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Elliptically polarized light in alkali amphibole from Poços de Caldas, Brazil

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An alkali amphibole crystal on a thin section of nepheline syenite from Poços de Caldas, Brazil is examined using an elliptically polarizing microscope equipped with a mercury lamp as a light source and a pair of rotatable quarter wave plates placed in the optical path. The nature of the light transmitted through the crystal is revealed to be an elliptically polarized light with an ellipticity of 0.23. This result is concordant with the works of T. Shoda (1957; 1958; 1961), who revealed the existence of elliptically polarized light not only in optically active crystals but also in some absorbing crystals. The difference between the nature of the elliptically polarized light resulting from absorption and that due to optical activity is discussed.

Introduction

The notion that light in a crystal is linearly polarized is not always appropriate. In general case, two sorts of elliptically polarized lights transmit in a crystal. This obvious fact has been explained both theoretically and experimentally. The very old textbooks (Pockels, 1906; Szivessy, 1928) have described it as the general theory for crystal optics. T. Shoda (Shoda, 1957; 1958; 1961) demonstrated this experimentally using some special species of alkali amphiboles and babingtonite from the Yakuki mine, as reported by one of the authors (Tokonami, 1996). We are afraid that these excellent articles have been overlooked by mineralogists and crystallographers. It is worth noting that not all alkali amphiboles show these phenomena. Even if the crystals show an optical anomaly, most of them are clearly explained by color dispersions. In fact, Enami et al. (2003), having enough knowledge of the elliptical nature of light passing through amphiboles from the Taguchi mine (Shoda and Bunno, 1972), described the optical characters without taking into account the ellipticity of light passing through the amphiboles in the Sanbagawa schist.

We intended to confirm the works of T. Shoda and found suitable amphibole crystals in a nepheline syenite rock specimen. We constructed a microscope for the observation of elliptically polarized light. This was done with a monochromatic light source and the insertion of two quarter wave plates in the appropriate sites in the optical path. Using the new apparatus, we observed the ellipticity of the transmitted light in a crystal.

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Construction of an elliptically polarizing microscope

Conventional polarizing microscopes are designed assuming that the light in a crystal is divided into two sorts of linearly polarized lights, and are equipped with two nicol prisms or polaroids; one is a polarizer and another is an analyzer. In this research, two quarter wave plates were inserted into the appropriate position in the optical path.

The light source for an elliptically polarizing microscope should be a monochromatic, because it can easily transform a linearly polarized light of a definite wave length into any grade of elliptically polarized light, or vice versa, by the aids of a quarter wave plate. However, there are no *ad hoc* instruments that are able to make the elliptically polarized white light, as far as the authors know.

(1) The main body of the new microscope is the system polarizing microscope BX-P made by Olympus Optical Co. Ltd. The photograph of the newly constructed microscope is shown in Figure 1.

(2) This system originally had the light source of a halogen lamp. We replaced this with a mercury lamp

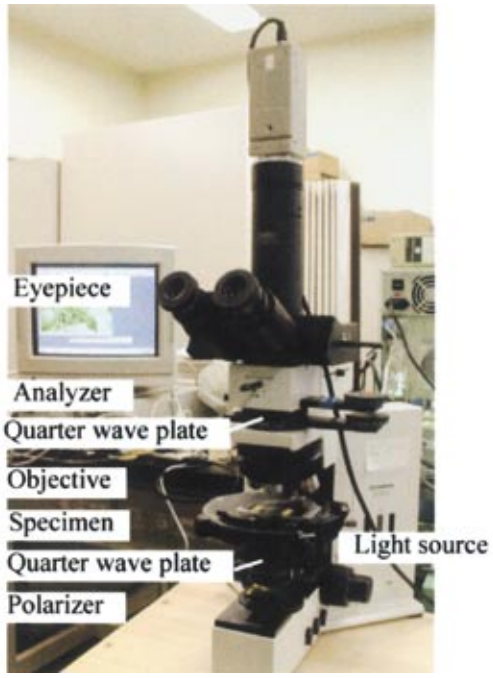

Figure 1. The elliptically polarizing microscope and its devices.

(SHL-100UVQ-2 produced by Harison Toshiba Lighting Corp. and sold by Toshiba Physics and Chemistry Co. Ltd). Then we put a narrow band interference filter (CJ43177 supplied by Edmund Optics Japan Co. Ltd.) into the light path to obtain a monochromatic light with a wave length of 546 nm.

(3) Two rotatable quarter wave plates were set; the lower one between the polarizer and the stage, and the upper one between the stage and the analyzer. The insertion of the lower plate proceeded as follows: There was, in the system microscope, a device called the U-POC for the adjustment of the diaphragm and rotation of the polarizer together with the attachment for the condenser lens under the stage. We removed this part and inserted a new device made by Mizoziri Optical Industry Co. Ltd., that consisted of a polarizer and a rotatable quarter wave plate. To set the upper plate, the optional device called U-OPA is inserted in the light path. We requested the Iwaken Co., Ltd. to remove a polaroid from a rotating device called U-AN360P and to mount a quarter wave plate on it. This device is used by fixing on the U-OPA.

(4) An analytical system was constructed: Although the images are directly observable with the naked eye through the eyepiece, they are displayed on a monitor cathode ray tube through a CCD camera (Sony CCD-IRIS DXC-108). The images are processed with a personal computer LPC-PT1GBS of Logitec Corp. The software used was an image analysis application program, Image-Pro Plus of the Cybernetics Company.

Rock specimen

The rock specimen (GSJ M37238) was lent from Geological Museum, Geological Survey of Japan, National Institute of Advanced Industrial Science and Technology.

Two of the authors (M.T. and N.A.) and the late Prof. Kenkichi Fujimori (Institute of Astronomy and Geophysics, São Paulo University) performed a field research as the 1st party of International Scientific Research Program of the Ministry of Education, Japan (Project number 62041024) and collected a piece of hypabyssal rock from Taquari, Poços de Caldas, Minas Gerais, Brazil on July 26, 1987. Afterwards, the specimen was dedicated to the Museum. The field name of the specimen was 'foyaite', but the name 'nepheline syenite' will be used in this manuscript hereafter.

The rock specimen contains two sorts of amphiboles (potassic magnesio-arfvedsonite with high potassium content and calcian arfvedsonite with no detectable potassium), K-feldspar, Na-feldspar, nepheline, aegirine, zircon, titanite and pyrite, and other accessary minerals.

The optical properties of the potassic magnesio-arfvedsonite in this rock are so unique that the visible rays in the crystal are not linearly, but elliptically polarized as described in previous reports (Tokonami et al., 2002a; b). The chemical composition is measured by an X-ray microanalyzer (EPMA) and determined to be Si 7.82, Ti 0.19, Al 0.21, Fe^{2+} 1.92, Fe^{3+} 0.56, Mn 0.49, Mg 1.74, Ca 0.32, Na 1.82, K 0.88 for 23 oxygen atoms, where the values of Fe^{2+} and Fe^{3+}-contents are calculated based on the average of the minimum and maximum ferric estimates (after Leak et. al., 1997).

Several thin sections for optical microscopy (30 μm thick with glass covers) and polished sections without covers for EPMA analyses were prepared by the Iwamoto Kosanbutu Syoukai (Iwamoto Mineral Products Company Ltd.).

Observation and measurement by EPMA

The back scattered electron images (hereafter BSEI) and the X-ray images of Na, Mg, Al, Si, K, Ca, Ti, Mn, Fe, Sr, Ba were collected from several areas on a polished section and both qualitative and quantitative analyses were performed at several positions. JXA-8900 electron probe microanalyzer made by JEOL. Ltd. and Shimadzu EPMA-8705 made by Shimadzu Corporation were used in this research.

A BSEI picture of the whole sheet of a polished section is shown in Figure 2, in which the dark parts correspond to the portion of strong electron-scattering. This figure consists of 10% of dark, 41% of intermediate and

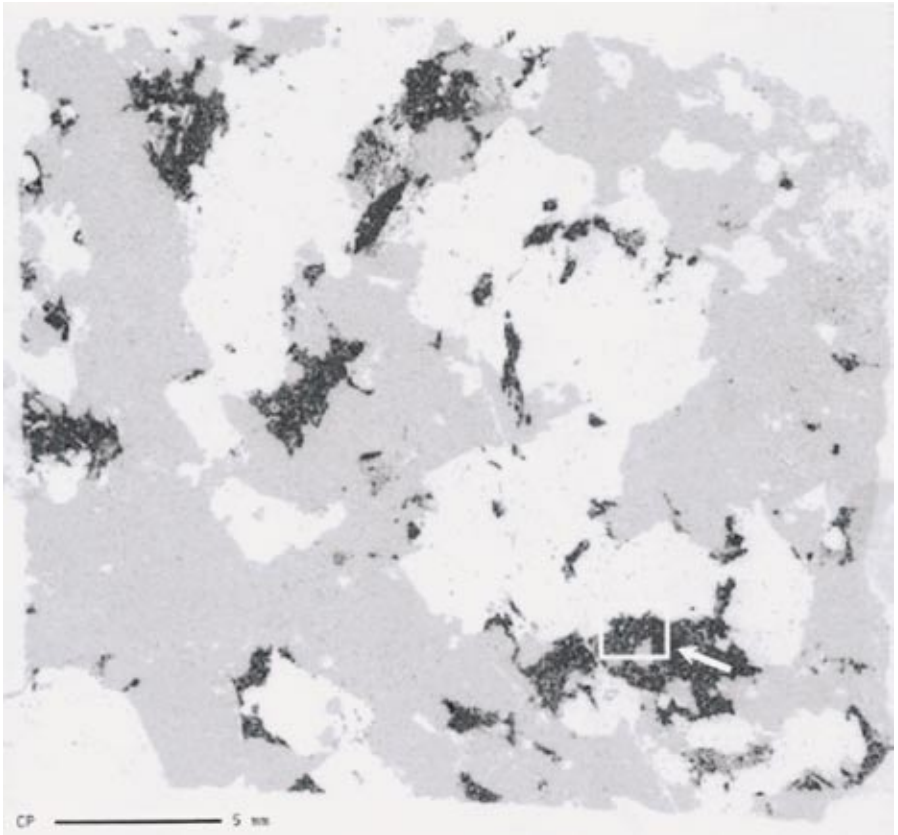

Figure 2. Back scattered electron image of the whole sheet of a polished section. Size of the view: 27 × 25 mm. The darker part corresponds to the strong electron scattering position. The area shown in Figures 3, 4 and 5 is indicated with a rectangle.

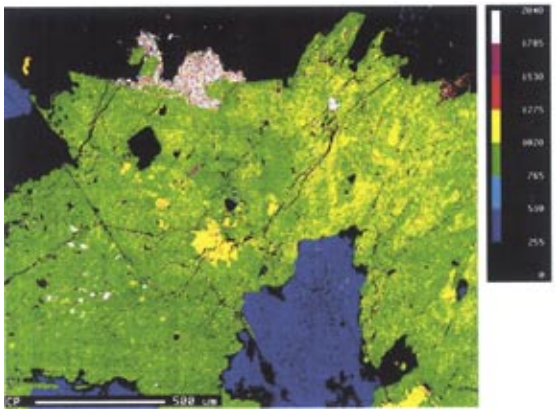

Figure 3. Intensity-colored BSEI of a local area mainly composed of amphiboles. Scale bar: 0.5 mm (size of the area: 1.8 × 1.5 mm).

45% of light areal parts.

Microscopic observation by visible light revealed the rock to be composed of 10% of amphibole, 52% of feldspar, 34% of nepheline and 4% pores. Accordingly, the dark parts in the figure are identified to be amphiboles, the intermediate parts are K-feldspars and the light parts are shared by 11% albite and 34% nepheline. Furthermore, the amiphbole is divided into two kinds of areas; those containing potassium as one of the major constituents, and the other-characterized by the lack of a detectable amount of potassium. In Figure 3, a BSEI picture clearly shows the nature of the amphiboles in the lower right box indicated in Figure 2.

Observation by a polarizing microscope

Figure 4 shows the microscopic image with the open nicol for the same area as Figure 3. Because the absorption depends on the frequency and the vibration direction of the light in crystals (pleochroism), the color of transmitted light changes by rotating the stage of microscope from yellow green to bluish green. The aggregate of the single crystals show almost the same tone in the BSEI. However, each of the individual grains show different behavior to the polarized light during the rotation of the stage.

Figure 5 is an image of the same area with crossed nicols. The images with crossed nicols change with the rotation of the stage sensitively even more than with the open nicol in which the extinction is not sharply recognized.

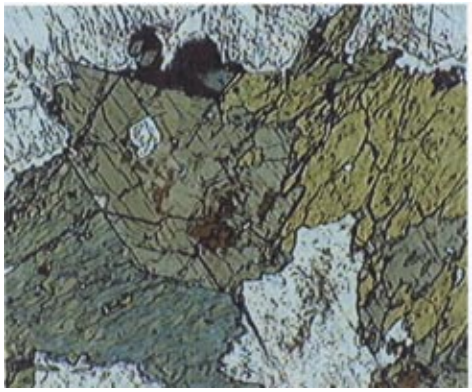

Figure 4. Open nicol micrograph of the same area as in Figure 3. Size of the area: 1.8 × 1.5 mm.

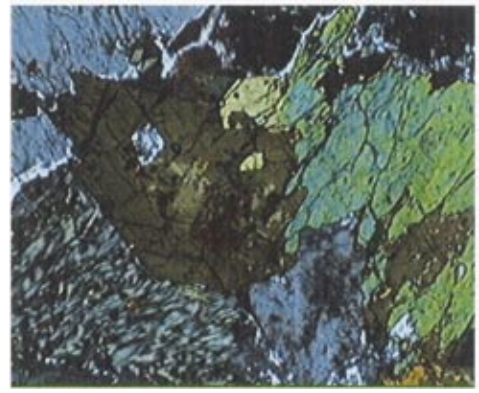

Figure 5. Crossed nicols micrograph of the same area as in Figure 4. Size of the area: 1.8 × 1.5 mm.

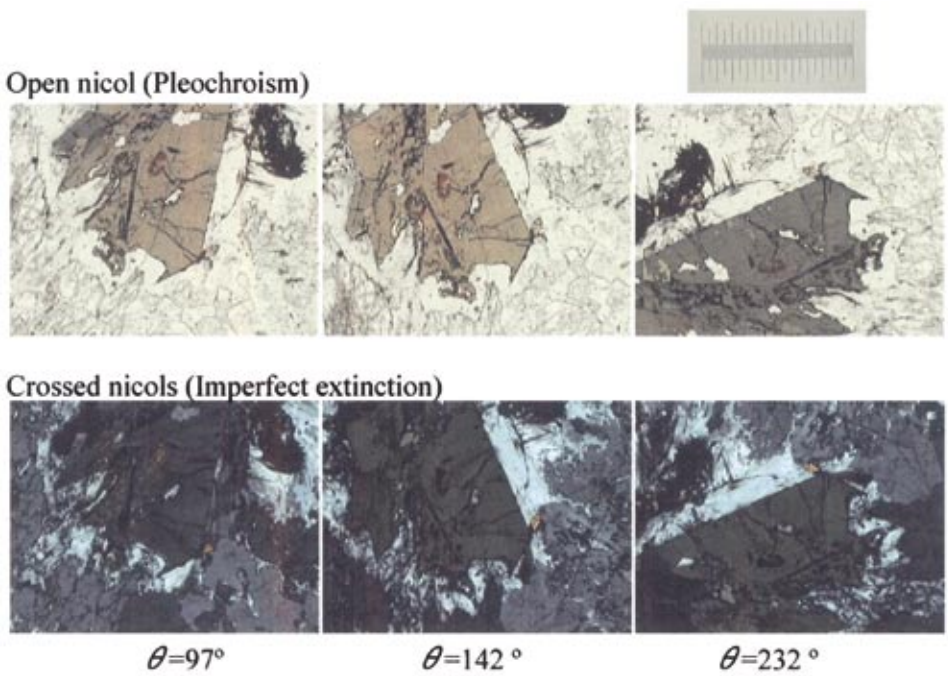

Figure 6. Images of a potassic magnesio-arfvedsonite at three different positions.

Microscopic observation of a crystal

Each of thin sections and polished sections included several amphibole crystals with abnormal extinction. Here we will describe the nature of a crystal along with the picture in Figure 6.

Observation by means of linearly polarized lights

Extinction is not clearly detectable, but is approximately recognized around the position of θ = 97° under crossed nicols. That is to say, the diagonal positions are supposed to be around θ = 142° and also 232°, where θ represents the angle of rotation of the stage graduated counterclockwise from an arbitrary origin. The images at these positions are shown in the lower row of Figure 6.

The images with the open nicol are shown in the upper row of Figure 6. They show remarkable pleochroism; dark green around θ = 232° and light brown around θ = 97° and 142°.

Observation using quarter wave plates

The direction of the lower quarter wave plate is shown with α. The origin of α is chosen to the point that the direction of vibration of fast rays in the plate coincides with

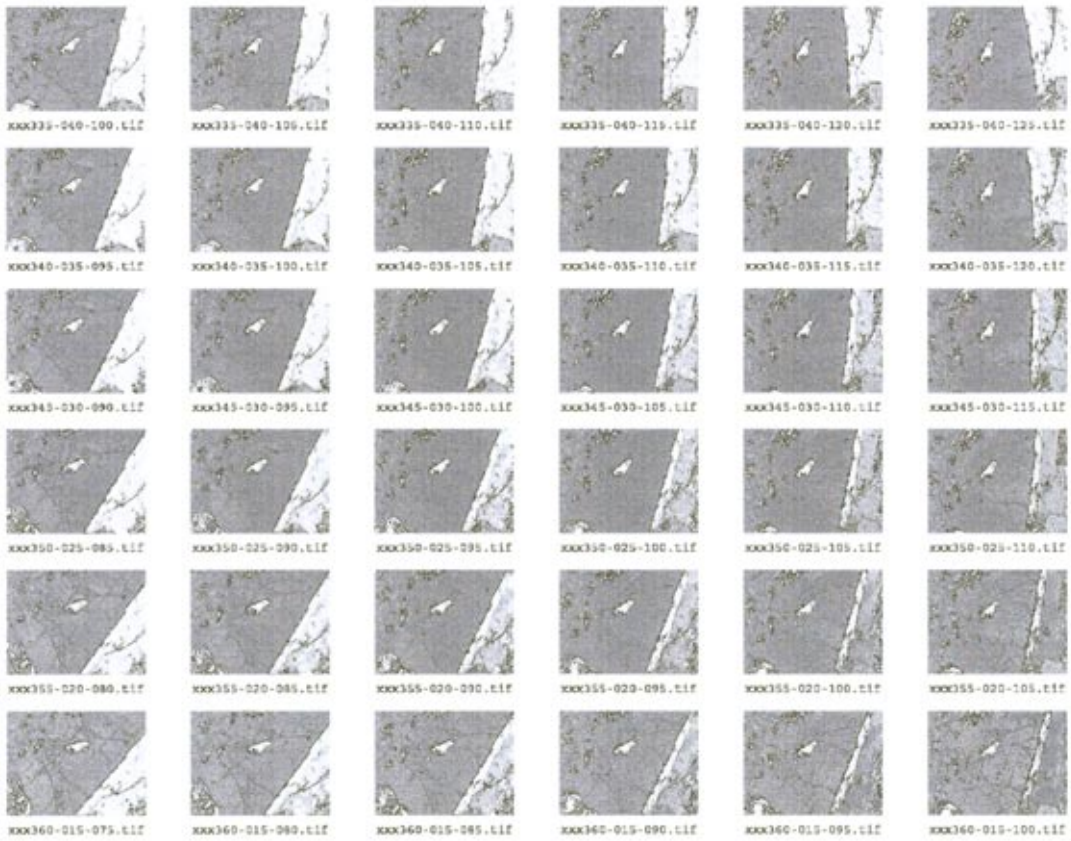

Figure 7. Images under the various orientations of two quarter wave plates and the stage. (See text.)

the direction of vibration of the polarizer, and graduated clockwise , contrary to the case of θ.

The direction of the upper quarter wave plate is shown with β. The origin of β is arbitrarily fixed, and its value increases counterclockwise. In the following observations, two quarter plates are set at subtractive positions of $\alpha + \beta = 375°$.

Several images with different values of α, β and θ are recorded as TIF file. The file name was given according to the combination of α, β and θ

(a) Observations at various angles with 5° steps

Figure 7 shows some of the recorded images. Six images of the bottom row are taken with $\alpha = 360°$ and $\beta = 15°$, which means none of the effect of the quarter plates occurs on the linearly polarized lights, and θ changes from 75° to 100° with a 5° step. Concerning the brightness at the central part of each image, they are fairly bright in the images on either end and fairly dark in the images in the center. However, no extinction position, where none of the transmitted rays are observed, is found for the linearly polarized light. Images of the second lowest row are taken at $\alpha = 355°$ and $\beta = 20°$, that means the incident beam is elliptically polarized lights whose ellipticity is equal to $|\tan 355°| = 0.087$, and θ changes from 80° to 105°. The central part of each image of this row is a little darker than that of the bottom row. Images of the other rows correspond to $\alpha = 350°$, $\alpha = 345°$, $\alpha = 340°$ and $\alpha = 335°$. The central parts of images of conditions around $\alpha = 345°$ and $\theta = 100°$ and appear to be fairly dark and found almost to be at the extinction positions.

To confirm this observation, it is listed in Figure 8a by the aid of Image-Pro Plus that the mean value of the darkness of 5502 pixels in the circle of 150 μm at the central part of each images in Figure 7, in which each line corresponds to α-value of 335° to 360° step 5° and each row corresponds to $(\alpha + \theta)$-value of 435° to 460° step 5°. The left small figure in Figure 9 shows the contour line of the distribution of the darkness in Figure 7. In this figure, there is one and only one peak near the center and the marginal parts are less dark compared to the inner parts.

(b) Observations at various angles with 2° steps

Figure 8b and the right large figure in Figure 9 are obtained through a similar procedure as described above, but α-value are selected between 340° to 354° and $(\alpha + \theta)$-value are between 440° to 454° with an interval of 2°. Because little differences are observed visually, figures

(a)

a/° \ $(\alpha+\theta)$/°	435	440	445	450	455	460
335	4148	4282	4316	4313	4232	4151
340	4338	4464	4555	4537	4442	4267
345	4459	4634	4686	4625	4489	4241
350	4534	4592	4641	4559	4409	4104
355	4149	4348	4402	4324	4121	3833
360	3773	3915	3982	3904	3688	3400

(b)

a/° \ $(\alpha+\theta)$/°	440	442	444	446	448	450	452	454
340	2657	2660	2662	2664	2661	2658	2654	2649
342	2664	2667	2669	2669	2667	2662	2657	2651
344	2672	2676	2677	2676	2673	2669	2660	2652
346	2674	2678	2681	2678	2675	2668	2660	2650
348	2675	2679	2681	2681	2678	2671	2661	2652
350	2674	2678	2679	2678	2674	2667	2656	2644
352	2669	2674	2675	2671	2667	2661	2650	2637
354	2661	2663	2667	2665	2657	2650	2643	2630

Figure 8. Darkness of the circle at central part of each image in Figure 7. (a) 5° steps, (b) 2° steps.

like Figure 7 would offer little information about the angular changes and the darkness. However, the quantitative analyses show distinct changes as seen in the table and contour lines. It is concluded that complete extinction is observed at the condition around $\alpha = 347°$ and $\alpha + \theta = 444°$, i.e. $\alpha = 347°, \beta = 28°$ and $\theta = 97°$. This means the ellipticity of the propagating light in the crystal is $|\tan 347°| = 0.23$ and elliptic vibration in the section is right handed under the present experimental conditions.

Discussion

The above result that complete extinction is observed at the condition around $\alpha = 347°, \beta = 28°$ and $\theta = 97°$ means that the propagating light in the crystal is elliptically polarized and its ellipticity of $|\tan 347°| = 0.23$. Although the crystallographic orientation for the crystal is not yet specified, this is concordant with the previous description of the ellipticity of 0.466 for the (010) section of heikolite from Kogansan in Korea, 0.212 for the (010) section of arfvedsonite from Nunarsuatsiak, Tunugdliarfik in Greenland, and 0.087 in the (010) section of riebeckite from St. Peter's Dome, El Paso County, Colorado (Shoda, 1957; 1958).

It is well known that the elliptically polarized lights exist in optically active crystals. Crystals, which belong to the following 15 point groups, C_1, C_2, C_3, C_4, C_6, D_2, D_3, D_4, D_6, T, O, S_4, D_{2d}, C_s and C_{2v}, have optical activity. Elliptically polarized rays had been observed near the direction of optical axes. Recently, a new optical method and apparatus 'HAUP' was developed (Kobayashi and

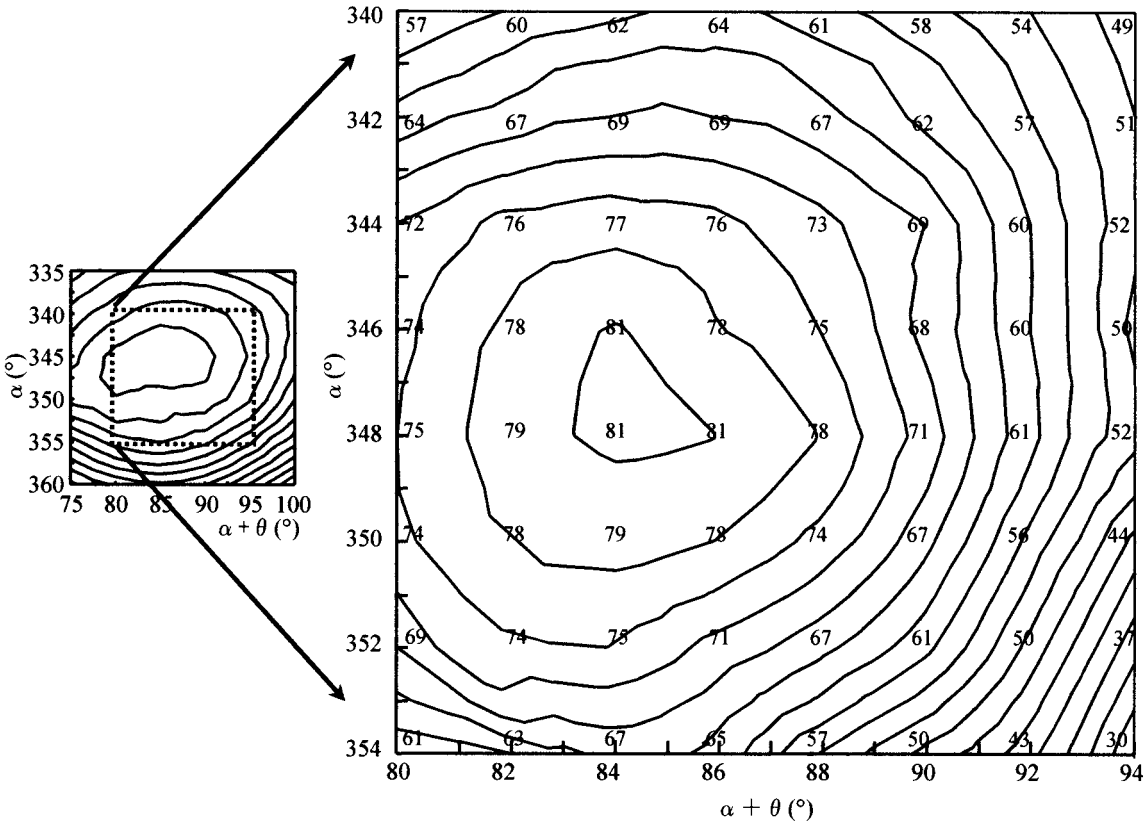

Figure 9. Contour lines of the distribution of the darkness in Figure 7.

Uesu, 1983; Kobayashi et al., 1983) and precise measurement of all optical properties including the ellipticity were able to performed for every desired orientation in such crystals. These phenomena are explained through the nature of the dielectric tensor ε. The relation between electric flux density D and electric field E can be written as

$$D = \varepsilon E \quad (1)$$

The principle of HAUP is originated in the assumption that ε is a function of not only the frequency but also the wave number vector of light. According to the developer's article (Kobayashi, 1995), if the absorption of light is ignored, equation (1) is straightforwardly rewritten to

$$D_i = \varepsilon_{ij} E_j + \mathrm{i}\varepsilon_0 G_{ij} E_j \quad (2)$$

where ε_0 is permittivity of vacuum. Because all of the components ε_{ij} of non-absorbing medium are real, the imaginary parts exist only in the second term. This means that the gyration tensor G is the origin of the elliptically polarized light in a non-absorbing crystal of appropriate symmetry.

In the case of the present study, equation (2) loses its meaning, because the crystals of alkali amphibole belong to C_{2h} symmetry, all components of G_{ij} are equal to zero. For the absorbing crystal like alkali amphibole, ε_{ij} themselves are not real numbers but complex ones, and the ellipticity of the light is originated from such nature of the dielectric tensor ε in equation (1).

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