Evolution from Quantum to Classical Fluctuation in the Soft Mode of KH_2PO_4

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The intensity ratio between Stokes (I_{S}) and anti-Stokes (I_{AS}) light scattering at frequency shift ω usually takes the Boltzmann factor $I_{\mathrm{S}}/I_{\mathrm{AS}} = e^{\hbar\omega/k_{\mathrm{B}}T}$, which is based on the quantum-mechanical detailed balancing in the light scattering process. Recently the deviation of the intensity ratio from the Boltzmann factor has been observed for quasi-elastic components in complex systems. In liquid carbon disulfide [1], $I_{\mathrm{S}}/I_{\mathrm{AS}}$ for cooperative rotational dynamics becomes unity in a frequency region less than several wavenumbers, while the usual relation holds for Boson peak. The symmetric spectral feature around $\omega = 0$ has also been reported for the quasielastic line in amorphous solids and supercooled liquids such as As_2O_3 [2]. Since the relation $I_{\mathrm{S}}/I_{\mathrm{AS}} = 1$ is expected for a classical fluctuation, these results mean that the dynamics in the frequency region loses the quantum character and behaves as a classical fluctuation. The classical character may be associated with Markovian nature resulting from dissipation in these cooperative fluctuations. In this report, we investigate the intensity ratio $I_{\mathrm{S}}/I_{\mathrm{AS}}$ of a paraelectric potassium dihydrogen phosphate (KH_2PO_4 or KDP) in which the quasi-elastic components appears originating from the polarization fluctuation which plays an important role as a soft mode in the ferroelectric phase transition (T_c=122.2 K) [3]. We expect that the analysis on the intensity ratio provides useful information on the soft mode which has dissipative and cooperative character.

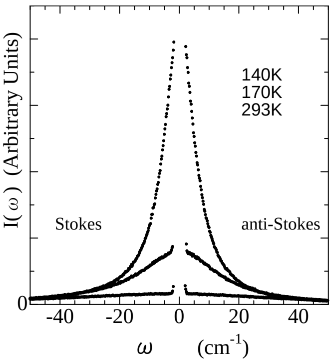

Figure 1: Light scattering spectra of KDP.

In Fig. 1, we show light scattering spectra of KDP in paraelectric phase measured with spectral resolution of 1.0 cm^{-1} under scattering geometry x(y,x)y, where x, y and z are the crystallographic axes. A Lorentzian-like spectrum of the soft mode centered at $\omega = 0$ grows up as temperature approaches T_{c}. The ratio $R \equiv (I_{\mathrm{S}}/I_{\mathrm{AS}})/\mathrm{e}^{\hbar\omega/k_{\mathrm{B}}T}$ is obtained from $I(\omega)$ as shown in Fig. 2. In the analysis, the spectral origin of each spectrum is adjusted to give $R = 1$ at $\omega = 40 \sim 50$ cm^{-1}. At 293 K, $R = 1$ holds in the whole frequency region within the experimental error. At 170 and 140 K, however, R deviates from unity systematically in $\omega < 30$ cm^{-1}, and $I_{\mathrm{S}}/I_{\mathrm{AS}}$ approaches unity as decreasing ω. It should be noted that the frequency of $\omega < 30$ cm^{-1} corresponds to the region where the spectral intensity rapidly increases as approaching T_{c}. These results may give us an useful information for investigating the evolution of the soft mode from microscopic quantum motions to the classical and macroscopic dynamics as approaching T_{c}. We will also discuss the comparison between the soft mode and the cooperative rotational dynamics in liquid from the viewpoint of Markovian nature of these cooperative dynamics.

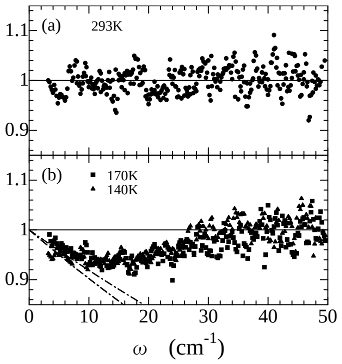

Figure 2: The ratio $R \equiv (I_{\mathrm{S}}/I_{\mathrm{AS}})/\mathrm{e}^{\hbar\omega/k_{\mathrm{B}}T}$. The dash-dotted lines in (b) show $R = \mathrm{e}^{-\hbar\omega/k_{\mathrm{B}}T}$ for $I_{\mathrm{S}}/I_{\mathrm{AS}} = 1$.

References

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