
GEMSTONES WITH ALEXANDRITE EFFECT

By E. Gübelin and K. Schmetzer

The term alexandrite effect refers to the apparent change of color in certain minerals from blue-green or greenish violet in daylight to red or reddish violet in incandescent light. This effect was discovered in chrome-bearing chrysoberyl from the Ural Mountains as early as the beginning of the 19th century. In more recent times, it has also been observed in certain varieties of garnet, corundum, spinel, kyanite, fluorite, and monazite. It has been determined that the absorption spectrum of all alexandrite-like minerals is characterized by transmission maxima in the blue-green and red regions and by a transmission minimum in the yellow region. The color of minerals with two such regions of transmission is determined in daylight (richer in blue and green) by the position of the transmission maxima and in incandescent light (richer in red) by the ratio of transmission in the red and transmission in the blue-violet sections of the visible spectrum.

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The alexandrite effect was observed for the first time in 1830, in a chromium-bearing variety of chrysoberyl from the emerald mines on the eastern slopes of the Ural Mountains. The new gem was named by the Swedish explorer Nils Nordenskiöld in 1838 (Koksharov, 1861), in honor of then-Tsarevitch Alexander Nikolayevitch, the future Tsar Alexander II (1818–1881). The alexandrite, famous for its conspicuous change of color—green in daylight and red in incandescent light—soon enjoyed great popularity as well. This gem was appreciated not only for its curious color change, which has only very recently been explained (Carstens, 1973; Hassan et al., 1974; Gübelin, 1976 a and b; and Schmetzer et al., 1980 a and b) but also because red and green were the colors of the tsarist army; thus the alexandrite was considered by many to be the national gem of tsarist Russia.

Most of the alexandrites found on the gem market today originate from deposits in Sri Lanka and those recently discovered in Zimbabwe, Brazil, and Tanzania. The Brazilian specimens usually display only a weak change of color, while those from Tanzania and Zimbabwe demonstrate a striking alteration from dark green and blue-green to pure or violetish red. Sri Lanka has been and still is the most important producer of alexandrite. The colors seen in the Sri Lankan specimens (brownish green in daylight and olive-tinged red in incandescent light) are more subtle than those commonly observed in their Russian counterparts (blue-green and deep red), and the change of color is less pronounced. Figure 1 illustrates the color change in a fine Sri Lankan alexandrite.

White et al. (1967) introduced the term *alexandrite effect* to the literature and identified that it was not just restricted to alexandrite, although alexandrite-like sapphire has been known to exist since at least the 1950s (the American Museum of Natural History in New York placed such a sapphire on display in 1960). A few garnets



Figure 1. Color change visible in an 8.07-ct alexandrite from Sri Lanka, from natural or fluorescent light (left) to incandescent light (right). Stone courtesy of the Los Angeles County Museum of Natural History. Photo by Tino Hammid.

from the gemiferous Sri Lankan district of Ratnapura displaying a very similar change of color came onto the market in the 1960s (the senior author acquired his first specimen, 5 ct, in 1968). Subsequently, alexandrite-like garnets were found in the Umba Valley of Tanzania as well as in Bohemia, Norway, and Yakutia. They were either chrome-rich pyropes or, more commonly, vanadium and/or chrome-bearing spessartite-rich pyropes (Amthauer, 1976; Schmetzer and Ottemann, 1979).

Although well-informed gemologists have known for some time that an alexandrite effect may be produced in synthetic corundum by adding 20% chromium oxide or traces of vanadium oxide (Barks et al., 1965), natural corundum from Thailand and Colombia displaying a similar effect has only recently been described in detail (Schmetzer et al., 1980). These gems contain V_2O_3 , Fe_2O_3 , and TiO_2 in addition to Cr_2O_3 . A few examples of natural spinel from Sri Lanka showing a change of color from bluish violet in daylight to red-violet in incandescent light have also appeared on the market. The one such crystal examined by the authors contains approximately 2% FeO as well as small amounts of Cr_2O_3 and V_2O_3 (Schmetzer and Gübelin, 1980). To complete this record, it may be mentioned that an alexandrite-like fluorite, from Cherbadung in Switzerland, was studied by H. A. Stalder (1978), an alexandrite-like kyanite from East Africa was described by Bosshart et al. (1982), and an unusual alexandrite-like monazite from North Carolina was described by Bernstein (1982).

The following discussion reviews the current knowledge on gemstones with the alexandrite ef-

fect and seeks to describe a common denominator for this unusual phenomenon. In our search for such a factor, we focused away from the question of how the eye perceives color (an important issue, but beyond the scope of this study) and concentrated on the conditions—the chemistry of the stone and how certain elements interact within a particular crystal to produce a characteristic spectrum—that produce the color the eye observes.

ALEXANDRITE

In the geochemical cycle, it is extremely uncommon that the structural component beryllium on one side and chromium as a coloring agent on the other should combine to form a crystal (Gübelin, 1976 a and b). The chromium-bearing variety of chrysoberyl ($BeAl_2O_4$) is thus a very rare gemstone. The petrology and paragenesis of alexandrite have been described by Bank and Gübelin (1976) as well as by Leithner (1980).

In alexandrite, part of the aluminum is substituted by chromium (Cr^{3+}). However, the chromium content varies within relatively broad limits depending on the occurrence. To date, the smallest trace of chromium, 0.03% Cr_2O_3 , has been determined in samples from Tanzania, whereas a concentration between 0.30% and 0.73% was found in Brazilian alexandrines (Schmetzer, 1978). The chromium ions produce the typical color and color change of alexandrite, whereas chrysoberyl normally owes its range of hues to traces of iron. Since Cr^{3+} has a slightly larger ion radius (0.63 Å) than Al^{3+} (0.51 Å), it naturally prefers the more spacious of the two available aluminum lattice sites (Al_2) within the chrysoberyl

structure; yet some Cr^{3+} ions are nevertheless incorporated into the second and narrower aluminum sites (Al_1). It will be shown that not only the amount of chromium in the chrysoberyl lattice, but also the distribution of the chromium ions between these two aluminum sites, as reflected in the spectrum, is responsible for the characteristic color change and especially for its intensity.

The spectrum for alexandrite (figure 2a) shows a transmission maximum at about 490 nm (i.e., in the blue to blue-green region), an absorption maximum at 570 nm (i.e., in the yellow region), and a second transmission maximum after about 600 nm in the red region and beyond in the adjacent infrared. This spectrum is primarily dependent on the presence of the Cr^{3+} ions, which occupy 65% to 80% of the Al_2 lattice sites (Solntsev et al., 1977; Matrosov et al., 1978). The presence of the Cr^{3+} ions in Al_1 sites effects the widening and eventual displacement of this absorption maximum, yet the corresponding Cr^{3+} bands remain weaker than the bands for the Cr^{3+} in Al_2 sites. The ratio of the Cr^{3+} ions in these two Al sites varies from crystal to crystal, and thus explains the variance in hue and intensity of the color change observed in different specimens from daylight to incandescent light.

The transmission of alexandrite for visible light is high in the blue-green as well as in the red region of the spectrum. In daylight, in which the blue short-wave components predominate (figure 3), green to blue-green results. This effect is amplified by the optimal sensitivity of the human eye for green light. In incandescent light, however, in which the long-wave spectral components predominate, the human eye perceives a red hue because of the transmission in the spectral section of minimal red absorption. The iron ion does not directly contribute to the alexandrite effect, although it enhances the brownish cast of certain alexandrites from Sri Lanka.

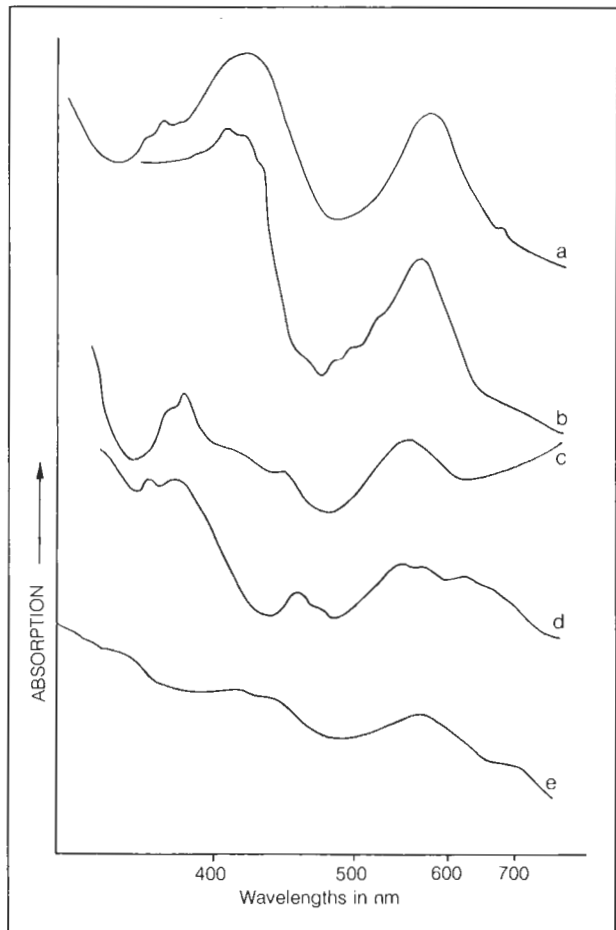
GARNETS WITH ALEXANDRITE EFFECT

Crowningshield (1970) was the first author to mention this new type of garnet. Garnets belong to the group of nesosilicates with the general formula $\text{X}_3^{2+}\text{Y}_3^{3+}[\text{SiO}_4]_3$, which indicates that the silicate anion is combined with a divalent metal ion X and a trivalent metal ion Y. The alexandrite effect is displayed only by some pyrope garnets ($\text{Mg}_3\text{Al}_2[\text{SiO}_4]_3$) and some mixed crystals between

the end members of the isomorphous series of pyrope-spessartite garnets (spessartite: $\text{Mn}_3\text{Al}_2[\text{SiO}_4]_3$). The color change of chromium-rich pyropes is from bluish green in daylight to wine red or reddish violet in incandescent light.

According to Yeremenko and Polkanov (1972), the alexandrite effect occurs in all pyropes that contain 4% or more Cr_2O_3 . The absorption maximum of pyropes with a lower chromium content lies in the green to yellowish green region of the

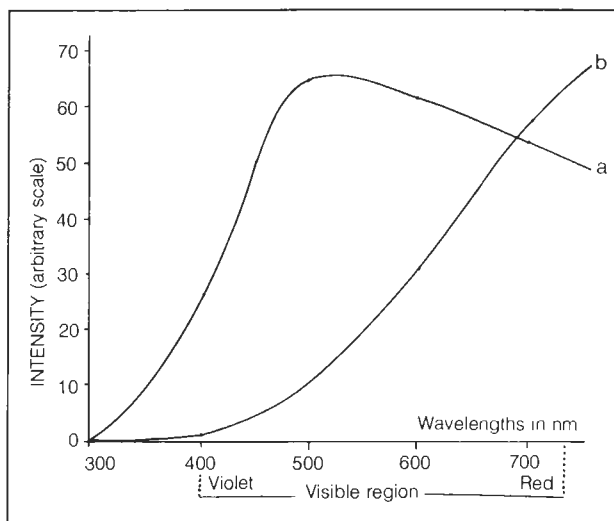
Figure 2. Absorption curves of gemstones with alexandrite effect: (a) alexandrite (from Lake Manyara, Tanzania); (b) Mn-rich pyrope (from Ratnapura, Sri Lanka); (c) corundum (in this instance, from Colombia, although similar sapphires may be found in Thailand); (d) spinel (from Ratnapura district, Sri Lanka); and (e) fluorite (from Cherbading, Switzerland). The absorption spectra for all of these stones have transmission maxima in the red and blue-green regions and an absorption maximum in the yellow region in common.



spectrum; hence, such pyropes appear red to bluish red in both natural and incandescent light. A greater amount of inherent chromium displaces the transmission from the blue-violet to the blue-green spectral region. Consequently, such garnets transmit the red rays of incandescent light (figure 2b). These circumstances are responsible for the alexandrite effect (Amthauer, 1976; Schmetzer and Ottemann, 1979).

Spessartite-rich pyropes from Tanzania, on the other hand, in which a larger portion of Mg is replaced by Mn, require only small amounts of vanadium or chromium (or both) to evoke an alexandrite effect (Gübelin, 1968; Schmetzer et al., 1980). The absorption spectrum for these types of garnets is characterized by a sharp maximum between 560 and 576 nm. The transmission occurs, just as with alexandrite, in the blue-green and red regions of the spectrum. Considerable differences in the coloration of individual garnets may be noted, caused by the situation of the absorption minimum in the blue-green region and a maximum in the yellow region of the spectrum. The iron content of the particular crystal is primarily responsible for this condition, inasmuch as some bands of Fe^{2+} are situated between 454 and 526 nm in the domain of the blue-green absorption minimum (figure 2b). The alexandrite effect oc-

Figure 3. Spectral composition of (a) daylight (with the sun 65° above the horizon), and (b) artificial light (50 W incandescent lamp). The different-intensity maxima (for daylight in the blue-green and for artificial light in the red spectral region) are very conspicuous.



curs in iron-rich pyropes with no spessartite molecules when they contain relatively high contents of chromium (e.g., 8% or more Cr_2O_3).

With regard to the absorption spectra for these stones, the chromium and vanadium ions behave similarly in all garnets. For instance, in Mn-rich crystals, the specific absorption is partially superimposed by Mn^{2+} bands; they appear between 408 and 422 nm. In the garnet crystal lattice, the Cr^{3+} ions (as well as the V^{3+} ions) substitute for some of the Al^{3+} , while Mn^{2+} replaces Mg^{2+} . Further details concerning the interpretation of the spectra of alexandrite-like garnets may be consulted in the articles by Amthauer (1976) and Schmetzer and Ottemann (1979). The article by Stockton (1982) illustrates some interesting color-change garnets from East Africa.

CORUNDUM WITH ALEXANDRITE EFFECT

Corundum is crystallized alumina ($\alpha-Al_2O_3$). Pure corundum is colorless; the partial replacement of Al^{3+} by one or another of the transition elements will introduce a wide variety of colors. Corundum forms a continuous isomorphous series of mixed crystals with chromium oxide, $(Al_{1-x}Cr_x)_2O_3$. Small amounts of chromium impart a red color; greater amounts impart green. Thus, a 20% Cr_2O_3 content renders corundum green in daylight and pink in incandescent light. The absorption maxima of synthetic alexandrite-like corundum lie at 415 and 568 nm. The spectrum of vanadium-bearing synthetic corundum is also conspicuous by two strong absorption maxima at 571 and 398 nm. The stones are gray-green in daylight and amethyst-like reddish violet in artificial light.

Natural corundum with alexandrite effect (figure 2c) develops transmission maxima at 633 nm (red) and 483 (blue-green). The alexandrite effect thus materializes the same as in alexandrite itself. Alexandrite-like sapphire from Thailand, which has less chromium, displays only a weak change of color; while the blue sapphire from the Umba Valley, which turns violetish red under incandescent light, possesses a greater amount of chromium and hence shows a stronger color change. This suggests that the intensity of the color change depends on the concentration of the transition-element atoms present (Schmetzer et al., 1980). Figure 4 illustrates the color change in a blue sapphire with alexandrite effect.



Figure 4. Color change evident in a 9.54-ct sapphire, purportedly from Sri Lanka, from natural or fluorescent light (left) to incandescent light (right). Stone courtesy of the Los Angeles County Museum of Natural History. Photo by Tino Hammid.

SPINEL WITH ALEXANDRITE EFFECT

Spinel is a combination of a divalent cation, typically magnesium in gem spinels, with alumina (MgAl_2O_4). Characterized by a dense cubic stacking of oxygen atoms, the tetrahedral interstices between the oxygens are filled in with bivalent metal ions (e.g., Mg^{2+} , Fe^{2+}), while trivalent ions (Cr^{3+} , V^{3+} , Al^{3+}) find their places in the octahedral coordinated lattice points.

The only alexandrite-like spinel described so far (Schmetzer and Gübelin, 1980) has a color change from violet-blue in daylight to red-violet in artificial light. This unusual spinel, which originated in Sri Lanka, contains 2.27% FeO as well as lesser concentrations of Cr_2O_3 (0.06%) and V_2O_5 (0.03%). Transmission maxima of equivalent intensities are observed at 490 and 440 nm; absorption maxima are at 571 and 633 nm (figure 2d). In contrast to the more typical blue spinel, which is rich in iron (without traces of Cr and V), the chromium and vanadium ions in the alexandrite-like variety strengthen the intensity of the absorption maximum at 571 nm, while the transmission maximum in the blue region is simultaneously weakened by the same ions. This means that the spectrum displays transmission in the blue-violet and red regions with an absorption maximum in the yellow regions, thus producing the alexandrite effect.

KYANITE WITH ALEXANDRITE EFFECT

Kyanite (Al_2SiO_5), which occurs in various shades

of blue and green, has become a highly appreciated collector's gem.

In their publication, White and White (1967) claimed that titanium was solely responsible for the typical blue color of kyanite. More recently, Bosshart et al. (1982) stated that "the chromophore pairs Fe + (Ti) and Cr + Fe produce the various blue colorations of kyanites rather than titanium traces" (see Schmetzer, 1978). Among the four kyanites examined by Bosshart and his colleagues was one greenish blue specimen that turned purple under incandescent light and was found to have an exceptionally high percentage of chromium (Cr_2O_3 , 0.50%) coupled with a low iron content (Fe_2O_3 , 0.35%) and a very small amount of titanium (TiO_2 , 0.04%).

In general, the absorption spectrum of the color-changing kyanite was similar in character to the spectra of all other minerals with an alexandrite effect examined thus far, in that the transmission maxima in the near ultraviolet (at 345 nm) and blue-green (at 488 nm) regions are combined with strong absorption in the violet (at about 410 nm) and in the green-yellow to red region (from 560 to about 650 nm). However, the decisive absorption of the α -ray, with transmission peaks at 345 and 488 nm and absorption maxima at 417 and 578 nm, is (according to Bosshart et al., 1982) governed totally by the chromium content, which imparts the α -ray with all four properties for releasing the color change. The β - and γ -rays do not directly participate in the color change, although their influence on the absorption enhances its general appearance. Thus,

in this rare gem, the conditions leading to an alexandrite effect are fulfilled by the particular spectral features generated by the chromium and assisted by the iron content, in that the α -ray mainly transmits the red rays of the incandescent light, whereas the β - and γ -rays transmit the strong greenish blue rays of daylight.

FLUORITE WITH ALEXANDRITE EFFECT

The gemstones with alexandrite effect described thus far are all oxides or silicates in which coloration and color effects are caused by ubiquitous ions such as Cr^{3+} and V^{3+} as well as Fe^{2+} and Fe^{3+} . The alexandrite-like fluorite cannot be fitted into this scheme because it is a calcium fluoride, occurring in nature as violet, blue, green, yellow, orange, red, pink, and colorless crystals, depending on the incorporated ion. The coloration of various fluorites was investigated by Bill and Calas (1978).

Stalder et al. (1978) referred to alexandrite-like fluorite from Cherbadung (VS, Switzerland). The absorption spectrum for this stone is similar to that of all other minerals with an alexandrite effect identified thus far; yet in the case of fluorite the alexandrite effect is caused by a superimposition of the bands projected onto one another by the ions of rare-earth elements such as yttrium (Y^{3+}), cerium (Ce^{3+}), and samarium (Sm^{3+}). As illustrated in figure 2e, the absorption maximum in the visible sector of the spectrum is situated at 578 nm and the absorption minimum occurs at 495 nm (Schmetzer et al., 1980). The fluorite illustrated in figure 5 shows a distinct change from blue in natural light to lavender in incandescent light.

MONAZITE WITH ALEXANDRITE EFFECT

The cerium-phosphide monazite, CePO_4 , is not usually considered a gemstone, although this mineral may be found among the oddities of some gem collections; yet it confirms that many other minerals—especially the chromium-, iron- and titanium-bearing ones—could theoretically display an alexandrite effect. According to Bernstein (1982), yellow-orange crystals of monazite from Montgomery County, North Carolina, appeared reddish orange under incandescent light and pale green under fluorescent light. These differing colors were found to be caused by the rare-earth element neodymium and the particular arrange-

ment of narrow absorption bands centered at about 800, 745, 580, 525, and 515 nm, with a cut-off below approximately 480 nm. The close coincidence of these bands, due to Nd^{3+} , and the transmission gaps between them create absorption features that are consistent with those observed in the aforementioned gems. The eye perceives different hues in response to changes in the wavelength compositions of different light sources; the narrow absorption bands of the monazite prove to be sufficient to generate the color change.

DISCUSSION AND SUMMARY

In reducing these observations to a common denominator, we wish to emphasize that the cause of the alexandrite effect shared by all of these stones lies not in the crystal structure or in the chemistry of the coloring agents, but rather in an equivalent correspondence of the position of the absorption maxima and minima in the visible region of the spectrum. Alexandrite and the alexandrite-like varieties of garnet, corundum, spinel, kyanite, and fluorite demonstrate a strong transmission as much in the blue-green to violet as in the red region of the visible spectrum. In addition, a strong absorption occurs in the yellow region, where it culminates in a pronounced maximum. Starting with 476 nm (blue region), the absorption increases gradually until it reaches the long-wave end of the visible spectrum in the red region (Schmetzer et al., 1980). Alexandrite-like spinel presents an exception to this general rule, developing three regions of strong transmission, in the red, blue-green, and violet. Consequently, the color change in this stone is from violet-blue (not blue or green) to red-violet.

The color of each mineral depends on a superimposition of the light components of all spectral regions not absorbed by the mineral. In minerals with an alexandrite effect, the occurrence of the absorption minimum in the green, bluish green, or blue region (i.e., between 560 and 480 nm) is decisive for their color in daylight because the red component of daylight is not strong (figure 3a) and, as mentioned before, the sensitivity of the human eye is greatest for green light. On the other hand, in incandescent light, which has a weaker blue and stronger red component (figure 3b), the transmission in the red region, from about 600 nm to the end of the visible spectrum, exerts a dominant influence on the resulting color.

The intensity of the color change and the hues

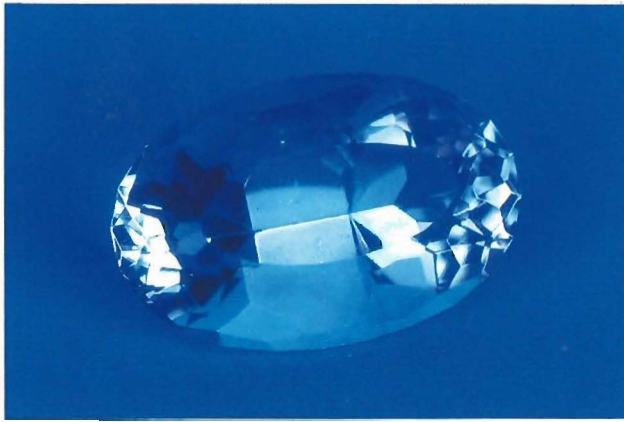


Figure 5. Color change apparent in a 28.55-ct fluorite from England, from natural or fluorescent light (left) to incandescent light (right). Photo by Tino Hammid.

observed in daylight and incandescent light in minerals with alexandrite effect depends on the particular position of the transmission and absorption regions as well as on their relative intensities. These in turn are dependent on the very complex absorption spectra, which themselves

are influenced by minute traces of chromophorous ions within the individual crystal. Nature possesses an enormous repertoire of such complex occurrences. It would, therefore, hardly be astonishing if more gemstones with such a color change were to appear in the future.

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