

Spectroscopic properties of an historical greenish yellow diamond

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Background

The vast majority of greenish-yellow type Ia diamonds available on the jewellery market resulted from the alteration of brown colours under high pressure and high temperature (HPHT) conditions. In the recent past, they were extensively documented and their colour modification explained on the basis of spectroscopic features [1].

On the contrary, naturally occurring greenish-yellow diamonds have long been recognised by collectors as being extremely rare. As a consequence of this scarcity, their spectroscopic properties have virtually not been published so far. Therefore, with respect to the distinction of greenish-yellow diamonds of natural and artificial coloration, some degree of uncertainty remains.

The spectroscopic characteristics of a natural-colour greenish-yellow diamond, being the property of the Natural History Museum of Vienna, are presented in this study.

Sample

The specimen was offered to the Natural History Museum of Vienna, Austria, in 1907 by His Majesty, the Emperor Franz Joseph the First. Since then, it has been on record in the museum archives under reference: no. H 5222.

Results of the investigation

Macroscopic and microscopic observations

This slightly distorted octahedral diamond rough weighs 1.118 ct. It displays an intense yellow body colour, combined with a fairly strong green luminescence to daylight. Its fluorescence under long-wave ultraviolet radiation is strong and greenish yellow. The intensity of the VIS-luminescence explains its greenish-yellow colour when observed in daylight or under an equivalent light source. A few minute green irradiation spots are detectable on the corroded surface of the crystal, together with some larger brown stains.

FTIR spectroscopy

The spectrum was recorded at room temperature on a Nicolet Nexus FTIR with a 5x beam condenser at a resolution of 0.5 cm⁻¹. Upon decomposition of the single-phonon region, the total concentration of nitrogen is estimated at 93.0 ± 7.4 ppm with 47.1 ± 2.9 ppm in the form of A aggregates, 43.1 ± 4.3 ppm of B aggregates and 2.8 ± 0.2 ppm of single nitrogen [2-5]. The platelet peak is centred at 1360.7 cm⁻¹ and has an absorption coefficient of 2.3 cm⁻¹ and an approximate Full Width at Half Maximum (FWHM) of 7.3 cm⁻¹.

UV/Visible and NIR spectroscopy

Spectra were recorded at cryogenic temperatures (ca -130°C) on a Varian Cary 500 spectrometer with a fixed bandwidth of 0.5 nm. The first scan from 800 to 200 nm showed a strong H3 (503.2 nm) zero-phonon line with an absorption coefficient of 0.7 cm⁻¹, a moderate N3 (415.2 nm) peak,

N4 to N7 (344.2 to 305.9 nm) peaks, a series of weak H11 to H14 peaks (374.7 to 364.3 nm) and a faint GR1₀ peak (741.1 nm). The second scan ranging from 1010 to 950 nm exhibited a weak H2 (986.0 nm) peak with an absorption coefficient of 0.06 cm⁻¹.

Photoluminescence spectroscopy

A low-temperature photoluminescence spectrum was recorded on a dispersive Renishaw 1000 Raman spectrometer using an ionised argon laser of 514 nm wavelength excitation. Major photoluminescence peaks and some of their characteristics are given in table 1.

Table 1

Peak position		Band name	Number of counts	FWHM (cm ⁻¹)	Peak position		Band name	Number of counts	FWHM (cm ⁻¹)
cm ⁻¹	nm				cm ⁻¹	nm			
773.9	535.8	-	204500	10.9	2071.0	587.9	-	17330	12.2
1332.0	552.4	Raman	48900	8.8	3737.8	637.0	NV	44200	15.9
2042.8	574.9	NV ^o	39200	18.8	5942.9	741.1	GR1	6140	17.2

Discussion

Successful application of the HPHT process was, at least theoretically, possible since the 1950's. The donation of this sample, as registered in the museum archives, predates this period and thus provides a high degree of certainty that the coloration of this diamond crystal is of natural origin. Nevertheless the specimen shows a very strong H3 colour centre and enables direct optical comparison with those green-luminescent yellow HPHT-treated diamonds.

The greenish and brownish surface stains are proof of the crystal having been irradiated and partly annealed in nature. The GR1 absorption is weak. Accordingly its influence on the overall colour appearance is minor. The irradiation damage causing GR1 absorption very likely is limited to the surface of the diamond rough as well. It would be affected to some extent or substantially removed by a cutting operation. If this absorption system would disappear entirely, the UV/Visible spectrum of the cut stone would look very similar to that of green-luminescent yellow HPHT-treated diamonds.

The presence of an H2 peak, here occurring naturally, and of a weak concentration of single nitrogen, accompanied by A and B aggregates, are characteristics typical of HPHT-treated diamonds.

Conclusion

This study describes a green-luminescent yellow diamond of unquestionably natural coloration. It appears that most of its spectroscopic properties are identical to those of HPHT-treated type Ia diamonds of the same colour. Therefore, unequivocal differentiation of these two identical kinds of diamond colour requires further testing of greenish-yellow diamonds of natural coloration.

Acknowledgments

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