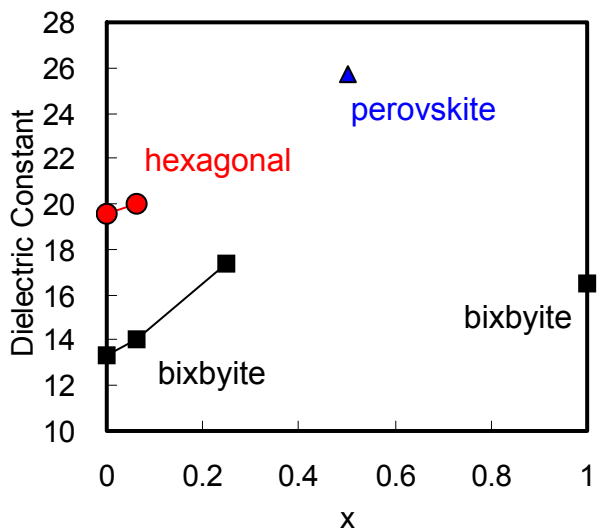
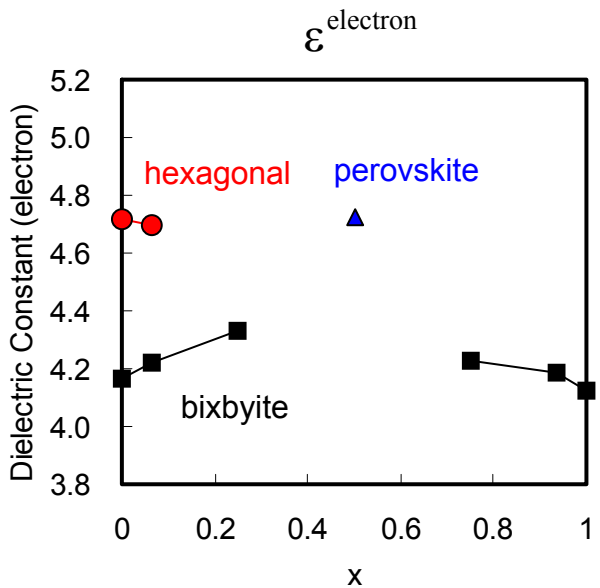
1. Dielectric Constant (ε) of $La_{2(1-x)}Sc_{2x}O_3$

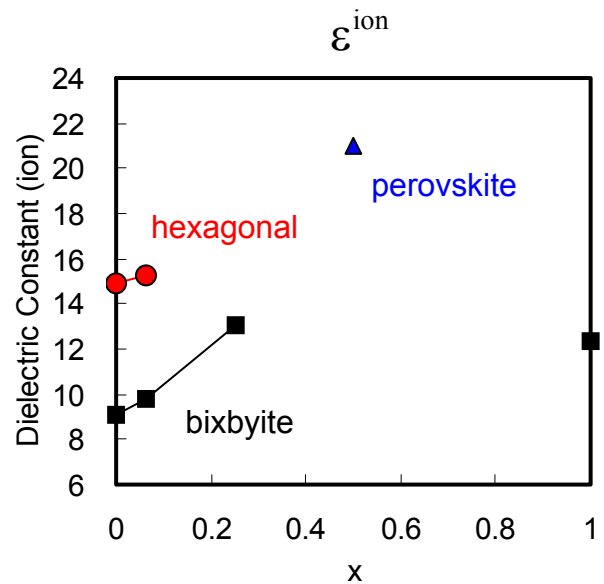
$$\varepsilon = \varepsilon^{electron} + \varepsilon^{ion}$$



Calculated dielectric constants increase with increasing Sc concentration. (x<0.5)

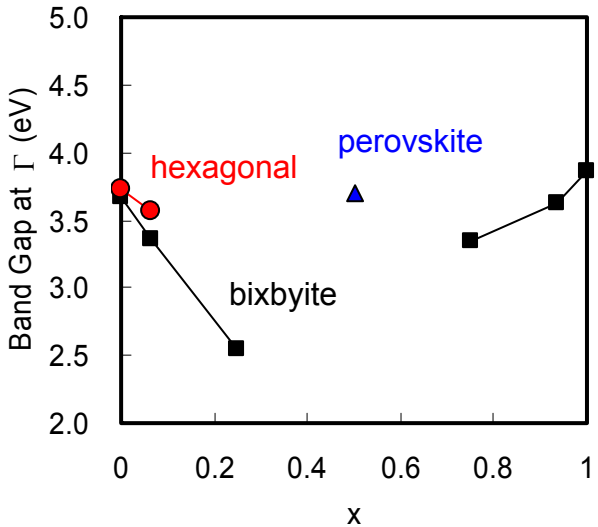
2. Electronic and Ionic Polarization Contributions ($\varepsilon^{electron}$, ε^{ion}) of $La_{2(1-x)}Sc_{2x}O_3$



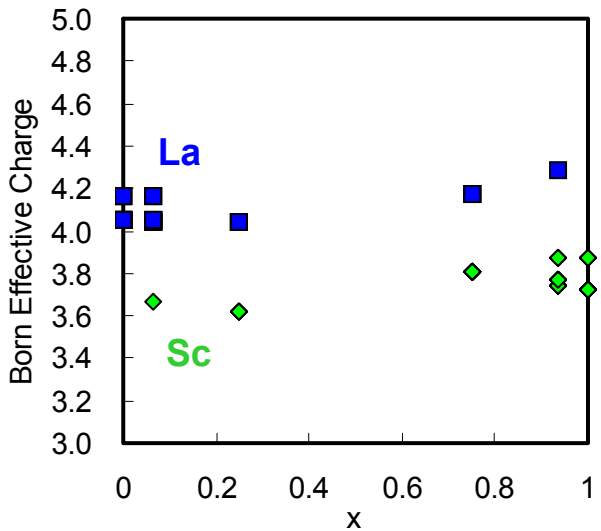


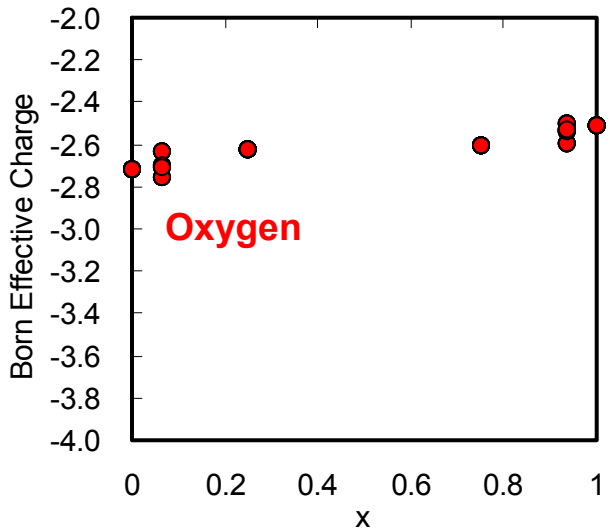
Both electronic and ionic polarization contributions increase with increasing Sc content. ($x<0.5$)
Ionic polarization contribution is larger than the electronic one.

3. Band Gap at Γ point of $La_{2(1-x)}Sc_{2x}O_3$



4. Born Effective Charge of $La_{2(1-x)}Sc_{2x}O_3$ (cubic bixbyite structure)

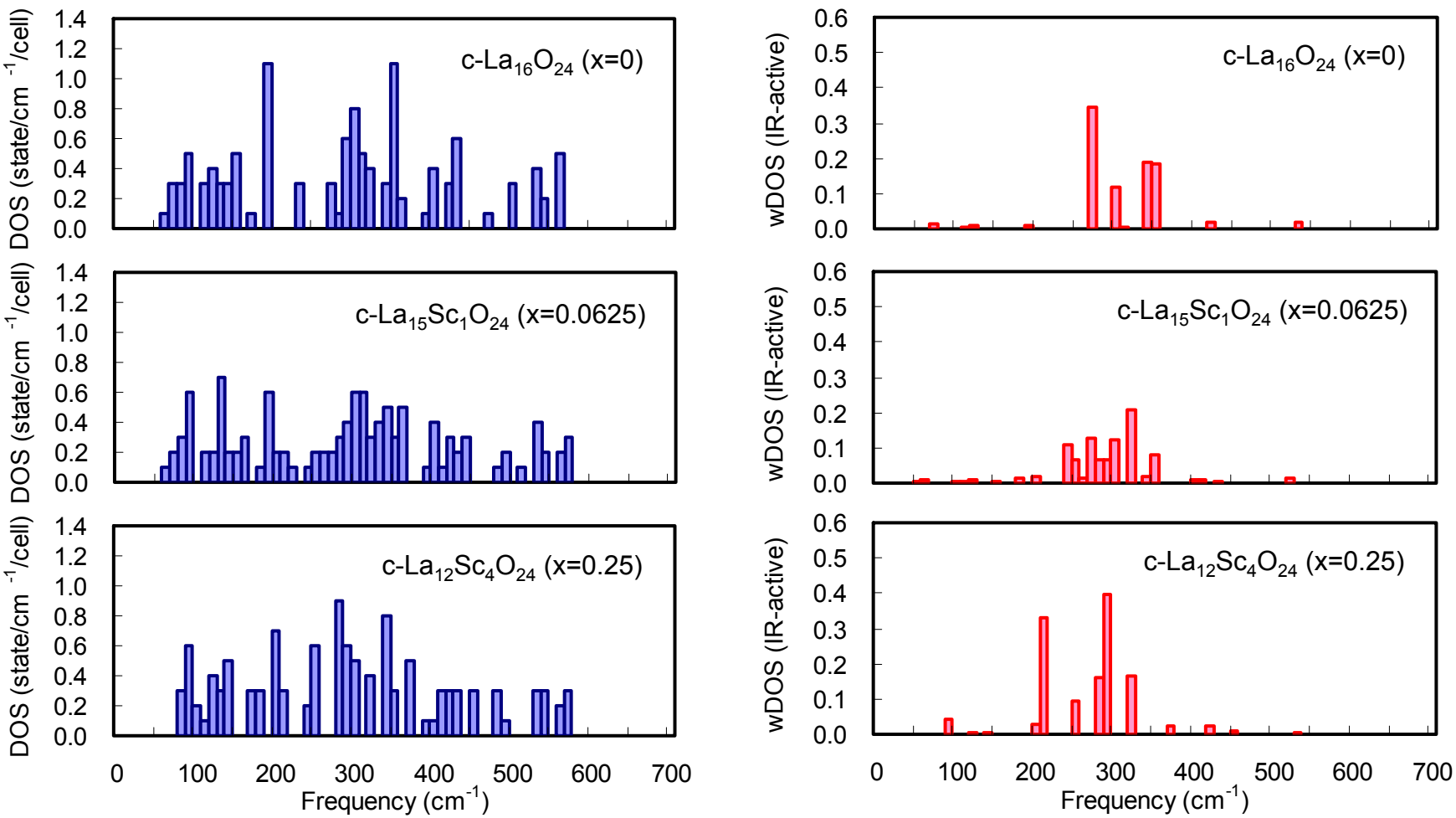




5. Phonon Density of States at Γ of $La_{2(1-x)}Sc_{2x}O_3$ (cubic bixbyite, x < 0.5)

$$\mathrm{DOS}(\omega) = \sum_{\lambda=1}^{3N-3} \delta(\omega_\lambda - \omega)$$

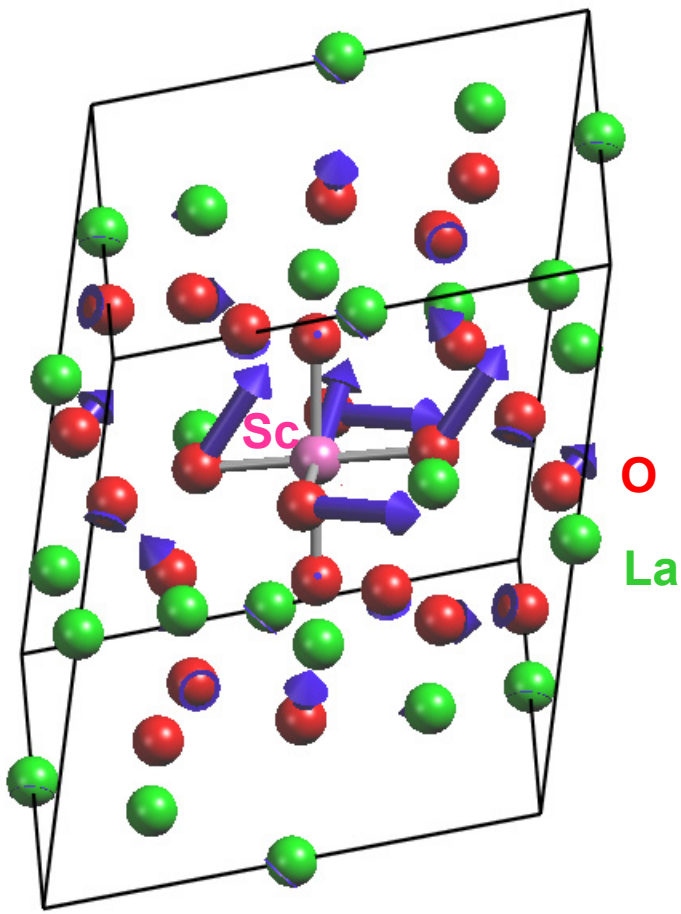
$$\mathrm{wDOS}(\omega) = \frac{4\pi e^2}{V} \sum_{\lambda=1}^{3N-3} \left(\frac{Z_\lambda}{\omega_\lambda} \right)^2 \delta(\omega_\lambda - \omega)$$



Lowering of IR-phonon frequencies by Sc doping into La_2O_3

6. Lowering of phonon frequency (due to Sc and neighboring O atoms)

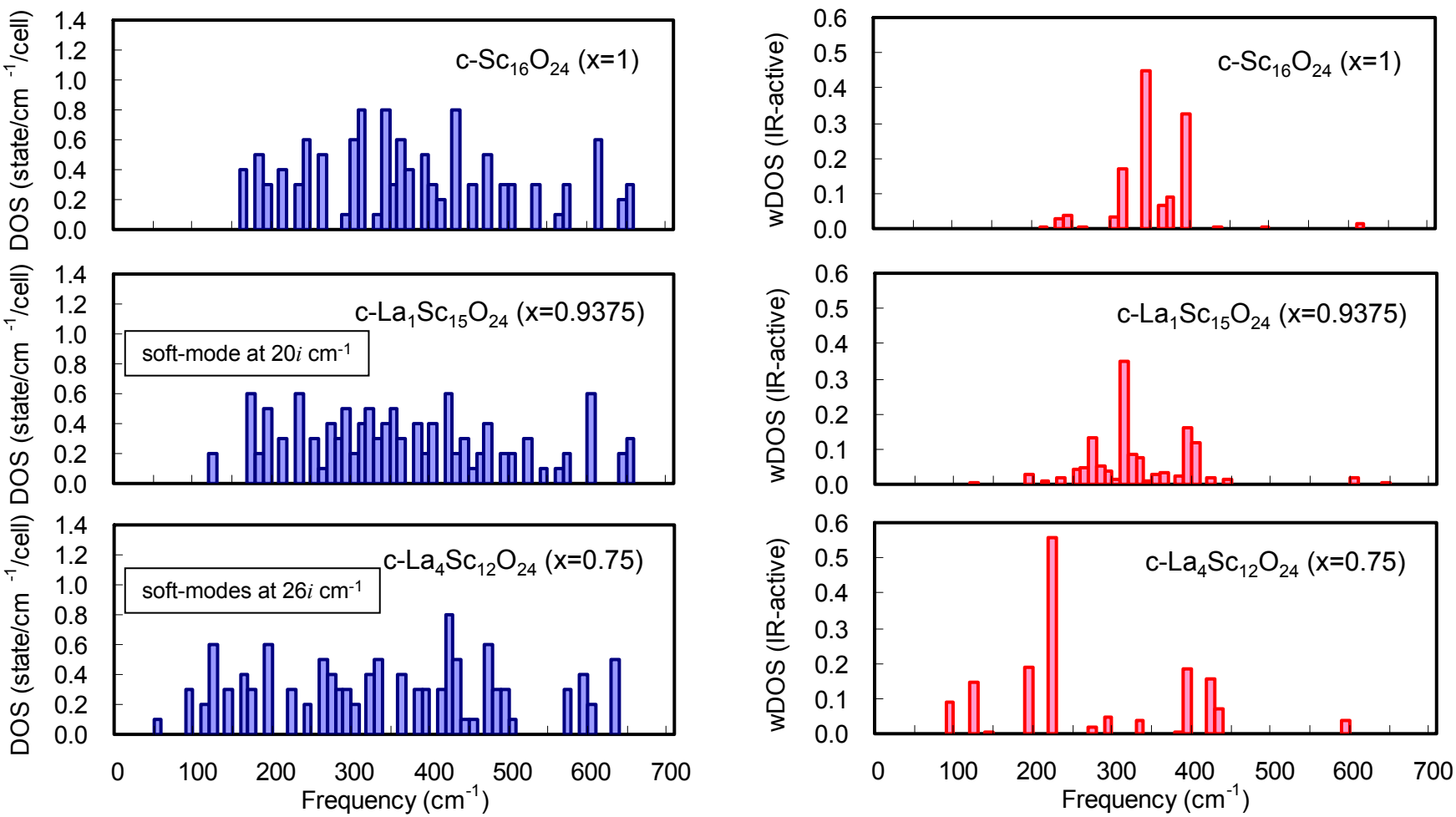
e.g. Phonon Mode of c-$La_{15}Sc_1O_{24}$ (x=0.0625) at 251cm^{-1} (IR active)



7. Phonon Density of States at Γ of $La_{2(1-x)}Sc_{2x}O_3$ (cubic bixbyite, x > 0.5)

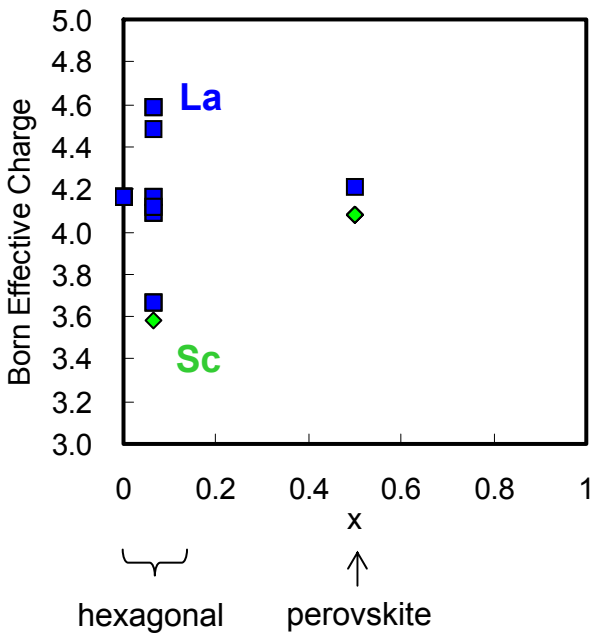
$$\mathrm{DOS}(\omega) = \sum_{\lambda=1}^{3N-3} \delta(\omega_\lambda - \omega)$$

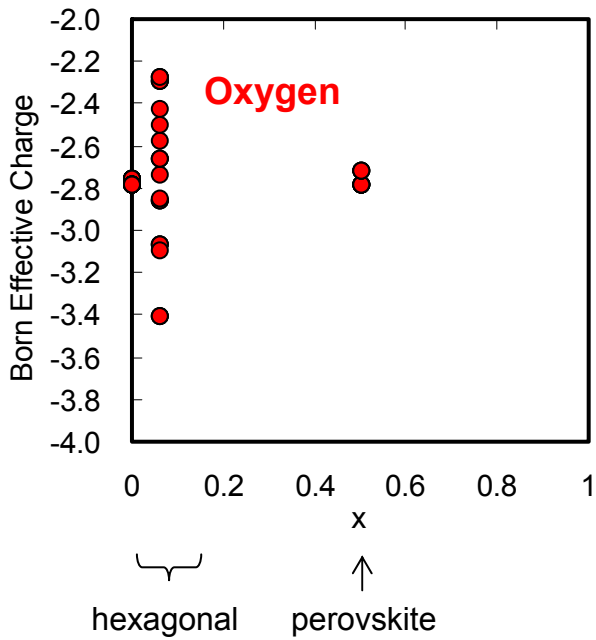
$$\mathrm{wDOS}(\omega) = \frac{4\pi e^2}{V} \sum_{\lambda=1}^{3N-3} \left(\frac{Z_\lambda}{\omega_\lambda}\right)^2 \delta(\omega_\lambda - \omega)$$



Lowering of IR-phonon frequencies by La doping into Sc_2O_3

8. Born Effective Charge of $La_{2(1-x)}Sc_{2x}O_3$ (hexagonal and perovskite)

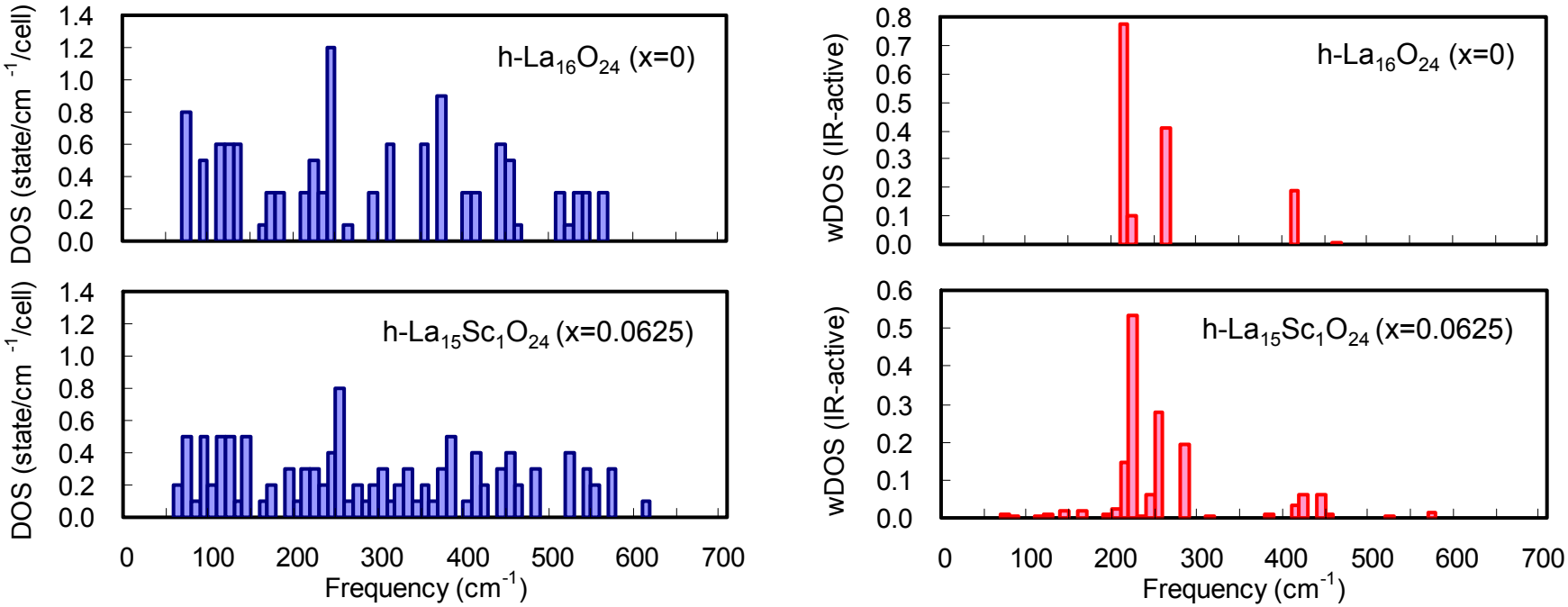




9. Phonon Density of States at Γ of $La_{2(1-x)}Sc_{2x}O_3$ (hexagonal)

$$\mathrm{DOS}(\omega) = \sum_{\lambda=1}^{3N-3} \delta(\omega_\lambda - \omega)$$

$$\mathrm{wDOS}(\omega) = \frac{4\pi e^2}{V} \sum_{\lambda=1}^{3N-3} \left(\frac{Z_\lambda}{\omega_\lambda} \right)^2 \delta(\omega_\lambda - \omega)$$

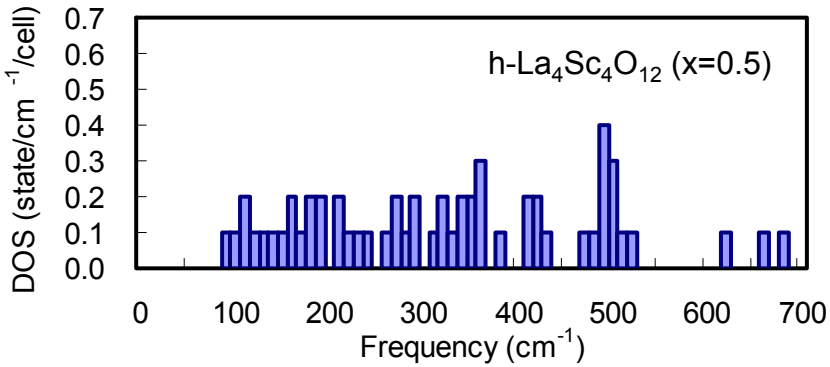


$$\int_{0\mathrm{cm}^{-1}}^{150\mathrm{cm}^{-1}} d\omega \left[\mathrm{wDOS}(\omega)\right] = 0.5$$

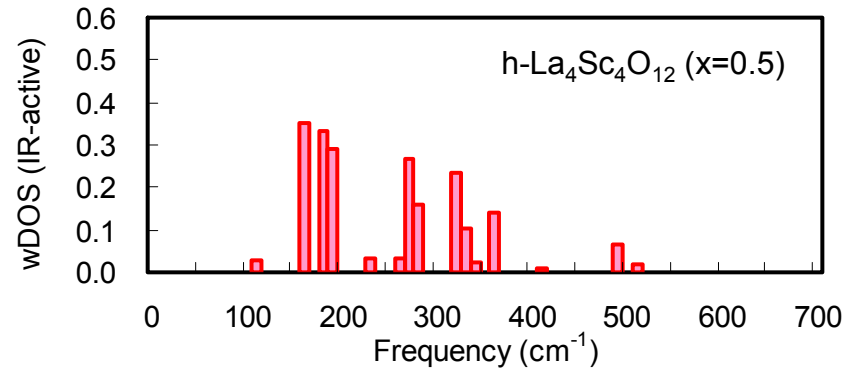
Integrated DOS up to 150cm^{-1} is 0.5 for $La_{15}Sc_1O_{24}$, while that is zero for $La_{16}O_{24}$.
This may account for the dielectric constant increase of 0.43 by Sc doping. (?)

10. Phonon Density of States at Γ of $La_{2(1-x)}Sc_{2x}O_3$ (perovskite)

$$\mathrm{DOS}(\omega) = \sum_{\lambda=1}^{3N-3} \delta(\omega_\lambda - \omega)$$



$$\mathrm{wDOS}(\omega) = \frac{4\pi e^2}{V} \sum_{\lambda=1}^{3N-3} \left(\frac{Z_\lambda}{\omega_\lambda} \right)^2 \delta(\omega_\lambda - \omega)$$



integration of the DOS gives

$$\int_0^\infty [\mathrm{DOS}(\omega)] d\omega = 3N - 3$$ number of phonon modes

$$\int_0^\infty [\mathrm{wDOS}(\omega)] d\omega = \varepsilon^{\mathrm{ion}}$$ dielectric constant (ion)