Nonlinear Energy Response to Oscillating Temperature in the Free Energy Landscape Picture

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The energy response has been studied as the measurement of the 1st order ac specific heat for the glass forming system. However, they are limited within the linear region. We analyse the nonlinear energy response to oscillating temperature for the system described by the free energy landscape picture, in which the time evolution of the system obeys the master equation [1] and show that the 2nd ac specific heat diverges at the glass transition point.

The 2nd order response consists of two terms; one is time dependent and the other is time independent. The former oscillates with frequency twice as large as the frequency of the perturbation, whose coefficient defines the 2nd order ac specific heat. The 2nd order ac specific heat has extrema which diverges as the temperature decreases toward the glass transition point. In addition, the latter diverges in the high frequency limit at the glass transition point.

Here, we show the 2nd order ac specific heat for a model landscape consisting of two basins, in which the barrier between basins grows up and diverges at the temperature $T_0 = 1.0$.

Figure 1 shows the frequency behavior of the 2nd order ac specific heat. As the temperature decreases toward T_0, the imaginary part of the 2nd order ac specific heat shows two growing extrema. Figure 2 shows the temperature dependence of the two extrema. As the temperature decreases toward T_0, they diverges. This is due to the divergence of the relaxation time at T_0.

With this result, we show the Cole-Cole plot of the 2nd order ac specific heat and the high frequency limit behavior of the time independent term of the 2nd order energy response.

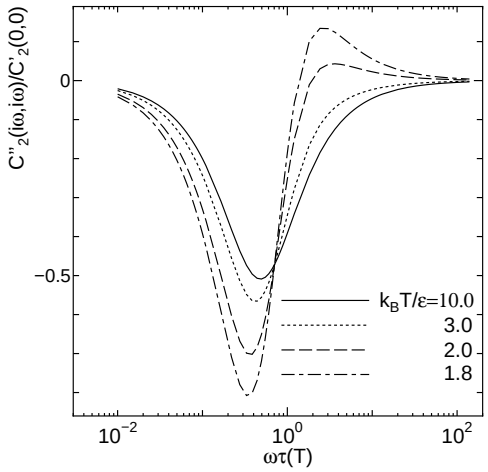

Figure 1: The imaginary part of the 2nd order ac specific heat: $\tau(T)$ is the relaxation time at the temperature T and ω is the angular frequency. As the temperature decreases toward T_0, the two extrema grows up.

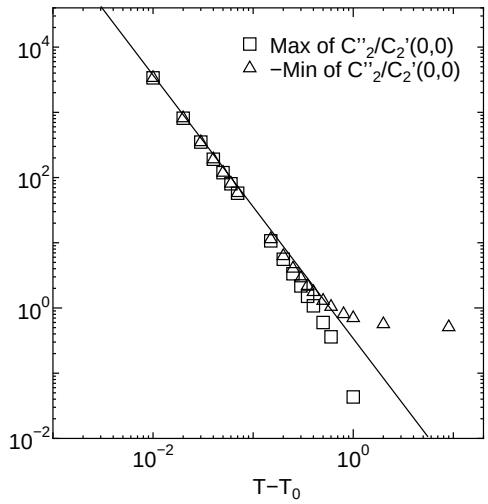

Figure 2: The temperature dependence of the two extrema of the 2nd order ac specific heat: Squares and triangles represent the maximum and minimum value, respectively. As the temperature decreases toward T_0, the two extrema diverge.

References

[1] T. Odagaki, T. Yoshidome, T. Tao, and A. Yoshimori, J. Chem. Phys. **117**,10151-10155(2002).