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J.J.CARROLL & T.A. GOLDMAN
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OPTICAL CHARACTERIZATION OF INTER-STARK ENERGY LEVELS AND EFFECTS OF TEMPERATURE ON SHARP EMISSION LINES OF Nd^{3+} in $CaZn_2Y_2Ge_3O_{12}$

Dhiraj K. Sardar and Raylon M. Yow
Division of Earth and Physical Sciences
The University of Texas at San Antonio
6900 N Loop 1604 West
San Antonio, Texas 78249

Abstract

A characterization of the Stark components of the $^4I_{9/2}$ and $^4I_{11/2}$ manifolds have been performed using the room temperature fluorescence spectra for the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ and $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transitions of Nd^{3+} in $CaZn_2Y_2Ge_3O_{12}$ laser crystal. The temperature dependencies of the linewidths and line shifts of the inter-Stark transitions of the 939.4 ($R_1 \rightarrow X_5$) and 1058.8 ($R_1 \rightarrow Y_1$) nm lines within the respective intermanifold transitions of $^4F_{3/2} \rightarrow {}^4I_{9/2}$ and $^4F_{3/2} \rightarrow {}^4I_{11/2}$ have been also investigated. The linewidths of these transitions were found to increase with increasing temperature. The 939.4 nm line shifted toward the shorter wavelength, while the 1058.8 nm line shifted toward the longer wavelength. The experimental results of the temperature-dependent widths and shifts of these emission lines are explained using the phonon-ion interaction theory which assumes the Debye model for phonons in crystalline solids.

I. INTRODUCTION

In the past, several investigations on a number of germanate laser hosts have shown that these materials possess better spectroscopic and laser characteristics over the conventional laser material such as Nd:YAG.[1-4] Unfortunately, most of these materials were unable to tolerate the thermal stress produced by the UV radiation present in the flashlamp. However, as the efficient and high power diode lasers operating at about 800 nm which corresponds to a strong Nd^{3+} absorption band, have become more readily available, there has been a renewed interest in the germanate laser hosts.

Because of the potential of the garnet structured calcium zinc yttrium germanate doped with trivalent neodymium ions, $CaZn_2Y_2Ge_3O_{12}$ (CAZGAR):Nd^{3+}, as a laser material, a thorough investigation of the spectroscopic and laser properties has been performed on this host recently by Sardar et al. and it has been shown that this material possesses some excellent spectroscopic and laser characteristics.[5,6] Recent studies on this host show that the broader absorption band near 800 nm is suitable for diode-pumping and the longer fluorescence lifetime of about 300 μs is capable of storing higher energy for Q-switch operation.[5] These properties along with the larger branching ratio of 40% for the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ transition of Nd^{3+} in CAZGAR are significant improvements over Nd^{3+}:YAG as a 0.94 μm laser.[6]

In this study, a detailed characterization of energy levels of Nd^{3+} ions in CAZGAR for the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ and $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transitions has been performed using the well-resolved, room temperature fluorescence spectra. For laser action, the thermal broadening and shift of the laser line have a significant meaning since these are closely related to the light amplification gain, output frequency stability, and thermal tunability of the laser. Owing to the potential of Nd^{3+}:CAZGAR as a laser material, the temperature dependencies of widths and shifts of the 939.4 and 1058.8 nm lines for the corresponding inter-Stark transitions of $R_1 \rightarrow X_5$ and $R_1 \rightarrow Y_1$ within the $^4F_{3/2} \rightarrow {}^4I_{9/2}$ and $^4F_{3/2} \rightarrow {}^4I_{11/2}$ intermanifold transitions, respectively, of Nd^{3+} ions in this host have also been investigated.

II. EXPERIMENTAL DETAILS

The samples used in this study were single crystals of Nd^{3+}:CAZGAR grown by A. Linz at the MIT Crystal Physics Laboratory using a top-seeded solution technique.[7] These samples were doped with 2 wt. % of Nd^{3+} in the melt.

The fluorescence spectra were measured on the spectroscopic sample at temperatures ranging from 10 to 300 K, by exciting the sample with the 514.5 nm line from a Spectra Physics model 2005 argon ion laser.

III. RESULTS AND DATA ANALYSIS

A total of 10 emission bands have been identified in the $^4F_{3/2} \rightarrow {^4I_{9/2}}$ transition and a total of 12 emission bands are observed in the $^4F_{3/2} \rightarrow {^4I_{11/2}}$ transition. The 939.4 nm line in the $^4F_{3/2} \rightarrow {^4I_{9/2}}$ transition and the 1058.8 nm line in the $^4F_{3/2} \rightarrow {^4I_{11/2}}$ transition are the most intense and well-resolved lines in their respective emission spectra.

The detailed characterization of the Stark energy levels of the $^4F_{3/2}$, $^4I_{9/2}$, and $^4I_{11/2}$ multiplet manifolds have been performed from the emission spectra for the $^4F_{3/2} \rightarrow {^4I_{9/2}}$ and $^4F_{3/2} \rightarrow {^4I_{11/2}}$ transitions of Nd^{3+} in CAZGAR at 300 K. The energy level diagram of the Stark components of the $^4F_{3/2} \rightarrow {^4I_{9/2}}$ and $^4F_{3/2} \rightarrow {^4I_{11/2}}$ transitions are shown in Fig. 1 The sharp, intense 939.4 nm line for the $R_1 \rightarrow X_5$ transition and the 1058.8 nm line for the $R_1 \rightarrow Y_1$ transition have been chosen to perform the measurements of linewidths and line shifts as a function of temperature.

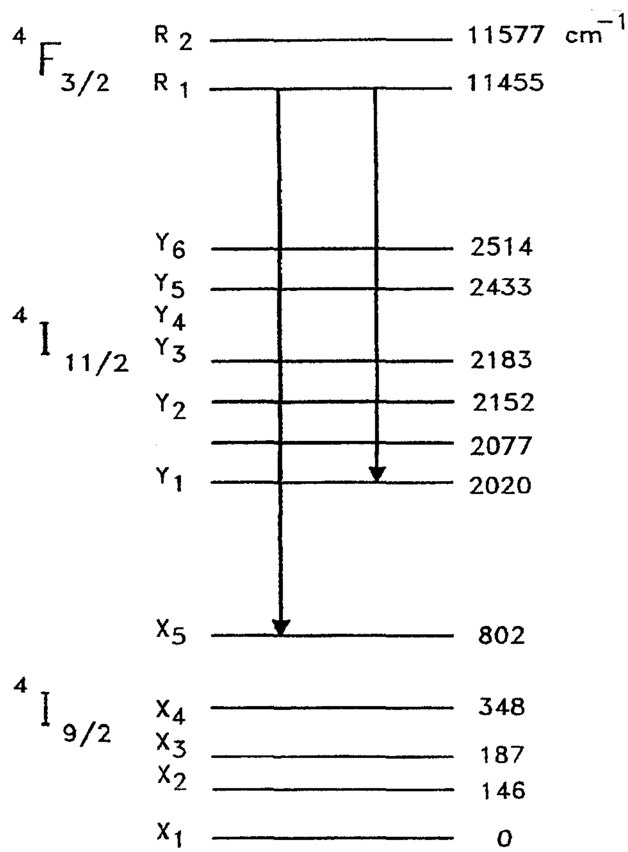

Fig. 1 Energy Level Diagram of Stark Components of the $^4F_{3/2} \rightarrow {^4I_{9/2}}$ and $^4F_{3/2} \rightarrow {^4I_{11/2}}$ Transitions

The fluorescence lines for the $R_1 \rightarrow X_5$ (939.4 nm) and $R_1 \rightarrow Y_1$ (1058.8 nm) transitions of Nd^{3+} in CAZGAR at 10 and 300 K are shown in Fig. 2. The intensities of these transitions are normalized to an arbitrary unit of 10. The temperature dependencies of width and shift of the sharp fluorescence lines are clearly observed in both transitions.

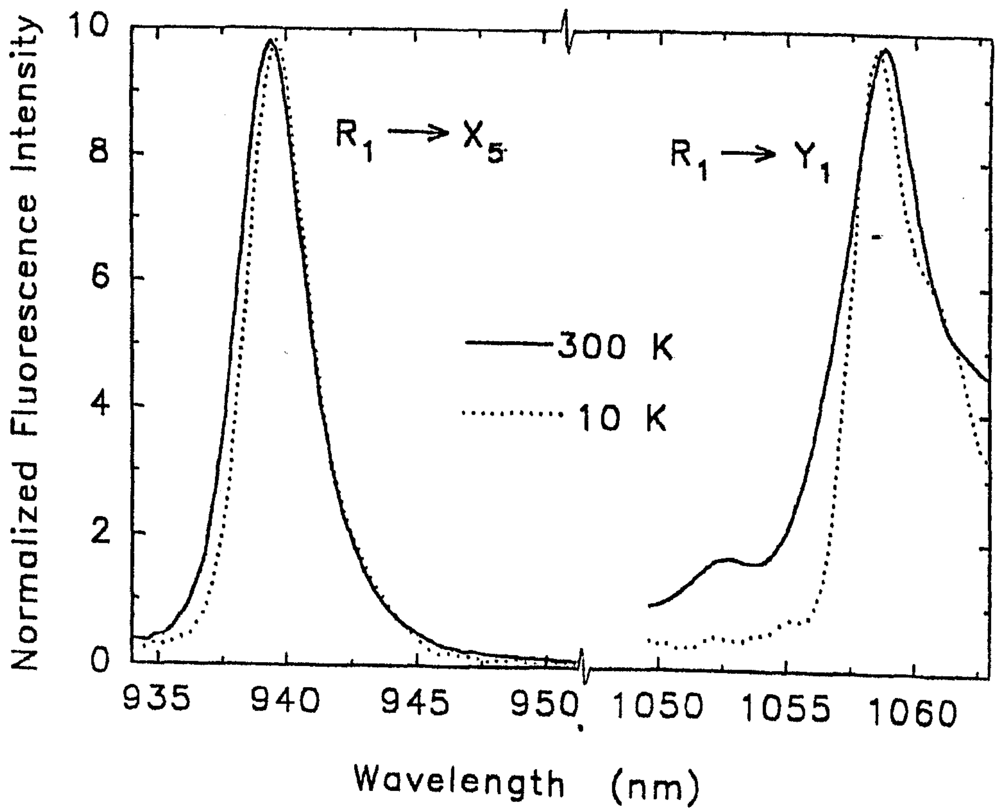

Fig. 2 Fluorescence Lines for $R_1 \rightarrow Y_1$ and $R_1 \rightarrow X_5$ Transitions as Function of Temperature

According to the presently available theory,[8-15] the linewidth of the inter-Stark transitions arises from (1) the crystal strain inhomogeneity, (2) direct one-phonon processes, (3) multiphonon processes, and (4) Raman phonon scattering processes. Even at the lowest temperature, the measurable linewidth of a radiative transition is in general accounted for the microscopic strains of the host crystal, which is inhomogeneous due to their random nature and therefore gives rise to a Gaussian line shape. Furthermore, as the temperature of the crystal is increased, the fluorescence linewidths of the Nd^{3+} ions increase.[12] In this article, we will consider only the temperature-dependent line broadening mechanism which arises from the interaction between ion and the crystalline host lattice vibrations. Therefore, the width (in cm^{-1}) of the *i*th energy level is governed by Raman multiphonon process. The Raman scattering process consists of the absorption of one phonon and the emission of another phonon without changing the electronic state of the ion and the width of the *i*th energy level is given by the following expression:[8]

$$\Delta \nu(T) = \Delta \nu_0 + \bar{\alpha} \left(\frac{T}{\theta_D} \right)^7 \int_0^{\frac{\theta_D}{T}} \frac{x^6 e^x}{(e^x - 1)^2} dx \quad , \qquad (1)$$

where $\Delta \nu_0$ is the temperature independent linewidth, $\bar{\alpha}$ is the coupling coefficient for the phonon-ion interaction , and Θ_D is the effective Debye temperature of the phonon distribution. The thermal broadening due to the direct one-phonon, multiphonon relaxation, and Raman phonon scattering processes is homogeneous and therefore yields a Lorentzian lineshape in the temperature range investigated. The measured linewidths for the $R_1 \rightarrow X_5$ and $R_1 \rightarrow Y_1$ transitions are plotted as a function of temperature in Fig. 3. The experimentally measured data are given in the table along with the fitting parameters.

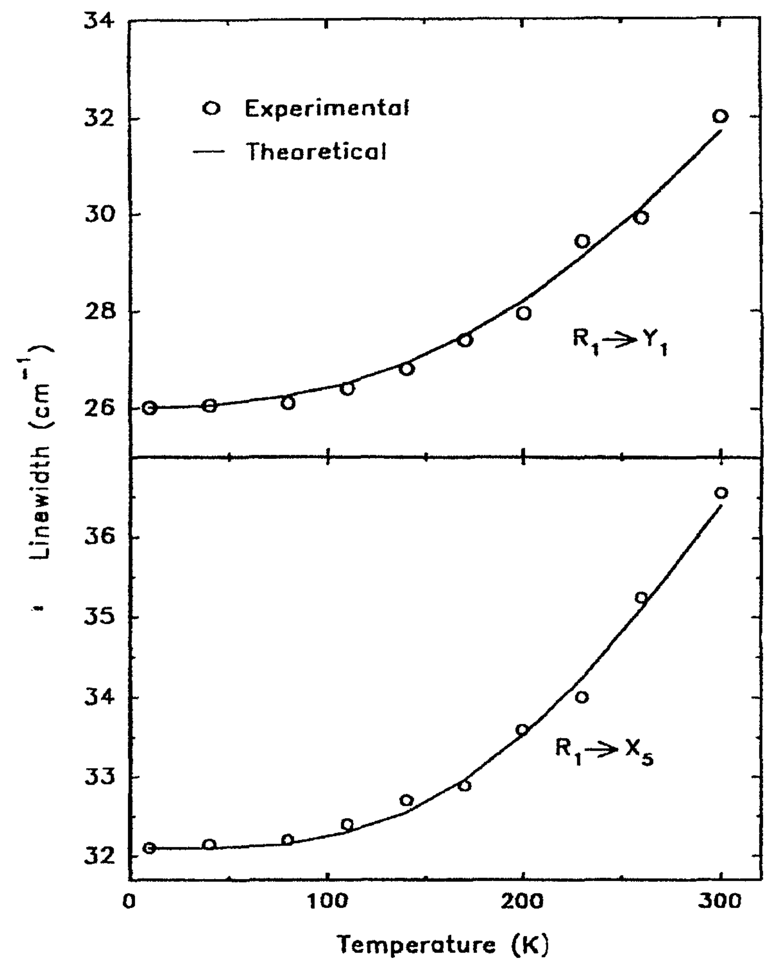

Fig. 3 Linewidth for $R_1 \rightarrow Y_1$ and $R_1 \rightarrow X_5$ Transitions as Function of Temperature

The 939.4 nm line position was found to shift toward the shorter wavelength, whereas the 1058.8 nm line shifted toward the longer wavelength as the crystal temperature was increased from 10 to 300 K. According to the phonon theory,[10-15] the thermal shift of a given transition is due to stationary effects of the phonon-ion interaction. In order to compare the thermal shift of the sharp fluorescence line, it is assumed that the thermal shift of a spectral line is the algebraic sum of the shifts of the two levels involved in the transitions. Therefore, the simplified theoretical expression for the line shift can be written in the following form:[8]

$$\delta\nu(T) = \delta\nu_0 + \alpha\left(\frac{T}{\Theta_D}\right)^4 \int_0^{\frac{\Theta_D}{T}} \frac{x^3}{e^x - 1}\,dx \qquad (2)$$

where $\delta\nu_0$ (in cm^{-1}) = ν(0 K) - ν(10 K) and ν(0 K) was obtained by extrapolating the experimental line position data to 0 K. The measured line shifts for the $R_1 \rightarrow Y_1$ transition are plotted as a function of temperature in Fig. 4. The fitting parameters are given in the table.

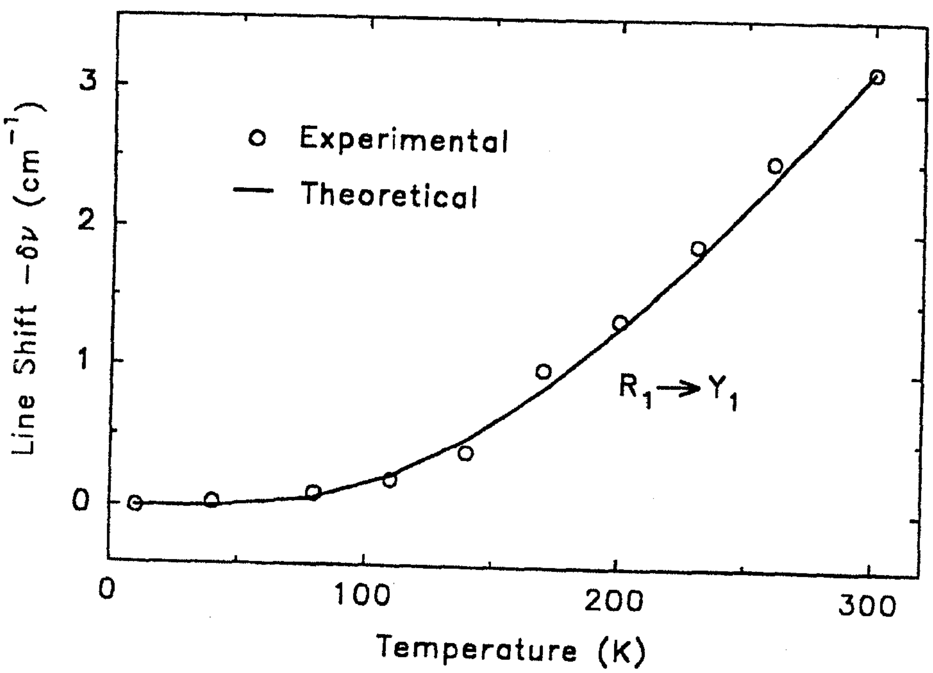

Fig. 4 Line Shift for $R_1 \rightarrow Y_1$ Transition as Function of Temperature

Table Experimental linewidth and line shift data and fitting parameters for the $R_1 \rightarrow X_5$ and $R_1 \rightarrow Y_1$ transitions of Nd^{3+} in CAZGAR.

	Experimental			Fitting Parameters					
	Linewidth $\Delta\nu$ (cm^{-1})		Line Shift $\delta\nu$(cm^{-1})	Linewidth			Line Shift		
Transition	10 K	300 K	10 - 300 K	Θ_D (K)	α (cm^{-1})	$\Delta\nu_o$(cm^{-1})	Θ_D (K)	α (cm^{-1})	$\delta\nu_o$(cm^{-1})
$R_1 \rightarrow X_5$	32.1	36.6	2.3	710	150	32.1	-	-	-
$R_1 \rightarrow Y_1$	26.0	32.0	-3.2	710	160	26.0	710	60	0

IV. CONCLUSION

Using the well-resolved, room temperature fluorescence spectra for the ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ and ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transitions of Nd^{3+} in CAZGAR, a detailed Stark components characterization has been performed for the three participating manifolds. The widths and line shifts of the sharp lines of the respective $R_1 \rightarrow X_5$ and $R_1 \rightarrow Y_1$ inter-Stark transitions have been investigated as a function of temperature. The experimental results have been quantitatively verified with the existing phonon-ion interaction theory which assumes the Debye phonon distribution in solids.

The sharp spectral lines in crystalline solids are usually observed to shift toward the longer wavelengths with increasing crystal temperature. The red shift of the 1058.8 nm line was fitted well with the theoretical expression given by Eq. (2). However, the blue shift of the 939.4 nm line with increasing temperature could not be fitted with the available theoretical expressions. It is worth noting that the blue shift has been observed in several other samples.[8,9] The blue shift of the $R_1 \rightarrow X_5$ transition may be attributed to the lowering of the terminal level X_5, the uppermost Stark-level of the ${}^4I_{9/2}$ manifold, due to the downward pushing by the upper Stark levels of the ${}^4I_{11/2}$ manifold.[10] Since the downward movement of the X_5 level with increasing temperature is not clear at this time, further studies on this potential laser host are needed in order to have a better understanding of the physical mechanisms of the blue shift.

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