

Compositional Tuning of Photoluminescence Properties in Nd-Doped YAG–YSGG Mixed Structures

Alberto Anedda, Carlo Maria Carbonaro, Daniele Chiriu, Pier Carlo Ricci, Mahmoud Aburish-Hmidat, Maurilio Guerini, Pier Giorgio Lorrain, and Emery Fortin

Abstract—The photoluminescence (PL) properties of neodymium-doped yttrium aluminum garnet and yttrium scandium gallium garnet mixed structures were investigated as a function of the relative concentration of the two garnets. The blue shift of the emission bands in the 930–950-nm range is ascribed to compositional tuning effect. An analytical model to estimate the variation of the PL position as a function of the compositional host structure is proposed.

Index Terms—Compositional tuning, mixed garnet, photoluminescence, yttrium aluminum garnet (YAG), yttrium scandium gallium garnet (YSGG).

I. INTRODUCTION

THE development of new solid-state lasers, especially those operating from 0.9 to 3.0 μm , has renewed general interest in the optical properties of rare-earth ions (R^{3+}) in garnet structure [1]–[3]. The energy levels of several R^{3+} ions inserted as dopants in different garnet compounds, like yttrium aluminum garnet (YAG) or yttrium scandium gallium garnet (YSGG), were successfully analyzed over the past years [4], [5]. Neodymium-doped YAG, for example, is one of the most important available laser systems for research and technological applications. The prospective of enhancing efficiency and tunability of solid-state lasers boosted the study of new materials able to meet specific technological purposes. Among the possible sources, mixed garnet host materials, like $\text{Y}_3\text{Al}_5\text{O}_{12}$ (YAG), $\text{Y}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$ (YSGG), $\text{Gd}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$ (GSGG), and $\text{Y}_3\text{Sc}_2\text{Al}_3\text{O}_{12}$ (YSAG), doped with lanthanides allow the so-called compositional tuning. By changing the material composition, the lattice parameters can be modified leading to a variation of the strength of the crystal field. As a consequence, the emission wavelengths of a selected R^{3+} ion can be tuned at request. Recently, Walsh *et al.* obtained over 100 mJ at 0.9441 μm in the Q-switched mode from

$\text{Nd:YAG}_{(0.18)}\text{YSAG}_{(0.82)}$ [6]. One of the main applications of mixed garnet materials is related to the remote sensing of the atmosphere. As an example, light detection and ranging (LIDAR) or differential absorption lidar (DIAL) techniques can be applied to determine molecular constituent concentrations present in the atmosphere, such as water vapor (H_2O) [7]. The possibility of tuning the laser system to the right wavelength and investigating the desired molecule easily explains the growing interest in compositional tuning of mixed garnet materials [8], [9].

In case of the mixed structure from $\text{Y}_3\text{Al}_5\text{O}_{12}$ (YAG) and $\text{Y}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$ (YSGG) garnets, Ga occupies tetrahedral sites, Sc is in octahedral positions, and Al can occupy both tetrahedral and octahedral sites. These mixed structures, doped with Nd^{3+} , generates a blue shift in the photoluminescence (PL) emission of Nd^{3+} with respect to the observed emission of the R^{3+} ion in YAG [10]. The aim of this paper is to investigate the PL emission of Nd^{3+} in samples with the mixed structure $\text{YAG}_x\text{YSGG}_{1-x}$ as a function of the concentration of YSGG. An analytical model to estimate the variation of the PL position as a function of the compositional host structure is proposed.

II. EXPERIMENTAL SETUP AND SAMPLES

PL measurements were performed with a single-pass spectrometer (Dilor XY800). An argon ion laser operating at 514.5 nm (Coherent Innova 90C-4) provided the excitation. The signal, dispersed with a 600-grooves/mm grating, was detected by a 1024 \times 256 LN2 cooled charge coupled detector (CCD). All PL measurements were performed at room temperature with a spectral resolution of 0.1 nm.

Mixed garnet samples, doped with 1% at. of Nd, were grown by Scientific Materials Europe, Tortoli, Italy, by the Czochralski method with a different composition of $\text{YAG}_x\text{YSGG}_{1-x}$ host materials. The stoichiometric term x was used as a simplification of the formula $\text{Y}_3\text{Al}_{(2-y)}\text{Sc}_y\text{Al}_{(3-x)}\text{Ga}_x\text{O}_{12}$ and it represents the relative concentration of Al, Sc, and Ga in the melt. The stoichiometric composition of samples was controlled by changing the amount of Ga_2O_3 and Sc_2O_3 oxides in the melt in order to obtain the desired relative concentration of YAG and YSGG. Table I summarizes the compositional structure of analyzed samples. Prime oxides of Al_2O_3 , Y_2O_3 , Ga_2O_3 , and Sc_2O_3 (all with 99.999% purity) were mixed and pre-sintered under pressure of 140 MPa and processed at 1400 $^\circ\text{C}$ for 24 h. Sintered tablets, with a first composition of $\text{YAG}_{0.55}\text{YSGG}_{0.45}$, were melted at 1970 $^\circ\text{C}$ in iridium crucible and pulled at 0.8

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Vibrational properties of mixed $(Y_3Al_5O_{12})_x-(Y_3Sc_2Ga_3O_{12})_{1-x}$ crystals

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The vibrational properties of yttrium aluminum garnet (YAG), yttrium scandium gallium garnet (YSGG), and mixed $(YAG)_x-(YSGG)_{1-x}$ structures ($0 < x < 1$) were investigated by means of Raman spectroscopy. By comparison with the vibrational spectrum of the YAG structure, the attribution of the Raman modes of the YSGG is proposed. The spectra were analyzed with a model of isolated polyhedra and the formation of mixed structures was verified. The Raman modes in the mixed structures lie between those of the pure garnets indicating the presence of truly mixed and not separated phases. An analytical model based on the free tetrahedral approximation to study the position of the vibration frequencies as a function of the relative concentration of the garnets is proposed and applied to the study of the A_{1g} breathing mode at 785 cm^{-1} in the pure YAG structure and downshifted at 752 cm^{-1} in the pure YSGG structure. The proposed analysis allows to verify both the quality of the grown crystal and to assess the relative concentration of the garnets. © 2006 American Institute of Physics. [DOI: 10.1063/1.2220473]

I. INTRODUCTION

The perspective of enhancing the efficiency and tunability of solid state lasers is still of great interest in scientific and industrial applications.¹⁻⁴ Among the possible technological applications, the atmosphere sensing and monitoring of environmental conditions affecting planet Earth is the driving field into the research of tunable solid state laser in the infrared region.^{1,2} Indeed, narrow band lasers operating in the Q-switched mode with a high energy per pulse are the ideal solution for such a type of applications, for example, in light detection and ranging (LIDAR) and differential absorption LIDAR (DIAL). In particular, DIAL technique can be used to determine molecular constituent concentrations in the atmosphere.⁵ However, H₂O absorption lines, for example, are very narrow and require that the lasing wavelength accurately matches the desired features, typically within picometric resolution. Recently, a new series of composite lanthanide laser materials was grown and investigated for applications in the wavelength range around 944.1 nm.⁴

It has been demonstrated that the lanthanide ion neodymium (Nd³⁺) operating on the $^4F_{3/2} \rightarrow ^4I_{9/2}$ transition produces a laser emission in the 940 nm range in several host materials.^{1,2,4} Conventional solid state laser hosts, such as yttrium aluminum garnet (YAG), yttrium gallium garnet (YGG), yttrium scandium aluminum garnet (YSAG), or

yttrium scandium gallium garnet (YSGG), can laser at specific wavelengths because of stoichiometric constraints and do not offer the requested tunability. A promising approach is the so called "compositional tuning" where the composition of the host material surrounding the neodymium ion is modified in order to exactly achieve the desired lasing wavelength.¹⁻³ By changing the host composition the lattice parameters change, leading to a variation in the crystal-field strength. As a consequence the position of the energetic levels of the dopant ion changes allowing for different emission wavelengths.^{2,3} It is well known that Nd ion undergoes laser emission at 945.87 nm in YAG and displays a luminescence peak at 937.75 nm when hosted in YSGG.^{1,2} The two crystals share the same symmetry but scandium and gallium atoms replace the aluminum atoms in YSGG. By changing the composition of the host material, YAG/YSGG mixed garnet crystals with different ratio between the two structures can be grown and emission laser in the wavelength range between the emission peaks of the pure structures can be expected. Indeed, it has been recently shown that the photoluminescence properties of Nd doped mixed crystals depend on the composition of the host material.³ In order to verify the presence of a mixed structure instead of two separated phases and to investigate the effects of the modified crystal structure on the dopant ions the analysis of the vibrational properties of the structure of mixed garnet crystals is a mandatory task. In this paper we present a room temperature Raman investigation of $YAG_x/YSGG_{1-x}$ mixed crystals. By

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changing the relative concentration of the two garnets the modifications of the Raman active modes are discussed. A rigid ion model is proposed to explain the reported data. The presented analysis allows to assess that we obtained truly mixed structures and shows that the Raman spectroscopy is a powerful technique to investigate, even *in situ*, the quality of the grown crystals.

II. EXPERIMENTAL SETUP AND SAMPLES

Mixed garnet samples were grown by Scientific Materials Europe (Italy) by Czochralski method with different compositions of $\text{YAG}_x/\text{YSGG}_{1-x}$ host materials. Pure oxides of Al_2O_3 (99.999%), Y_2O_3 (99.999%), Ga_2O_3 (99.999%), and Sc_2O_3 (99.999%) were mixed and presintered under pressure of 140 MPa and processed at 1400 °C for 24 h. Sintered tablets, with composition of $\text{YAG}_{0.55}/\text{YSGG}_{0.45}$, were melted at 1970 °C in an iridium crucible and pulled at 0.8 mm/h with $\omega_r=15$ rpm under Ar atmosphere. The obtained boule was used as a seed for pulling the samples with a different relative concentration of the two compounds. Sample composition was controlled by changing the amount of Ga_2O_3 and Sc_2O_3 oxides in the melt in order to obtain the desired concentration of YAG and YSGG. Samples were cut along the [111] direction and lapped to optical finish.

Raman scattering measurements were carried out in backscattering geometry along the [111] crystal direction by using the 514.5 nm line of an argon-ion laser. Measurements were performed in air at room temperature with a triple spectrometer Jobin-Yvonne Dilor integrated system with a spectral resolution of about 1 cm^{-1} . Spectra have been recorded in the Stokes region by a 1200 grooves/mm grating monochromator and a charge-coupled device (CCD) detector system.

III. RESULTS AND DISCUSSION

The crystal structure of YAG belongs to a body centered cubic, $O_h^{10}-Ia_3d$, with eight formula units in the primitive cell.⁶ Twenty-four aluminum atoms are tetrahedrally coordinated to oxygen atoms while the remaining 16 Al atoms are coordinated to the O atoms with an octahedral symmetry.⁶ A factor group analysis of the YAG structure shows that there are 98 vibrational modes at $k=0$ belonging to the following irreducible representation:

$$5A_{1u} + 3A_{1g} + 5A_{2u} + 5A_{2g} + 10E_u + 8E_g + 14T_{1u} + 18T_{1g} + 14T_{2g} + 16T_{2u},$$

where the A_{1g} , E_g , and T_{2g} modes are Raman active, the T_{1u} modes are IR active, and the others are silent modes.⁶ In Fig. 1 the Raman spectrum of YAG crystal in the 200–1000 cm^{-1} range is presented; the main peaks can be easily identified and assigned according to Hurrell *et al.* (Table I).⁶ The Raman spectrum of the YSGG crystal is also reported in Fig. 1 and the peak positions in the 200–1000 cm^{-1} range are listed in Table I with the proposed attribution according to the discussion reported below.

The YSGG crystal is characterized by the body centered cubic symmetry and the vibrational modes at $k=0$ belong to the same irreducible representation reported for the YAG

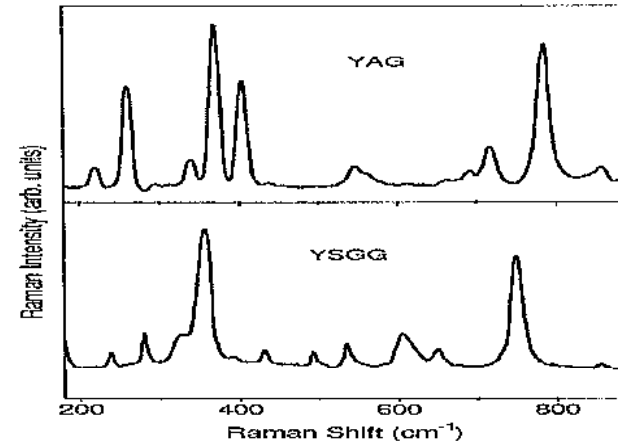


FIG. 1. Raman spectra of pure YAG and YSGG single crystals.

crystal. The YSGG structure can be obtained from the YAG structure by substitution of aluminum 2+ ions at the octahedral sites with scandium ions and by substitution of aluminum 3+ ions at the tetrahedral sites with gallium ions.^{1,7} It has been reported that the vibrational spectrum of YAG crystals can be interpreted on the basis of characteristic vibrations of the AlO_4 tetrahedral and AlO_6 octahedral sites.^{6,8–10} According to this interpretation, the Raman spectrum can be divided in three different parts: the bands in the 400–600 cm^{-1} range are related to the vibrations of Al–O in quasi-isolated AlO_6 octahedral sites while the peaks between 600 and 800 cm^{-1} belong to vibrations of Al–O in quasi-isolated AlO_4 tetrahedral. On the contrary the Raman bands in the low wave number region are related to the motions of the cations with respect to the whole structure.^{6,8,9} The

TABLE I. Raman frequencies and relative assignments in YAG and YSGG single crystals.

Vibrational mode	YAG	YSGG
E_g	161	179
T_{2g}	219	237
T_{2u}	261	279
E_g	339	Unresolved
A_{1g}	372	
E_g	403	
E_g	530	394
T_{2g}	545	430
A_{1g}	560	492
T_{2u}	690	536
T_{2u}	714	604
T_{2g}	720	650
A_{1g}	785	752

proposed interpretation is based on the comparison with the vibrational frequencies of a number of molecules and allows to describe the oxygen vibrations in the high frequency range while those in the low wave number region are strongly mixed to the vibrations of the cations and cannot be interpreted by means of vibrations of specific structures.^{9,11} On the basis of the proposed model, the Raman modes of the YSGG structure can be assigned to specific vibrations by comparison to those of the YAG crystal. The symmetric stretching mode (A_{1g}), due to the isolated octahedron in YAG crystal, is reported at 560 cm^{-1} , while the E_g and T_{2g} vibrations are detected at 530 and 545 cm^{-1} , respectively. By substituting the central cation with a heavier and larger atom a shift of the octahedral vibrational modes to lower wave numbers is expected. As already reported for the free polyhedra structure,¹² the shift is not rigid, it is larger for the T_{2g} mode than for the A_{1g} mode, and a larger separation of the two bands in the YSGG Raman spectrum can be observed. Thus we propose to assign the vibrations at 492 and 430 cm^{-1} in the YSGG Raman spectrum to the T_{2g} and A_{1g} modes, respectively. Concerning the E_g mode its intensity is very low in both the investigated structures and a shift from 530 cm^{-1} in the YAG crystal to 394 cm^{-1} in the YSGG can be hypothesized.

As regards the three vibrations at 339 , 372 , and 403 cm^{-1} detected in YAG crystals, their counterparts can be tentatively identified in the composite vibrational structure peaked at 358 cm^{-1} with a shoulder at 327 cm^{-1} . It has been reported that the 372 cm^{-1} mode corresponds to the rotational vibration of the AlO_4 units in the YAG structure around two perpendicular axes, while the octahedra vibrate in a similar way around another set of perpendicular axes.⁹ The vibrations at 403 and 339 cm^{-1} possess the E_g symmetry but cannot be ascribed to the vibrations of an isolated structure (tetrahedron or octahedron). As deduced from the partial phonon density of the YAG structure, calculated by Papagelis *et al.*,¹⁰ the contributions of isolated tetrahedron and octahedron overlap within this spectral region. The substitution of the central cation with a heavier atom causes a blueshift of the vibrational modes of the two isolated structures. Thus in the Raman spectrum of the YSGG the bands overlap leading to the composite vibrational structure peaked at 358 cm^{-1} with a shoulder at 327 cm^{-1} .

Concerning the low frequency modes ($<300\text{ cm}^{-1}$) of the Raman spectrum of the YAG crystal, they are associated with mixed modes due to the coupling between the octahedral and tetrahedral structures in the garnet crystal.^{6,9,11} It has been reported that the low wave number modes blueshift while increasing the unit cell dimensions in rare-earth doped aluminum garnets.⁹ The lattice constant of the YAG structure is 12.01 \AA and it increases to 12.45 \AA in the YSGG garnet crystal.^{13,14} Thus the bands at 179 , 237 , and 279 cm^{-1} in the Raman spectrum of the YSGG garnet structure correspond to the vibrations found at 161 , 219 , and 261 cm^{-1} in the YAG Raman spectrum and share the same attribution. In addition, the increase of the unit cell dimensions and the substitution of the Al^{3+} ions with Ga ions generate a redshift in the vibration modes associated with the tetrahedral sites ($>650\text{ cm}^{-1}$).^{10,11} The modes at 690 , 714 , 720 (T_{2g}), and

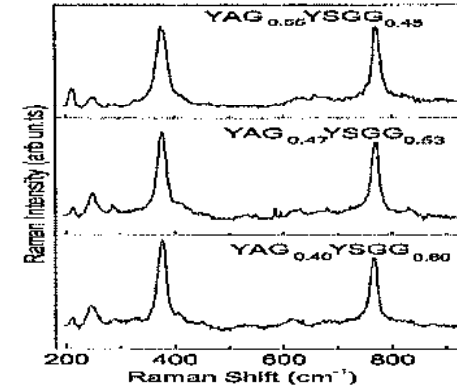


FIG. 2. Raman spectra of mixed garnet crystals ($\text{YAG}_x/\text{YSGG}_{1-x}$).

785 cm^{-1} (A_{1g}) in the YAG structure redshift to 536 , 604 , 650 , and 752 cm^{-1} , respectively, in the YSGG. As already discussed indeed the A_{1g} mode of the free tetrahedron presents a downshift lower than the T_{2g} mode when the cation is replaced with a heavier ion.¹²

The Raman spectra of different mixed garnet crystals ($\text{YAG}_x/\text{YSGG}_{1-x}$) are reported in Fig. 2. The main variations can be observed in the Raman peaks between 650 and 800 cm^{-1} where a progressive redshift of the peaks is detected. On the contrary, the vibrational modes at about 400 cm^{-1} are less affected by the variation of the relative concentration of the two garnets. The Raman bands of the mixed samples show a larger width than the bands of pure YAG and YSGG crystals. However, the spectrum does not show the presence of new vibrational bands. In order to verify this hypothesis, the Raman spectra of mixed garnets were fitted with Lorentzian bands by means of a least-square fit procedure. As an example, the deconvolution of the Raman spectrum of the $\text{YAG}_x/\text{YSGG}_{1-x}$ ($x=0.55$) sample is reported in Fig. 3. The fitting procedure was carried out by introducing the same number of bands observed in both the YAG and YSGG Raman spectra. The peak positions of the vibrational modes of the mixed garnets lie between the positions of the corresponding bands of the single crystals. The good agreement between the fitting procedure and the experimental data (square correlation factor $R^2 \approx 0.99$) allows to confirm the substitutional role of Sc and Ga ions in the mixed garnets and that the symmetry of the structure is preserved. The Raman spectra of mixed garnets show that the Raman bands in the central part of the spectra present a partial or total overlapping of the different vibration modes, making difficult a systematic study of the Raman shift as a function of the variation of the relative concentration of the two garnets. On the contrary, the nondegenerated A_{1g} mode (the breathing of the free tetrahedral molecule) is very intense and does not overlap with other Raman bands, allow

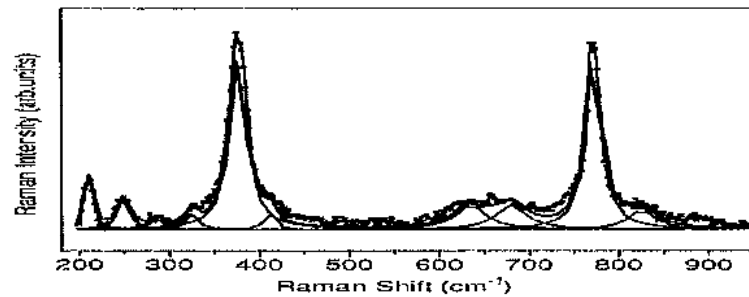


FIG. 3. Deconvolution of the Raman spectrum of $\text{YAG}_{0.55}\text{YSGG}_{0.45}$ mixed garnet.

ing for a detailed analysis of its Raman shift as a function of the mixed garnet structures. Due to the large cation-anion bonding interaction in the garnet structure, the analysis was carried out in the framework of the free tetrahedron model.^{6,10,11} In principle, the analysis could be performed upon the modes related to isolated octahedron. However, the low intensity of these modes does not allow to precisely identify them in the garnet structures. On the contrary, the vibrational modes in the 400 cm^{-1} range (at 339, 372, and 403 cm^{-1} in the pure YAG) display a larger intensity but cannot be interpreted in the framework of the single molecule model. Indeed the spectrum of the phonon dispersion in this spectral region indicates the overlapping of the octahedron and tetrahedron structures preventing the analysis with a single molecule model.⁹

The application of the model to the analysis of the A_{1g} Raman shift as a function of the relative concentration of the YSGG structure is supported by the observation of the A_{1g} stretching mode of tetrahedral sites in YSGG crystal at 752 cm^{-1} , very close to the vibration frequency of the same mode in YGG single crystal (758 cm^{-1}).^{15,16} Indeed YSGG and YGG crystals have the same crystal symmetry with gallium atoms located in the tetrahedral positions in both the crystals while the occupancy of the central anions in the octahedral sites is different (gallium in YGG, scandium in YSGG). Thus, the tetrahedral vibration modes are slightly influenced by the neighboring atoms and the free tetrahedron model can be applied. The A_{1g} mode is at 783 cm^{-1} in the Raman spectrum of the YAG crystal and progressively downshifts as the relative concentration of YSGG in mixed garnets increases. In the pure YSGG crystal the A_{1g} mode is at 752 cm^{-1} . In Fig. 4 the peak position of the A_{1g} mode for different mixed crystals is reported as a function of the relative concentration of YSGG garnet. In order to study the shift of the A_{1g} mode the variation of the potential in the free tetrahedral structure was investigated according to the rigid ion model.¹⁰ The rigid ion model can explain the motion of the atoms in the garnet structure and allows to calculate the frequencies of the Raman active modes. In this model, the atoms are considered as charged points which interact via

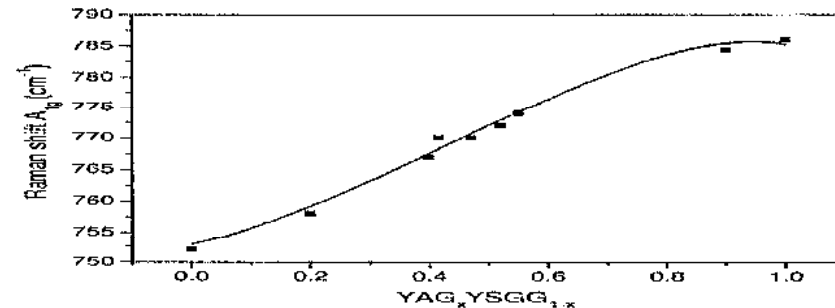


FIG. 4. Experimental data of the Raman position of the A_{1g} vibrational mode vs YAG relative concentration in the mixed garnet. The solid line represents the theoretical curve.

short- and long-range forces. The potential affecting the motion of the atoms and the frequency of the total symmetric stretching mode in the mixed crystals is calculated starting from the potential in the pure YAG and YSGG structures. The Born-Mayer potential was applied to model the short-range repulsive interaction potential.^{10,17} The potential is of the form

$$V^{\text{SR}}(|r(kk')|) = V_0 e^{-(|r(kk')| - |r(kk')|)/(\sigma_k + \sigma_{k'})},$$

where $r(kk')$ is the position vector of the k atom in kk' primitive cell, and V_0 and $\sigma_k + \sigma_{k'}$ are the parameters describing the strength and the range of the repulsive interaction, respectively. The potential depends on the distance between the central atom and the nearest neighbors.

In the approximation of free tetrahedron for the A_{1g} vibration mode, it is possible to consider only the motion of the oxygen atoms with respect to the central cation; a simplified form of the potential of the pure YAG or YSGG crystal¹⁷ is

$$V_{\text{cryst}} = \sum_j A_{O-j} e^{-b_{O-j}/r_{O-j}}, \quad (1)$$

where the terms A_{O-j} and b_{O-j} represent the strength and the range of the repulsive interaction between the oxygen and the other atoms of the tetrahedron, respectively. For a given pair of atoms, A_{O-j} and b_{O-j} are constant values and can be calculated from the single atom parameters:¹⁷ $A_{O-j} = (A_j A_O)^{1/2}$ and $b_{O-j} = (b_j + b_O)/2$. By using the harmonic approximation, this potential can be applied to calculate the vibrational frequencies of the breathing mode of a free tetrahedron. Starting from the values reported by Abrahamson¹⁷ for the interaction of free atoms, the parameters for the ions constituting the YAG and YSGG structures were calculated by fitting the A_{1g} symmetric stretching in the two garnets.

In the rigid ion model framework the atoms can be considered as charged point interacting with short-range forces, where the force constant is related to the second derivative of the potential

$$k = \frac{d^2 V_{\text{Mayer}}(r)}{dr^2} \quad (2)$$

By using the expression (1) for the Born-Mayer potential and the harmonic approximation to fit the frequency of the A_{1g} stretching mode the A_{O-j} and b_{O-j} parameters were calculated for the YAG and YSGG garnet structures. The obtained values are $b_{O-Al} = 3.0 \text{ \AA}^{-1}$ and $b_{O-Ga} = 2.4 \text{ \AA}^{-1}$ and 736 eV and 1520 eV for A_{O-Al} and A_{O-Ga} , respectively. It can be noted that the parameters calculated by Abrahamson for the free atoms are larger than the values here calculated: indeed in the present model the atoms are not free but linked in the tetrahedral structure. In addition one should consider that the position of the A_{1g} stretching mode is fitted by assuming the tetrahedron as isolated while it is linked into the garnet structure.

As previously discussed, the mixed structure is realized by gradually substituting the Al cations of YAG structure with the Sc and Ga cations of the YSGG, without changing the crystalline structure and the unit cell symmetry. Then it can be hypothesized that the interaction potential for the mixed garnet can be expressed as a linear combination of the interaction potentials of the two separated phases. The obtained parameters b_{O-j} and A_{O-j} were used to calculate the A_{1g} frequency in the mixed garnets as a function of the relative concentration of the two compounds (Fig. 4),

$$V_{\text{mix}} = [cA_{O-YAG}^0 + (1-c)A_{O-YSGG}^0] \times e^{-[cby_{YAG} + (1-c)by_{YSGG}]} [ery_{YAG} + (1-c)ry_{SGG}] \quad (3)$$

where c ($0 < c < 1$) is the relative concentration of the YAG phase in the mixed structures.

The very good agreement between the experimental data and the theoretical curve reported in Fig. 4 indicates that the proposed free tetrahedron model is able to represent both the single and mixed structures allowing to predict the A_{1g} frequency for a given relative concentration of the garnets. On the other hand, the reported investigation clearly shows how the Raman spectroscopy is a powerful diagnostic tool which can be applied to verify the quality of the grown crystal: the peak position of the A_{1g} frequency can be analyzed to assess the effective mixing of the two garnets and the relative concentration of the grown crystal.

IV. CONCLUSIONS

By means of Raman spectroscopy the analysis of the vibrational properties of yttrium aluminum garnet (YAG), yttrium scandium gallium garnet (YSGG), and mixed $YAG_x/YSGG_{1-x}$ structures ($0 < x < 1$) was carried out. By comparison with the vibrational spectrum of the YAG structure, the attribution of the Raman modes of the YSGG is proposed. The formation of mixed structures was verified by analyzing the spectra within the isolated polyhedra framework. The Raman spectra of the mixed structures do not show the presence of new vibrational bands: the modes of mixed structures lie between those of the pure garnets indicating the presence of truly mixed and not separated phases. In the approximation of free polyhedra, the position of the vibration frequencies of the A_{1g} tetrahedron breathing mode as a function of the relative concentration of the garnets was investigated by modeling the interaction potential as a linear combination of the contribution due to the pure structures. The proposed analysis allows to verify both the quality of the grown crystal and to assess the relative concentration of the garnets.

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