An interferometric survey of the micas.

(With Plates VII–IX.)

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Introduction.

In a group of earlier publications[1] details have been given of the application of multiple-beam interference methods to the study of mica. The characteristic structure of this mineral is exhibited by the Fizeau fringes which are produced when one side of a mica slip is silvered and placed against a similarly coated optical flat. The fringes show that the surface is violently contorted and crossed by cleavage steps, the heights of which appear to be integral multiples of the crystal-lattice spacing perpendicular to the basal plane of cleavage. If, however, a mica slip (thickness *circa* 1/100 mm.) is silvered on both sides then fringes of a totally different appearance are observed. It is frequently found that, despite the distortions on the surface, the Fizeau fringes are large *areas* of uniform illumination bounded by cleavage 'lines'. It has already been shown by multiple-beam Fizeau fringes and by the multiple-beam fringes of equal chromatic order that: (1) the contortions on one side are faithfully followed by those on the other to within molecular dimensions; (2) the mica can cleave true to a single molecular lattice over a very extensive area (many square centimetres); (3) inclusions (solid, liquid, or gaseous) which are normally not visible are shown up in detail; (4) variations in chemical composition are revealed; (5) the birefringence is shown over an extensive wave-length range.

In the earlier experiments a few samples of muscovite of unknown origin were studied, and because of its widespread use and ubiquity in rock distribution it seemed desirable to undertake a systematic survey of micas from different regions. For this purpose we have secured from the Mica Control Board twenty-five samples of known origin. These

[1] S. Tolansky, Proc. Roy. Soc. London, Ser. A, 1945, vol. 184, p. 51; ibid., 1946, vol. 186, p. 261; Phil. Mag., 1946, ser. 7, vol. 37, p. 390. [M.A. 10–134.]

micas have been examined by the multiple-beam methods formerly described. They have been graded according to their optical perfection, and at the same time measurements have been made on dielectric power loss, but a report on this technological aspect is not given here. Local structure features have been investigated in some instances.

Description of Materials Examined.

The varieties of mica available comprised 18 muscovites, 4 phlogopites, 2 biotites, and 1 lepidolite. There was a sufficient quantity of each to permit of the examination of many different samples belonging to any group. The micas as received were graded commercially in accordance with the following scheme. In this grading the micas are selected for their optical clarity and freedom from flaws and inclusions when held against a diffuse white light source. Most of the muscovites would be graded as of high quality commercially.

Table I. Mica varieties.

No.		Designation and origin.	General description (white light).
1	Muscovite.	Clear green, Madras	Slightly mottled, feather-like inclusions
2	,,	Clear green, East Africa	Very clear, stained green
3	,,	Dark ruby, East Africa	Very clear, ruby
4	,,	Ruby A.Q., Madras	Clear, surface scratches
5	,,	Clear green, Madras	Pale green, very clear
6	,,	Ruby B.Q., Madras	Clear, mottled, inclusions
7	,,	Ruby C. and S.S., Rhodesia	Very clear, ruby, surface scratches
8	,,	C. and S.S., Brazil	Clear, colourless, very uniform
9	,,	C. and S.S., Tanganyika	Clear
10	,,	C. and S.S., Calcutta	Clear, few inclusions
11	,,	F.S., Rhodesia	Clear, light ruby, surface scratches
12	,,	F.S., Brazil	Clear, few inclusions
13	,,	F.S., Tanganyika	Clear, few stains
14	,,	F.S., Bihar	Clear
15	,,	G.S., Calcutta	Very clear
16	,,	S., Brazil	Clear, slightly mottled, included crystals
17	,,	S., Calcutta	Clear, mottled inclusions
18	,,	Brazil	Clear, mottled
19	Phlogopite	Amber, Madagascar	Transparent, green-brown, stained, slightly mottled
20	,,	Amber, Madagascar	Almost opaque, black, masses of inclusions
21	,,	Dark amber, Travancore	Dark brown, fairly clear, mottled, bubble chains

TABLE I (*continued*)

No.		Designation and origin.	General description (white light).
22	Phlogopite	Ontario	Opaque, metallic, buckled lump
23	Biotite	—	Black, stained, almost opaque, many bubbles
24	,,	—	Black, mottled, 'dusty' appearance, many small bubbles
25	Lepidolite	—	Quite clear, brittle and stiff

Abbreviations of gradings kindly supplied by F. F. Chrestian & Co.:

C. & S.S. Clear and slightly stained. F.S. Fair stained. G.S. Good stained. S. Stained. H.S. Heavily stained. D.S. Densely stained. A.Q. First quality. B.Q. Second quality.

The muscovites were all initially selected as of good commercial optical quality and although a number were stained or coloured, all showed large areas which were apparently flawless visually. As shown below, the sensitive multiple-beam methods reveal the existence of inclusions and structures not visible by direct means.

EXPERIMENTAL.

Thin sheets of mica (*circa* 1/100 mm.) were cleaved from the samples supplied and silvered on both sides by evaporation *in vacuo* in accordance with a technique formerly described, with reflecting coefficients sometimes approaching 0·94. As shown elsewhere,[1] with such silverings a multiple-beam Fizeau fringe set-up leads to the formation of uniformly tinted areas when a mercury-arc source is used. A simple calculation shows that a change in thickness of *but a single crystal-lattice layer* (20 Å.U.) can lead to a 50 % change in the intensity, hence adjacent regions which are separated by even only one lattice layer show easily distinguishable areas on the interferogram. The micas were examined with Fizeau fringes employing magnifications varying from $\times 3$ to $\times 200$ and using various isolated and mixed monochromatic radiations from a mercury-arc source. A number of features of special interest were examined with the white-light fringes of equal chromatic order and these reveal local details and birefringence effects.

So complex are the features revealed and so rich is the detail often exhibited even on a single interferogram, that any attempt at a full description of what has been observed would occupy a prohibitive amount of print. We therefore reproduce typical Fizeau fringe interferograms in pl. VII, figs. 1–9, since they are much more revealing than

[1] S. Tolansky, Proc. Physical Soc., 1946, vol. 58, p. 654. [M.A. **10**–134.]

simple descriptions. Explanations of the plates are given at the end of this paper.

It was first proved that any interferogram is indeed typical of a given kind of mica. Each variety, as received, contained a number of fairly thick sheets of material. For a given variety a random selection of a dozen pieces was taken from various sheets. The interferograms were examined, and it was observed without any doubt that the random samples from any one variety closely resembled one another and can all differ distinctly from some other variety. It can be confidently asserted that the reproductions in figs. 1–9 are typical for the varieties indexed.

In three of the photographs secondary fringes of low visibility can be seen within the uniform tint areas. In these cases the mica slips have been mounted between glass plates both to protect the silver from atmospheric attack and to assist manipulation (a freely suspended thin sheet is sensitive to air shocks). These secondaries are formed in the air films between the silvered mica and the unsilvered glass surfaces. They show that although an area is of uniform tint yet the corresponding mica surface is highly distorted, from which it follows that identical distortions occur on both faces of the mica. The secondaries are eliminated when the mica is freely suspended, and for particular investigations this was frequently done.

Discussion of Fizeau Fringes.

The degree of optical uniformity of the various mica samples is shown by the fringes in figs. 1–9 inclusive. As indeed may be expected, the white-light insensitive grading only correlates crudely with this. It will be shown elsewhere that a precision grading can be made on the basis of the interferograms and it has in fact been found possible to derive some correlation between the character of the inclusions and the dielectric power loss. We shall here survey only the general characteristics shown by the interferograms, discussing in particular some specific points of interest, whilst recognizing that we are leaving completely untouched a mass of material already readily available for study on these photographs. In particular we have made estimates of: (1) the average area of uniformity; (2) the largest area of uniformity found for the given variety; (3) inclusions visible per square centimetre. The data are given in table II for all the samples studied. The number of inclusions observed depends, of course, on the volume, but since all the samples selected were roughly 1/100 mm. thick, the visual number count (being in any case only an approximation) can be taken as a measure of the number of inclusions per cubic centimetre of mica. The figure given is then approximately the number of inclusions per cubic millimetre of mica.

TABLE II (For sheets *circa* 1/100 mm. thick.)

No.	Mica.	Average area of uniformity sq. cm.	Largest uniform area sq. cm.	Inclusions per sq. cm.	Remarks.
1	Muscovite	0·06	1	17	Large network of fine bubbles
2	,,	0·9	14	—	—
3	,,	0·7	6	—	—
4	,,	0·6	5	—	—
5	,,	0·7	6	—	—
6	,,	0·1	1·5	20	Large fluctuations in area
7	,,	0·3	1	—	—
8	,,	0·9	13	—	—
9	,,	0·5	10	1	A few large inclusions
10	,,	0·7	6	2	Non-uniform patchy areas of bubbles
11	,,	1·0	20	—	—
12	,,	0·5	5	1	A few large bubbles
13	,,	1·3	26	—	—
14	,,	1·4	11	—	A few large bubbles
15	,,	0·7	5	11	—
16	,,	0·5	4	4	—
17	,,	0·7	8	7	—
18	,,	0·4	3	3	—
19	Phlogopite	0·06	0·5	15	Dense local patches of inclusions
20	,,	0·01	0·5	200	—
21	,,	0·06	0·5	52	Many pin-point inclusions
22	,,	—	—	200	Many bubbles, boundaries are obscured
23	Biotite	0·11	0·25	55	Ragged areas dotted with bubbles
24	,,	0·1	0·04	100	—
25	Lepidolite	0·45	1	—	—

Muscovites.—Almost without exception the muscovites exhibit considerable regions of uniform tint, areas which are free from inclusions and over which cleavage is probably true to a single molecular lattice.

The white-light visual commercial grading is only a rough index. Thus, to select but one instance, whilst no. 3 is clearly of high quality, no. 8 is equally good and both are much better than no. 1.

On the basis of these observations the micas can be regraded into the following order in which the samples are tabulated in accordance with the value of the *average* of the area of uniformity, the first in the list having the largest *average*, namely, 1·4 square centimetres: nos. 14, 13, 11, 2, 8, 3, 5, 10, 17, 15, 4, 12, 9, 16, 18, 7, 6, 1. As seen, since the commercial grading is in sequence 1, 2, 3, 4 . . . 18, correlation is virtually non-existent within the muscovite group, nor does grading according to the number of inclusions offer a better correlation. Clearly the commercial grading is inadequate.

Of striking crystallographic interest is the fact that certain of the muscovites exhibit relatively enormous areas which have cleaved true to a single molecule *on both sides of the sheet*. The largest single area we have observed is 26 square centimetres and over such extensive regions it must be considered that one has true perfect cleavages on either side of a single crystal. Since such mica sheets are clearly ideal for functioning as α-particle absorbing screens, windows, &c., we list below in order of merit the four varieties which we have found the best from this point of view.

TABLE III. Muscovites with large uniform areas.

No.	Origin.	Average uniform area (sq. cm.).	Largest uniform area (sq. cm.).
13	F.S., Tanganyika	1·3	26
11	F.S., Rhodesia	1·0	20
2	Clear green, East Africa	0·9	14
8	C. & S.S., Brazil	0·9	13

Figs. 2 and 5 are samples of specimens whose Fizeau fringes show good uniform tint areas. Fig. 2 is shown here with strongly developed secondaries to illustrate the nature of these, whilst in fig. 5 the mica is freely suspended.

Phlogopites.—The phlogopites examined, although from widely different geographical regions, show interferograms resembling one another and all totally distinct from the typical muscovites. Extensive areas of uniform tint are absent, the specimens often consisting of a confused mass of small elements. The number of inclusions is considerable and several samples exhibit more than 200 inclusions per cubic millimetre. It is clear that the natural phlogopites are less perfect crystals than are the muscovites.

It may be noted that one particular phlogopite (a light variety of Travancore amber) showed a few moderately large uniform tint areas (one of 4 sq. cm.) and this is possibly a phlogopite showing some muscovite interpenetration.

Biotites.—The two biotites examined do not differ much in appearance from the phlogopites.

Lepidolite.—One good-quality lepidolite sample was available. The interferogram somewhat resembled a muscovite and showed that the crystal was chemically extremely homogeneous although the areas of uniform thickness were comparatively small.

Some Specific Features.

From amongst the considerable detail presented we have selected some features of interest for discussion.

(1) *Bubble Chains.*—Chains of bubbles have been found on at least four muscovites, two phlogopites, and one biotite, appearing therefore to be ubiquitous in micas; pl. VII, fig. 7 shows a typical example. A portion of the same chain with higher magnification is shown in pl. VIII fig. 12. It can be seen that the chain consists of a series of discrete ellipsoidal bubbles. The curvature is so high that the fringe localization in the mica film no longer exists. Hence the fringes within the bubbles are completely out of focus and thus abnormally broad.

(2) *Filament Channels.*—The origin of these bubble chains is possibly associated with the detail shown in fig. 11 ($\times 51$) which is from a muscovite. This is a filament channel, probably liquid and about 0·03 mm. wide, which is just about the width of a typical bubble chain. Clearly such a liquid channel could break up into a bubble chain through continued crystal growth into the channel.

Similar channel-like markings are common features on other samples; fig. 4 is an example. Some of these filaments are almost in the stage of breaking up, and some are so fine as to give interference colours in white light.

(3) *Crystalline Inclusions and Bubbles.*—Crystalline inclusions in mica are, of course, well known. We illustrate in fig. 10 (magnification $\times 42$, insets $\times 230$ approximately) several particularly well-marked crystal plates (A and B) which are clearly visible with the increased contrast produced by multiple-beam interference. The visibility has been further enhanced for the small details by using a high-pressure mercury-arc source. This increases the fringe width and reduces the risk of the fringe maximum missing the detail under observation. In

many mica specimens inclusions of the nature of 'dust' are observed. As in the upper part of fig. 10 these are often too small for structure detail to be resolved. We cannot give any information concerning the chemical nature of the inclusions.

Regarding the character of the many bubble-like inclusions observed, little can be said. It is probable that the nearly circular inclusions (as in pl. IX, fig. 14) are liquid. In some cases crystalline material appears to be present, whilst other bubbles are so perfectly circular that they almost certainly consist of liquid drops held under high surface tension forces.

Many curious effects have been noted. Pl. VIII, fig. 13 shows the Fizeau fringes produced when one bubble lies beneath another, and similar confused fringes can always be seen in biotites.

(4) *Birefringence.*—It has already been shown that the birefringence of mica in the direction perpendicular to the cleavage plane can be demonstrated over an extensive wave-length range with white-light fringes of equal chromatic order. We show in pl. IX, figs. 16, 17, 18 a series of such fringes for different specimens which are optically biaxial. The fringe doubling $d\lambda$ is related to the birefringence $d\mu$ by the simple expression $d\lambda = (\lambda/\mu)\cdot d\mu$. This is independent of the thickness of the mica sheet used, and these fringes could be used to convert easily measured $d\lambda$ values into absolute values of $d\mu$. However, our use for the fringes has been to demonstrate differences in cleavage quality. Those micas (as in figs. 2 and 5) which give Fizeau fringes over large areas have smooth straight fringes of equal chromatic order (fig. 16). Other types show highly ragged broken fringes, as in figs. 17 and 18.

Explanation of Plates VII–IX.

Plate VII. Fizeau fringes of doubly silvered mica sheets.

Fig. 1. Muscovite, green, Madras (specimen 1). A number of small inclusions. ×55.

Fig. 2. Muscovite, ruby, Madras (specimen 6). Uniform tint areas crossed with low-visibility secondary fringes. ×24.

Fig. 3. Muscovite, Bihar (specimen 14). Large uniform tint areas. Inclusion showing fringe doubling due to birefringence. ×7.

Fig. 4. Muscovite, Calcutta (specimen 15). Showing bubble chains. ×55.

Fig. 5. Muscovite, Brazil (specimen 18). Areas of high uniformity. ×51.

Fig. 6. Phlogopite, amber, Madagascar (specimen 20). Numerous inclusions. ×31.

Fig. 7. Phlogopite, light amber, Travancore (specimen 21). Inclusions and series of bubble chains. ×55.

FIG. 8. Phlogopite, dark amber, Travancore (specimen 21). Large inclusion. Does not exhibit birefringence doubling. ×55.

FIG. 9. Biotite (specimen 24). Many irregular inclusions. ×51.

PLATE VIII. Fizeau fringes of doubly silvered mica sheets.

FIG. 10. Phlogopite, dark amber, Travancore (specimen 21). Showing large inclusion. ×42. Inset A (×230) enlarged portion showing crystal inclusion. Inset B (×230) enlarged portion showing crystal plate inclusion and 'dust'-like specks of included matter.

FIG. 11. Muscovite, Calcutta (specimen 15). Showing filament channel. ×51.

FIG. 12. Phlogopite, light amber, Travancore (specimen 21). Showing enlarged detail of bubble chains as ellipsoidal features. ×83.

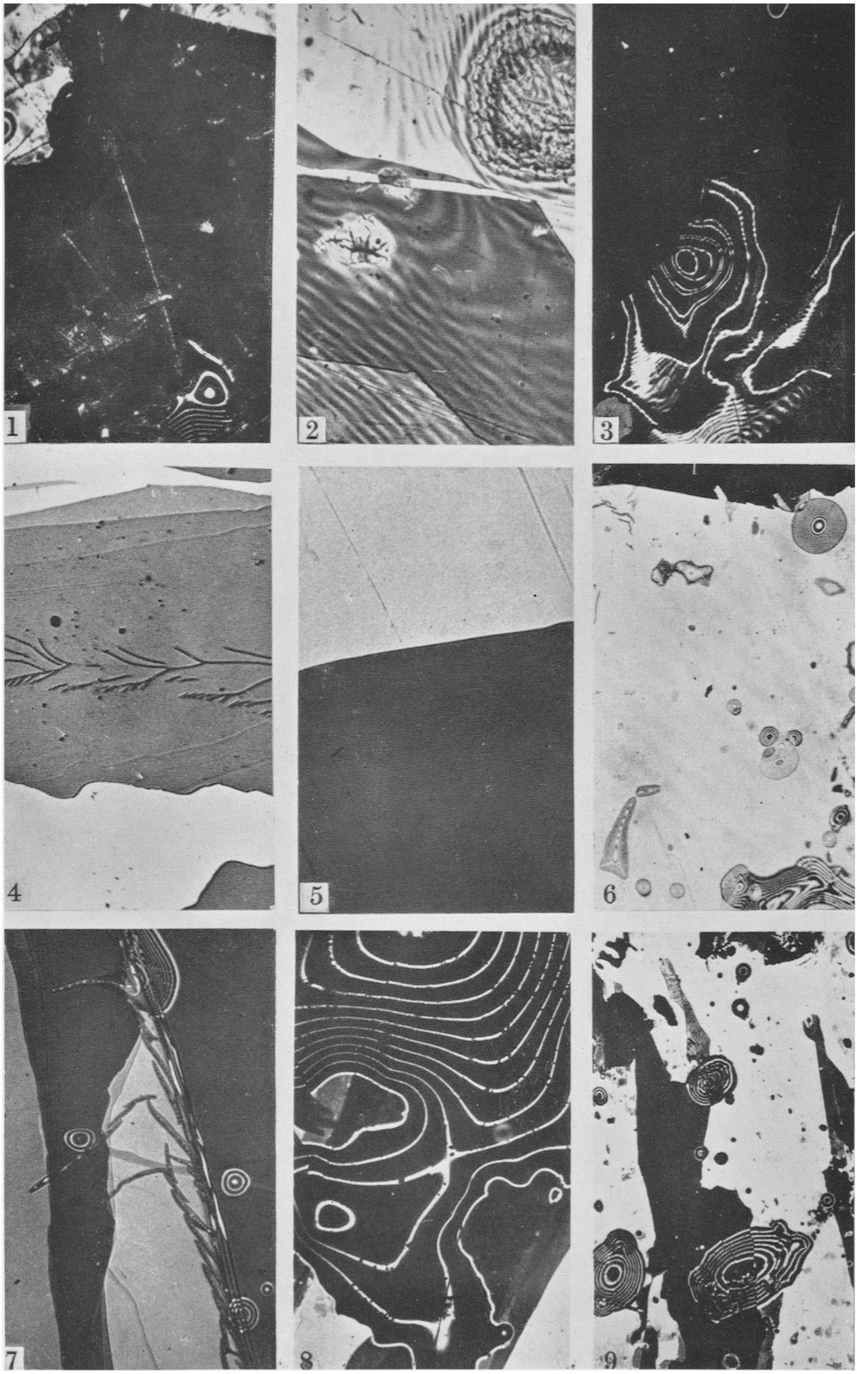
FIG. 13. Phlogopite, dark amber Travancore (specimen 2). Overlapping inclusions. ×130.

PLATE IX, FIGS. 14, 15. Fizeau fringes of doubly silvered mica sheets.

FIG. 14. Biotite (specimen 23). The circular rings suggest liquid inclusions. ×40.

FIG. 15. Muscovite, Bihar, F.S. (specimen 14). Large bubble-like inclusion, showing birefringence effect and also crystal-like nucleus. ×15.

FIGS. 16, 17, 18. Fringes of equal chromatic order for three different muscovites, showing differences in cleavage behaviour. The fringes are double because of birefringence. Fig. 16 is the type of fringe given by the majority of the muscovites.



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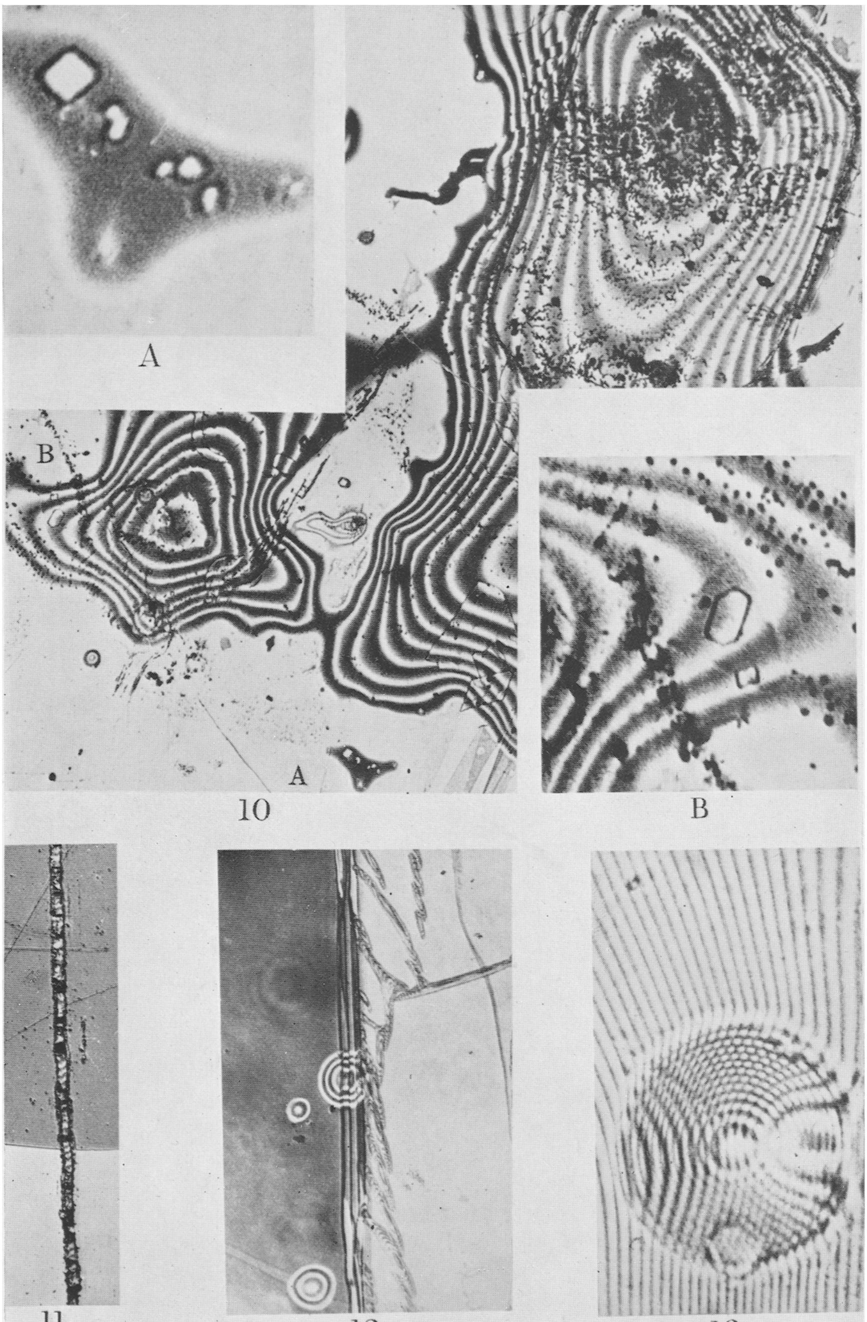

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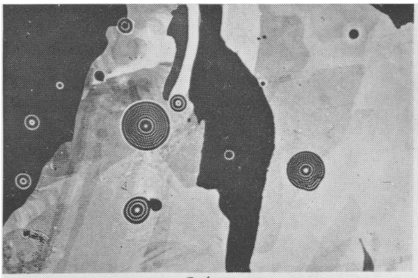
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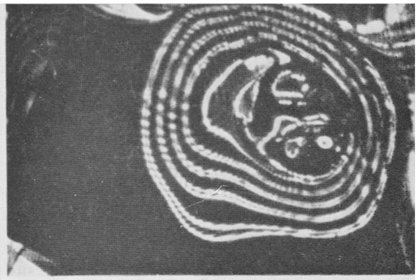
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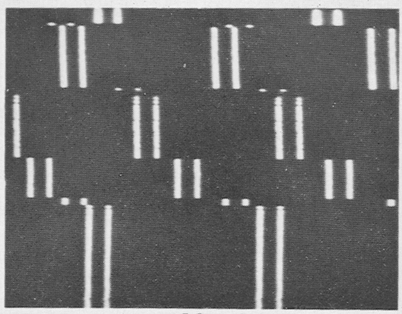
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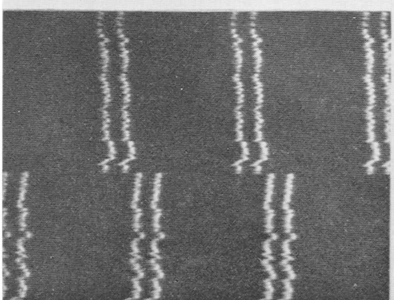
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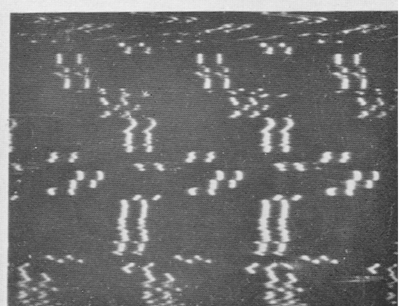
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