Gems & Gemology

GIA

FALL 1976

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ISSN 0016-62X

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gems &
gemology

VOLUME XV NUMBER 7

FALL 1976

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How to Define Non-Single-Crystal Synthetics

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Introduction

When dealing with single crystal synthetic equivalents of gem materials there are quite clear-cut guidelines as to terminology. To be termed "synthetic," the gem material must have essentially the same chemical composition and the same crystal structure. This should automatically ensure an appearance essentially identical with the natural gem. (Note that chemical composition alone does not automatically guarantee structural identity, as with diamond and graphite, where both have the same composition but different structures.) In a previous article(1) the distinctions between "Natural, Treated, Synthetic and Imitation Gems" were discussed in some detail (including some exceptions).

However, there are peculiar problems which are now becoming apparent in connection with non-single-crystal* materials, those that would normally be termed amorphous, microcrystalline, polycrystalline, glassy, and so on.

Until recently, gem synthesis had been successfully accomplished only for single crystal materials such as ruby, emerald, quartz, etc. The achievements of what appear to be the synthetic counterparts of natural opal,(2,3) turquoise,(4,5,6) and lapis lazuli (made by P. Gilson, France) raise the question of how to define certain subtleties involved in the "synthetic" designation and also how to determine whether or not these materials meet the definition.

In the years to come, undoubtedly, yet other non-single crystal materials such as malachite, rhodochrosite, the jades, the agates, etc., may well join this list. It would thus seem to be desirable to clarify the problems involved in the designation "synthetic" and, to this end, a possible set of definition criteria is presented below.

The aim of this discussion is to suggest guidelines for the examination (by what may be called the gemologists' gemologists) of a new material of the non-single-crystal type when it

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*The author apologizes for this rather clumsy hyphenated designation. It is, however, concise, the alternative being "polycrystalline, amorphous, vitreous and partly ordered" (the last being necessary for materials such as opal and amazonite).

first appears. As part of this examination, the type of information discussed below will enable the decision to be made whether the material deserves the "synthetic" label or not. Another part of this initial examination will determine those characteristics, usually listed in gemology texts, by which the practicing gemologist can recognize the material without the necessity of any of the elaborate testing discussed below. As always, a careful watch will be needed to ensure that a modification of the manufacturing process does not alter the material sufficiently so that a change in classification will be necessary.

The Five Levels of Examination

Having stated the chemical composition and the crystal structure, the specification of a single crystal equivalent of a gem material is essentially complete. There are two additional subtleties: 1) different colors caused by different valence states of the impurity, or from impurities located in differing sites, with the possibility of color centers as well; and 2) disorder, as in metamict zircon.

In non-single-crystal materials the structural properties can be of significance at no less than five levels of magnification. Proceeding up the size scale from the smallest items we have:

(1) Individual atoms. Here there are the chemical composition, including significant impurities, valence state specification of variable valence ions, and color centers.

(2) The atomic structure. This is revealed, for example, by an X-ray powder diffraction pattern which reflects the structural features at the 0.1 to 10 nanometer (1 to 100 Ångstrom) level. Absence of any signs of structure at this level may indicate an amorphous or glassy nature.

(3) The sub-micro-structure level may be taken as the region accessible only to the electron microscope, with features at the 100 to 1000 nanometer (0.1 to 1.0 micron) level. An example is the alignment of the spheres in opal to form the diffraction grating structure which produces the color phenomenon. The lamellar structures in materials such as labradorite also range down to this size.

(4) The micro-structure, as seen under the optical microscope, and even under the magnifying glass, reveals structures from the 1000 nanometer up to the 1 millimeter level; here considerable variation may exist from, say, one specimen of turquoise to another. This range also includes the needles and inclusions in many eye and star materials. At the upper end of this scale occur gross inclusions, such as the pyrite seen in much lapis lazuli, as well as matrix present in much turquoise, for example.

(5) Macro-structure, visible to the naked eye. Here again there is considerable variation from one specimen to another, yet all specimens of a given species, say gem opal, have that appearance which makes the material instantly recognizable as being gem opal.

A Proposed Working Definition

To be considered the synthetic counterpart of a natural non-single-crystal gem material, it is proposed that four criteria should be met in the strictest sense:

(a) The chemical composition must be the same, including valence states. There may be some substitutional variation, as in natural material, as well as contributions from the matrix. The synthetic must fall within the range found to occur in nature; no foreign binder, filler, or coloring matter can be present. (This may present analytical problems since a small amount of binder may be difficult to detect.)

(b) The crystal structure as seen by X-ray diffraction must be the same as that of the natural equivalent (several structures may be possible for a given chemical composition, e.g. graphite and diamond, rutile and anatase, etc.).

(c) Any sub-micro-structures, particularly those leading to characteristic optical effects, must be essentially identical (e.g. the diffraction grating arrangement in opal, perthitic structures, etc.).

(d) Finally, it is essential that the synthetic non-single-crystal material give to the naked eye the same overall appearance as the naturally occurring counterpart.

The only way in which this type of synthetic can be permitted to differ from the natural material is at the microscopic level, involving grain size and micro-structure, inclusions, and matrix. It is in this range where natural materials also show considerable variation from one locality to another, and even among specimens from a single locality. (Some restriction on the range of variation possible within this group may be necessary. However, such a limitation cannot be construed in any narrow sense as the following analogy would indicate: if a new natural rhodochrosite, jade, opal, etc. were to be found which shows a somewhat different grain structure from any found previously but is otherwise identical, one would nevertheless have no hesitation in applying the conventional designation. One would be justified in coining a new mineral name only if a really spectacular difference were to be noted, e.g., the difference between chalcedony and opal!)

These proposals are summarized in concise form in *Table I.* Any non-single crystal materials not meeting the four congruency requirements should then be designated an "artificial" or "imitation" product rather than a "synthetic" if this scheme is followed.

It should be noted that as a consequence of congruency by these four criteria, the usually measured gemological parameters will be approximately the same as in the natural material, as should be expected. This includes refractive index, specific gravity, color (but not necessarily fluoresence), hardness (but not necessarily fracture), behavior with respect to heat, acids, and so on. If any of these parameters are not approximately correct (e.g. see under Gilson Lapis below) then the case must be decided on its merits consistent with other current criteria.

Microscopic examination will therefore have to be the major technique for distinguishing synthetic from natural non-single-crystal gem materials.

Based on the published examinations and the above criteria one would draw the following conclusions:

Gilson Opal: appears to meet all the criteria(2,3) and is therefore a true "synthetic opal," by these standards.

Gilson Turquoise: an examination by W. F. Eppler(5) indicates that a cement is present which would negate the synthetic designation *if it is a foreign material.* In view of Mr. Gilson's statement that no binder is used, detailed investigation is required to confirm that this binder is merely finely divided turquoise, which would be consistent with the designation "synthetic," and not a foreign ingredient, which would make the material an "imitation." Such an investigation is under way in several laboratories.

Syntho Turquoise: only a partial report has appeared, which ". . . shows a chemical analysis but does not give any of the gemological information such as refractive index and specific gravity, nor does it give any indication of crystal structure or other factors which are usually the basis for determining a synthetic gem material."(6)

Gilson Lapis: the results of examinations have not yet appeared, although preliminary reports raise some questions: although giving essentially the same spectrochemical and X-ray diffraction data as natural lapis, a low S. G. and strong reaction with dilute hydrochloric acid seems inappropriate for lapis and the material may therefore not

TABLE I

Phenomena Observed at Different Magnification in Non-Single Crystal Materials

Level	Approximate Size Range	Examination Technique	Property Observed	Near Congruency* Necessary?
1.	Atom level	elemental analysis, optical spectroscopy	chemical composition, valence states, impurity sites, color centers	Yes
2.	0.1 nm to 10 nm	X-ray diffraction	crystal structure	Yes
3.	100 nm to 1000 nm	electron microscope	diffraction structure, etc.	Yes
4.	1000 nm to 1 mm	optical microscope, magnifying glass	grain structure, inclusions, matrix, etc.	No
5.	1mm to overall	naked eye	visible appearance	Yes

*The word "congruency" is used to describe the necessity for near complete agreement at a given level of examination of the synthetic with the natural material. In the absence of the necessary congruency, the material may only be designated "artificial" or "imitation."

be up to "synthetic" standards.(7,8)

Conclusions

A working definition for a non-single-crystal synthetic equivalent of a natural gem material is proposed on the basis that it must duplicate in essence the equivalent natural material at all levels of magnification (chemical analysis, X-ray structure, electron microscope structure if applicable, naked eye appearance) except in the micro-structure region (say 1000 nm to 1mm) where the grain structure, inclusions, etc., may vary over a wide range much in the same way as natural material does, but not necessarily restricted to the range observed in the natural material. This will generally, but not always, ensure that the usual gemological tests give values close to those of the equivalent natural material, as they of course should.

The distinction between the synthetic and the equivalent natural non-single-crystal gem materials thus has to be based predominantly on microscopic examination supplemented by specialized tests where indicated, as is also the case with the single crystal materials.

Acknowledgements

Helpful comments from Mr. B. W. Anderson, Mr. A. Farn, and the late Mr. R. Webster of Great Britain, Mr. R. Crowningshield and Mr. R. T. Liddicoat, Jr. of the Gemological Institute of America, and particularly from Dr. E. Gübelin of Switzerland are gratefully acknowledged.

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Gem Notes

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Madagascar Hambergite

The recent acquisition of a set of ten cut hambergite gems (0.7 to 2.9 cts.) adds yet another facet to the U.S. National Gem Collection at the Smithsonian Institution. This set of gems, ensconced in a specially designed case, was part of the mineral collection of Dr. Carl Bosch, and may be the largest set of Madagascar hambergites in existence. Since little has been written of gem hambergite, a mention of these gems seems warranted.

Hambergite, $Be_2(OH)(BO_3)$, from Anjanabanoana, Madagascar, was originally described by Lacroix (1909) and Goldschmidt and Müller (1910). The colorless orthorhombic crystals are found in sizes suitable for the cutting of small gems (1-3 cts.). Although the hardness (7-1/2) and refractive indices (1.553 - 1.628) might qualify it as a useful gem material, the lack of color, low dispersion, perfect cleavage on $\{010\}$, and extreme rarity of gem quality material have relegated it to the obscure status of a gem for museums and collectors of rare gems.

Measurements of the densities of these 10 gems, using a modified Berman-type balance and temperature correction yielded an average value of 2.35. The refractive indices, measured in sodium light, taking care to observe readings on many facets of each gem, were quite constant at $\alpha = 1.554$, $\beta = 1.588$ and $\gamma = 1.628$ (± 0.002). These are in good agreement with the values given by Palache *et al.* (1951): $\alpha = 1.5536$, $\beta = 1.5873$ and $\gamma = 1.6278$ which are averages of values obtained by Lacroix (1909) and Goldschmidt and Müller (1910). This obvious constancy in refractive indices could be due to all ten gems having been cut from the same crystal, or to a consistency in composition for the Madagascar material. The high birefringence of the hambergite causes an obvious doubling of the pavilion facets.

The gemologist might encounter some hambergite with lower refractive indices. Switzer *et al.* (1965) have shown that there is a substantial decrease in the refractive indices (to $\alpha = 1.543$, $\beta = 1.580$ and $\gamma = 1.617$) with isomorphous replacement of hydroxl by fluorine, as occurs in the hambergite from The Little Three mine in San Diego County, California.

Inclusions in the examined stones consisted of thin cigar-shaped libellae consisting of a liquid and a gas or vacuum bubble, and thin non-parallel fibrations which appear to be randomly oriented. These fibrations, or tubelets, appear similar to those seen in beryllonite but are thinner and interrupted along their length.

There was no discernible luminescence in either long- or short-wave ultraviolet or x-radiation. Exposure to intense CuKα x-radiation did not effect any color change in these gems.

The author is indebted to Dr. George Switzer for a critical reading of the manuscript.

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Inclusions in Sapphires From Yogo Gulch, Montana

Fine sapphires from this American locality have been known for some time. The deposit was originally discovered, according to legend, in 1895. For an excellent description of the area and overall production, the reader is referred to the work of Sinkankas (1959). The deposit was also noted by Crocker (1956).

Sapphire crystals from the Yogo Gulch mines are small and yield small accessory gems for the most part. The crystals are frequently flattened on {0001}, further limiting the size of a gem with proper proportions. The color of the material varies from blue-green to a rich cornflower-blue and violet-blue. Some of the sapphires, particularly the violet-blue varieties, do exhibit a dull violet-red fluorescence in long-wave ultraviolet radiation.

Inclusions in these crystals are not common, but are quite distinctive. These inclusions were separated from gem rough and X-rayed utilizing CuKα x-radiation and Gandolfi powder cameras.

The light brown partially resorbed crystals are spinel. The color of these spinel inclusions is consistent in several samples. The bright red resorbed blebs are rutile (TiO_2) and to be expected in a corundum colored by titanium. Both of these minerals have been previously noted as inclusions in sapphire and their occurrence here was anticipated. The long, very irregular inclusions of a brownish color give an X-ray pattern indicative of a member of the mica group, but the exact nature of the mica is unknown.

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Green Fluorite From New Hampshire

Long noted as a mineral deposit which has produced large etched octahedra of green fluorite, the Will Wise Mine in Westmoreland, Cheshire County, New Hampshire, has recently produced some fine facettable green fluorite. The deposit was reworked in the autumn in 1974.

The fluorite occurs as veins in quartz and is found as resorbed green octahedra up to 15 cm on an edge. The material is very evenly colored, without the unsightly color-zoning so common in fluorite. One gem of 70 carats and many gems of 30 carats have been cut from this flawless material. The color is a very uniform and pleasing pastel green. It fluoresces a weak violet in short-wave ultraviolet and a strong violet in long-wave. There is no phosphorescence.

Opalescent Sandstone From Louisiana

A sandstone comprised of quartz grains cemented together by precious opal was discovered in Central Western Louisiana by Mr. Gary Moore of 205 East Union Street, Leesville, Louisiana 71446, who sent the material to the Smithsonian Institution.

The sandstone is quite pure and is comprised almost entirely of clear quartz with some minor black and grey quartz.

The original sand is poorly sorted with an average grain size of 0.1 to 1.0 mm. The grains are quite angular and exhibit little rounding.

The cementing opal is, for the most part, bluish-violet or colorless. Pieces with a bluish-violet base color have a blue to violet opalescence. Pieces which have no base color show red and green opalescence. The opal is quite homogeneous and unfractured. Zones of oxidized iron run through the material and are quite porous and friable.

The material has some gem potential. Although the gemologist is conditioned to seeing opal in sizeable fragments, the pinpoint effect of hundreds of tiny precious opal fragments resting in interstices between colorless quartz grains is quite attractive. Polished surfaces are somewhat grainy due to the undercutting of the opal, but the well-colored material is quite tough and easily worked.

Inclusions in Brazilian Andalusite

Fine waterworn crystals of gemmy andalusite, Al_2SiO_5, have been known to occur in the area around Minas Novas, Minas Gerais, Brazil. The crystals occasionally attain dimensions of 8 x 8 x 30 mm but most are considerably smaller.

Isolated euhedrons *(Figures 1 and 2)* have been observed within the gems and have been identified. The inclusions were exposed by grinding and polishing rough gemmy fragments and then analyzed with an ARL-SEMQ

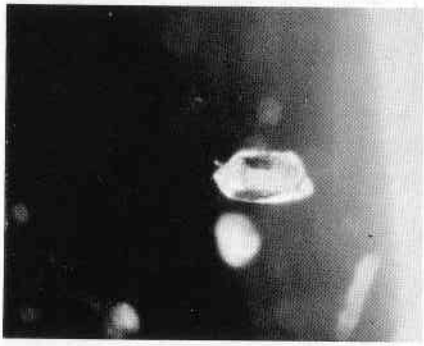

Figure 1.

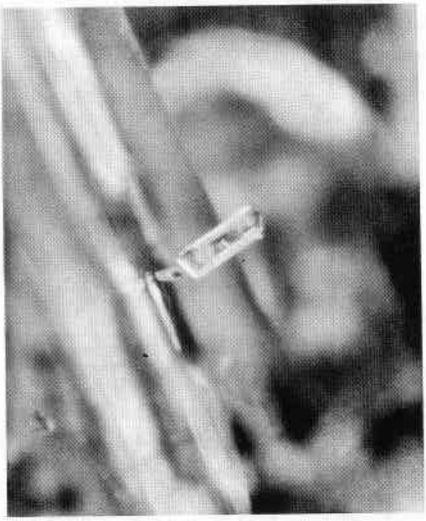

Figure 2.

Figure 3.

electron microprobe using an operating voltage of 15 kV and a beam current of 0.15 μA. A preliminary analysis indicated the inclusions were not a silicate and had an appreciable calcium content. Subsequent analyses indicated the inclusions were apatite, and a final analysis of this fluorapatite is given as *Table 1.* The included crystals are colorless, and easily observable. They appear to be somewhat resorbed or corroded and not all exhibit the rough hexagonal cross section so common in apatite inclusions in almandine. Also noted were some ladder-like arrangements of acicular tubelets *(Figure 3).*

TABLE I

ANALYSIS OF FLUORAPATITE IN ANDALUSITE

	Brazil	Theoretical
CaO	54.63	56.10
P_2O_5	42.15	40.30
F	3.46	3.60
TOTAL	100.24	100.00

Alexandrite From Lake Manyara, Tanzania*

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Introduction

The far-flung emerald deposit at Lake Manyara was mentioned by THURM (1972) and described more precisely by BANK (1974), by the present author (1973 & 1974), and more recently by MWAKISUNGA (1976). BANK (1974) reported a small piece of mother rock, on which, associated with feldspar and mica, alexandrite, emerald and ruby occurred, i.e. three extremely rare gemstones all of which owe their color to an admixture of chromium. From the mineral paragenesis he also surmised that the Manyara deposit was analogous to the famous occurrence of beryllium minerals at Tokowaya in the Ural mountains (described by FERSMANN, 1929), where the same gem minerals are found in close association. Towards the end of his paper "The Emerald Deposit at Lake Manyara, Tanzania" *(Lap. Journ.*, May, 1974) the writer refers to several other gems such as apatite, garnet, spinel and chrysoberyl, of which the detection of alexandrite is the most intriguing *(Figure 1)*.

Since then further investigation could be carried out on the alexandrite from Lake Manyara in its quality as a crystal and precious stone as well as on its mode of occurrence, justifying a special publication on this fascinating gem.

The mother rock of the Manyara alexandrite has meanwhile been carefully analyzed. Each individual mineral component was X-rayed. The red grains which can easily be seen in the rock all proved to be chondrodite. In thin sections they are yellow. Contrary to the previous information (GÜBELIN, 1974), garnet seems to be rather scarce or then very sparsely dispersed; at least it has not been encountered in a more recent and more thorough analysis. The mineral components which were definitely identified are in sequence of frequency: actinolite, enstatite, fayalitic olivine, chondrodite, chrysotile and pleonaste. Consequently the name *actinolite-schist* is proposed. In contrast to the former conjecture it is not a peridotite, be-

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*Manuscript received July, 1976.

cause it does not contain enough olivine. The association of actinolite and alexandrite indicate a metamorphic rock, hence: it is a schist.

According to BANK (1976) and OKRUSCH (1971) the possibilities of formation are more limited for the alexandrite than for ordinary chrysoberyl. Alexandrite owes its great rarity to the fact that the geochemical conditions are highly accidental because beryllium and chromium do not normally occur within the same rock suites. Beryllium is concentrated in the pegmatitic and pneumatolytic phases which themselves are devoid of chromium. The latter, on the other hand, is more frequently present in mafic rocks. Therefore the formation of alexandrite may only be expected in places where the ingredients of these rocks can meet, that is to say, where the pegmatitic and pneumatolytic residual phases carry the beryllium into contact with the chromium in such ultramafic rocks as actinolite, peridotite, pyroxenite, and their metamorphic derivates (particularly serpentinites). These were the favorable conditions found in the deposit at Lake Manyara manifesting close similarity to the deposit of Tokowaya as well as of the Girdlestone Farm at Novello, Rhodesia (BANK, 1964).

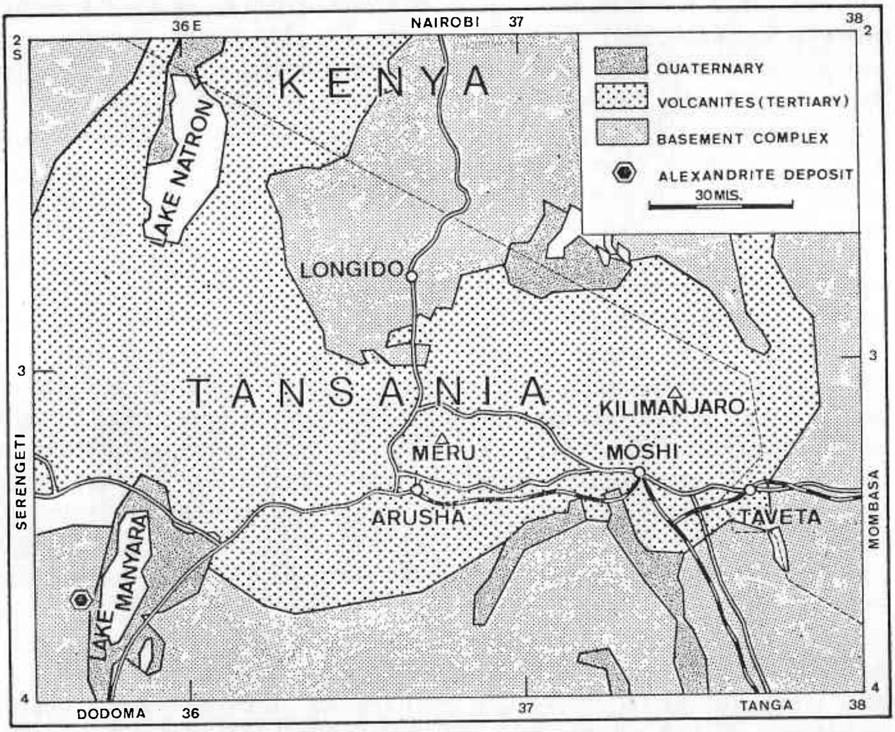

Figure 1. This simplified geological map of northern Tanzania shows the situation of the alexandrite deposit at Lake Manyara, where alexandrite occurs together with some other chromium-bearing gem minerals.

External Appearance

Considering this geochemical similarity of occurrence, the author was not astonished to observe that all the Manyara alexandrites which he has seen so far looked puzzlingly similar to those from Tokowaya and from Novello. In daylight they all displayed the same exquisite bluish-green hue with a remarkable change of color to pronounced raspberry red in incandescent light.

With a few exceptions, most of the specimens seen or investigated displayed those characteristic formations of three crystals intersecting each other, which yield the well known trillings. The habit of the individual crystal was mainly determined by the prism. Some were just broken-off crystals and others merely fragments. Some were absolutely clear and of exceptional gem quality and when cut would have resulted in high grade precious gems up to 5 carats. However, many crystals and fragments were intensely etched and marred by too many mineral and liquid inclusions as well as cracks.

Chemical Composition

By means of the electron microprobe, an analysis was carried out on two crystals, the results of which are communicated in *Table I.* The calculation program was started off in assuming Be0 to amount to 20% (theoretical value), because due to the low atomic number (Z=4) of Be the microprobe does not register this element. In the alexandrite from Novello Be0 amounts to 19% and thus very closely approaches the theoretical value.

TABLE I

	Alexandrite	
	No. 1	No. 2
	%	%
Be 0	20	20
$A1_2 0_3$	77	78
$Cr_2 0_3$	0.25	0.20
$Fe_2 0_3$	0.7	0.7
$V_2 0_3$	—	—
$Ti\,0_2$	0.12	0.08
$Si\,0_2$	0.13	0.4

The percentage of the trace elements present reveals that chromium and iron are responsible for the color. Vanadium seems to be entirely absent. However, as will be explained further down, the striking alexandrite effect is caused solely by chromium, while iron is of secondary importance and mainly acting as an inhibitor (see later).

Physical Properties

The physical properties do not manifest any peculiarities – the one exception of course being the chameleon color change – and may therefore be mentioned hereafter merely for statistic purposes.

Density:

The frequency medium of numerous measurements taken results in

$$d = 3.71 g/cm^3 \text{ at } 4°C$$

R. I. and birefringence:

Again the medium data of numerous readings are:

$$n_\alpha = 1.745$$
$$n_\beta = 1.750$$
$$n_\gamma = 1.754$$

whereas the birefringence varied from 0.008 to 0.010. The optical character is of course negative.

Trichroism:

This property is very distinct and depending upon the three main optical directions produces the colors shown in Table II.

TABLE II

Optical direction	Daylight	Incandescent light
n_α	reddish	carmine red
n_β	yellow green	orange red
n_γ	blue green	greenish

For this accentuated pleochroism the transition-metal chromium alone is responsible.

Absorption:

The absorption spectrum is a typical chromium spectrum whose spectral image is characterized by two prominent bands and marked by several individual absorption lines in the red region *(Figure 2)*. The two bands are designated A and B. They range astride the critical wavelengths of 580 nm and 415 nm. If they trespass these values the alexandrite is green, otherwise it is red. The A band extends from approximately 550 nm to roughly 610 nm and culminates at 571.4 nm. It is accompanied on its long-wave side by the telltale chromium lines at 680, 678, 665, 655, 649, and 645 nm which result from spin-forbidden transitions. The B band is somewhat narrower ranging from 395 nm to 443 nm with its peak at 415 nm. It is chaperoned by four satellite lines at 470 nm and 465 nm on the long-wave side and at 385 nm and 375 nm on the short-wave side. The latter two lines are caused by trivalent iron. The narrow lines in the red and in the short-wave section become more conspicuous when a polaroid is used. The absorption minimum is situated at 494 nm and very closely corresponds to the intensity maximum of the solar spectrum in the green (at 500 nm) imparting the alexandrite a green color in daylight. Incandescent light excels in a predominance of longer wavelengths which are transmitted by the alexandrite's second absorption minimum at 725 nm thus conferring the gem a red appearance.

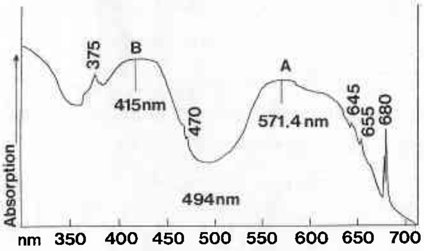

Figure 2. Absorption spectrum of the alexandrite from Lake Manyara in unpolarized light ranging from 325 nm to 700 nm at room temperature.

The breadth of band A suggests that it must be provoked by two overlapping absorption spectra of two differing color centers. This becomes evident when polarized light is applied. In unpolarized light the breadth of the A band complex is produced by the superposition of two bands with maxima slightly displaced against each other. The polarized absorption parallel to the z-direction ($A_{\parallel z}$) culminating in longer wavelengths at 571.4 nm is responsible for the green color, while the polarized absorption perpendicular to the z-direction ($A_{\perp z}$) sweeps to a maximum among shorter wavelengths at 551 nm and induces red coloration *(Figure 3)*. This change of color is called "Alexandrite Effect."

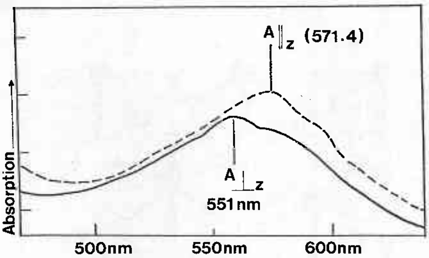

Figure 3. Polarized absorption spectra of band A parallel and perpendicular to the z-direction (Hassan et al., 1974).

The alexandrite-effect:

This phenomenon is an exceptionally well-pronounced specialty of the Manyara alexandrite, for the color change from green to red and vice versa is very definite and so complete, that the beholder cannot help wondering about the nature of its cause. The attempt to explain essence and cause of this unusual feature was undertaken in numerous publications. For a very long time it was simply connected with the mineral's strong trichroism and it was merely regarded as an effect provoked solely by the different spectral composition of daylight and incandescent light. None of these older and rather one-sided explanations of the very complex process were convincing. Nowadays the contexts are better understood, and hence the interpretation of the causal conditions acquire a new significance. Today's apprehension is based upon a much more profound comprehension of the intrinsic structure of the alexandrite. While POOL (1964) and partly also WHITE *et al.* (1974) attributed the color change to a rather psychological effect of the human eye and brain than to the inherent structure and properties of the alexandrite, FARREL *et al.* (1963 & 1965), NEWNHAM *et al.* (1964), WHITE *et al.* (1967), and HASSAN *et al.* (1974) investigated the structural background of the question. Their results provide an instructive insight into the mechanisms which take place, when light falls into an alexandrite. The author feels that his present account of the Manyara alexandrite might be a welcome opportunity of making some integrating reflections on these more recent studies.

Alexandrite is isostructural with olivine and sinhalite, i.e. it has an identical structure as these two minerals (FARRELL, NEWNHAM, 1965 and STRUNZ 1968). The structure of the alexandrite is determined by a dense packing of the relatively large oxygen ions in a hexagonal arrangement. Within this lattice the tetrahedral interstices are occupied by Be while the Al partly sits inside the coordination octahedra. This results in a combination of tetrahedral BeO_4 groups and octahedral AlO_4 groups. Numerous chemical analyses as well as recent refinements of the structure of chrysoberyl have lead to the conviction that the transition element chromium (Cr^{3+}) (NASSAU, 1975) is the chromophorous ion, yet not vanadium. The aforementioned chemical analyses did not furnish any alibi for the existence of vanadium.

It is not the mere presence of Cr^{3+} ions within the structure of the alexandrite which is responsible for the color change but rather their array in the crystal lattice. Within the structure of the chrysoberyl there are various lattice sites with octahedral symmetry, which are held by Cr^{3+} instead of Al^{3+} ions. However, normally only two of them are being claimed by Cr^{3+}. The refinement of the structure of chrysoberyl by FARRELL *et al.* (1963) disclosed that these two octahedral lattice sites preferred by the Cr^{3+} ions differ with regard to size – not very much but significantly enough to produce the color change *(Figure 4)*. Half of the $A1^{3+}$ ions are perched in coordination polyhedra with inversion symmetry = $A1^{3+}$ (1) and the other on mirror plane positions = $A1^{3+}$ (2). The average distances to the nearest oxygen ions measure 1.890Å for the $A1^{3+}$ (1) sites and 1.934Å for the $A1^{3+}$ (2) sites (the mean value between the two (= 1.914Å) happens to be almost identical to the distance A1 – 0 in alpha-corundum). If trivalent cations replace others they favor one place to another depending upon symmetry and size.

The Cr^{3+} ion (ϕ = 0.615Å) is slightly larger than the $A1^{3+}$ion (ϕ = 0.530Å) and hence prefers the more spacious $A1^{3+}$ (2) sites. Yet, if introduced under pressure and high temperature some Cr^{3+} ions may accommodate themselves on narrower $A1^{3+}$ (1) sites. The dispersion of the Cr^{3+} ions onto transition-metal sites differing in size and symmetry within the crystal field cannot happen without drastic influence on the color properties of the alexandrite, especially so because the absorption spectrum of the alexandrite consist of two individual spectra of the Cr^{3+} (1) and the Cr^{3+} (2) color centers overlapping one another in the A band.

The intrinsic cause of the chameleon effect of the Cr^{3+} ions on their two different lattice sites is their different distance to the oxygen ions. On the smaller octahedral sites (1) of the alexandrite lattice the Cr – 0 distance is shorter, the Cr^{3+} ion finds itself in a compressed position and thus promotes the transposition of the absorption maximum from 571.4 nm (assigned to the more numerous Cr^{3+} (2) centers) to 551 nm (ascribed to the less populated Cr^{3+} (1) centers). The denser the population of the Cr^{3+} (1) centers the greater the spectral dislocation and hence the more intense the red coloration. This condition concurs exactly with observations made in gemstones of the ruby type (ruby, spinel) where the red color also results from compressed space at the Cr^{3+} lattice sites.

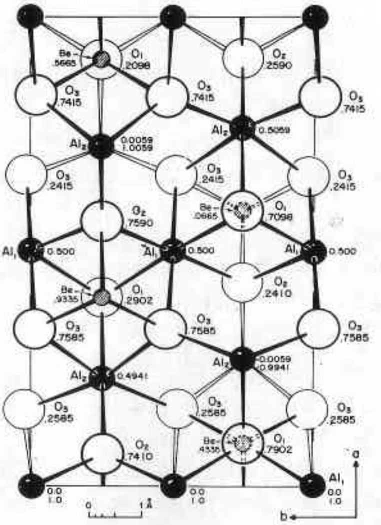

Figure 4. The refined structure of chrysoberyl projected on (001). Heights of the atoms are expressed in cell fractions (Farrell et al., 1963).

The investigation of the two different coordination sites elucidates that the Cr^{3+} (1)/Cr^{3+} (2) population ratio decides the color itself as well as the intensity of the color change. The greater the Cr^{3+} (1)/Cr^{3+} (2) ratio the more pronounced is the color of the alexandrite in daylight as well as in incandescent light. Due to the denser population of the Cr^{3+} (2) centers, but also because of the polarizing influence of the adjacent Be ion acting as a central ion, the conditions for the Cr^{3+} ions are very similar as in the Cr^{3+} doped silicates of the emerald type (emerald, demantoid, hiddenite etc.) as long as the alexandrite is impelled by the intensity as well as by the spectral character of the light in which the gem is being viewed. Daylight or any other white illumination with an identical distribution of wavelengths interacts with both Cr^{3+} centers imparting the green as well as the red hue in dependence on the Cr^{3+} (1)/Cr^{3+} (2) population ratio. In incandescent light on the other hand, which is relatively poor in short (blue, high energy) wavelengths but rich in long (red, low energy) wavelengths the alexandrite appears red, because the strong band B with its culmination at 415 nm absorbs the majority of the scanty short wavelengths up to about 443 nm thus blotting out almost all of the blue region. The residual light is being controlled by the broad A band complex at 571.4 nm, i.e. it absorbs mainly in the green. On account of its complex nature the band A also blacks out parts of the bluish green and of the yellowish green between 625.5–516.6 nm. This extensive absorption of almost all the shorter wavelengths results in the complementary purplish red color: the characteristic raspberry red of the alexandrite in incandescent light. From this it may be concluded that the smaller the Cr^{3+} (1)/Cr^{3+} (2) population ratio, i.e. the more concentrated the Cr^{3+} centers, the more more important the absorption effect of band A.

After all these reflections it must, however, be considered that the A band complex is neither the sole nor the principal factor for the quality of the color change but rather the correlation of both bands A and B. Above all the absorbence of band B, that is to say, the sum of the absorbing centers in band B must apparently reach a certain critical value to completely absorb the shorter blue and green wavelengths which are in any way quite scanty in incandescent light.

In summarizing it may be recapitulated that the alexandrite effect materializes in that chromoforous chromium atoms in the state as Cr^{3+} ions occupy octahedral coordination sites in the crystal which are otherwise held by Al^{3+} ions. These sites differ in size; some of them are of normal largeness, others are constricted. The color change is provoked when in comparison to the Cr^{3+} (2) ions on regular spacious sites sufficient Cr^{3+} ions can slip onto the compressed sites Cr^{3+} (1), that is when a certain critical value of the Cr^{3+} (1)/Cr^{3+} (2) ratio is reached. The distribution of the Cr^{3+} ions onto the different transition-metal sites certainly depends upon the growth conditions of the alexandrite; in order to force enough Cr^{3+} ions onto the compressed lattice sites either very high temperatures and low pressures or low temperatures with high pressures are necessary. Such extreme conditions happened to occur very rarely in the birth chambers of the alexandrite (not to mention the extreme rarity of the chromium) so that natural specimens with a good color change are purely accidental!

The foregone explanation may be preliminary and perhaps it is an oversimplification but it had to be adapted to different grades of scientific education of the readers. To understand the miracle does not deny its existence!

In the above considerations the iron was not mentioned because it exerts no influence on the alexandrite effect. If present in sufficient quantity as to affect the color by imposing its absorption upon that of the chromium, Fe^{3+} may impair the purity of the hues in day and incandescent light. Thus iron is often responsible for the brownish tint of some alexandrites from Sri Lanka. In addition, iron acts as a powerful inhibitor in that it prevents the alexandrite from emitting luminescence.

Behavior under short wave radiation:

Table 1 reveals that iron participates with 0.7% of the total amount of chemical elements in the composition of the Manyara alexandrite. This is approximately three times that of chromium. Yet, it appears to be insufficient for completely suppressing the luminescence, for contrary to alexandrites from other sources which are normally inert to shortwave illumination, the Manyara alexandrite glows faintly in UV light but does not respond to X-rays. This distinct luminescence serves as a welcome virtue of distinction from (a) genuine alexandrites from other deposits and (b) both types of synthetic alexandrite presently known. *Table III* offers a clear survey of the behaviour under

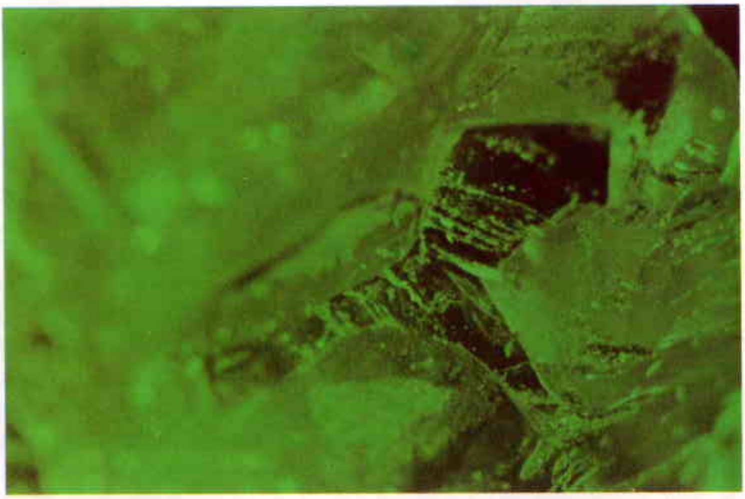

Figure 5. A "parcel" of dark blackish brown biotite flakes (32x).

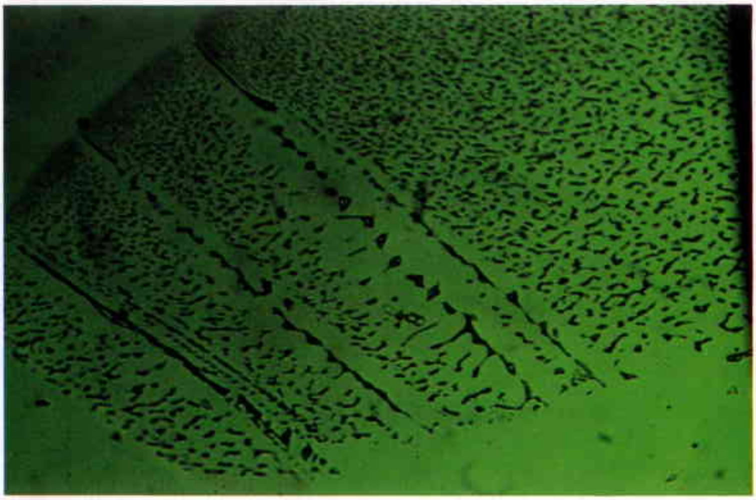

Figure 6. A section of a "fingerprint" inclusion, i.e. a partially healed fracture marked by a system of intercommunicating channels of residual liquid (40x).

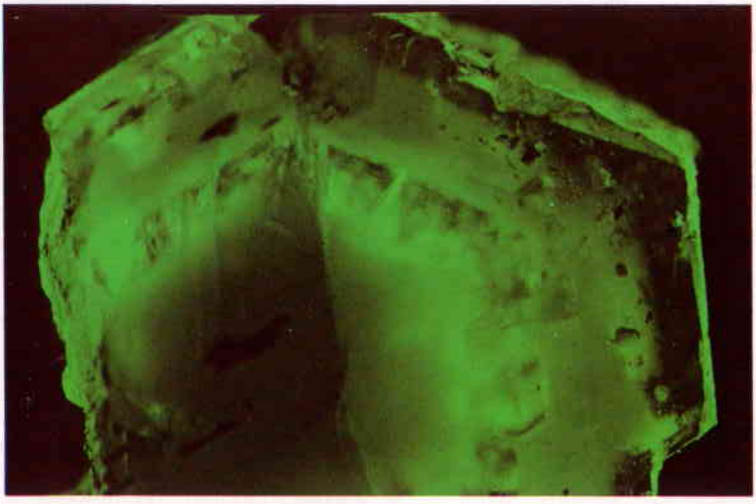

Figure 7. Pseudohexagonal arrangement of color zones alternatively dark and pale green (32x).

short wave radiation of the various genuine and synthetic alexandrites.

The immediate deduction to be assumed from the positive response of the Manyara alexandrite to UV radiation would be that it contains less iron than the alexandrites from the other localities. This observation is also confirmed by the high degree of transmission of the Manyara alexandrite in long wave UV light. Under short wave UV radiation all natural alexandrites as well as flux grown synthetic alexandrites are opaque, while the Czochralski pulled synthetic alexandrites are transparent.

Inclusions

The internal paragenesis of the Manyara alexandrite is typically moulded by its mode of formation and the component minerals of the mother rock. Therefore it is not surprising to encounter some minerals of the external mineral association such as actinolite and biotite. The actinolite occurs in the form of fibres or stalks either discretely distributed or then concentrated in irregular masses or as slightly divergent sheaves. Close and dense arrangement of fine actinolite fibres may cause chatoyancy in some specimens. The biotite also either assembles in the well known form of so-called "books" or hovers as individual flakes in the body of its host gem *(Figure 5)*. In one small alexandrite an individual, well formed crystal of apatite was observed.

The majority of those Manyara alexandrites excelling in inclusions contain mainly fanciful liquid inclusions which traverse the whole crystal

TABLE III

ALEXANDRITE FROM:	LUMINESCENCE			Transparency in long wave UV light 365 nm	Opacity in short wave UV light 254 nm
	long wave UV light 365 nm	short wave UV light 254 nm	X-Rays		
Lake Manyara Tanzania	medium dull red	weak yellowish	inert	good	opaque
Novello Rhodesia	inert	inert	inert	medium	opaque
Ratnapura Sri Lanka	inert	inert	inert	medium	opaque
Tokowaya Ural Mts. Siberia	inert	inert	inert	medium	opaque
Synthetic Alexandrite flux grown (Patterson)	distinct orange red	very weak reddish	inert	very good	opaque
Czochralski pulled (Dr. Morris)	strong red	strong red	inert	excellent	transparent

at random and display the characteristic appearance of secondary inclusions or partly healed fractures. Their pattern is decided by liquid filled tubes or channels which are more loosely disseminated or then by dense and irregular arrangement of ramifying veins and capillaries *(Figure 6)*. No small number of Manyara alexandrites display periodic growth either by a layered distribution of the color or a pseudohexagonal zonal accord of light and darker shades *(Figure 7)*.

Unfortunately the supply of this magnificent precious stone from Lake Manyara will presumably always remain rather uncertain or at least very irregular and in any case too scarce as to satisfy a receptive market, and the exhorbitant prices which were asked from the beginning did not help to promote the gem's popularity.

Acknowledgements:

The author wishes to extend his gratitude to Prof. Dr. M. Weibel for the chem. analyses of the native rocks and the alexandrite crystals as well as to G. Brunner for running the absorption curves (both at the Institute of Crystallography and Petrology in Zurich) and to C.A. Schiffmann (Gem. Lab. Gübelin, Lucerne) for his assistance with the gemological experiments.

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Developments and Highlights at **GIA**'s Lab in Santa Monica

By RICHARD T. LIDDICOAT, JR.

Assembled Hornbill

Recently, a snuff bottle reputed to be hornbill came in for identification. It is shown in two views in *Figures 1 and 2.* The body of the material is light brown and the wings of what apparently represent a cicada are a dark brownish-red, typical of hornbill casque. Chuck Fryer's examination under magnification with strong overhead light revealed spherical bubbles underneath the brownish-red wings, proving to his satisfaction that the wings had been cemented to the body. *Figure 3*, taken at 25x, shows a portion of the wing (darkest area) and the zone where the top of the body has been built up from some type of


Figure 1.

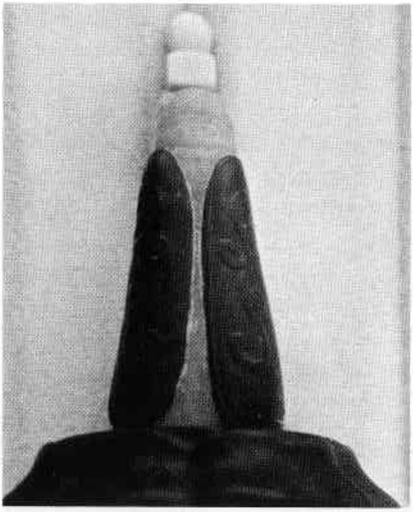


Figure 2.

Figure 3.

casein cement (the area that is slightly darker than the under body). This cement area is full of spherical bubbles. *Figure 4* is the top view of the bottle (taken at 15x) showing that the tip of the beak was probably cut off. The area was filled with casein cement and apparently then drilled for the stopper. The cement portion is the darker area shown in *Figure 4.* The bottle appears to have been assembled partially from hornbill.

Rare Spodumene Cat's-Eye

One of the most interesting stones encountered recently in the Santa Monica Laboratory was a loose stone that was semitranslucent and that had a reasonably good eye. The surprising thing about it was that it turned out to be spodumene. We had never seen a spodumene that even closely resembled chatoyant material. To make doubly certain that the identification was correct, we resorted to X-ray diffraction to prove it. This unusual stone is shown in *Figure 5.*

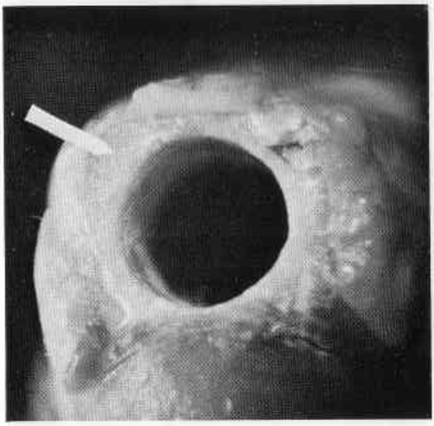


Figure 4.

African Alexandrites?

We received a group of alexandrites which in their reactions to ultraviolet, both long and short wave, seemed very similar to the reactions we expect from synthetic alexandrite. The transparency to the shortwave test suggested that they were synthetic, but the inclusions looked natural. Tested on the Luminoscope, they showed a distinctly different reaction from synthetic material. They were identified as natural, without any question. What it proves is that the transparency to shortwave testing is no

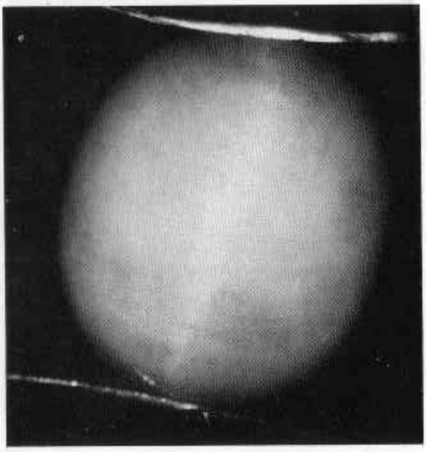


Figure 5.

longer viable. One stone glowed red to longwave, and yellow to yellow-green to shortwave ultraviolet; it was color-zoned red and green in its fluorescence to the Luminoscope. Some of the inclusions are shown in *Figures 6* through *10. Figures 9* and *10* show two views of a very fluxlike inclusion that was obvious in one of the stones. Hexagonal appearing inclusions with stress cracks are seen in *Figure 6.*

Figure 6.

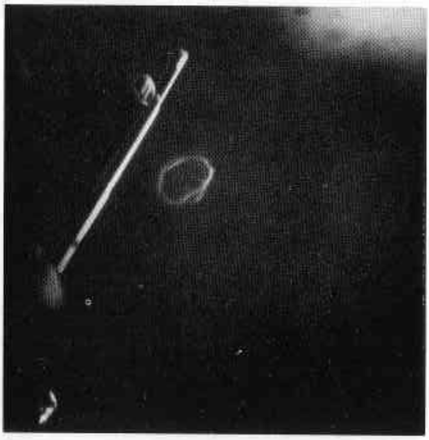


Figure 7.

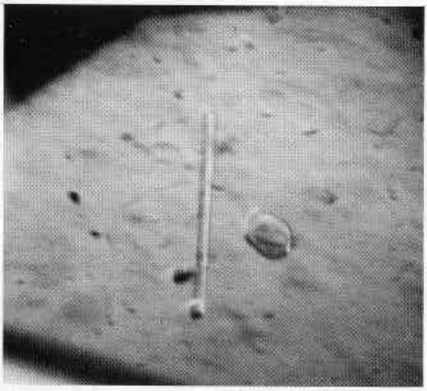


Figure 8.

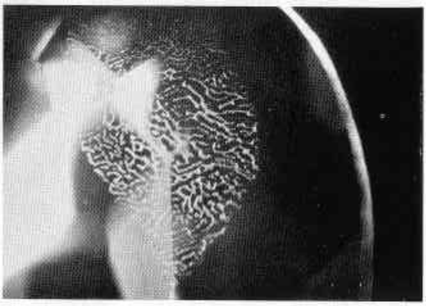


Figure 9.

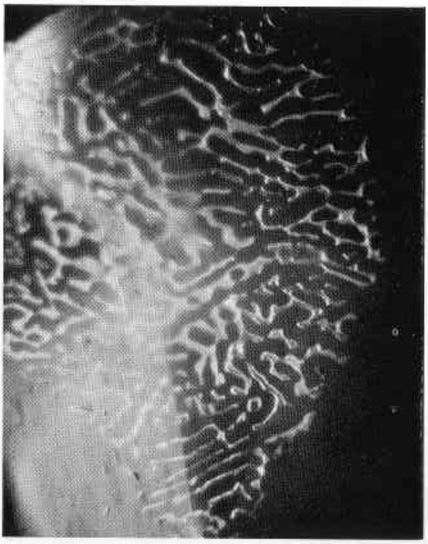


Figure 10.

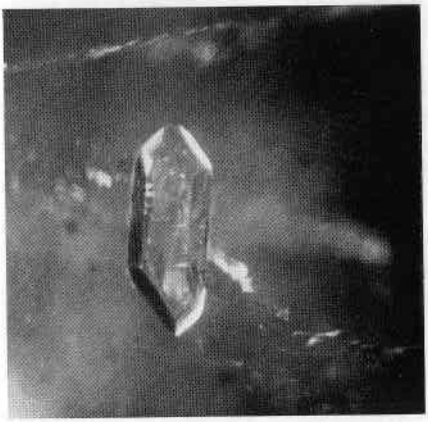


Figure 11.

Figure 7 shows transparent light green included crystals at 126x. Another view of the same inclusions is shown in *Figure 8. Figures 9* and *10*, the first taken at 48x and the second at 126x, illustrate the fluxlike inclusions.

Interesting Quartz? Inclusion in Emerald

On another occasion, we encountered an interesting crystal inclusion in a natural emerald from Colombia. It appeared to be a doubly terminated quartz crystal. The inclusion is shown in *Figure 11*, taken at 63x.

A Knotty Problem In a Diamond

We received for identification a ring set with a greenish-brown diamond weighing slightly over 3 carats. That the color was natural was easily proved to our satisfaction. Surprisingly, there was no X-ray fluorescence. *Figure 12* is a picture of the diamond table showing that it was really full of knots. This is the most knot-filled diamond in memory.

Botryoidal Opal

In *Figure 13*, we see an inclusion in a botryoidal opal. The photograph is taken at 63x. We do not know the nature of the inclusion. In the Spring 1967 issue of *Gems & Gemology* we showed another picture of the botryoidal structure in a type of opal. This unusual opal appears under magnification to have sections of curved striae *(Figure 14).* We have seen this only in

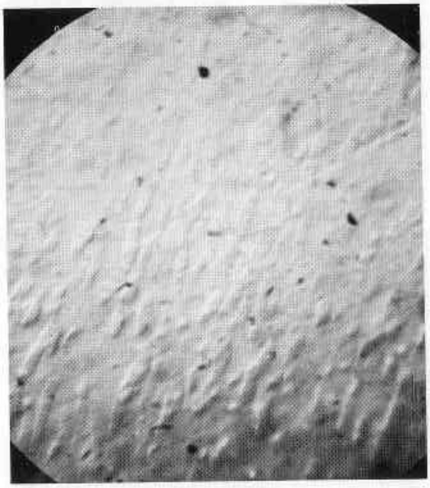


Figure 12.

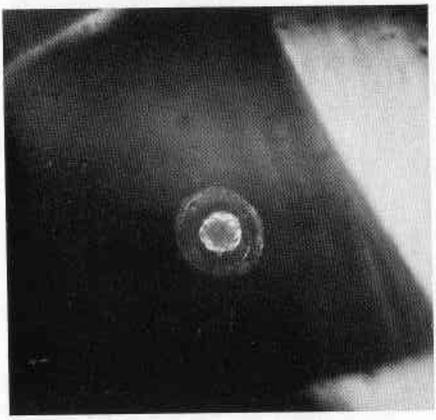


Figure 13.

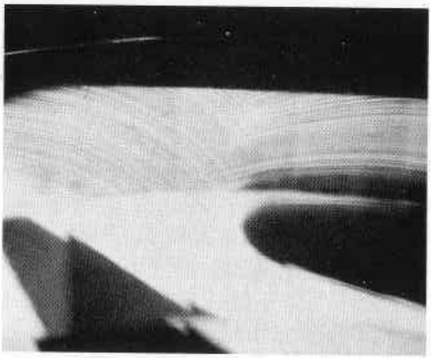


Figure 14.

opal from one locality in northern Mexico. The surface of the rough opal shown in *Figure 15* is botryoidal in nature. *Figure 16* shows one more inclusion in botryoidal opal. This resembles a crablike nebula.

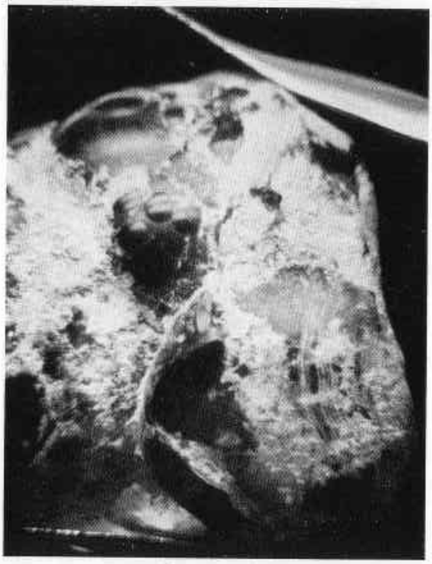


Figure 15.

Unknown Inclusion In Synthetic Emerald

We received a synthetic emerald for identification which had in it a large crystal with an appearance similar to that of a spodumene crystal. We never heard of a spodumene inclusion in synthetic emerald. Even though the flux used probably contains some lithium, it seems an unlikely possibility. See *Figure 17.*

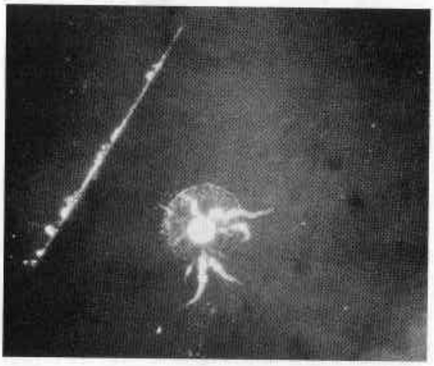


Figure 16.

Devitrifying Glass

A spherical gas bubble in glass about which devitrification is taking place and distinct zones of crystallization

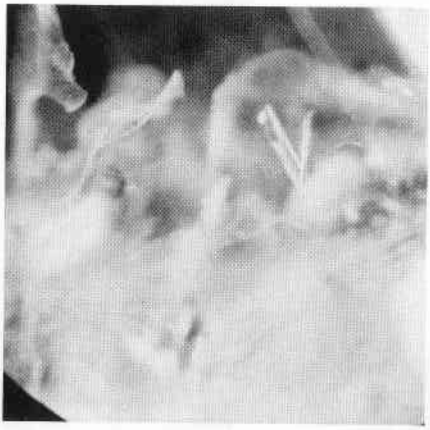


Figure 17.

Figure 18.

are visible is shown in *Figure 18.* To have the crystallization appear to nucleate around the bubble is new in our experience.

Acknowledgements

We wish to express our sincere appreciation for the following gifts:

To *Afghan Malaya Trading Company*, Singapore, for a collection of rough and cut emeralds.

To *Joe Best*, G.G., F.G.A., of Gilchrist Jewelers, Santa Barbara, California, for a specimen of rough Linde synthetic emerald.

To *Loreen Haas*, G.G., Crown Gems, Sherman Oaks, California, for a selection of gem materials including a large assortment of natural opals, amethyst and coral.

To *Fred Howlett*, Diamond Correspondence Course student from San Antonio, Texas, for six partially faceted Montana sapphires.

To *William F. Larsen*, Pala Properties, Fallbrook, California, for donating an extensive collection of fine quality rare and unusual rough gem materials including herderite and chrome diopside from Brazil, phosphophyllite and vivianite from Bolivia, boleite from Mexico, cuprite from South-West Africa, red beryl from Utah and numerous other specimens.

To *Mr. and Mrs. V. R. Martin* of Glendale, California, for a generous gift of a large fine quality transparent oval brilliant cut blue topaz weighing 213.39 carats and measuring 42x33x21 mm. for our new gemstone museum collection.

To *Digby Matheson*, GIA Santa Monica Resident student, of Crystal Pty., Ltd., Sydney, Australia, for a generous selection of faceted stones from Burma including red and blue spinels, peridots and pink sapphires.

To *R. Wm. Sampson*, GIA Correspondence student, of Ingeborg's Goldsmiths, Inc., Aiea, Hawaii, for a very useful assortment of miscellaneous cut and tumbled stones.

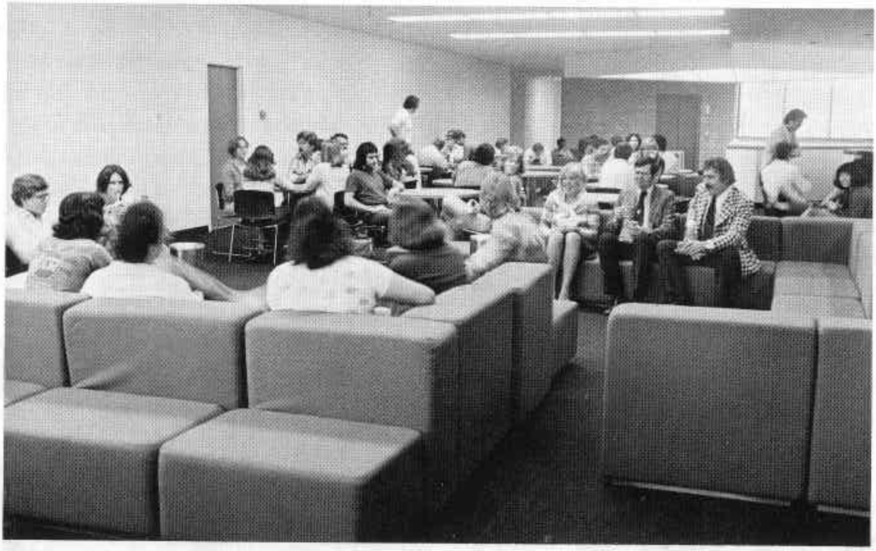
To *Horace Simon,* G.G. Dales Jewelers, Shreveport, Louisiana, for a collection of opal cabochons, broken diamonds and natural rubies.

To *Stephen L. Singer*, Gemologist, of Finlay Departments, New York City, for a collection of 128 gold-plated sterling rings set with a variety of gemstones including opal, ruby, pyrope, turquoise, amethyst, sapphire, jade and emerald.

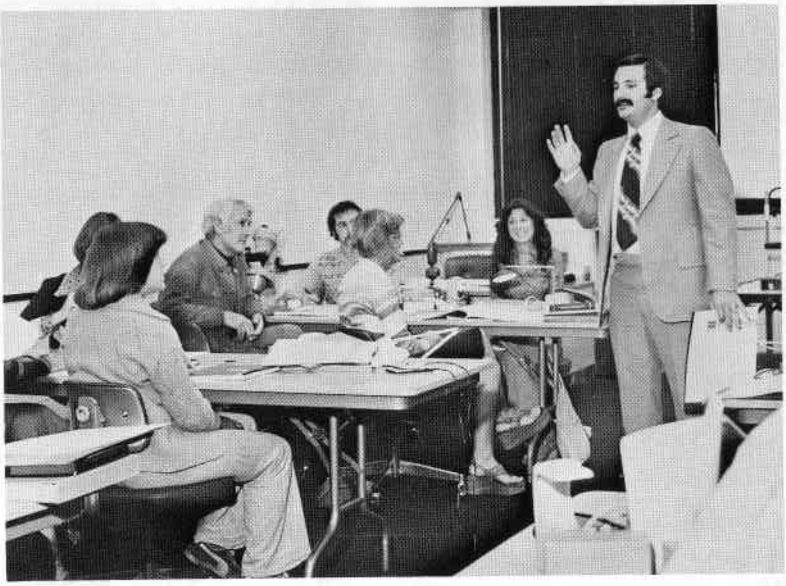
To *Gerald Winer*, GIA Diamond Correspondence student, Brentwood, Tennessee, for a specimen each of emerald, sapphire and ruby.

Inside and Outside of GIA's New Headquarters in Santa Monica

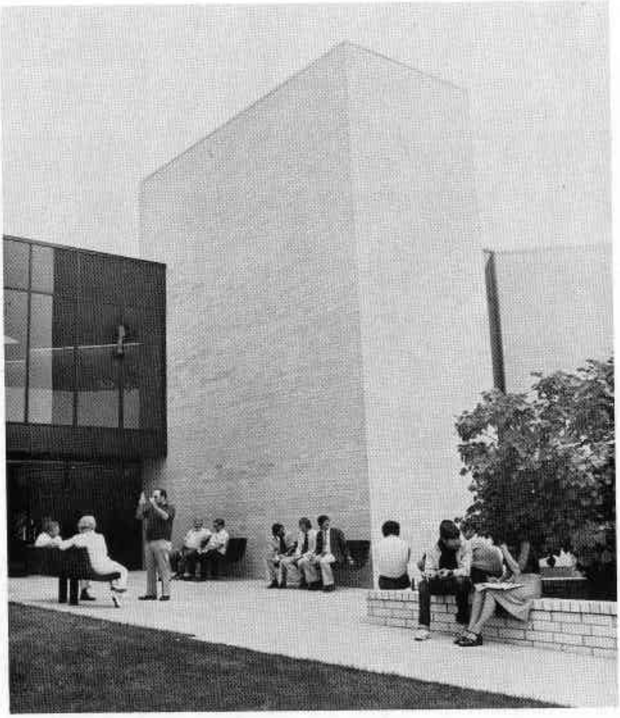
New GIA Headquarters Building, Santa Monica, California



A view of the popular student lounge on the second floor of the Institute.



Popular one-week class in Diamond Appraising is held in Santa Monica. Classes are limited in size to give students close personal attention.



Students and staff enjoy relaxing in the southern California sunshine at the side entrance during break time.

Developments and Highlights at GIA's Lab in New York

By ROBERT CROWNINGSHIELD

Diamonds-Diamonds-Diamonds

The overwhelming majority of the hundreds of diamonds examined each week in GIA laboratories are relatively free of flaws so that any stone with photographable inclusions is a rarity. A few such stones have been seen in New York recently among which is the laser-damaged 10-carat pear-shaped stone shown in *Figure 1*. Branching from the main drill hole is an erratic pattern of subsidiary holes shown in *Figure 2*. Without doubt the damage contributed to the I_1 grade. We have seen small cleavages radiating from a laser hole before but not this type of

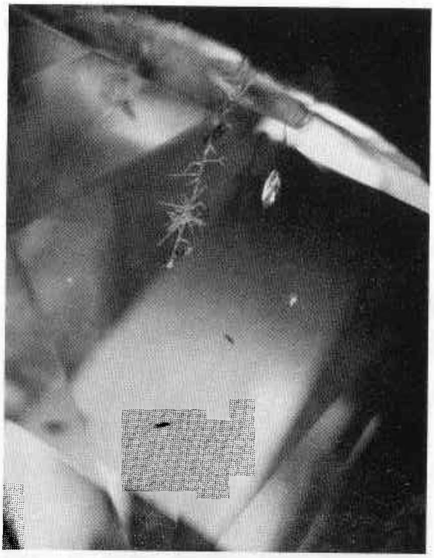


Figure 1.

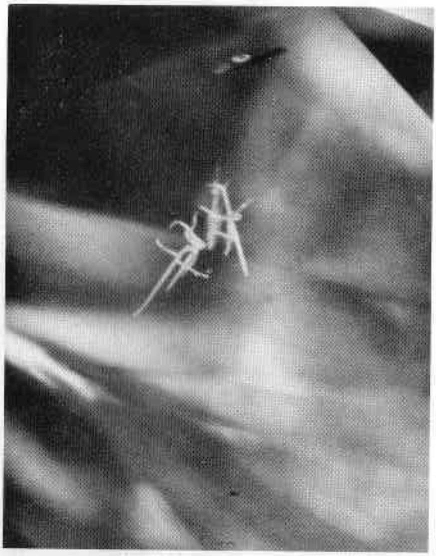


Figure 2.

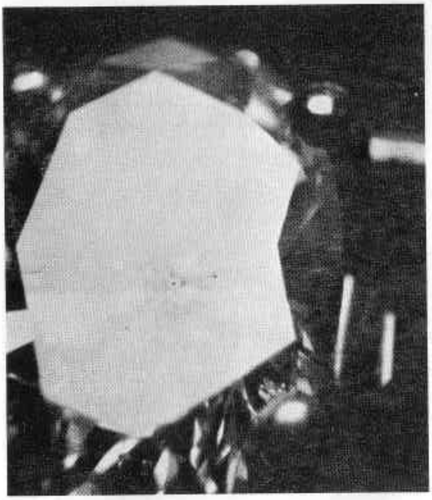


Figure 3.

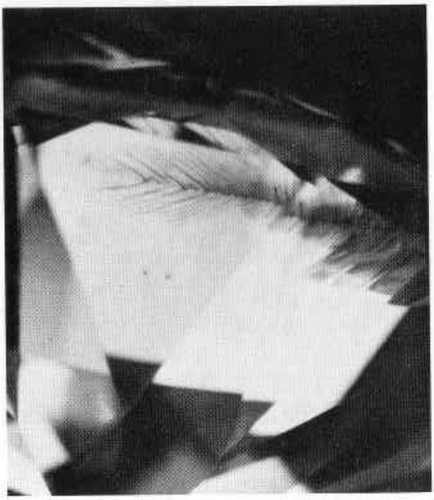


Figure 4.

damage. We have no history of the stone as to when the technique was applied. In *Figure 3* we see a lasered stone of which some history is known. It was presumably done early in the application of the process and the surface of the table has become brown. The owner repeatedly tried to clean the stone not knowing that it was a laser drilled stone and that the technique had resulted in a burned surface. Although we advised that the surface could be polished and the discoloration removed, there was a risk of creating pronounced drag lines leading away from the hole. This fact we failed to mention when describing the diamond with 17 laser drill holes in the Winter 1975-1976 issue of *Gems & Gemology*. (*Figure 4*, page 124)

Unusual "Fish Skeleton" Cleavage

Another rather weird inclusion photographed recently appeared to be an excellent rendition of a fish skeleton. It was actually a cleavage on the table which reflected the striations along the cleavage, giving the whole a remarkable symmetry. See *Figure 4.*

A Simple Test

The "tilt test" for getting some idea of the relative refractive index of diamond substitutes has been recommended and does have a certain application in gem testing. The idea behind it is that in comparison with diamond with its refractive index of 2.417, most gem materials of lower index will leak light as seen when the stone is tilted so that one observes the back of the stone through the table but at an angle. An attempt to illustrate this is shown in *Figure 5.* Of course, there are limiting factors in both making and photographing the test. One factor is that stones are not all equally well proportioned for the index involved nor are they equally well polished. However, in the photograph we can

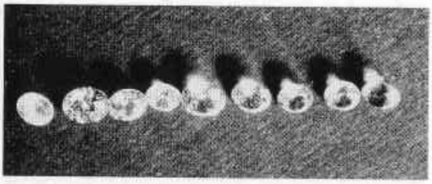


Figure 5.

see that there is more leakage of light with the stones of low refractive index. Quartz is "blackest" as shown in the stone on the right. Next is synthetic spinel, synthetic sapphire, "YAG," "GGG," Zircon (note the effect of abraded facet junctions), strontium titanate, diamond and last, synthetic rutile. Note the preponderance of first surface reflection in the synthetic rutile.

Emeralds-Emeralds-Emeralds

With emeralds apparently at an all-time high in demand and popularity, we continue to see new imitations and substitutes. For a change, we illustrate an excellent natural substitute in *Figure 6* which proved to be aventurine quartz. In appearance it closely resembled the type of Colombian material known as "trapiche." Even the black flecks illustrated in the picture contributed to the resemblance. The glass imitation of emerald shown in *Figures 7* and *8* was a real fooler. Under a loupe no clearly discernible gas bubbles were seen but indistinct "flaws" typical of natural emeralds were observed. There was even a layered effect resembling planes of calcite *(Figure 8).* Another imitation rough emerald crystal is shown in *Figure 9.* This concoction was not easy to determine as to components. Green plastic (which was red under the filter) provided the color. The deception was heightened by the natural appearing "matrix" consisting of an earthy micaceous material held in a mastic or cement. Again, the client was amazed to find out that his "investment" for which he was offered

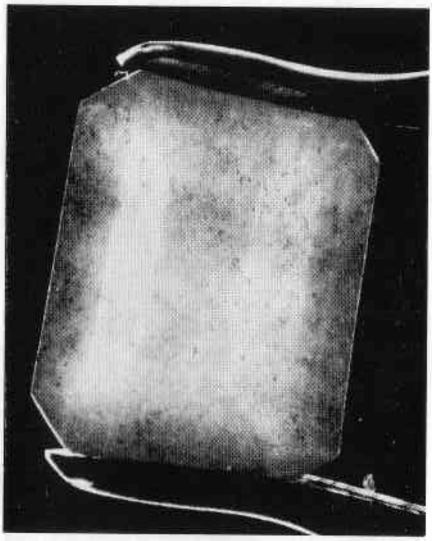


Figure 6.

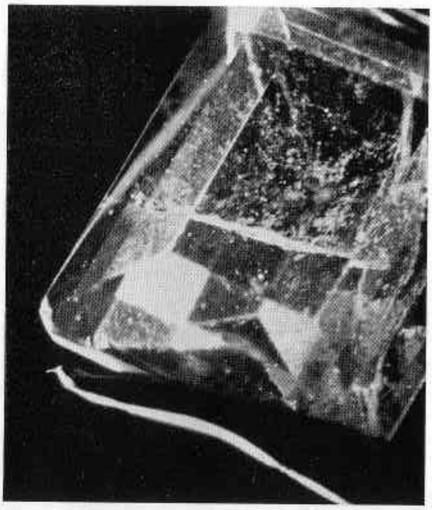


Figure 7.

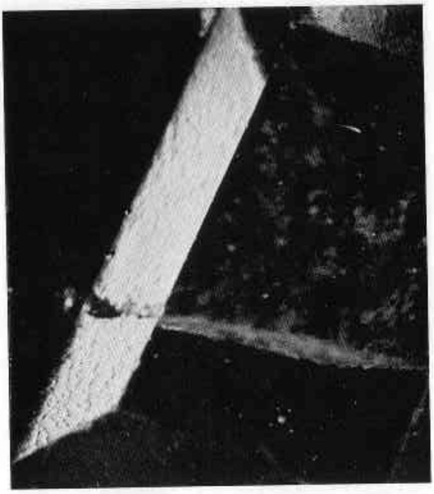


Figure 8.

a "profit" if a satisfactory laboratory report could be secured was in fact worthless. (Except back in Colombia where it could be used again!)

Recently we examined a very nice translucent green stone with two phase inclusions in which the gas bubbles were enclosed in cubes reminiscent of the inclusions we have seen in fluorite. The stone proved to be emerald but we certainly did not recognize a source *(Figure 10).*

Odd Diamond Inclusions

A very shallow cleavage in a diamond submitted for grading resembled a Trojan Horse head *(Figure 11).* The orange-brown natural color diamond shown in *Figure 12* contained a dense white cloud that resembled a marshmallow with a six ray star.

Zirconium Oxide

We are grateful to Mr. Marvin S. Bankoff of MSB Company, New York, for a sample round brilliant of the zirconium oxide described in the

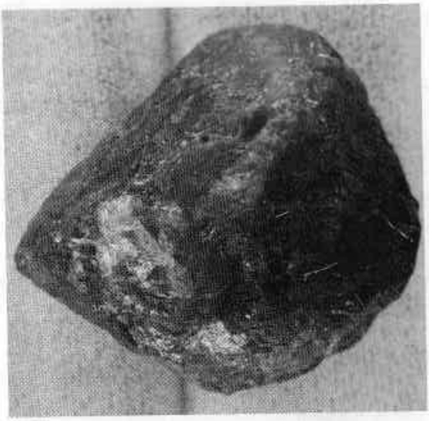


Figure 9.

Spring 1976 issue of *Gems & Gemology* by Dr. Kurt Nassau. Tentatively, Mr. Bankoff plans to market it under the trade marked name Diamonique III. (Diamonique I was his name for YAG and Diamonique II the name for GGG.) A fingerprint inclusion runs across the table of the stone *(Figure*

Figure 10.

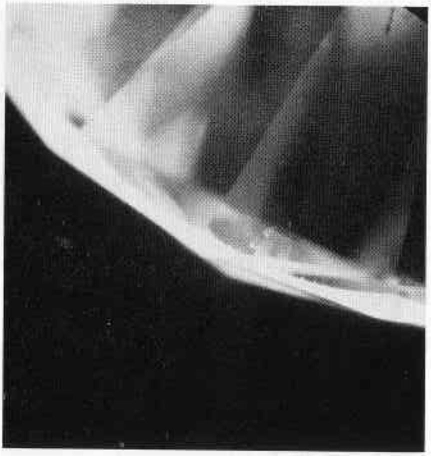


Figure 11.

13) and resembles a reflective grain line.

Some Foolers

The irregularly oriented black needles in the translucent chatoyant cabochon shown in *Figure 14* took us by surprise as the host is cat's-eye chrysoberyl.

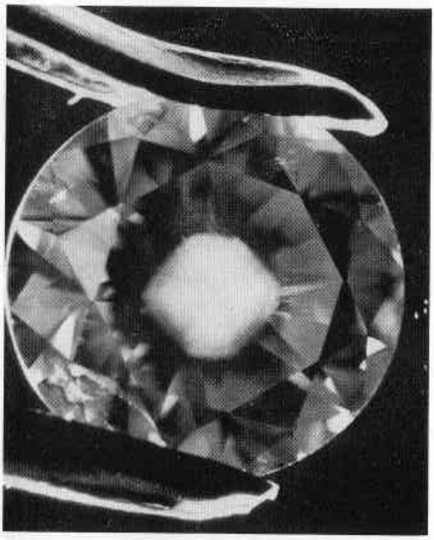


Figure 12.

Figure 15 illustrates what for all the world appears to be the engine-turned appearance one associates with carved ivory. In fact, though, the piece is a plastic netsuke.

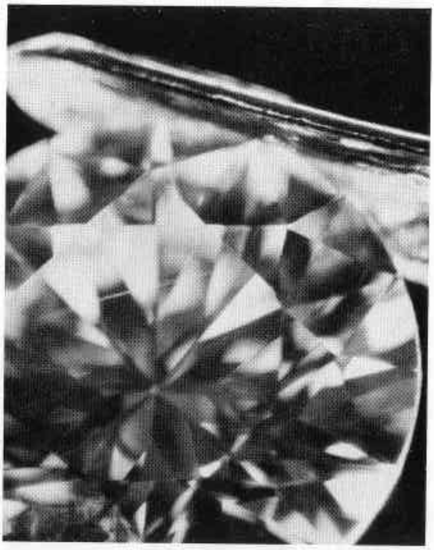


Figure 13.

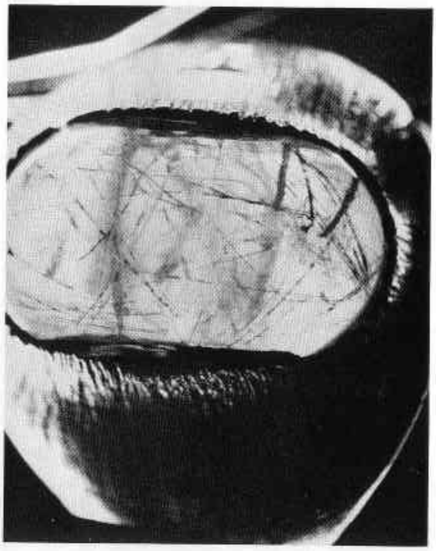


Figure 14.

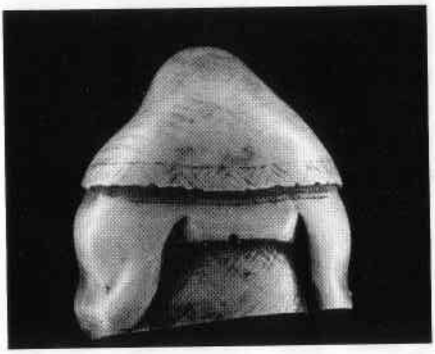


Figure 15.

A fine quality translucent jadeite was selected for the hololith ring shown in *Figure 16.* It was then selectively dyed green on the top.

Use of a fine-grained calcite similarly selectively dyed can only be looked on as reprehensible. *Figure 17* shows what can happen the first time a customer wears such material in a hololith bracelet.

Acknowledgements

We wish to express our sincere thanks for the following gifts and courtesies:

To current resident class student *Menashe Dror*, Israel, for a fine example of so-called Eilat stone (malachite, quartz chrysocolla, and related copper minerals) and two intricate opal mosaic for our display case.

To old friend *Eric Engel* of Joema Indústria Comércio e Exportaçao Ltda, Rio de Janeiro G.B., for a fine Brazilian rhodochrosite crystal and fine red cabochon of the same material. Also a faceted transparent lazulite and an especially gemmy opal and iolite for the collection.

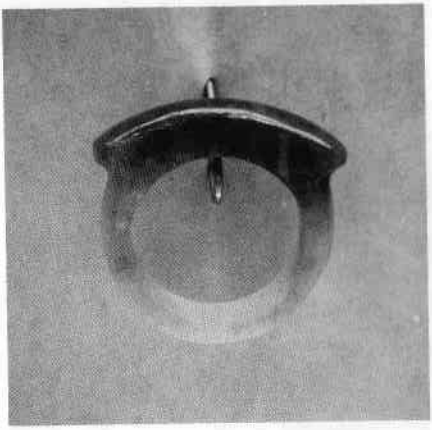


Figure 16.

To graduate *Sam Koulish*, New York (Meyer Koulish Co., Inc.,) for numerous small calibre cut natural and synthetic stones used in their line of fine guard rings and very useful in training students to identify these small stones.

To *Mr. Ivan Malnekoff*, San Francisco dealer in jades, for a selection of

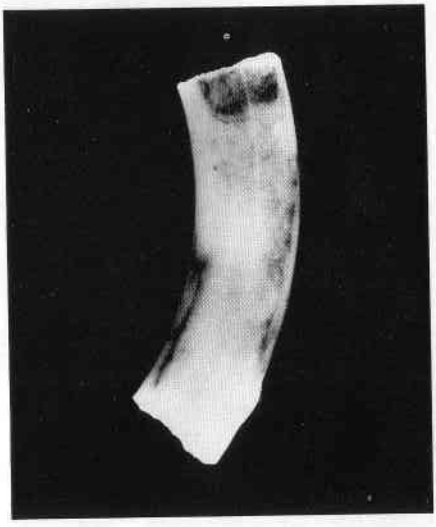


Figure 17.

carved serpentine figurines for our collection and display cabinet. These were recently secured during a buying trip to mainland China. He spoke about the trip before the New York Guild of the American Gem Society in September.

To the gentlemen at *New England Lapidary* for a natural ruby that will find use in classes.

To *Mr. Harold A. Oates*, F.G.A. of Glen Ellyn, Illinois, for a polariscope and interference figure rod of his own design together with a mica plate for determining optic character.

To *Mr. J. C. Pinnavaia*, New York City, for two transparent yellow orthoclase cut stones of great value for use in residence classes.

To *Mr. Joel Schechter* of HONORA Jewelry, New York, for numerous natural stones including two nicely carved malachites and an unusually fine imitation peridot. All will find use in classes and display.

To *Mr. R. L. Scott*, Meridian, Mississippi, for a light brown round brilliant cut "GGG" which is the first we have heard that changed to this darker color after only exposure to sunlight. Heretofore we were aware that some changed color after exposure to strong artificial ultraviolet radiation.

To the following students and friends for gifts to the GIA Library: *Janet Jones*, Ruppenthal America, for a beautiful copy of "Edelstein and Mineralien"; to *Mr. Benjamin Zucker* for copies of his controversial book, "Investing in Gems"; to resident class student *Vernon Crawford* for a copy of "Edelstein"; to graduate *Irving Michaels*, New Haven, Connecticut, for a selection of numerous common stones as well as a small lot of much needed transparent green andalusites. To student *Sy Ellenhorn*, New York City, for a nice group of natural red spinel crystals and a fine crystal of the uncommon mineral gaylussite. At 2 to 3 hardness, nearly colorless and very brittle, it offers no interest to the gemstone wearer.