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ON THE ORIGINS OF HOMOGENEOUS AND INHOMOGENEOUS LINEWIDTHS OF  
THE  $^5D_0-^7F_0$  TRANSITION OF  $Eu^{3+}$  IN AMORPHOUS SOLIDS

by

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## ABSTRACT

Fluorescence line narrowing techniques have been used to measure the homogeneous widths of the  ${}^5D_0$ - ${}^7F_0$  transition of  $\text{Eu}^{3+}$  in a variety of glasses at room temperature. The homogeneous width is found to vary linearly with the modifier concentration for the borate glasses and also linearly with the transition frequency within the inhomogeneous profile in all of the glasses studied. These results, together with the observed changes in the inhomogeneous width in the borate glass with modifier concentration, are discussed in terms of dynamical processes associated with phonon modulation of the so-called two level systems in glasses, and the static chemical structure of the glass.

## I. INTRODUCTION

There are a number of properties of amorphous solids that differ from crystalline substances, yet which do not seem to depend on the detailed chemical identity of the amorphous material itself. The low-temperature heat capacity is dominated<sup>1-3</sup> by a term linear in temperature, and the low-temperature thermal conductivity of amorphous solids is insensitive to the presence of impurities and exhibits a  $T^2$  dependence. These low-temperature properties of amorphous solids have successfully been accounted for by a model which postulates the existence of a tunneling-type motion of atoms or molecules between two inequivalent minima. These are known as the "two level systems" (TLS) model of Anderson et al.<sup>4</sup> and Phillips.<sup>5</sup>

The optical properties of trivalent rare earth ions in amorphous solids have also recently been studied. The homogeneous linewidths of the intra f shell transitions of  $\text{Eu}^{3+}$  in a silicate glass are found to increase as  $T^{1.8 \pm 0.2}$  between 1.7 and 90K,<sup>6</sup> with a similar temperature dependence observed over the 100 to 350K temperature range.<sup>7,8</sup> One also finds  $\text{Pr}^{3+}$  in other glasses<sup>9</sup> exhibits a  $T^{1.9 \pm 0.2}$  dependence in the 10 to 300K temperature range. The low temperature behavior is different from that observed for crystals, and cannot be accounted for by the usual one- or two-phonon processes. Recently, Reinecke<sup>10</sup> proposed a mechanism for the homogeneous width involving the TLS which predicts an optical linewidth proportional to  $T$  at low temperatures. At the same time Lyo and Orbach<sup>11</sup> proposed a mechanism for the homogeneous width involving the TLS which predicts a quadratic temperature dependence for the optical linewidth at low temperatures. Both theories predict a linear temperature dependence at high temperatures.

Both of these theories for the homogeneous linewidth involve parameters which also are important for the inhomogeneous broadening. In order to

see if they are relevant, we have undertaken a systematic investigation of the homogeneous width of the  ${}^5D_0$ - ${}^7F_0$  transition of  $\text{Eu}^{3+}$  at ambient temperature in a number of different glasses, and as a function of the modifier concentration in the same (borate) glass.

Previous measurements<sup>6,7</sup> have shown that the value of the homogeneous linewidth of the same  $\text{Eu}^{3+}$  transition in a silicate glass changes smoothly as one probes different packets within the inhomogeneous line, increasing as the transition frequency increases across the inhomogeneous profile. A similar increase in the radiative decay rate with transition frequency has also been reported.<sup>12,13</sup> In the present work we have examined the variation in the homogeneous width with transition frequency for a number of glasses, and for the same type of glass with different modifier concentrations. We have found that the homogeneous width changes essentially linearly with both transition frequency ( $\nu$ ) as well as modifier concentration in the borate glasses. Qualitative discussion of these observations, as well as the variation of the inhomogeneous width with modifier concentration, in terms of the existing theories<sup>10,11</sup> of optical homogeneous linewidth in glasses and the glass structure, is given.

## II. EXPERIMENTAL TECHNIQUE

The silicate and calcium metaphosphate glasses were prepared by melting the corresponding constituents at 1200°C for 12 hours and 1000°C for 8 hours respectively. The melts were quenched at about 300°C and annealed for 2 hours at about 600°C with slow cooling. The  $\text{Na}_2\text{O}$  and  $\text{BaO}$  were obtained from their respective carbonates. The borate glasses were prepared by melting anhydrous  $\text{B}_2\text{O}_3$  with  $\text{Na}_2\text{CO}_3$  and  $\text{Eu}_2\text{O}_3$  according to the concentrations indicated at about 880°C for 10 hours, quenched to about 200°C, and annealed at about 490°C for 6 hours. After annealing the samples were checked with a polariscope.

Measurements of the homogeneous linewidths were carried out using laser fluorescence line narrowing techniques.<sup>14</sup> A Quanta-Ray DCR-1 Nd:YAG pumped pulsed dye laser with a spectral width of  $\sim 0.3 \text{ cm}^{-1}$  and a 7ns pulse width was used as the excitation source. The emission was dispersed by a 1M monochromator with a narrow slitwidth to give a total instrumental width of  $0.7\text{-}1.5 \text{ cm}^{-1}$ . Because the measurement of homogeneous fluorescence linewidths requires detection resonant with the excitation source, a special gated phototube<sup>15</sup> was used to block the scattered laser light. A Princeton Applied Research (PAR) Model 162 boxcar averager with a  $500 \mu\text{s}$  window and a 1 ms delay was used to signal average. Homogeneous widths were calculated from the observed widths using the approximation  $\Delta\nu_{\text{hom}} = \frac{1}{2} \Delta\nu_{\text{m}}$ , where  $\Delta\nu_{\text{m}}$  is the measured width after careful deconvolution of the instrumental width from the data.<sup>16</sup> Reported values are fwhm. All measurements were carried out at 298K.

The observed fluorescence line narrowed lineshapes were found to be Lorentzian for the samples whose results are reported here. In other samples at higher  $\text{Eu}^{3+}$  concentration, spectral diffusion resulted in distorted lineshapes. The results from these samples will not be used in our analysis.

Measurements of the inhomogeneous linewidths were done in the same manner as the homogeneous linewidths except that a mercury arc lamp with a 500nm UV pass filter was used instead of the laser as the excitation source. The inhomogeneous lineshapes had similarly asymmetrical shapes. The high energy sides fall off more slowly than the low energy sides. The inhomogeneous linewidths reported are half width at half maximum (hwhm) measured on the high energy half.

Both longitudinal and transverse ultrasonic velocities were measured at 5 MHz at room temperature, by a travel-time technique.<sup>17</sup>

### III. EXPERIMENTAL RESULTS

We present the results of the dependence of the homogeneous linewidth,  $\Delta\nu_{\text{hom}}$ , on the transition frequency for different oxide glasses, and on the added network modifier concentration in borate glasses. Fig. 1 exhibits the linear dependence of the homogeneous linewidth on the transition frequency within the inhomogeneous profile of the  ${}^5\text{D}_0\text{-}{}^7\text{F}_0$  transition of  $\text{Eu}^{3+}$  for a number of glasses at 298K. Table 1 gives the slope and intercept of the fit of the data to the form  $\Delta\nu_{\text{hom}}(\nu) = m(\nu - 17300 \text{ cm}^{-1}) + b$  where  $\nu$  is the transition frequency in wavenumbers and  $17300 \text{ cm}^{-1}$  corresponds to the center of the inhomogeneous profile in these glasses. The value of the slope is largest for the silicate glass, followed by the phosphate, and then the borate glass.

The dependence of the homogeneous and inhomogeneous linewidths on the mol% of  $\text{Na}_2\text{O}$ , acting as a modifier in the borate glasses, is exhibited in Fig. 2. The homogeneous linewidth at the peak ( $17300 \text{ cm}^{-1}$ ) of the  ${}^5\text{D}_0\text{-}{}^7\text{F}_0$  fluorescence line of  $\text{Eu}^{3+}$  is found to vary linearly with a slope of  $0.1 \text{ cm}^{-1}/\text{mol}\% \text{ Na}_2\text{O}$  over the concentration range in which a stable borate glass can be formed. The inhomogeneous width exhibits a double maximum structure, with maxima at 16 and 24 mol%  $\text{Na}_2\text{O}$  and a minimum at 20 mol%  $\text{Na}_2\text{O}$ .

The velocity of sound data are reported in Table 1. The longitudinal velocity is found to increase smoothly with the addition of  $\text{Na}_2\text{O}$  in the borate glasses up to  $\sim 30 \text{ mol}\% \text{ Na}_2\text{O}$ . The transverse velocities are roughly half those of the corresponding longitudinal velocities in all of the glasses measured.

The velocity of sound and homogeneous linewidth data reported in Table 1 show that both quantities increase with the addition of  $\text{Na}_2\text{O}$

in the borate glasses. This behavior is in marked contrast to the  $\Delta\nu_{\text{hom}} \sim v^{-2.5}$  dependence observed by Pelligrino et al.<sup>18</sup> for  $\text{Nd}^{3+}$  in glass hosts, where  $v$  is the rms sound velocity. However, they also report deviations for  $\text{B}_2\text{O}_3$  containing glasses from the behavior observed in all other glasses they studied.

#### IV. DISCUSSION

In the introduction we have mentioned the observed  $T^2$  dependence of the homogeneous linewidth for  $\text{Eu}^{3+}$  and  $\text{Pr}^{3+}$  in glasses over a wide range of temperatures.<sup>6-9</sup> This led to theoretical proposals<sup>10,11</sup> involving TLS to explain yet another unusual property of amorphous solids. It is expected that there will be a distribution of coupling parameters for the different oxygen atoms bonded to the different  $\text{Eu}^{3+}$  ions in the glass. We shall argue that an oxygen ligand could conceivably be part of a TLS, and that those TLS with dynamical motion contribute to the origin of the homogeneous linewidth. If the tunneling barrier for the TLS is very high, the phonon induced transition rate will be very small, and only a static contribution will result, which we associate with a significant part of the inhomogeneous width. The characteristic rate which separates the two contributions for the process described in Ref. 11 is the radiative or natural width. The origin of the homogeneous linewidth arises from TLS with smaller barriers. Phonon modulation of the individual energies of the TLS wells will phase interrupt the optical transition, giving rise to a homogeneous linewidth. The details of this process are given in Ref. 11.

One aspect of this model is that the strength of the coupling between the oxygens which are supposed to form part of the TLS (see below) and the

rare earth impurity is the same for the inhomogeneous linewidth and the homogeneous linewidth. The difference between these contributions primarily involves the time scales associated with phonon modulation of the energies of the two wells making up the TLS state. If this is true and everything else remains constant, the homogeneous linewidth should scale as the square of the inhomogeneous linewidth.<sup>11</sup> The explicit expression is

$$\Delta\nu_{\text{hom}} = \frac{(k_B T)^2 \eta D \langle V^2 \rangle}{96 \hbar^4 v^5 \rho B} \quad (1)$$

where  $\eta$  is a constant of order unity,  $D$  is related to an average of the phonon coupling to the two level systems (taken to equal  $0.5 \text{ (eV)}^2$  in Ref. 11),  $\langle V^2 \rangle$  is the average of the square of the coupling between the TLS and the rare earth impurity and is assumed to be on the order of the square of the inhomogeneous width,  $v$  is the acoustic phonon velocity,  $\rho$  is the mass density, and  $B$  is the TLS bandwidth. For our cases we have taken values of  $\rho$  and  $v$  either from the literature or from measurements we have made ourselves (see Table 1). We assume  $B = 0.1 \text{ eV}$ . We find for the silicate glass ( $v = 3.622 \text{ Km/sec}$ , the rms value from the data in Table 1) with  $\langle V^2 \rangle = (60 \text{ cm}^{-1})^2$ ;  $\Delta\nu_{\text{hom}} = 7.1 \text{ cm}^{-1}$ , in general agreement with the range of values  $4 - 8 \text{ cm}^{-1}$  measured by us as a function of wavelength.

In order to examine the predicted relationship between  $\Delta\nu_{\text{hom}}$  and  $\Delta\nu_{\text{inh}}$ , we must measure these quantities in different glasses. We find, however, that the homogeneous linewidth, taking the changes in the sound velocity into account, does not simply scale with the square of the inhomogeneous linewidth and inversely as the fifth power of the sound velocity as one changes the glass host, as would be predicted by Eq. (1). Instead, the other parameters in (1) appear to be changing by factors of two to three.

We believe that this shows that significant structural changes occur as we change the glass host. As a consequence we chose to focus on a single system, the borate glasses, and discuss the changes in inhomogeneous and homogeneous linewidth as a function of frequency for different concentrations of  $\text{Na}_2\text{O}$  modifier over the range of stability, 10 - 36 mol%. The results are shown in Fig. 2.

Fig. 2(a) shows that as the modifier concentration increases,  $\Delta\nu_{\text{hom}}$  in the borate glasses increases. NMR studies<sup>19</sup> have shown that the addition of modifier to the borate glasses changes the hybridization of the boron atoms from trigonal to tetrahedral. For each modifier oxygen added, two trigonally bonded borons (with oxygen and boron atoms lying in the same plane) become tetrahedrally bonded. The mole fraction of borons in tetrahedral coordination (R) is thus  $R = \frac{x}{100 - x}$ , where x is the mol% of  $\text{Na}_2\text{O}$ . This behavior is observed for  $\text{Na}_2\text{O}$  concentrations up to 40 mol%. This might suggest that oxygen atoms bonded tetrahedrally around the network forming atom give rise to a larger homogeneous width for the  $\text{Eu}^{3+}$  optical transition. This conclusion is supported from the results shown in Fig. 1 and Table I in which the silicate and phosphate glasses (with tetrahedral oxygen structure around Si and P respectively) have larger homogeneous widths than the borates at low modifier concentration. The observed linear dependence of the homogeneous linewidth on modifier concentration in the borate glasses may therefore be interpreted as an increase in the number of oxygen atoms tetrahedrally bonded to a boron. It is conceivable that multiple potential minima exist in the tetrahedral unit structure, or perhaps localized in the Eu-O-B bond and dependent on the bonding of the oxygen to a tetrahedrally bonded network former, and that this may be the origin of the TLS responsible for the modulation of the  $\text{Eu}^{3+}$  transition frequency.

Fig. 2(b) shows the dependence of the inhomogeneous width ( $\Delta\nu_{inh}$ ) on the  $\text{Na}_2\text{O}$  concentration in the borate glass. While the homogeneous linewidth and velocity of sound both show a smooth increase as one increases the  $\text{Na}_2\text{O}$  concentration, the inhomogeneous width shows a double maxima. This suggests that the direct relationship between  $\Delta\nu_{inh}$  and  $\Delta\nu_{hom}$  assumed in Ref. 11 may be more complex. While both  $\Delta\nu_{inh}$  and  $\Delta\nu_{hom}$  depend on the coupling between the  $\text{Eu}^{3+}$  ion and the surrounding oxygens,  $\Delta\nu_{inh}$  also depends on the site distribution in the glass, a fact that was not explicitly considered in Ref. 11. In order to verify this, we examine the behavior shown in Fig. 2(b) in terms of the expected variation in the site distribution in the borate glass with  $\text{Na}_2\text{O}$  concentration. Inherent to this interpretation will be the assumption that the site distribution, as reflected by the inhomogeneous width, is directly related to the uniformity of the glass structure.

The structural model of alkali-borate glasses proposed by Krogh-Moe<sup>20</sup> involves structural groupings larger than the basic  $\text{BO}_3$  and  $\text{BO}_4$  units and has been substantiated by a wide variety of measurements.<sup>21</sup> The behavior shown in Fig. 2(b) can thus be interpreted<sup>22</sup> by the known sequential appearance of three structural groupings in the glass: the boroxol ring and  $\text{BO}_3$  units at low  $\text{Na}_2\text{O}$  concentration, the tetraborate group at intermediate concentrations, and the diborate group at higher modifier concentration. For  $R = 0.10 - 0.20$  the boroxol network is disrupted by the presence of tetraborate groups. This gives rise to relatively more disorder, and the inhomogeneous width is expected to increase as shown. For  $R = 0.20 - 0.25$  the tetraborate group begins to dominate, giving rise to a relatively more "ordered" structure; the inhomogeneous width is observed to narrow, reaching a minimum at  $R \approx 0.25$  (corresponding to adding 20 mole percent of  $\text{Na}_2\text{O}$  and the observed minimum at this concentration in Fig. 2b) where the mole fraction of borons associated in tetraborate groups approaches unity. A similar process occurs at a higher modifier

concentration where the tetraborate network is disrupted by increasing numbers of diborate groups resulting in an increase in the inhomogeneous width. At the highest modifier concentrations used, the diborate group begins to dominate with a concomitant decrease in the inhomogeneous width (due to increase in the relative order).

Along the same line of reasoning one can explain the frequency dependence of the homogeneous linewidth, as exhibited in Fig. 1. As shown by Brawer and Weber,<sup>23</sup> the strength of the crystal field splitting of the  ${}^7F_1$  increases as one goes to higher frequency within the  ${}^5D_0$ - ${}^7F_0$  transition. Thus, packets excited at higher frequencies have larger crystal field strengths. If the TLS coupling strength tracks the crystal field strength, then excitation at higher frequency corresponds to larger  $\langle V^2 \rangle$  in (1), leading to larger homogeneous linewidths. Let  $V_0$  be the strength of TLS coupling at the low frequency edge ( $\nu_{\min}$ ) of the line, and take

$V(\nu) = V_0 + \delta V(\nu)$ ,  $V_0 > 0$ , as one moves to higher frequencies within the inhomogeneously broadened transition. We assume  $V_0 \gg \delta V(\nu_{\max})$ , where  $\nu_{\max}$  is the high frequency edge of the  ${}^5D_0$ - ${}^7F_0$  emission line, and that  $\delta V = V_1(\nu - \nu_{\min})$ ,  $V_1 > 0$ . This assumption is roughly borne out experimentally.<sup>18</sup> Then

$$\begin{aligned} \langle V(\nu)^2 \rangle &= \langle (V_0 + \delta V(\nu))^2 \rangle \\ &\approx \langle V_0^2 \rangle + 2\langle V_0 \delta V(\nu) \rangle \\ &= \langle V_0^2 \rangle + 2\langle V_0 V_1 \rangle (\nu - \nu_{\min}) \end{aligned} \quad (2)$$

We predict, therefore, that the homogeneous linewidth should increase linearly with increasing  ${}^5D_0$ - ${}^7F_0$  energy. The behavior in Fig. 1 is consistent with this picture. Though our arguments are rather qualitative, they do appear to be consistent with our observations, and those of Brawer and Weber.

Phase separation is known to occur in alkali-borate glasses<sup>24</sup> and may affect the spectral properties at high  ${}^5D_0-{}^7F_0$   $\text{Eu}^{3+}$  transition energies.<sup>13</sup> The smooth undistorted inhomogeneous lineshapes observed for the glasses used in this study suggest that the  $\text{Eu}^{3+}$  ion resides in a continuum of sites. Furthermore, our smooth linear frequency dependency of the homogeneous linewidth seems to continue in the energy region previously proposed to have spectral properties from the new phase. This might suggest that the homogeneous width is similar in this energy range for the two phases.

#### V. CONCLUSIONS

We have performed fluorescence line narrowing experiments on a variety of  $\text{Eu}^{3+}$  doped glasses, and on borate glasses with controlled changes in  $\text{Na}_2\text{O}$  modifier concentration. The importance of the tetrahedral unit structure of the glass network in determining the homogeneous width has been shown by the monotonic increase in the homogeneous width of a given  ${}^5D_0-{}^7F_0$  packet with increasing  $\text{Na}_2\text{O}$  concentration in the borate glasses. The variations in the inhomogeneous width in the borate glasses with the  $\text{Na}_2\text{O}$  concentration has been discussed in terms of the variation in the distribution of trigonal and tetrahedral units around the  $\text{Eu}^{3+}$  site. The linear frequency dependence of the homogeneous linewidth for different packets within the  ${}^5D_0-{}^7F_0$  inhomogeneously broadened line can be explained in terms of the variation of the TLS coupling constant with the crystal field

of the different sites. These explanations are consistent with experiment and exhibit the close relationship between the static structural and dynamical properties of glasses, as seen through the properties of optical impurity transitions.

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TABLE 1

Base glass composition (mol%)	Eu <sub>2</sub> O <sub>3</sub> (mol%)	m <sup>(a)</sup>	b <sup>(a)</sup> (cm <sup>-1</sup> )	v <sub>sound</sub> <sup>(b)</sup> (Km/sec)	
				longitudinal	transverse
15% Na <sub>2</sub> O, 5% ZnO 5% BaO, 75% SiO <sub>2</sub>	0.25	0.037 ± .004	4.90 ± .17	4.89 ± .05	2.78 ± .05
100% Ca(PO <sub>3</sub> ) <sub>2</sub>	1	0.025 ± .004	3.21 ± .17	4.77 ± .05	2.72
10% Na <sub>2</sub> O      90% B <sub>2</sub> O <sub>3</sub>	0.5	0.013 ± .003	1.43 ± .12	4.40 ± .05	2.38
12%            88%	0.5	0.015	1.49	4.50	2.22
14%            86%	0.5	0.011	1.68	4.69	2.37
16%            84%	0.5	0.015	1.77	4.75	2.42
18%            82%	0.5	0.014	2.03	4.89	2.68
20%            80%	0.5	0.013	2.17	4.94	2.60
24%            76%	0.5	0.012	2.67	5.11	2.98
30%            70%	0.5	0.015	3.34	5.31	2.95
36%            64%	0.5	0.021	4.14	5.27	3.05

(a) parameters of fit to  $\Delta v_{\text{hom}} = m(\nu - 17300 \text{ cm}^{-1}) + b$  where  $\nu$  is the transition frequency (cm<sup>-1</sup>) and 17300 cm<sup>-1</sup> corresponds roughly to the center of the inhomogeneous profile.

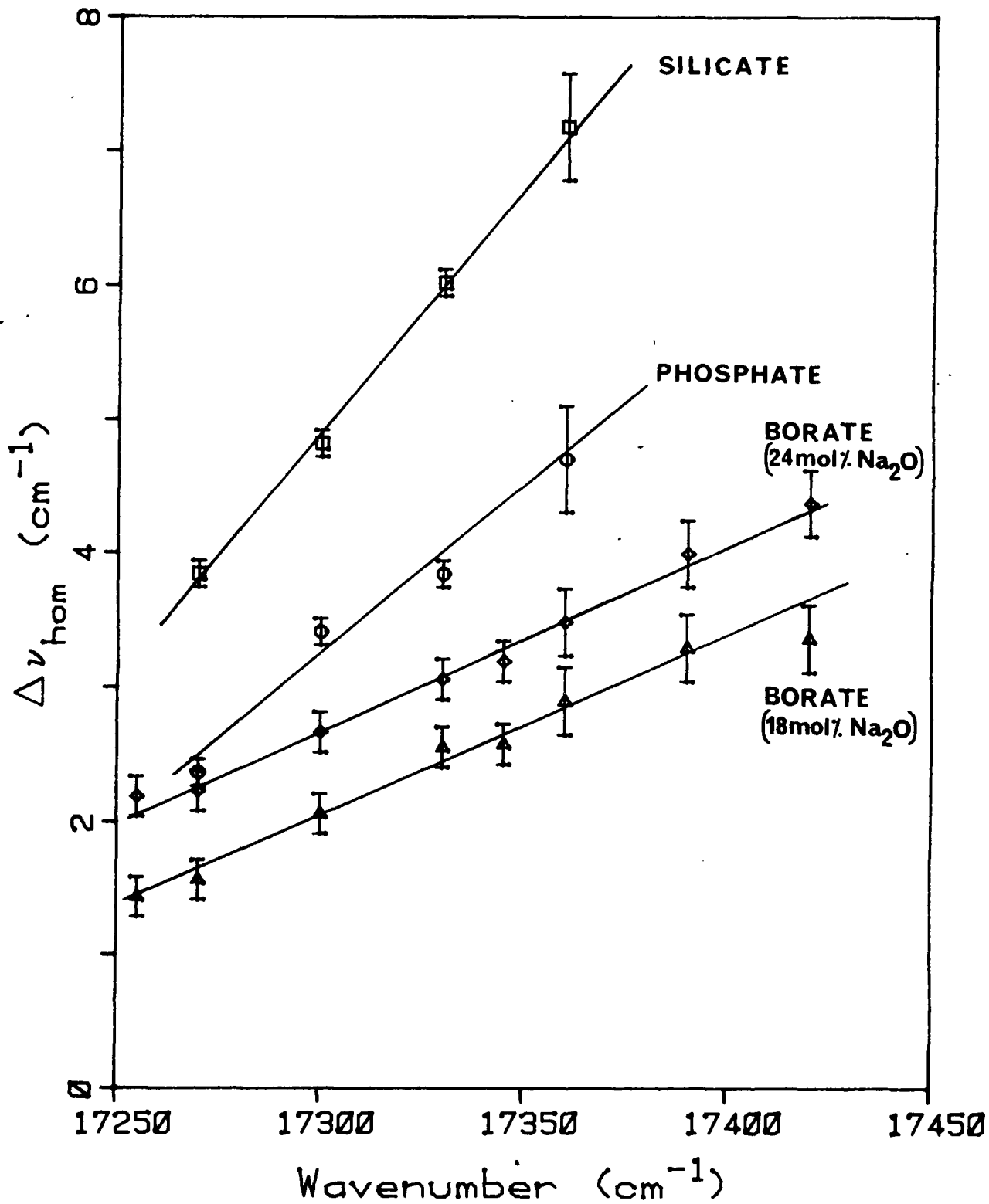
(b) sound velocity at 5 MHz and 298°K

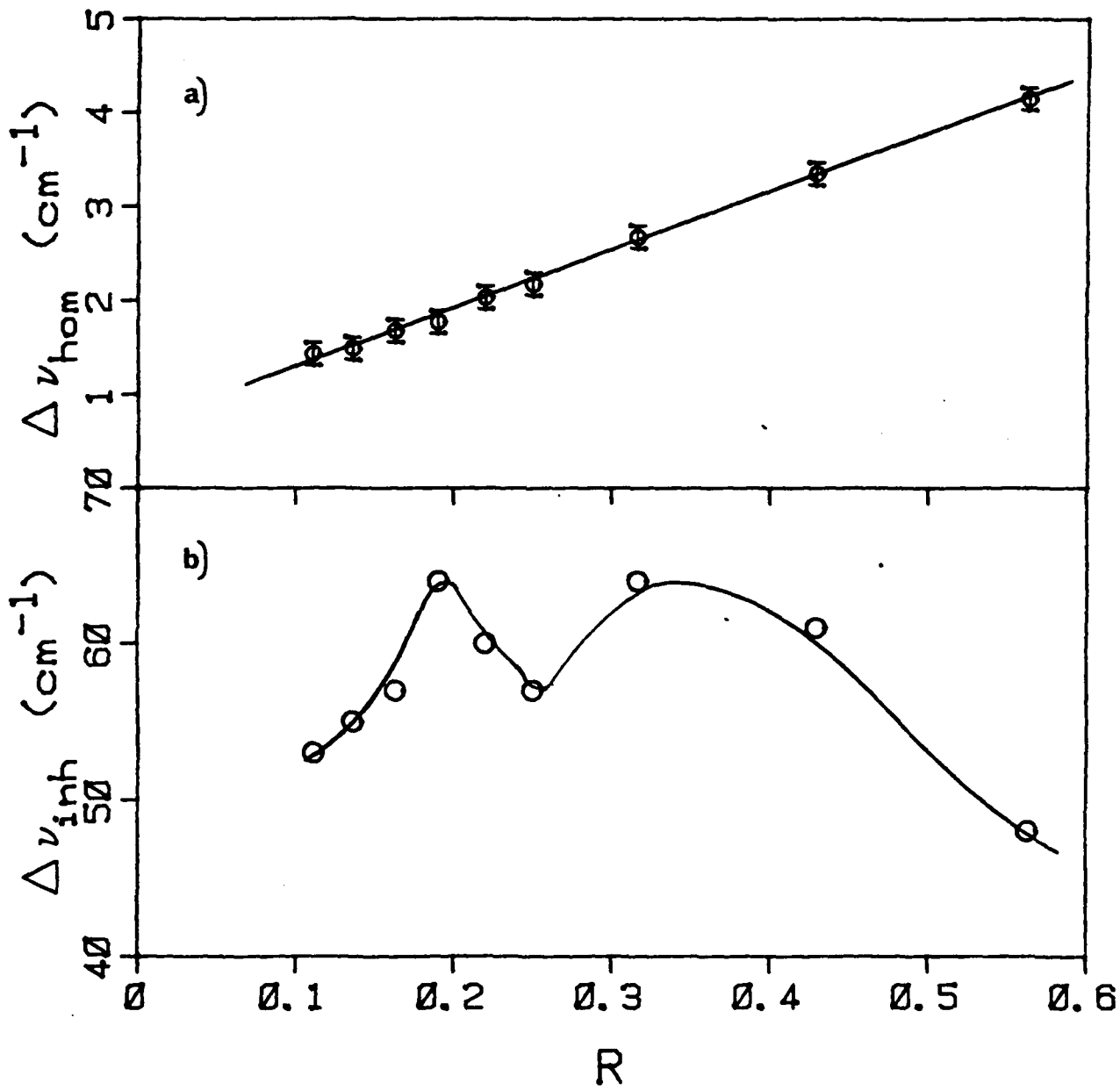
#### FIGURE CAPTIONS

Fig. 1 The linear relationship between the homogeneous width of the  ${}^5D_0-{}^7F_0$  transition of  $\text{Eu}^{3+}$  and the transition frequency within the inhomogeneous profile for several glasses. See Table 1 for glass compositions.

Fig. 2 Homogeneous and inhomogeneous linewidths of the  ${}^5D_0-{}^7F_0$  transition of  $\text{Eu}^{3+}$  in  $\text{B}_2\text{O}_3$  glasses as a function of the mole fraction of borons in tetrahedral coordination  $R = \frac{x}{100-x}$ , where  $x$  is the mol%  $\text{Na}_2\text{O}$  concentration.

- a) Homogeneous linewidth at  $17300 \text{ cm}^{-1}$
- b) Inhomogeneous width





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