Low-Frequency Raman Study of Glass-like Properties of Mixed Crystals with the Fluorite Structure

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Mixed crystals with the fluorite structure display the characteristic two level system (TLS) spectrum associated with glasses. Our investigation of the temperature dependence of the low frequency Raman intensity between 1.4 K and 20 K suggests that inelastic scattering from the TLS distribution is the dominant mechanism. This occurs both for $(ZrO_2)_{0.91}(Y_2O_3)_{0.09}$ and $(CaF_2)_{1-x}(YF_3)_x$ as well as for the series $(CaF_2)_{1-x}(LaF_3)_x$, $(SrF_2)_{1-x}(LaF_3)_x$ and $(BaF_2)_{1-x}(LaF_3)_x$ for $0.05<x<0.45$. The overall cubic symmetry of these mixed crystal systems permits the Raman scattering types to be identified. We observe that the scattering from TLS only appears in A_{1g} symmetry while the intensities in the other two symmetries (E_g and T_{2g}) are at least a factor of 100 weaker. The Boson peak in the fluorite mixed crystals is observed to have E_g symmetry. In contrary to the suggestion that there is a direct correlation between the TLS spectrum and the Boson peak in glasses, we find that the intensity due to TLS in the glassy fluorite mixed crystals does not scale with the Boson peak scattering intensity. The spectral width of the Raman active T_{2g} phonon mode of the fluorite crystal gives a measure of the disorder in these systems. This disorder can be used to determine an effective temperature which when incorporated in the law of mass action provides a connection between the total number of Raman active TLS and the disorder [1]. In conclusion, our measurements show that the total number of TLS is a feature of the dynamical equilibrium of a system at high temperatures. The mixed crystal disorder simply permits the observation of the high temperature law of mass action properties at low temperatures.

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[1] J. J. Tu and A. J. Sievers, Phy. Rev. Lett. **83**, 4077 (1999).