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THE ORIGIN OF THE COLOURS IN PRECIOUS OPAL*

1. INTRODUCTION

THE opal has been, since ancient times, one of the most admired of all gem-stones. It is certainly unique amongst them all in many respects. The description by the Roman writer Pliny of a famous opal is so graphic that it may be quoted here. "In it," he says, "you shall see the burning fire of the carbuncle, the glorious purple of the amethyst, the green sea of the emerald, all glittering together in an incredible mixture of light. Some opals by their refulgent splendour rival the colours of the painters; others the flame of burning sulphur or of fire quickened by oil". From this by no means over-enthusiastic word-picture, it is clear that no pictures in black and white could do justice to the beauty of the opal. Nevertheless, we reproduce below enlarged photographs of the two finest specimens in the museum of my Institute. One is a flat tablet purchased from a jeweller in Bangalore and shows brilliant green and blue reflections with a few yellow patches. The characteristic lamellar structure of the gem-stone can be seen in places, though the magnification is inadequate to show it up properly. The water-opal pictured to the right presents an altogether different appearance. It is of rounded form, and though beautiful enough in broad daylight, is a most attractive object when viewed under a bright source of artificial light. Numerous little spangles of reflected light of varied colours sparkle here and there within its volume, shifting their positions as the specimen is turned round.

Explanations of the remarkable optical effects exhibited by precious opal have not been wanting. But we shall not recount them

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here, since they are conjectural and have no factual support. The problem has, however, at last been definitely solved. In recent papers by myself and Mr. Jayaraman published in the *Proceedings* of the Academy, experimental facts revealed by optical and X-ray investigations are set out which enable us to state in precise terms the nature of the structure which is responsible for the iridescence of precious opal.

Gem-Opal FIG. 1 Water-Opal

2. X-RAY STUDY OF OPAL

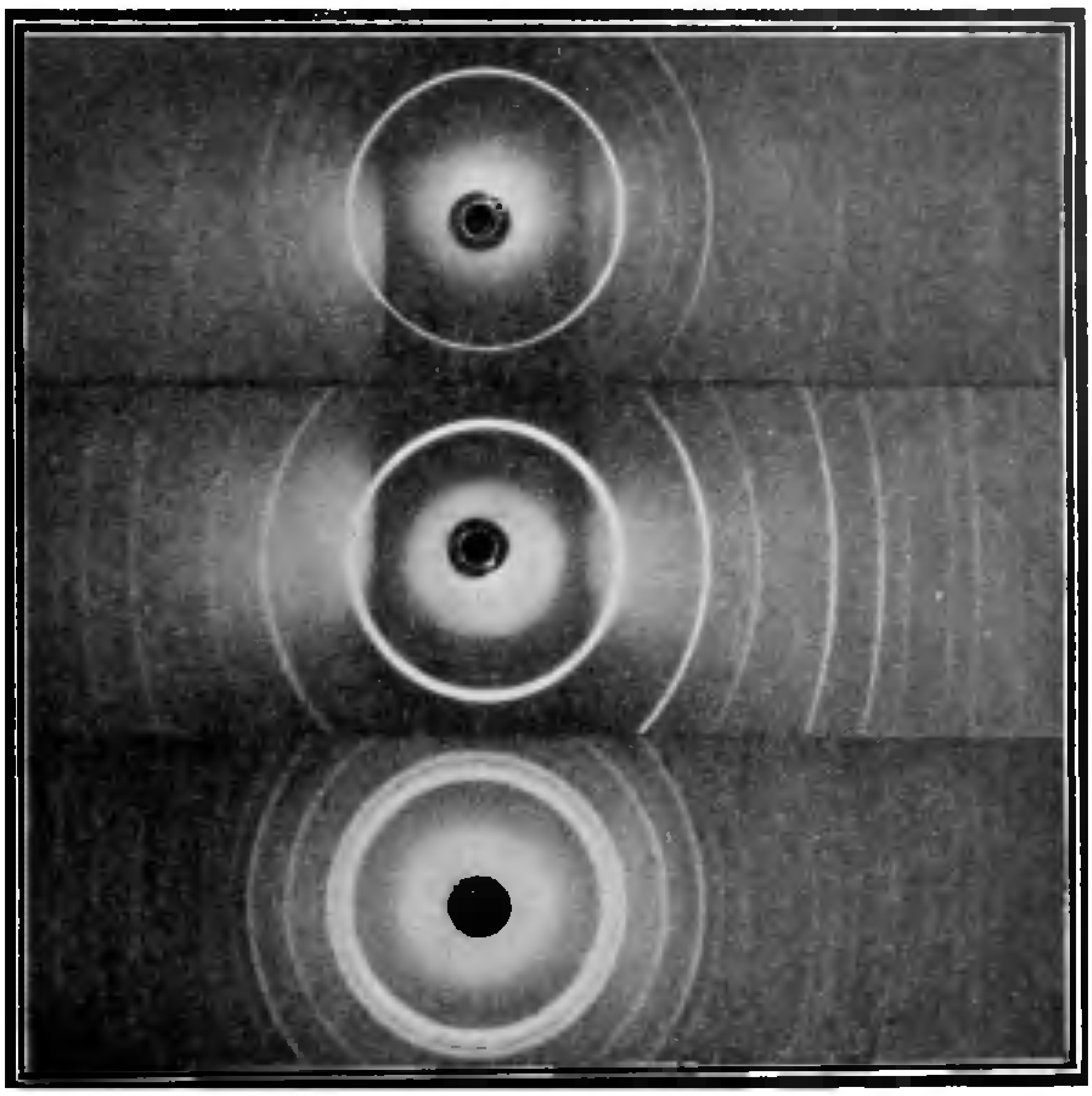
Any substance consisting predominantly of silica which has a density too low for its being identified with chalcedony and is "amorphous" in appearance is designated by mineralogists as opal. Materials which are different in their ultimate structure and even in their external appearance and physical properties are thereby lumped together under a common name, and as a consequence, there has been much confusion in the literature of the subject. A proper basis for the classification of siliceous materials is evidently to be found in their X-ray diffraction patterns. The study of the latter is accordingly the first step towards an understanding of the true nature of opal and the discovery of the origin of the colours which the precious varieties exhibit.

Well-developed crystals of the forms of silica known as tridymite and cristobalite are something of a rarity. It might, however, be anticipated that massive or cryptocrystalline forms of tridymite and cristobalite might be found to occur in nature rather more frequently. Indeed, specimens collected by me some years ago on the open terrain in Central India not far from Indore exhibit in association with agate a white porcelain-like material, examination of the X-ray diffraction pattern of which reveals it to be pure cristobalite in its low-temperature form, see Fig. 2 (*a*). On heating it to about 300° C., the material changes over to the high-temperature form of cristobalite with fewer lines in its X-ray pattern, see Fig. 2 (*b*). On cooling, it reverts again to the low-temperature form.

3. THE STRUCTURE OF HYALITE

Totally different from the naturally occurring cristobalite in its appearance and physical properties is the material known as hyalite or Muller's glass. It is a transparent solid, but examination of a specimen available in the museum of my Institute has revealed that the passage of a pencil of light through it results in the production of a well-defined diffraction halo with a hexagonal outline, thereby showing that the material is not optically homogeneous but has a stratified structure. Hyalite, in fact, appears to be the true prototype of iridescent or precious opal. X-ray examination reveals that the structure of hyalite is totally different from that of the naturally occurring cristobalite and exhibits certain characteristic features. The latter are to be found also in other materials resembling it in transparency, density and refractive index. The three X-ray diffraction patterns reproduced below as Fig. 3 (*a*), (*b*) and (*c*) are respectively those of a semi-transparent waxy opal, of hyalite itself, and of a "fire-opal" which is a gem-stone exhibiting a very pretty orange red colour. The water-opals give X-ray patterns almost identical with those reproduced in Fig. 3.

A clue to the understanding of these patterns is to be found on a careful comparison of the same with the diffraction patterns reproduced in Fig. 2 (b) and (c) above which are respectively those of the high-temperature form of cristobalite and of tridymite at room temperature. While in their outer parts, the patterns are closely parallel to that of high-cristobalite, in other features they resemble that of tridymite, especially in the fact that the principal ring of the opals is a multiplet. The demonstration that the structure of the opals includes both tridymite and high-cristobalite is completed when the patterns are recorded with X-ray cameras of higher resolution. The principal ring is then seen resolved into three components; the positions and relative intensities of these agree with those to be expected as a result of the superposition of the single ring of high-cristobalite with the three rings of tridymite.



(a) Low—Cristobalite

(b) High—Cristobalite

(c) Low—Tridymite

FIG. 2. X-Ray Diffraction Patterns

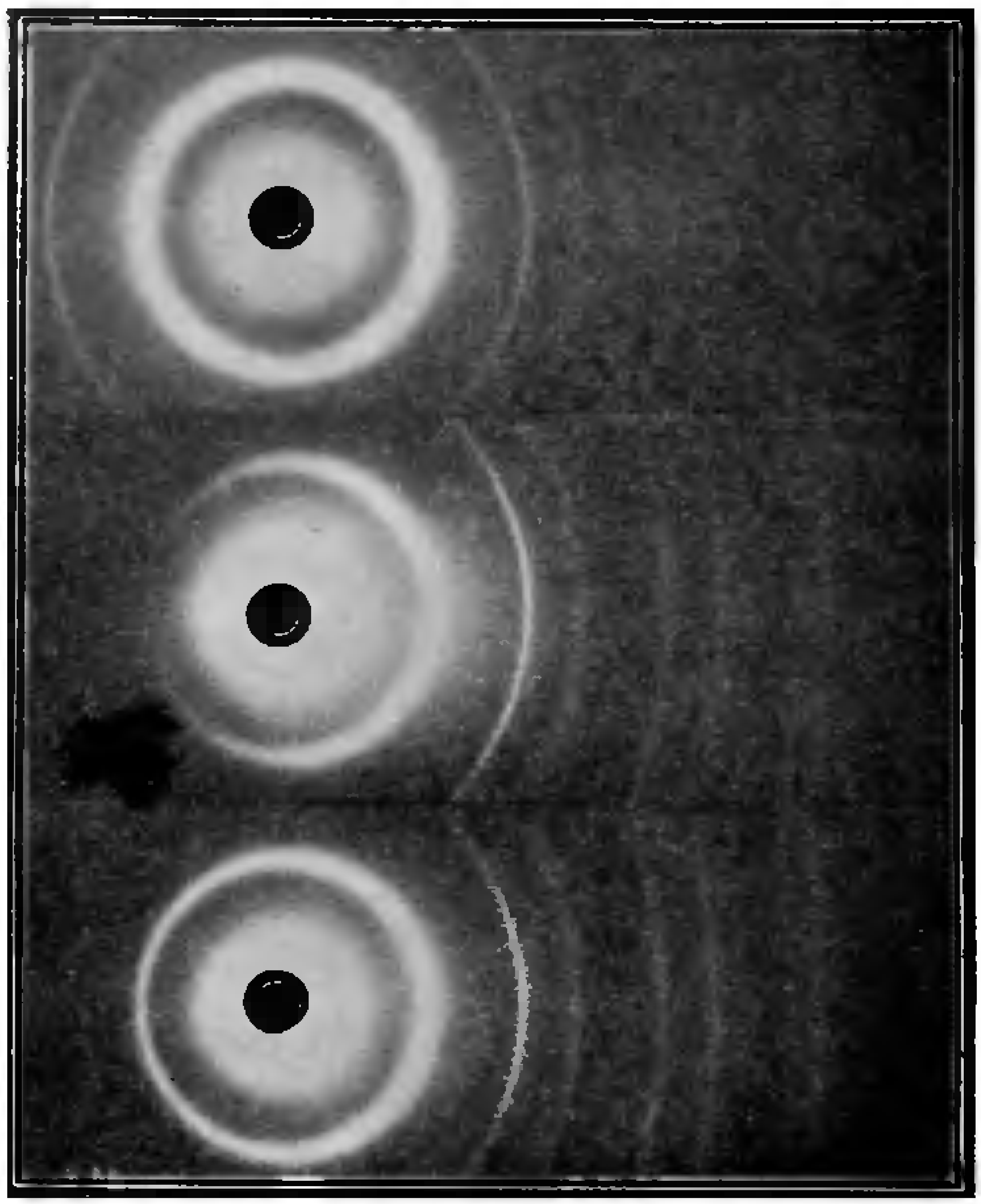
4. The Optics of Stratified Media

The optical phenomena which result from the passage of a beam of light through a regularly stratified medium are determined by the spacing of the stratifications and the direction in which they are traversed by the light. A variety of effects may be observed in different circumstances, examples of which are actually met with in the study of the optical behaviour of opals. As the spacing of the stratifications is progressively diminished, a stage is reached when the diffraction haloes or spectra observed in the forward or transverse directions disappear, and we have, instead, monochromatic reflections backwards towards the source of light. The water-opal illustrated in Fig. 1 (b) represents this middle stage. Many of the coloured internal reflections that it exhibits are yellow and red, indicating that the spacings of the stratifications are still large. With finer stratifications, even the backward reflections disappear for the longer wavelengths of the spectrum, only the blue and violet reflections being then possible. The size of the domains covered by the stratifications also plays a role. If they are small enough, we obtain a diffusion of the light in all directions instead of a regular reflection in any particular direction.

The extraordinarily perfect monochromatism of the reflections and of the corresponding extinctions displayed by fine opals is a proof that their stratifications fulfil simultaneously four conditions, (a) that their number in any given domain is large, (b) that their spacing is uniform, (c) that the difference of refractive index between the alternate stratifications is constant, and (d) that such difference in index is very small. If any one of these four conditions is violated, the reflections would cease to be monochromatic. The only reasonable hypothesis that would cover these requirements is that the alternate layers in the material consist of two distinct crystalline modifications of silica. The X-ray data indicate that these two modifications are respectively high-cristobalite and low-tridymite and thus furnish the clue to the understanding of the observed optical behaviour of precious opal.

5. Some Further Remarks

High-cristobalite, as is indicated by its X-ray pattern, is a cubic crystal with a structure resembling that of diamond in its general plan. It reverts immediately to low-cristobalite at temperatures below 270° C. It seems at first sight, therefore, surprising that it should continue to exist as such as a constituent of opal. The answer to the puzzle is furnished by its close association with low-tridymite. Evidently, the presence of the latter stabilises the situation and prevents the reversion of the high-cristobalite to the low-temperature form. The X-ray patterns indicate that the high-cristobalite forms the larger proportion of the material, and since it is optically isotropic, one can readily understand why opal is reasonably transparent. On the other hand, since low-tridymite is a crystal possessing a pseudo-hexagonal symmetry, it is not surprising that the stratifications in opal often exhibit geometric patterns in three dimensions.



(*a*) Waxy Opal

(*b*) Water Opal

(*c*) Fire Opal

FIG. 3. X-Ray Diffraction Patterns

It is important to remark also that not all opals give beautifully well-defined X-ray patterns of the kind illustrated in Fig. 3. Indeed, it is found that many opals give diffuse patterns resembling those of vitreous silica, though there are recognizable differences. A careful comparison of the X-ray patterns and of their optical behaviour, however, leaves no room for doubt that they are essentially similar to hyalite in their structure.

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