect of a diffraction-grating, and if it is sensibly corrugated, would give rise to a series of diffraction spectra with more orders on one side than on the other. As was shown in the case of iridescent shells and illustrated by a series of photographs in a recent paper by the present authors,[4] the sequence of spectra thus resulting would include the light internally reflected by the layers of the material as one of them, usually the last of the series. Further, the order of that spectrum would be the order of the interference of the reflections from the successive strata. In the present case, we are concerned with a convex diffraction grating and the spectra to which it gives rise would be focussed by a lens to a series of spots or streaks.

The foregoing consequences of the theory are in agreement with what is actually observed and are illustrated by figures 1, 2, 3 and 4 in plate I already referred to. In the case of cultured pearls of good quality, the stratifications are usually so nearly parallel to each other and to the external surface that the separation of the different orders in the reflection-diffraction spectrum is very small. Careful focussing is then necessary to enable them to be seen clearly separated.

4. Nature and origin of the chromatic halo

We may here appropriately refer to the phenomenon[3] of the body colour exhibited by nacreous iridescent shells when illuminated and observed in directions other than of the internally reflected light. This body colour is most vivid in directions not too remote from that of the internal reflection and then exhibits a complementary tint. It fades away in intensity and also changes colour as the direction of observation is further removed from that of regular reflection. The angle of incidence of the light on the surface of the material influences the colour of the internal reflection as also the body colour, but their complementarity when viewed in adjacent directions persists.

The chromatic diffusion halo exhibited by pearls is a phenomenon of the same general nature as the body-colour of nacreous shells referred to above, but the shape of the pearl and the curvature of its reflecting layers have important consequences. When the pearl is illuminated by a light-source of small angular dimensions and is viewed in the opposite direction, the angle of incidence of the light on the surface alters progressively from the centre outwards, and the light rays reaching the observer also make increasingly larger angles with the normal to the surface. Hence, what is observed would be an ensemble of effects; at the centre, the internal reflection would appear as a bright spot, and surrounding it would appear the body colour with its intensity and tint changing progressively from the centre outwards consequent on the changing angles of incidence and observation.

The foregoing analogy between the chromatic diffusion halo of pearls and the body colour exhibited by iridescent nacre enables us to predict various features of the former phenomenon. The diffusion halo should in the immediate vicinity of the reflected light exhibit the complementary colour. Then again, the larger the pearl the greater would be the apparent size of the halo as observed on its surface. If the pearl is not spherical in shape, but has a different curvature in different planes, we should expect the halo to spread further out from the centre in the plane of least curvature. Complications may also be expected if the stratifications of the material are not parallel to its external surface. For, instead of a simple reflection we would then have streams of diffracted radiations emerging in different directions and these would be superposed on the diffusion halo, thereby modifying the effects observed. Hence, the chromatic diffusion halo is best studied with pearls in which the separation between the various orders of the reflection-diffraction spectrum is either zero or very small.

The origin of the chromatic diffusion halo is evidently to be sought for in the fact that the reflecting layers in the pearl are not optically continuous but consist of individual crystallites of aragonite held together in a network of conchyolin. Each crystallite would therefore function as a diffracting particle, and the light returned by an individual layer would not be confined to the direction of geometric reflection, but would be spread out over a cone whose angular dimensions would be determined by the size and shape of the individual crystallites. The distribution of intensity of the diffracted rays within such cone would necessarily also be modified by the interference of the rays diffracted by adjacent crystallites. The manner in which the crystallites are disposed in the individual layers would therefore also have to be considered. The grouping of the crystallites in parallel linear rows which appears to be a common feature in cultured pearls (see figure 22, plate V) is a factor which needs to be taken into account in this connection. We may, in fact, expect to find that the arrangement of the crystallites in parallel and roughly equidistant rows would give rise to specific features in the chromatic diffusion halo, as it actually does in the diffusion haloes observed in transmitted light.[1] Finally, we have also to consider that the light emerging from the material is not a simple summation of the light diffused by successive layers. The progressive changes in phase and intensity of the incident light as it advances into the material and the mutual interferences of the radiations diffused backwards by the successive layers would have to be taken into consideration. Indeed, it is only on some such lines that we could hope to explain the complementarity of colour in adjacent directions of the reflected and diffused radiations.

5. Some illustrative examples

Reproductions in natural colour would be necessary to exhibit the beauty of the chromatic haloes of pearls in a satisfactory manner. However, by using panchromatic films and when necessary also colour filters, it has been possible to obtain pictures which show a fair degree of photographic contrast and exhibit the characteristic features of the halo. Twelve such photographs showing typical examples are reproduced as figures 9 to 20 in plates III and IV. Individual figures amongst them illustrate the various features referred to above. For example, the influence of the shape of the pearl on the configuration of the halo is shown clearly by the circular haloes reproduced in figures 9, 10 and 13 which were obtained with spherical pearls. On the other hand, figure 11 which exhibits an elliptic halo was recorded with a spheroidal pearl, its major elongation being in the same direction as the elongation of the halo itself. Figure 14 which was recorded with a barrel-shaped specimen exhibits a certain amount of parallelism with the shape of the pearl. A characteristic feature which frequently appears is a bar of diffuse radiation along a diameter of the halo having the same colour as the iridescence, while the curved arcs on either side exhibit the complementary colour. A distinct concentration of intensity is also generally seen in such cases at the two ends of the bar. Figures 11, 15 and 16 exhibit these features and it seems highly probable that they are a consequence of the arrangement of the crystallites of aragonite in linear rows perpendicular to the direction of the bar observed.

A certain measure of optical perfection in the stratifications of the pearl and of its surface is necessary in order that the halo be regular and exhibit vivid chromatic effects. Irregularities show up quite prominently in the character of the diffusion halo. A typical example is figure 19 which shows a patchy appearance. Cases are also not lacking in which the halo exhibits comparatively little colour. In such cases the reflection also exhibits little iridescence, suggesting that these features are interrelated.

The diffusion of light which manifests itself in the chromatic halo clearly plays an important role in the general appearance of a pearl. It extends over a substantial area of the pearl and hence when the illuminating source is also of extended area, the reflected and diffused streams of radiation overlap. Thus, an observer would no longer perceive sharply defined images of the external luminous objects but would instead regard the pearl itself as a lustrous object. The complementarity of the colours of the reflected and diffused radiation would compensate each other and result in a silvery white lustre.

6. The whispering gallery effect

Figures 5, 6, 7 and 8 in plate II recorded with a natural pearl exhibit a noteworthy phenomenon characteristic of pearls. They illustrate the fact that light incident on the surface of the pearl in any direction diffuses around within the material in the plane of the stratifications with very little loss.

Figure 5 illustrates a case in which a narrow pencil of light was incident normally on the pearl and the latter was viewed from the remote side in the opposite direction. It will be seen that the pearl nevertheless appears luminous, the periphery being much brighter than the centre. Hence the effect observed does not arise from a penetration of the light *through* the body of the pearl.

In figure 6, the light was incident almost normally on the surface of the pearl in a direction transverse to that in which it was observed. The actual area of illumination was small and was located just behind the bright edge seen in the figure. It is evident from the photograph that the light has travelled along the periphery of the pearl in all directions transverse to the direction of incidence of the light. The slow fall in intensity of the apparent luminosity of the pearl as we move away from the illuminated spot is a noteworthy feature.

In figure 7, the illuminating pencil grazed the surface of the pearl horizontally at a point a little to the rear of the top of the picture. A remarkable feature of this photograph is that it exhibits no very marked asymmetry of illumination to the right and to the left. The fall of intensity of the luminosity from the top to the bottom of the picture is quite slow.

Figure 8 was photographed in the same circumstances as figure 5 except that the illuminating pencil grazed the surface of the pearl instead of falling on it normally. Here again, it is significant that there is little difference in intensity between the right and left halves of the picture.

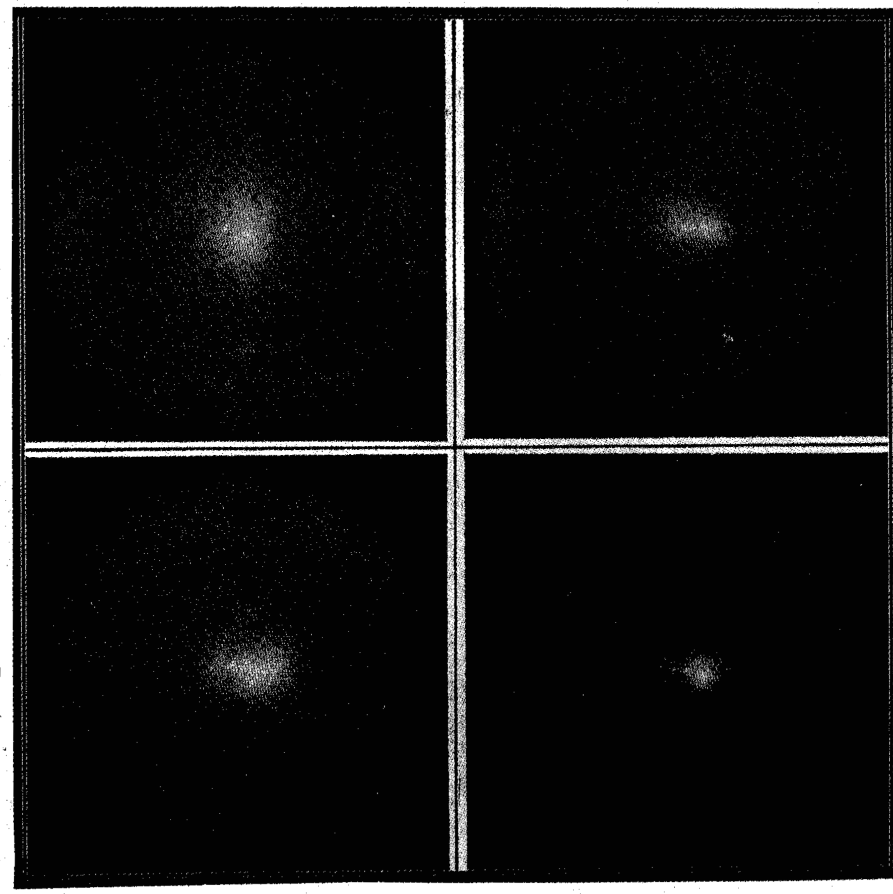
The spherical shape of the pearl plays a not unimportant role in these phenomena, but it is not the essence of the matter. Long ago, it was shown by one of us[3] that when light falls on a plane nacreous shell, it diffuses far more freely in directions parallel to the stratifications than normal to them. We need not here enter into an explanation of this fact, but will content ourselves with pointing out that essentially the same property of the nacreous substance operates in the case of pearls, enabling light falling in any direction on any part of the surface to find its way around with relatively small attenuation, thereby making the pearl visible even in regions where no light is directly incident.

7. Summary

The paper discusses the origin and character of the coloured diffusion halo exhibited by pearls surrounding the reflected image of a light source seen at their surface. Twelve photographs illustrative of various types of halo are reproduced. Other aspects of the optical behaviour of pearls are also described and discussed with illustrative photographs.

References

1. Raman C V and Krishnamurti D, *Proc. Indian Acad. Sci.*, 1954, **39A**, 215.
2. Raman C V, *Ibid.*, 1935, **1A**, 574.
3. *Ibid.*, 1935, **1A**, 859.
4. Raman C V and Krishnamurti D, *Ibid.*, 1954, **39A**, 1.

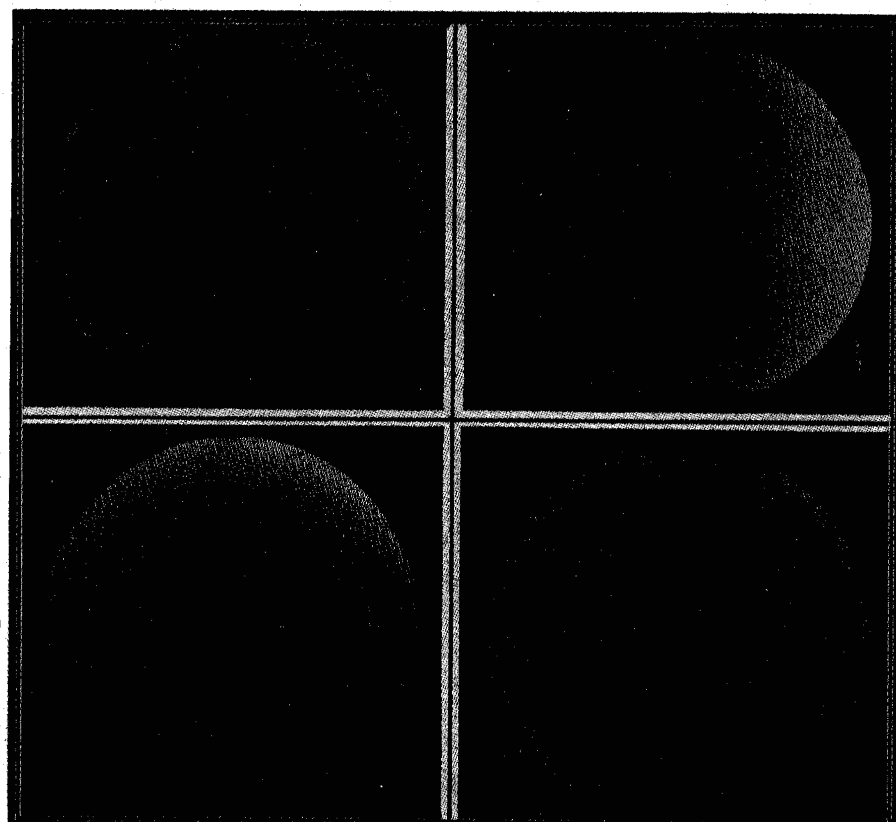


(1) (2) (3) (4)

Figures 1–4. The reflection-diffraction spectra of a natural pearl.

Plate I

(5) (6)

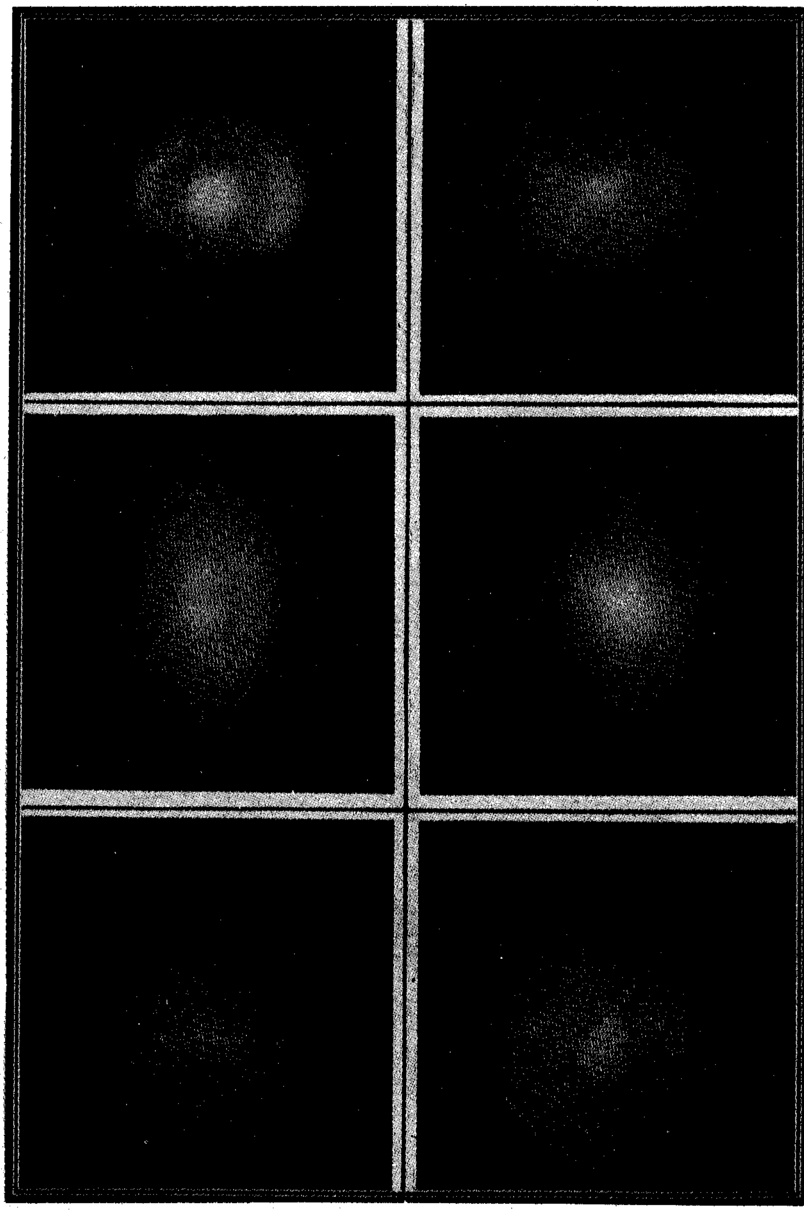


(7) (8)

Figures 5–8. The lateral diffusion of light in a natural pearl.

Plate II

(9) (10)
(11) (12)
(13) (14)



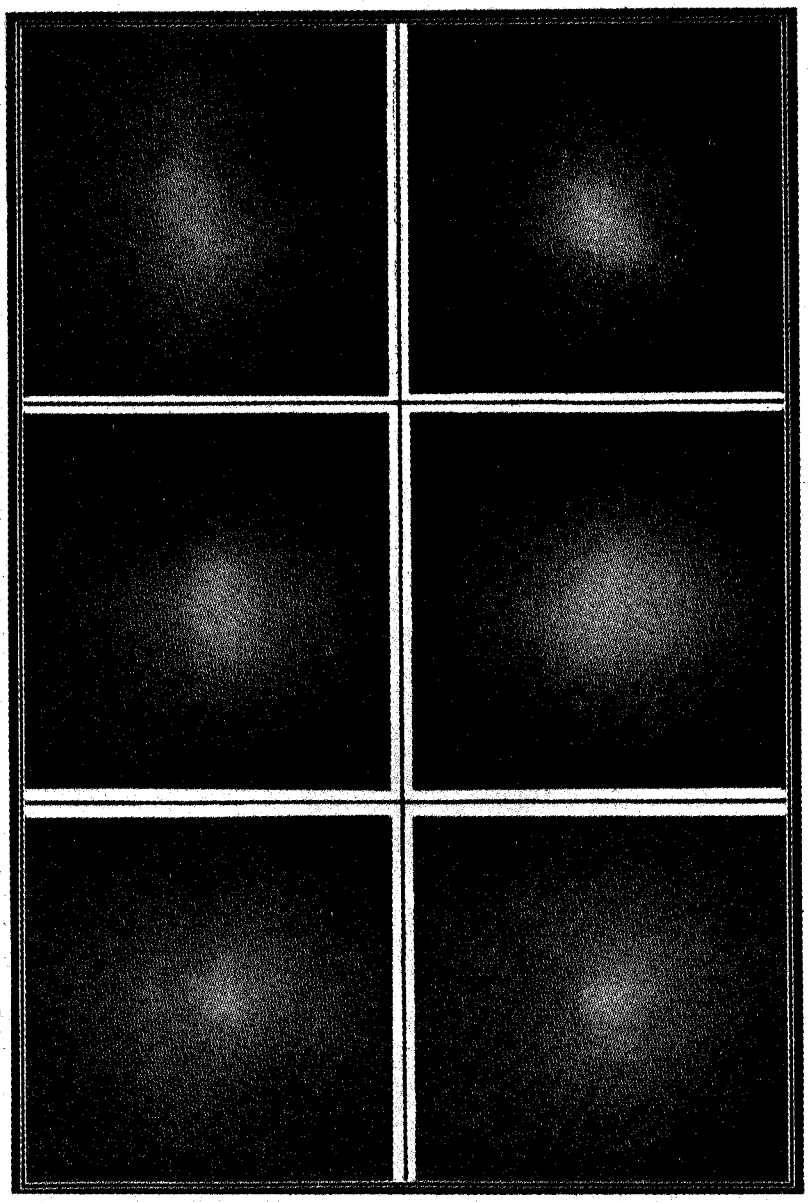
Figures 9–14. The chromatic diffusion halo in cultured pearls.

Plate III

(15) (16)

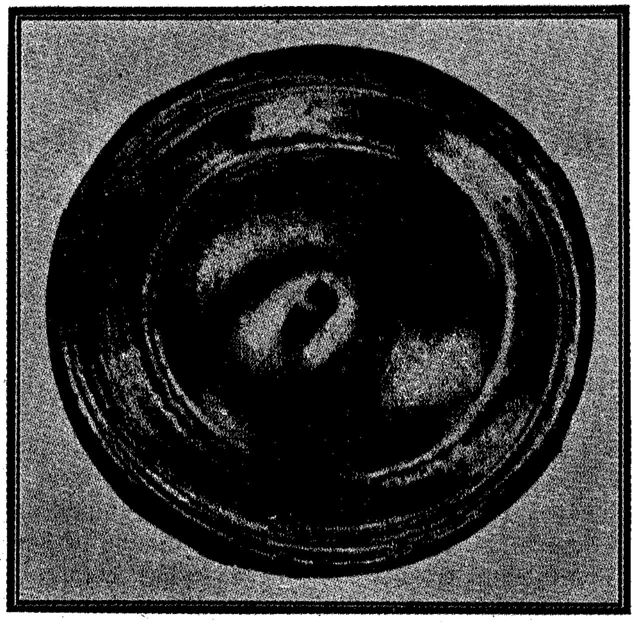
(17) (18)

(19) (20)

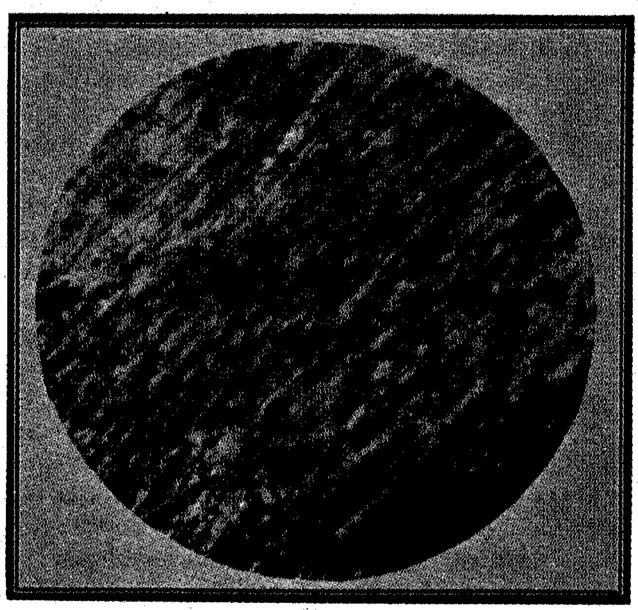


Figures 15–20. The chromatic diffusion halo in cultured pearls.

Plate IV



(21)



(22)

Figures 21 and 22. Illustrating the structure of the material in a cultured pearl.

Plate V