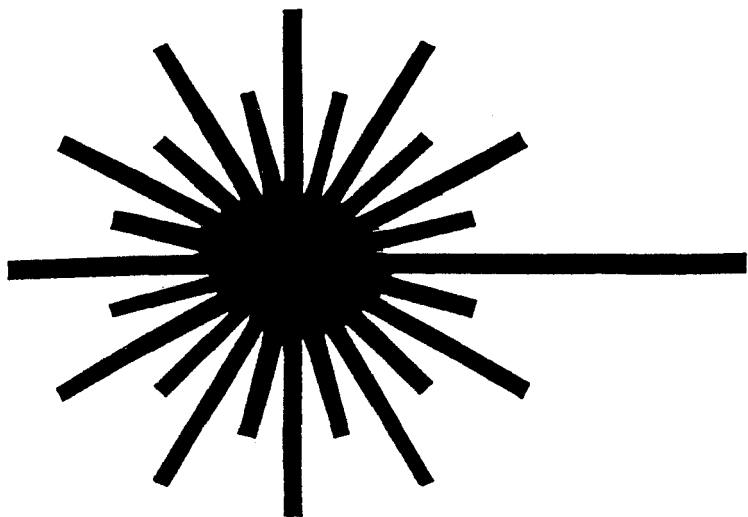


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Phonon effects on sharp spectral lines for inter-Stark transitions of Nd³⁺ in Sr₅(VO₄)₃F (S-VAP)

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ABSTRACT

A detailed investigation of phonon effects on the widths, positions, and shapes of sharp inter Stark transitions of Nd³⁺ in Sr₅(VO₄)₃F has been performed. The sharp lines studied correspond to the inter-Stark transitions of R₁ - X₁ (886.2 nm) and R₁ - X₅ (942.8 nm) within the ⁴F_{3/2} - ⁴I_{9/2} intermanifold transition, and of R₁ - Y₁ (1065.2 nm) within the ⁴F_{3/2} - ⁴I_{11/2} intermanifold transition. The widths of these lines and their shifts have been measured as a function of temperature. The linewidths are found to increase with increasing temperature of the host material and the spectral lines shift to the longer wavelength with increasing temperature. The theory of phonon-ion interactions has been employed to explain the temperature dependencies of the linewidths and line shifts. These interactions give rise to phonon relaxation processes which are responsible for the broadening and shifts of the spectral lines.

I. INTRODUCTION

The laser crystal, Nd³⁺:Sr₅(VO₄)₃F, also known as Nd:S-VAP, has become a subject of intense study for the past several years, because of its excellent spectroscopic and laser properties.^{1,4} The strong absorption of Nd:S-VAP at 809 nm is particularly important for efficient pumping by the currently available high power laser diodes or appropriate tunable solid state lasers. This newly developed material possesses unique laser properties, e.g., strong, highly polarized, and very narrow line emission at 1.06 μm, with a high gain cross section.¹ The radiative lifetime of the upper laser level, ⁴F_{3/2}, has been predicted to be 248 μs by the Judd-Ofelt theory and the fluorescence lifetime of this level has been measured 230 μs,² which is adequate for Q-switching and many other applications that require higher energy storage. In addition to the 1.06 μm emission from the ⁴F_{3/2} - ⁴I_{11/2} transition, Nd³⁺:S-VAP exhibits another intense and well-defined emission around 0.94 μm due to the ⁴F_{3/2} - ⁴I_{9/2} transition. The branching ratio of the ⁴F_{3/2} - ⁴I_{9/2} transition has been found to be 58% compared to 37% for the ⁴F_{3/2} - ⁴I_{11/2} transition.² Owing to the higher energy storage capability for Q-switching operation and the larger branching ratio of the ⁴I_{9/2} transition, Nd³⁺:S-VAP can be an excellent candidate for a 0.9 μm, Q-switched laser system. Although a number of excellent studies have been performed on the spectroscopic and laser properties of this material, to our knowledge, there has been no investigation on the thermal effects on the sharp spectral lines of Nd³⁺ in S-VAP laser host. The thermal broadening and shift of the sharp laser lines have profound significance, because these properties are closely related to gain, output frequency stability, and thermal tunability of the laser.

A detailed characterization of the temperature dependencies of widths, shifts, and positions of the fluorescence lines due to the R₁ - X₁ (886.2 nm), R₁ - X₅ (942.8 nm), and R₁ - Y₁ (1065.2 nm) inter-Stark Transitions of Nd³⁺ ions in S-VAP host, as shown in Fig. 1,³ has been reported here. The first two inter-Stark transitions belong to the ⁴F_{3/2} - ⁴I_{9/2} and the last one belongs to the ⁴F_{3/2} - ⁴I_{11/2} intermanifold transitions. The observed temperature dependencies of the spectral widths and shifts of these lines are explained with the help of the existing theory of phonon-ion interaction as a perturbation.

II. EXPERIMENTAL RESULTS AND DISCUSSION

The spectral linewidths and line shifts of Nd³⁺ ions in S-VAP were measured in the temperature range from 10 to 300 K. Even at the lowest temperature, the linewidth of a radiative transition is in most crystalline solids accounted for the random microscopic strains due to the impurity ions in the host crystal.^{11,14} The statistical nature of the strains in solids is random, and therefore gives rise to an inhomogeneous broadening which predicts a Gaussian line shape. However, as the temperature of the impurity-doped solid increases, the optical linewidths of the ions invariably increase. The line broadening is caused by direct one phonon, multiphonon relaxation and Raman scattering processes.

Therefore, for the i th inter-Stark energy level, the net contribution to the linewidth (in cm^{-1}) due to the ion-phonon interaction is given by

$$\Gamma_i = \Gamma_i^D + \Gamma_i^M + \Gamma_i^R. \quad (1)$$

The first term Γ_i^D on the right hand side of Eq. (1) is due to direct or one-phonon process between the i th energy level and other nearby level. The second term Γ_i^M is the contribution from the multiphonon emission processes, which can take place between levels having energy difference greater than the greatest available phonon energy; and the transition probability per unit time has been shown to be temperature independent.⁸ Therefore, the multiphonon absorption processes are negligible in temperature ranges of interest.¹¹ The third term Γ_i^R is due to the Raman multiphonon process associated with phonon scattering by the impurity ions. 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 is given by the following expression:^{8,12}

$$\Gamma_i^R(T) = \bar{\alpha}_i \left(\frac{T}{\Theta_D} \right)^7 \int_0^{\frac{\Theta_D}{T}} \frac{x^6 e^x}{(e^x - 1)^2} dx, \quad (2)$$

where $x = h\nu/kT$ (here, ν is the phonon frequency, and h and k have their usual meaning), α_i is the coupling coefficient for the phonon-ion interaction, and Θ_D is the effective Debye temperature of the phonon distribution.

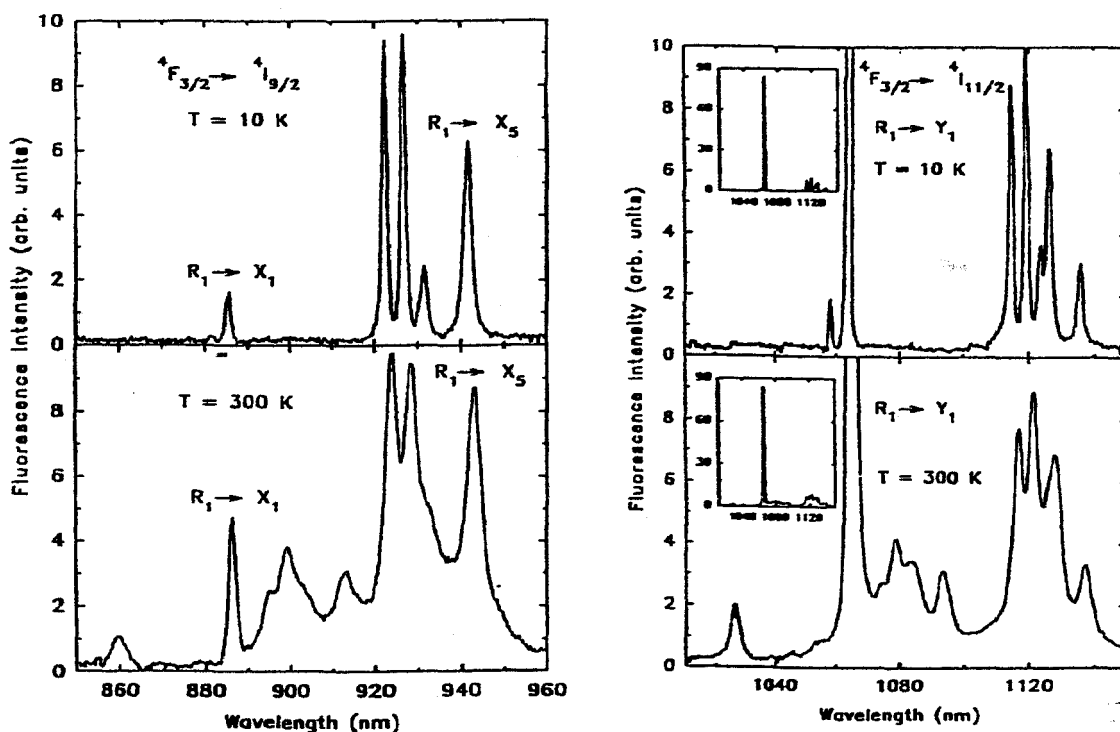


Fig. 1. Fluorescence spectra of Nd^{3+} in S-VAP for the ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ and ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transitions at 10 and 300 K.

If the total contribution to the linewidth due to microscopic strain, direct one-phonon, and multiphonon processes is assumed to be temperature independent, the spectral width, $\Gamma(T)$, of the line is, therefore, the sum of the energy spread of the two Stark energy levels involved in that particular transition, and can be written in the following form:

$$\Gamma(T) = \Gamma_0 + \bar{\alpha} \left(\frac{T}{\Theta_D} \right)^7 \int_0^{\frac{\Theta_D}{T}} \frac{x^6 e^x}{(e^x - 1)^2} dx \quad (3)$$

where Γ_0 is the temperature independent linewidth and α is the simplified coupling coefficient. The simplified model described by Eq. (3) has been successfully used in the past by many authors to explain their experimental results.¹¹⁻¹³ The thermal broadening due to the direct one-phonon, multiphonon relaxation, and Raman phonon scattering processes is homogeneous and therefore gives rise to a Lorentzian lineshape in the temperature range of our interest.

The temperature dependence of the line broadening and shift are clearly observed in these transitions. The strong, well-resolved inter-Stark transitions identified as $R_1 - X_1$ at 886.2 nm and $R_1 - X_5$ at 942.8 nm, and $R_1 - Y_1$ at 1065.2 nm in Fig. 1, were measured at temperatures from 10 to 300 K. These

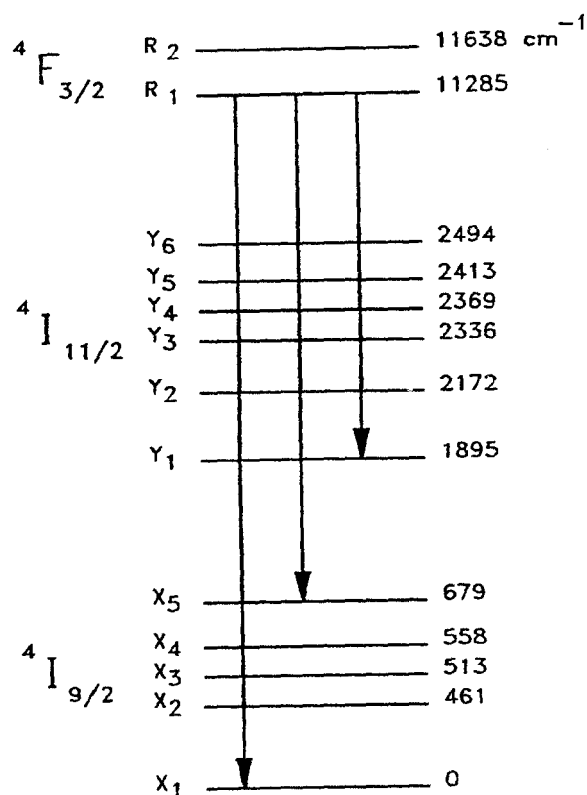


Fig. 2. Energy level diagram of Nd³⁺ in S-VAP for the inter-Stark transitions among the ⁴F_{3/2}, ⁴I_{9/2}, and ⁴I_{11/2} manifolds at 300 K.

inter-Stark transitions are also shown in Fig. 2.³ The measured widths Γ (in cm^{-1}) of the 886.2, 942.8, and 1065.2 nm lines are plotted as a function of temperature in Fig. 3. The circles represent the experimental data and the solid curves are the theoretical fit. The measured data were best fitted to Eq. (3) with the fitting parameters that are given in the Table I.

According to the phonon theory,^{5,10} thermal shift of a given transition is due to stationary effects of the phonon-ion interaction. In order to compare the experimental line shift, 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 given in the following form:^{11,12}

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

where $\delta\nu_0$ (in cm^{-1}) = $\nu(0\text{ K}) - \nu(10\text{ K})$ and $\nu(0\text{ K})$ was obtained by extrapolating the measured line position to absolute zero temperature.

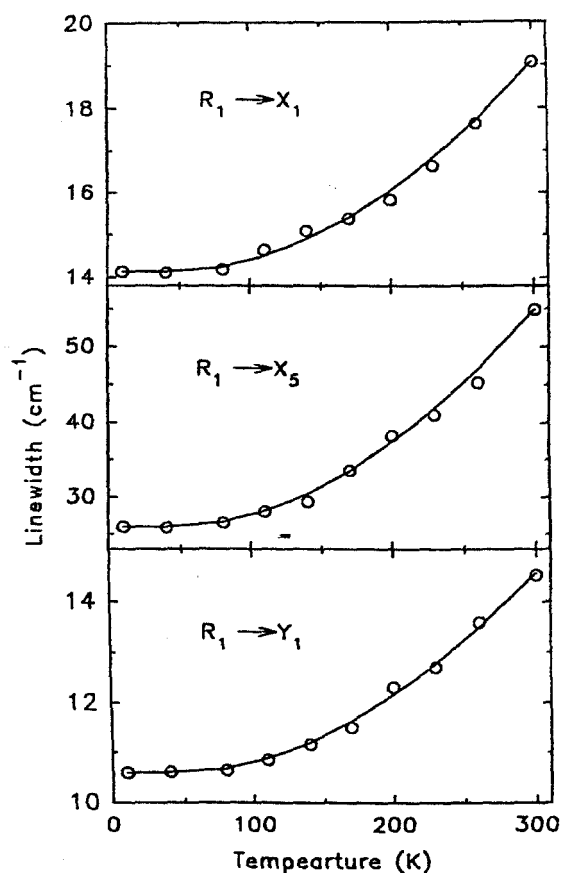


Fig. 3. The linewidths for the $R_1 \rightarrow X_1$, $R_1 \rightarrow X_5$, and $R_1 \rightarrow Y_1$ transitions of Nd^{3+} in S-VAP as a function of temperature (see text for explanation).

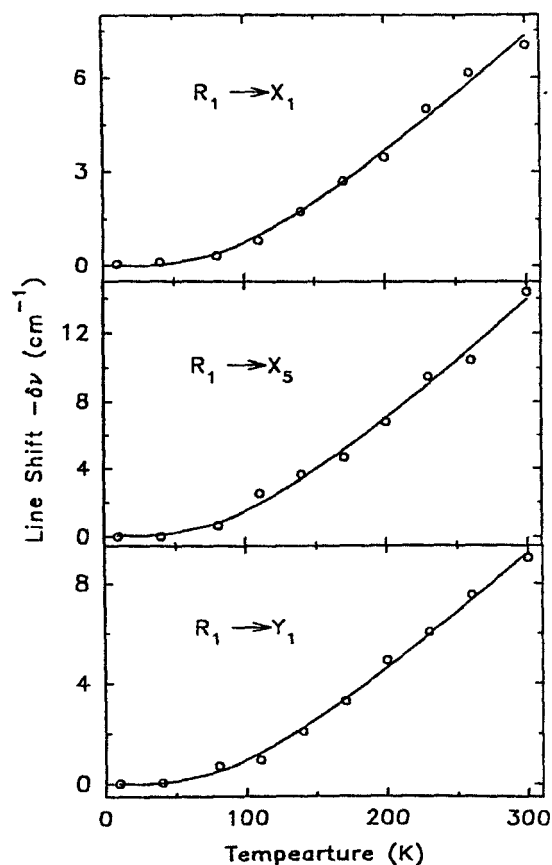


Fig. 4. The line shifts for the $R_1 \rightarrow X_1$, $R_1 \rightarrow X_5$, and $R_1 \rightarrow Y_1$ transitions of Nd^{3+} in S-VAP as a function of temperature (see text for explanation).

The measured line shifts for these transitions are plotted as a function of temperature in Fig. 4. The circles are the experimental data and the solid curves represent the theoretical fit. The measured line shifts have been best fitted to Eq. (4) with the fitting parameters given in Table I. All three lines were found to shift towards the longer wavelength with increasing temperature.

Table I. Some experimental linewidth and line shift data and fitting parameters for the R_1-X_1 , R_1-X_5 , and R_1-Y_1 transitions of Nd^{3+} in S-VAP.

Transition	Experimental			Fitting Parameters					
	Linewidth Γ (cm^{-1})		Line shift $\delta\nu$ (cm^{-1})	Linewidth			Lineshift		
	10 K	300 K	10 - 300 K	Θ_D (K)	α (cm^{-1})	Γ_0 (cm^{-1})	Θ_n (K)	α (cm^{-1})	$\delta\nu_0$ (cm^{-1})
R_1-X_1	14.1	19.1	-7.0	405	48.7	14.2	407	51.8	0.05
R_1-X_5	26.0	54.9	-14.4	409	294.5	25.9	409	98.8	0.01
R_1-Y_1	10.6	14.5	-9.0	411	42.0	10.6	402	63.3	0.02

III. SUMMARY

Using the well-resolved fluorescence spectra of the Nd^{3+} ${}^4F_{3/2} - {}^4I_{9/2}$ and ${}^4F_{3/2} - {}^4I_{11/2}$ manifold transitions in S-VAP laser host, the linewidths and line shifts of the inter-Stark transitions of $R_1 - X_1$ (886.2 nm), $R_1 - X_5$ (942.8 nm), and $R_1 - Y_1$ (1065.2 nm) have been investigated as a function of temperature. The experimental results have been quantitatively verified with the existing theory of phonon-ion interaction which assumes the Debye phonon distribution in solids. Although temperature-dependent one-phonon contribution has been ignored in this work, it would provide better fitting of the experimental data to the theory when this contribution is taken into account.¹⁷

The sharp spectral lines of the trivalent neodymium ions in crystalline solids are usually observed to shift toward the longer wavelengths with increasing crystal temperature. The observed red-shift for the 886.2, 942.8, and 1065.2 nm lines of Nd^{3+} with increasing temperatures in S-VAP host were found to agree well with the available theory. The fluorescence intensity of the 886.2 nm line at 10 K decreases by more than 50% of that at 300 K and the fluorescence intensity of the 942.8 nm line in the same manifold of ${}^4F_{3/2} - {}^4I_{9/2}$ decreases by about 50%, but the fluorescence intensity of the 1065.2 nm line in the ${}^4F_{3/2} - {}^4I_{11/2}$ manifold remains almost the same at both 10 and 300 K.

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REFERENCES

1. S.A. Payne, B.H.T. Chai, W.L. Kway, L.D. DeLoach, L.K. Smith, G. Lutts, R. Peale, X.X. Zhang, G.D. Wilke, and W.F. Krupke, Conference on Lasers and Electro-Optics (CLEO), Baltimore, postdeadline paper CPD12 (1993).
2. D.K. Sardar and P.D. Bella, J. Appl. Phys. **76**, 5900 (1994).

3. D.K. Sardar and P.D. Bella, Phys. Rev. B **55**, 2859 (1997).
4. P. Hong, X.X. Zhang, R.E. Peale, H. Weidner, M. Bass, and B.H.T. Chai, J. Appl. Phys. **77**, 294 (1995).
5. A. Kiel, Phys. Rev. **126**, 1292 (1962).
6. R.H. Silsbee, Phys. Rev. **128**, 1726 (1962).
7. R.J. Adler, Bull. Am. Phys. Soc. **7**, 600 (1962).
8. D.E. McCumber and M.D. Sturge, J. Appl. Phys. **34**, 1682 (1963).
9. G.F. Imbush, W.M. Yen, A.L. Schawlow, D.E. McCumber, and M.D. Sturge, Phys. Rev. **133**, A 1029 (1964).
10. D.E. McCumber, Phys. Rev. **133**, 163 (1964).
11. W.M. Yen, W.C. Scott, and A.L. Schawlow, Phys. Rev. **136**, A271 (1964).
12. X. Chen and B. Di Bartolo, J. Appl. Phys. **76**, 1710 (1994).
13. X. Chen and B. Di Bartolo, J. Lumin. **54**, 309 (1993).
14. A.L. Schawlow, A.H. Piksis, and S. Sugano, Phys. Rev. **122**, 1469 (1961).
15. T. Kushida, Phys. Rev. **185**, 500 (1969).
16. L.D. DeLoach, S.A. Payne, B.H.T. Chai, W.L. Kway, G. Loutts, Appl. Phys. Lett. **65**, 1208 (1994).
17. D.K. Sardar and S.C. Stubblefield, Phys. Rev. (to be submitted).