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# Anisotropism in Garnets

Arthur B. Merkle

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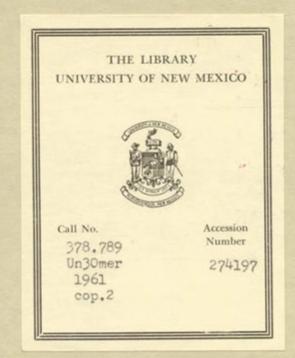
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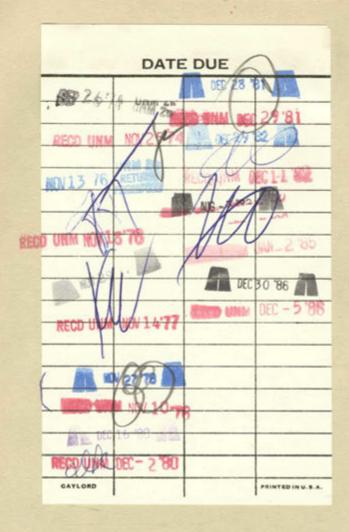
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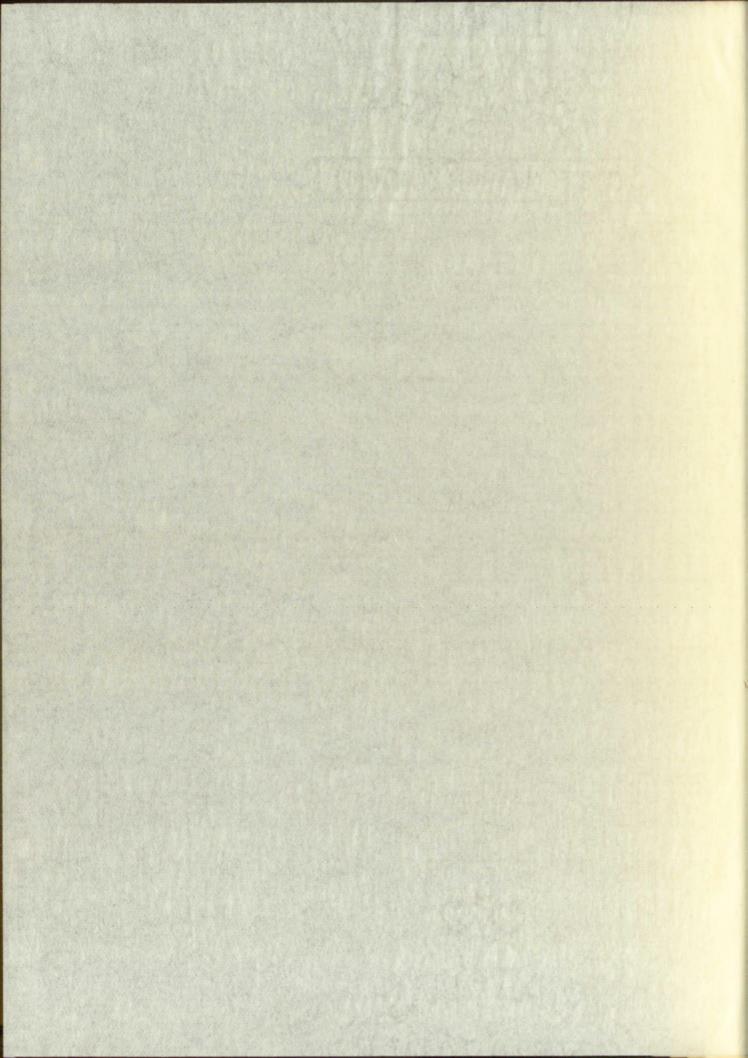


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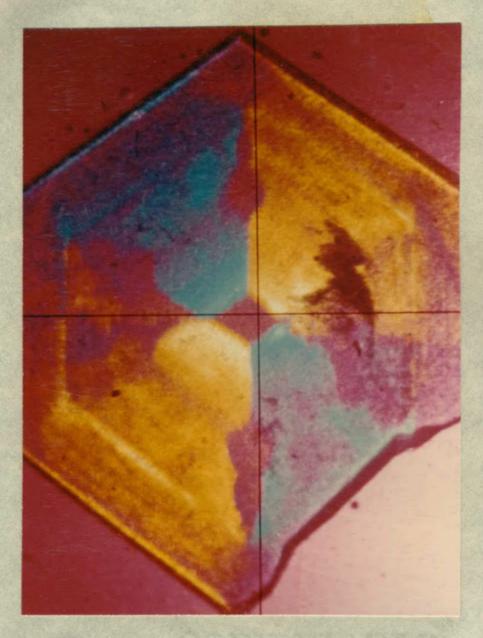
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Garnet VI. C. Anisotropic crystal, view parallel twofold axis, before heating, X-nicols, first order red plate, 26X.



### ANISOTROPISM IN GARNETS

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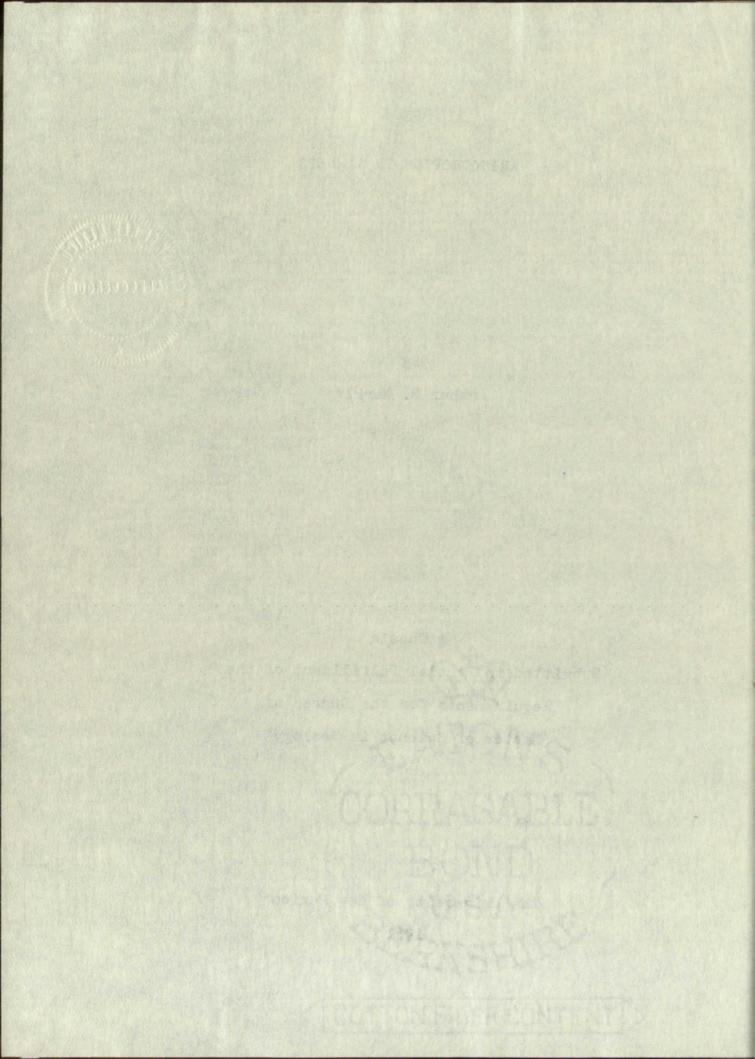
By

Arthur B. Merkle

A Thesis

Submitted in Partial Fulfillment of the Requirements for the Degree of Master of Science in Geology

The University of New Mexico



This thesis, directed and approved by the candidate's committee, has been accepted by the Graduate Committee of the University of New Mexico in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

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Date

Thesis committee

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Wolfgang E. Elston

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### ABSTRACT

Six anisotropic garnets are studied by optical, thermal, and X-ray diffraction methods in an attempt to determine the cause of anisotropism. The garnets are all ugrandites with compositions ranging from chromian grossularite to nearly pure andradite; the composition having been estimated from indices of refraction and unit cell dimensions. In all cases the birefringence is 0.004 or less. Both positive and negative uniaxial and biaxial figures are sometimes observed. Heating of the garnets at 900°C for periods of 96 to 264 hours reduced the anisotropism in five of the six garnets studied; this observation is in agreement with previously reported studies. Precession and powder X-ray diffraction photographs show no detectable change from unheated to heated samples. Precession photographs of several sectors of a single anisotropic garnet yield identical patterns with isometric Ta3d symmetry, suggesting that anisotropism is not caused by twinning. I feel the cause of anisotropism in garnets is due to a combination of strain and rate of crystal growth in the metamorphic environment. A technical garnet bibliography of over 300 papers, written from 1900 to 1960 is presented; papers on anisotropic garnets written before 1900 are also included.

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### INTRODUCTION

### The Garnet Group

The term garnet is derived from the Latin granatus, meaning grainlike. This mineral crystallizes in the hexoctahedral class of the isometric system and forms a group of isomorphous silicates whose general formula is R<sub>3</sub><sup>+2</sup> R<sub>2</sub><sup>+3</sup> (SiO<sub>14</sub>)<sub>3</sub>. Winchell (1933, p. 174) has divided the garnet group into two species with the following names and compositions:

Specie	Type	Composition
Pyralspite	Pyrope	Mg3Al2(S104)3
	Almandite	Fe3Al2(S104)3
	Spessartite	Mn3A12(S104)3
Ugrandite	Uvarovite	Ca3Cr2(S104)3
	Grossularite	Ca3Al2(SiO4)3
	Andradite	Ca3Fe2(S104)3

According to Dana (1932, p. 591-592) the two common habits of garnet are dodecahedral and trapezohedral.

The hardness ranges from 6.5 to 7.5; the specific gravity varies from 3.15 to 4.3.

Menzer (1928, p. 300-304) described the structure of garnet as follows: space group Ia3d; eight formulas to the unit cell.

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### Lattice Parameters

Ion	Position	Parameters
R+3	16	(0,0,0)
S1+4	24	(1/4,3/8,0)
R+2	214	(1/4,1/8,0)
0-2	96	(x,y,z)

In aluminum garnets the oxygen parameters are, x=0.0\psi.01, y=0.055\psi.01, and z=0.6\psi.015; in calciumiron and calcium-chrome garnets the oxygen parameters are, x=0.035\psi.01, y=0.0\psi.01 and z=0.655\psi.015.

Dana (1932, p. 591) describes the structure of garnet as follows, SiO<sub>4</sub> tetrahedra are independent of each other; R+3 atoms lie in the center of a group of six oxygen atoms, and R+2 atoms in the center of a group of eight oxygen atoms.

Substitution in garnets may occur freely within each species, but is limited to about 20% between the two species.

Garnets are normally isotropic. Some, however, show optical anisotropism. Winchell (1933, p. 180) states anisotropism is usually restricted to ugrandite, but is sometimes seen in spessartite.

### Total Persons and the

(0,0,0)	
(0,8/8,0/0)	
(0,8/1,1/1)	
(2,7,2)	5-0

In sluminum garmets the oxygen parameters are, x=0.06.2.01, y=0.0532.01, and a=0.66.2.015; in calcium-tron and calcium-chrome garmets the oxygen parameters are, x=0.0352.01, y=0.062.01 and n=0.6552.015.

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Purpose, Scope, and Method

The three main purposes of this thesis are:

- 1. The description of anisotropism in garnets.
- 2. A study of the effect of heat on anisotropic garnets.
- 3. A study of the cause of anisotropism in garnets.

Experimental work of three types, optical, thermal, and X-ray, was done between November 1959 and March 1961.

Anisotropic garnets from California, Canada, New Mexico, and Tasmania were studied. A literature search was made, and a selected bibliography of all technical garnet papers from 1900 to 1960 prepared; papers dated prior to 1900 were included only if they pertained directly to anisotropism in garnets.

### Acknowledgments

The author wishes to express appreciation to

Dr. Abraham Rosenzweig, of the University of New Mexico

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for supplying garnet samples used in this study. The
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I wish to express my appreciation for the research assistantship under a Sandia Corporation contract which provided me with financial support during my graduate study. The precession camera used in the X-ray studies was obtained through a Research Corporation grant.

### Previous Work

Since anisotropism was first observed in garnets, many attempts have been made to define its cause. In these attempts two methods of study have been used, optical and X-ray diffraction. Previous studies indicate two main causes of garnet anisotropism:

- 1. Twinning of elements with a lower symmetry.
- Stress and pressure caused by isomorphous substitution during formation of the garnet.

The following statements are those of the authors; their terminology is used in all cases.

The first optical study of anisotropic garnets was carried out by Mallard in 1876. His studies led him to believe anisotropism was caused by twinning of individuals with a lower symmetry.

Klein (1898) studied various garnets from Vilui,
Siberia and decided they were composed of twinned monoelinic or triclinic individuals showing vicinal forms.

He also found anisotropism was not dependent on composi-

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"optical structure" and external form. Anisotropism is also affected by pressure differences and the relative size of atoms in the structure. Anisotropic garnet crystals are built up by overgrowths of twinned monoclinic or triclinic individuals in various orientations upon a small single crystal. This twinning produces an orthorhombic symmetry which is secondary and caused by either tensions due to isomorphous substitution or twin laminae. The external symmetries of the garnets vary with crystal growth and need not all be the same.

Kirganov (1941) said anisotropism in garnets is caused by the presence of intergrown twin laminae.

Corin (1943) stated anisotropic garnets are caused by leucite-type polysynthetic twinning.

Lehmann (1888) said anisotropic garnets are a "mechanical mixture" of two or more chemically different garnet types. The different thermal coefficients of expansion of these types cause stress within the crystal, that varies with crystallographic direction. Homogeneous crystals have similar coefficients of expansion in all directions, whereas in mixed crystals the coefficients of expansion vary with the crystallographic directions. This could be the cause of anisotropism in garnets.

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This could be the cause of anisotropism in darnets.

Brauns (1891) stated that optical observations show anisotropism in garnets caused by an isomorphous substitution of two or more chemically different garnet types. This substitution creates internal stress and tension causing anisotropism. The amount of anisotropism is varied by a variation of stress with crystallographic direction. This is analagous to phenomena seen in sylvite and flourite.

Rinne (1925) was the first person to study anisotropic garnets by the use of X-ray diffraction (the Laue and powder methods). His experiments showed anisotropic garnets are caused by variations in the electron paths due to a slight shifting of the atomic nucleus. This shifting was probably caused by stress and pressure due to twinning and isomorphous substitution within the crystal.

Starkov (1950) described three distinct types of optical anomalies in garnets:

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thickness are variable. All bands extinguish parallel to the 110 trace of the crystal. This extinction appears in six regular sectors corresponding to the faces of a hexagon.

- 2. Crystals are characterized by a system of bands in each sector, generally trending at an angle near 60° to the trace of the (110) face for the given sector and parallel to the trace of the (110) face in one of the adjoining sectors. These bands are uniform in width and situated in a triangle between the central core and the peripheral part of the crystal (Starkov fig. 4, p. 285).
- 3. Crystals with birefringent bands arranged perpendicular to the trace of the (110) face. These bands do not have sharp boundaries and are generally found near the edge of the crystal. In cross section, these bands form a "rectangular lattice with the concentric layers."

Starkov felt these features were caused by isomorphous replacement of Fe<sup>+2</sup> by Ca<sup>+2</sup>. The difference in the ionic

The expression in quotation is a direct translation from the Russian. Starkov provides no illustration of this garnet type, and consequently the description of this anisotropism remains ambiguous.

2. Drystair and change to recent by a larger of the THE THE RESERVED AND THE PROPERTY AND

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radii of these two ions will cause a distortion "of the electron shell surrounding ions of andradite" thus creating internal stress. Impurities in the crystallizing solution were systematically arranged in layered overgrowths. These also could cause anisotropism.

Merwin (in Wright, C. W., 1915, p. 108) performed the first thermal experiments with anisotropic garnets. He observed that they became isotropic when heated to 800° for a few hours.

Soloviev and Nikogosyan (1938) found that anisotropic garnets lost their birefringence within the temperature range 7500-8500C and a heating time of 72-168 hours.

Kozu (1940) observed that anisotropic zones in garnets decreased in intensity with increased temperature.

Yoder and Keith (1951) working with artificially prepared garnets, found a complete solid solution between spessartite and yttrogarnet

Mn<sub>3</sub>Al<sub>2</sub>(SiO<sub>4</sub>)<sub>3</sub> → Y<sub>3</sub>Al<sub>2</sub>(AlO<sub>4</sub>)<sub>3</sub>

Yttrogarnet has been shown to be isometric. However, at 1970°C±50° a solid state transition occurs between yttrogarnet and yttroalumite. X-ray studies by Donnay show yttroalumite to be tetragonal with a primitive lattice.

Keith and Roy (1954) presented further evidence substantiating the experiments of Yoder and Keith. edit to thing the loss of any office of sold to the time time of any office of sold and the sold to the sold and any office of the sold the sold to the sold the sold

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### EXPERIMENTAL WORK

#### Procedures

The purpose of the experimental work of this thesis is three-fold:

- 1. Identification of the garnet type.
- Relation of the crystallographic properties to anisotropism.
- 3. Relation of the thermal behavior to anisotropism.

The garnets studied in this thesis were unoriented garnet masses (garnets I, II, and III), small single crystals (garnets IV and V), and orientated thin sections of large single crystals (garnet VI).

All garnets were heated in an electric furnace, under oxidizing conditions and atmospheric pressure, to temperatures of 900°C for intervals ranging from 96 to 264 hours. The results of this work are presented in table 2.

The index of refraction of the garnets was determined by the immersion method on crushed fragments. Optic signs and figures were obtained with the polarizing microscope. The 2V was approximated by the curvature of the isogyres. In each garnet studied the birefringence is approximated as 0.004 or less. The universal stage was not used in any of the optical observations.

X-ray powder photographs were taken of garnets I, II, III, and IV; Buerger precession photographs were taken of

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garnets IV, V, and VI. Filtered molybdenum and copper radiations were used. The unit cell dimensions and the symmetry of these garnets were determined from the reflections present on these photographs. The unique space group Ia3d was selected because the systematic extinctions of this group impose conditions that are fulfilled by the garnets studied. That is, reflections of the type hkl are present only if h+k+l is even; hhl reflections are present only when l is even; furthermore, for hhl, 2h+l is a multiple of four, and hk0 reflections are present only when both h and k are even.

The crystals of garnet VI were large enough to isolate various portions of the crystal in the X-ray beam, permitting an examination of the orientation of various crystal segments in order to detect twinning. In general, the procedure is as follows: the crystal is centered in the X-ray beam and lined up on a symmetry axis. A precession photograph is then taken. The crystal is then shifted, so the X-ray beam strikes the outer edge of a portion of the crystal. Another precession photograph is taken. This procedure is repeated at regular intervals around the crystals edge. If detectable twinning is present it will be seen as a change in the orientation of the diffraction pattern. No detectable twinning was observed in garnet VI.

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Garnets seen in plates I. B, I. D, and II. C are not the same areas seen in plates I. A, I. C, and II. B; these areas are of adjacent thin sections in the rock sample.

# Composition of the Garnets

The composition of the garnets studied has been determined from triangular diagrams plotted by Sriramadas (1957, p. 294-298). These diagrams relate unit cell dimension and index of refraction to composition. The compositions of these garnets are shown in table 1. . . .

# Anisotropism of the Garnets

Garnet I.

Location:

One and a quarter miles north of Darwin, near the Thompson Mine, Darwin mining district, Inyo County, California.

Megascopic description:

The garnet occurs as a yellowish-green mass in a tactite.

Optical properties:

The thin-section color of the garnets is light yellowish-gray. The masses are made up of zoned dodecahedra. Numerous inclusions are present. The crystals are composed of anisotropic and isotropic zones (Pl. I. A). Wavy

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Table 1 .-- Garnet composition

Garnet number Location		Index of refraction	Composition (after Sriramadas, 1957)
I Darwin, California	11.9Å	1.830±.005	53% Andradite 34% Grossularite 16% Others
II Silverhill Mine, New Mexico	12.0Å	1.865±.005	83% Andradite 12% Grossularite 5% Others
Unknown	12.0Å	1.870±.005	85% Andradite 7% Grossularite 8% Others
IV Iron King Mine, New Mexico	12.04	1.880±.005	90% Andradite 2% Grossularite 8% Others
V Tasmania	12.0%	1.875±.005	88% Andradite 5% Grossularite 7% Others
VI Orford, Canada	11.8Å	1.730±.005	90% Grossularite 10% Others (probably uvarovite in part)

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extinction is seen in some crystals. Faint uniaxial and biaxial figures are seen. The optic sign varies from (+) to (-). The optical anomalies resemble Starkov's type 1.

Garnet II.

Location:

Southeast of the Silver Hill mine, NW2 sec. 3, T. 25 S., R. 21 W., Hidalgo County, New Mexico.
Megascopic description:

The rock is a solid mass of green, brown, and red garnet. Some euhedral crystals are present. These crystals show zoning which can be distinguished by color differences.

Optical properties:

The thin-section color is a yellowish-green. The garnets are massive, anhedral, and cloudy with numerous inclusions. Wavy extinction is present. The optic sign and optic figure grenot determinable. Anisotropism is present in the entire mass (Pl. I. C).

Garnet III.

Location:

Unknown.

Megascopic description:

Garnet crystals occur in a calcite mass with chalcopyrite, malachite, and magnetite. The garnet is greenbrown and zoned. extinction is seen in some organic, in this file that is a seen and biaxis I figures are seen. The tiping of a security of the (-) to (-). The optical anguist on remaining against type i.

Carnet II.

Locations

Southeast of the Silver Hill mine, Not see. ...
T. 25 8., R. 21 W., Hidalgo Cownby, New Yorks...
Negascopic description:

The rock is a colld mass of green, brown, and red gurnet. Some suhedral organizate and process. They organizate above coning which can be distinguished of uplandid differences.

Optical properties;

The thin-section solar is a pollori elegant. The garacte are usually and closery with investigations in classic, who cathocides is presume. The critic atm and optic figure arenet determinable. Library of the present in the entire mass (Fl. T. C).

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Regescopie description:

Garnet orretals occur in a calcife speak planta deprint de capacite, "The senses is grown or brown and somed,

Optical properties:

Thin-section color of the garnets is greenish-brown.

The garnet is massive with some euhedral crystals. Zoning present in the garnets parallels dodecahedral faces

(Pl. II. A). Some wavy extinction is observed. These garnets are mostly isotropic; anisotropism is seen in zones of varied widths, which outlines dodecahedral faces

(Pl. II. B). The anisotropism resembles Starkov's type 1; uniaxial and biaxial figures are observed.

Garnet IV.

Location:

Iron King mine, Jarilla Mountains, sec. 34, T. 21 S., R. 8 E., Otero County, New Mexico.

Megascopic description:

Euhedral, yellowish-brown garnets occur in a granular marble with specular hematite. These garnets are zoned and are both dodecahedral and trapezohedral in habit.

Optical properties:

The crystal studied is a yellowish-brown dodecahedron 0.5 mm in size. The crystal is anisotropic and contains some inclusions. Anisotropism varies with crystallographic direction. Due to the small size of the crystal, optic figures and signs are not observed.

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Garnet V.

Location:

Tasmania (precise locality and mode of occurrence not known).

Megascopic description:

The crystals are greenish dodecahedrons. Their size ranges from 0.5 mm to 1.0 mm; the crystals are intergrown with some inclusions.

Optical properties:

The crystal studied is a greenish dodecahedron 0.7 mm in size. Anisotropism seen in the crystal varies with crystallographic direction. Because of the crystal size, optic figures and signs are not observed.

Garnet VI.

Location:

Orford, Sherbrooke County, Quebec, Canada. Megascopic description:

Emerald green garnet crystals occur in a soft white tale.

Optical properties:

Three dodecahedral garnet crystals were studied.

Their color in thin section is greenish; their size is about 3.5 mm. Three orientated thin sections were made.

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A. perpendicular to a three-fold exis; B. perpendicular to a four-fold exis; and C. perpendicular to a two-fold exis.

Crystal A. Anisotropism in this crystal is a combination of Starkov's type 1, and type 3 (Pl. III. A and B). Anisotropic zone boundaries are not sharp. The crystal shows biaxial negative optic figures. A 2V of 60° is estimated from the curvature of the isogyres.

Crystal B. This crystal is mainly isotropic with some small anisotropic bands (Pl. III. C and D). The anisotropism is similar to Starkov's type 1. Faint traces of similarly orientated biaxial (-) figures are observed.

Crystal C. Thin isotropic sections are observed.

The central part of the crystal is biaxial (-);

2V is about 60°. Six triangular areas are arranged about the central portion. These sectors are uniaxial; their optic signs alternate (+) and (-).

This crystal is shown on Plate IV. The anisotropism is a combination of Starkov's type 1 and type 3.

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## Symmetry of Anisotropic Garnets

Based on procedures stated in the beginning of the experimental work section, the symmetry of the six anisotropic garnets is found to be Ia3d. There are no observable deviations from this. No detectable twinning is seen in garnets IV, V, and VI. X-ray patterns of garnets IV, V, and VI are in no way altered with heat.

### Thermal Treatment and Results

The six garnets studied have been heated in an electric furnace, under oxidizing conditions and atmospheric pressure, to a temperature of 900°C for intervals ranging from 96 to 264 hours. Optical and X-ray properties remain constant after heating. Only the amount of anisotropism in the garnets is changed. Where possible to observe, as in garnet VI, the optics of the anisotropic zones are in no way altered with heat. X-ray patterns are also unaltered. Results of this treatment are shown in table 2.

#### CONCLUSIONS

Thermal treatment of five of the six garnet samples show either a reduction in or a destruction of the anisotropism of the garnets. In these five cases the samples were heated to 900°C for intervals ranging from 96 to 264 hours. The sixth sample (garnet I)

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Table 2 .-- Results of thermal treatment of the garnets

Garnet number	ture		Observed results
Location	attained	nould	
Darwin, California	900°C	226	Thin section color changed from light-yellow-gray to yellow-brown. Heat had no observable effect on anisotropism (Pl. I. B).
Silver Hill mine, New Mexico	900°C	226	Thin section color changed from yellowish-green to brown-yellow. Faint traces of anisotropism remain in the mass (Pl. I. D). Heat reduced the amount of anisotropism.
III.			
Unknown	90000	264.	Thin section color changed from greenish-brown to brown- yellow. Thin anisotropic zones remain (Pl. II. C). Amount of anisotropism was reduced with heat.
IV. Iron King mine, New Mexico	900°C	168	Thin section color changed from yellowish-brown to red- brown. The amount of aniso- tropism was greatly reduced.
V. Tasmania	900°C	96	Thin section color changed from greenish to yellowish-brown. Heating reduced the anisotropism present. However, it did not produce a completely isotropic crystal.
VI. Orford, Canada	900°C	168	Thin section color changed from greenish to brown-green. Zones remaining anisotropic became yellow-green. Anisotropism is reduced in amount in each crystal (Pls. III and IV).

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destroy the anisotropic effect is probably in some way proportional to the amount of strain that the garnet has undergone during its formation.

With further study this could probably be developed into a tool to define the temperature and pressure of formation of a metamorphic deposit containing anisotropic garnets.

This "strain" hypothesis does not take into account the sharp boundaries and distinct crystallographically orientated zones of anisotropism in the garnets. A possible but unproven reason for these could be the rate of growth of the crystal. The faster the growth the more pronounced the anisotropic effect. These anisotropic zones are usually parallel to the outer faces of the crystal and would represent growth surfaces.

The anisotropic effect seen in garnets could be due to a combination of strain and growth rate of the crystal.

The absence of idetectable twinning does not rule out
the possibility that twinning is the cause of anisotropism in garnets. One needs to use other and better
methods for determining the presence of twinning in
garnets.

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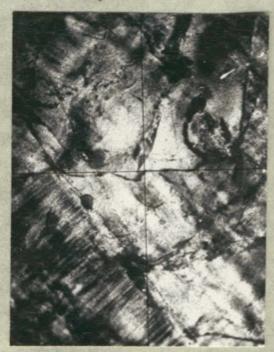
The chaonds of detectable twinning does not rule out the pessibility that twinning is the cause of amisotropism in garnets. One needs to use other and better methods for determining the presence of twinning in garnets.

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A. Garnet I: Anisotropic crystals, before heating, X-nicols, 80X



C. Garnet II: Anisotropic mass, before heating, X-nicols, 26X



B. Garnet I: Anisotropic crystals, after heating for 226 hours, X-nicols, 80%



D. Garnet II: Anisotropic mass, after heating for 226 hours, X-nicols, 26X





A. Garnet III: Anisotropic crystals, before heating, plane polarized light, 26X



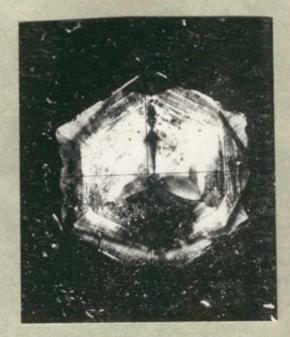
B. Garnet III: Anisotropic crystals, before heating, X-nicols, 26X



C. Garnet III: Anisotropic crystals, after heating for 264 hours, X-nicols, 26X



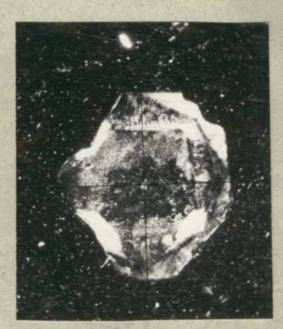
### PLATE III



A. Garnet VI.A: Anisotropic crystal, view parallel three-fold axis, before heating, X-nicols, 26X



B. Garnet VI.A: Anisotropic crystal, view parallel three-fold axis, after heating for 168 hours, X-nicols, 28X

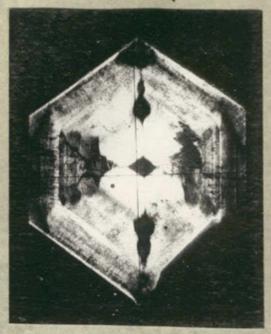


C. Garnet VI.B: Anisotropic crystal, view parallel four-fold axis, before heating, X-nicols, 26X



D. Garnet VI.B: Anisotropic crystal, view parallel four-fold axis, after heating for 168 hours, X-nicols, 28X

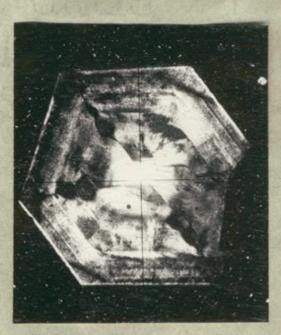




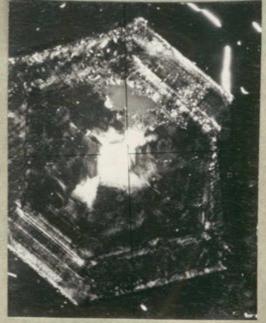
A. Garnet VI.C: Anisotropic B. Garnet VI.C: Anisotropic crystal, view parallel two- crystal, view parallel twofold axis, before heating, X-nicols, 26X



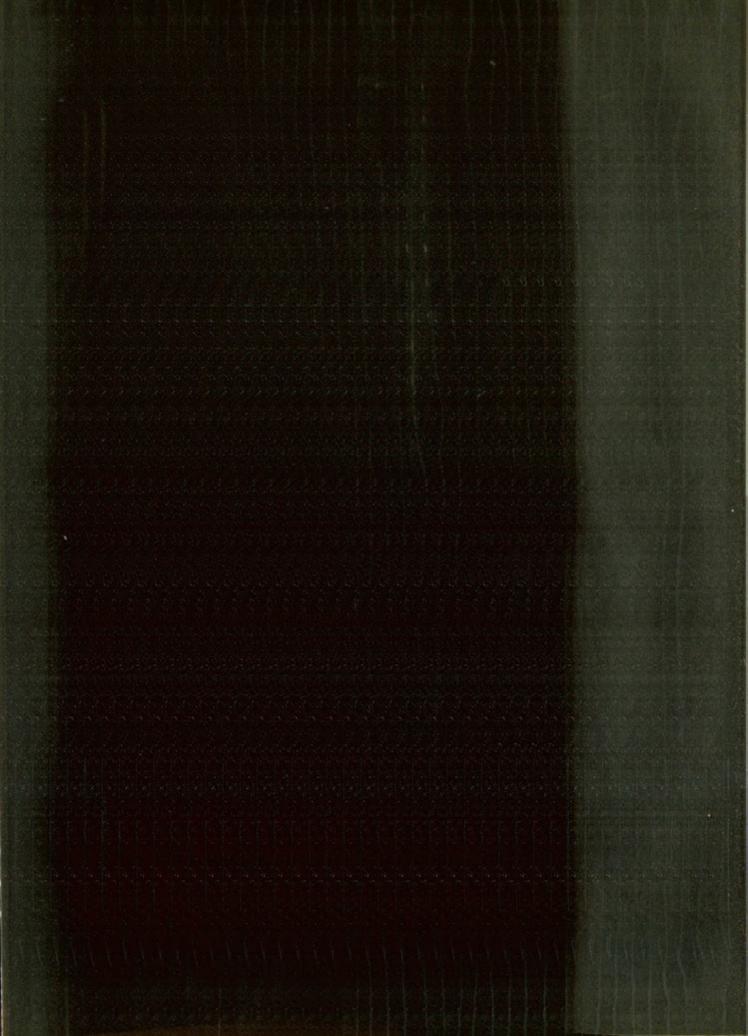
fold axis, after heating for 144 hours, X-nicols, 28X



C. Garnet VI.C: Anisotropic D. Garnet VI.C: Anisotropic nicols, 26X



crystal, view parallel twofold axis, rotated 45° from
A., before heating, X
Crystal, view parallel twofold axis, rotated 45° from
A., after heating for 144 hours, X-nicols, 28X



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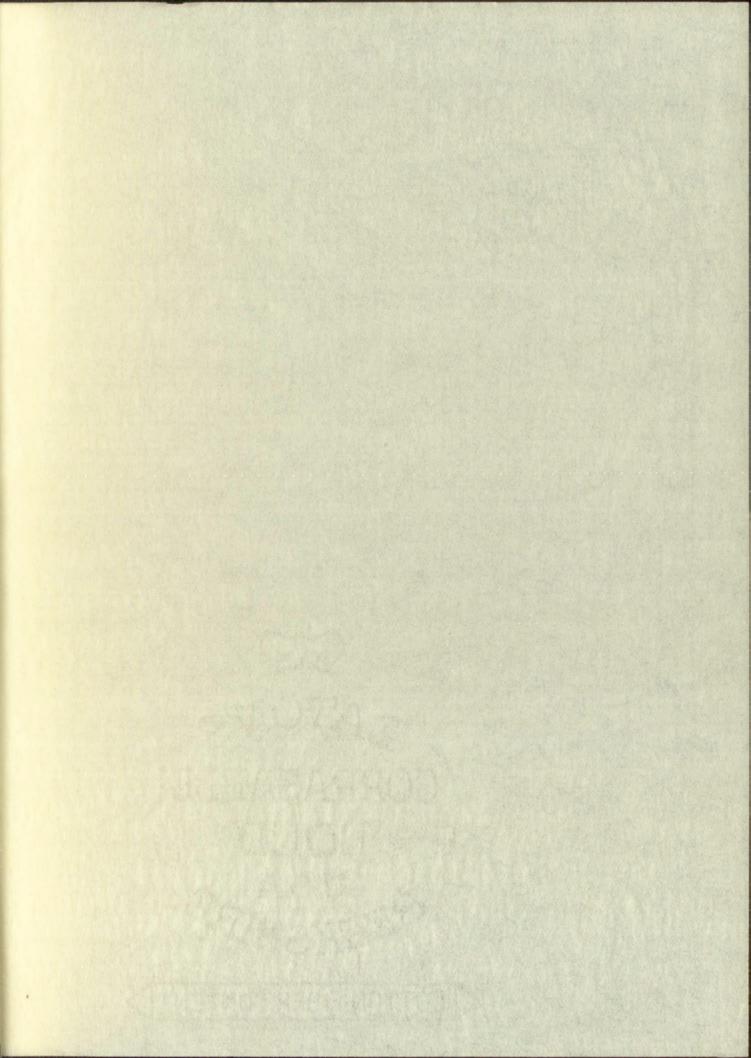
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