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Reply to ''Comment on 'Depolarized light scattering study of molten zinc chloride' ''

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Our reply to Ngai and Roland's comment emphasizes that our analysis of the $ZnCl_2$ depolarized light scattering spectrum was only designed for proving the existence of a boson peak and revealing its thermal evolution. This analysis, as well as Ngai and Roland's one, are based on a separation between relaxational and vibrational contributions, which has up to now no theoretical justification. [S1063-651X(96)10312-3]

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In their Comment, Ngai and Roland have reanalyzed our depolarized light-scattering susceptibility data obtained at various temperatures on the intermediate glass-forming liquid $ZnCl_2$ [1]. Let us first recall our own goal. As can been seen on Fig. 1 of the preceding Comment, there are three distinct spectral features appearing in the experimental window, the α relaxation peak being by far the most visible. The region of the boson peak is less clear and the boson peak itself appears only as a rather poorly defined shoulder. As our only goal in this fitting was to show that a softening of the boson peak takes place as the temperature increases, we attempted a very simple fitting procedure that did not make use of sophisticated fitting functions recently designed for the description of this peak [2]. Therefore, our analysis consisted of a phenomenological decomposition of these spectra into the sum of three terms: a high-frequency part, a ''boson peak'' and an α-relaxation process, principally visible at low frequency. We represented this last term by a Cole-Davidson function and the two former ones by simple Lorentzians following previous approaches cited in our paper [1].

In their Comment, Ngai and Roland show that, by replacing the Cole-Davidson function by the coupling model, given by the equations,

$$\Phi(t)=\begin{cases}\exp(-t/\tau_0) & t<t_c \qquad (2)\\ \exp[-(t/\tau^*)^{\beta}] & t>t_c, \qquad (3)\end{cases}$$

where $\tau_0(T)=t_c^{[1-\beta(T)]}\tau^*(T)^{\beta(T)}$, $t_c \sim 2\times10^{-12}$ s, and where β has the same value as the one we used ($\beta=0.8$), apparently equally good fits are obtained. The small differences between the susceptibility $\chi''(\omega)$ of the coupling model and of the Cole-Davidson function appear only in the frequency domain above 100 GHz, which is dominated by the boson peak.

Ngai and Roland conclude that the coupling model description of the fast dynamics of $ZnCl_2$ is consistent with the data and that it is impossible to distinguish between the coupling model and our simple phenomenological superposition approach because of uncertainty in the form needed to describe the boson peak. We would add that one should be careful about the significance of either fit, apart from the information provided on the existence of a boson peak softening, because the separation of the spectrum into separate vibrational and relaxational contributions is only an assumption that does not rely on any firm theoretical basis.

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