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LOW-TEMPERATURE THERMAL EXPANSION OF YTTRIUM ALUMINUM GARNET

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ABSTRACT

Coefficients of thermal expansion of pure and Nd-doped yttrium aluminum garnet were measured from room temperature down to liquid nitrogen temperature by the *x*-ray diffraction method. No phase transformations were observed in this range and the coefficients of both crystals were identical, being 4.3×10^{-6} cm/cm/° C. at −173° C.

INTRODUCTION

The rare-earth aluminum garnets have technological interest because of their use in the device known as the laser (acronym for light amplification by stimulated emission of radiation). These optical devices, using large single crystals, often have to cycle between room temperature and that of liquid nitrogen. For this reason the mechanical properties of a laser crystal are of interest. The existence of a low-temperature phase transformation or an anomaly in the expansion coefficient curve could be a cause of shattering of the crystal. One crystal of particular interest in current laser work is yttrium aluminum garnet ($Y_3Al_5O_{12}$) (Yoder and Keith, 1951), doped with Nd. This paper reports the low-temperature thermal expansion of both pure and Nd-doped yttrium aluminum garnet grown in our laboratory by the Czochralski technique. Both the pure and doped crystals were analyzed by emission spectroscopy. They both contained traces (<0.001%) of Fe, Ca, Mg, Si and Mn. The doped crystal contained 1.6% of Nd by weight.

Coefficient of thermal expansion values were calculated from *x*-ray powder diffraction measurements of the unit-cell dimension for a range of different temperatures. The classic examples of measurements of this type are the study of quartz (Jay, 1933a) and of silver (Jay, 1933b). There is an extensive amount of literature on the subject since the original work, but all of it is in the temperature region from 20° C. to 2000° C.

EXPERIMENTAL

The *x*-ray measurements were made on a Norelco high-angle diffractometer using nickel-filtered copper radiation from a high-intensity tube (40 Kv and 40 ma). A divergence angle of 4° was used and the detector was a scintillation counter feeding to a pulse-height analyzer.

The *x*-ray sample stage is a silver block 1 cm ×2 cm and 0.1 cm thick, with a copper-constantan thermocouple imbedded in it. The sample under study was ground in acetone and painted on the surface of the

TABLE 1. PROPERTIES OF YTTRIUM ALUMINUM GARNET—PURE

Temperature °C.	Lattice Parameter Å	Linear Expansion %	Coefficient of Expansion cm/cm° C.
+ 22°	12.008	+0.017	7.7×10^{-6}
− 47°	12.002	−0.033	7.0×10^{-6}
− 92	12.000	−0.050	5.4×10^{-6}
−127	11.997	−0.074	5.8×10^{-6}
−163	11.997	−0.074	4.5×10^{-6}
−170	11.997	−0.074	4.3×10^{-6}

block making a layer approximately 0.05 cm in thickness. The coolant consists of a stream of pre-cooled dry nitrogen, concentrically surrounded by a stream of room-temperarure dry nitrogen to prevent condensation on the sample. The temperature is adjusted by regulating the gas flow rate and by diluting the cold nitrogen with warm dry nitrogen. During each measurement the sample temperature is controlled to within $\pm 1°$ C.

Since the sample is cubic the unit cell determination is straight-forward. The x-ray reflections measured for the determination at different temperatures are 14·4·0 (12·8·2), 14·3·3 (13·6·3) and 14·4·2 (12.6·6; 10·10·4). Average values derived from the α_1 component were used. Calibration of the instrument was made using a value determined on the same sample in a standard room temperature sample holder. A high value of Bragg angle ($2\theta\sim140°$) was selected to minimize the sample height adjustment error and the general systematic error. At the small changes measured in these experiments, the error in expansion coefficient is on the order of 5%.

TABLE 2. PROPERTIES OF YTTRIUM ALUMINUM GARNET—1.6% Nd

Temperature °C.	Lattice Parameter Å	Linear Expansion %	Coefficient of Expansion cm/cm° C.
+ 23	12.010	+0.017	7.4×10^{-6}
− 2	12.008	—	—
− 58	12.004	−0.033	5.7×10^{-6}
− 83	12.002	−0.050	6.0×10^{-6}
−103	12.002	−0.050	4.9×10^{-6}
−137	12.000	−0.066	4.8×10^{-6}
−156	12.000	−0.066	4.3×10^{-6}
−173	11.999	−0.075	4.3×10^{-6}

Conclusion

Phase transformations were not found in either the pure or doped samples. The unit-cell dimension plotted as a function of temperature makes a smooth (almost linear) curve in both cases. The unit-cell dimensions, linear expansion, and coefficient of thermal expansion are presented in Tables 1 and 2. It is to be noted that the larger lattice constant for the Nd-doped yttrium aluminum garnet is consistent with the larger ionic radius of Nd as compared to Y. The coefficients of thermal expansion of both the pure and doped crystals are identical within the experimental error. Hysteresis was not observed in any of the measurements.

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