

Experimental report

25/01/2024

Proposal: 6-05-1066

Council: 10/2022

Title: Study of the Collective Density Fluctuations in Glass-Forming Liquids

Research area: Physics

This proposal is a resubmission of 6-05-1041

Main proposer: Peter FALUS

Experimental team: Antonio FARAONE
Lucas TROJANOWSKI

Local contacts: Peter FALUS

Samples: ZnCl₂

Instrument	Requested days	Allocated days	From	To
WASP	5	5	04/09/2023	09/09/2023

Abstract:

This proposal was accepted under proposal number 6-05-1041, but was backlogged before the long shutdown. Hence the resubmission. Recent theoretical and computational studies have suggested that low energy collective modes with shear nature indeed, exist in liquids and are enhanced in glasses. Furthermore, the appearance of these modes in collective density fluctuations characterized by the coherent dynamic structure factor, is intimately related to the fragility of the liquids. Our recently developed ViscoElastic Hydrodynamic (VEH) theory predicts that the common origin of the anomalous low-energy excitations in strong liquids and collective fast relaxation in fragile liquids, is the manifestation of transverse collective modes. We have some preliminary experimental evidence of this on two glass-forming ionic compounds, K₃Ca₂(NO₃)₇ (CKN) and ZnCl₂, fragile and strong glass formers respectively, from INS and NSE measurements. We now request WASP to measure the collective relaxation modes (both the slow and an additional fast mode) ZnCl₂ over a wider Q range than previously achievable and further test our theory.

Experiment 06-05-1066 Report:

We examined the relaxation dynamics of ZnCl_2 ($T_g=380$ K, $T_m=563$ K) via WASP, but the data we collected proved inconclusive to study the T and Q dependence of different relaxation mechanisms due to a number of factors. Primarily, the furnace on WASP can only reach temperatures of 620 K, limiting not only the number of temperatures we can study, but also stopping us from seeing the full decay line shape for large t . Additionally, our data indicates that our signal to noise ratio is low, likely as a result of incoherent scattering off of Cl-35 in our sample and from the quartz and vanadium sample holder.

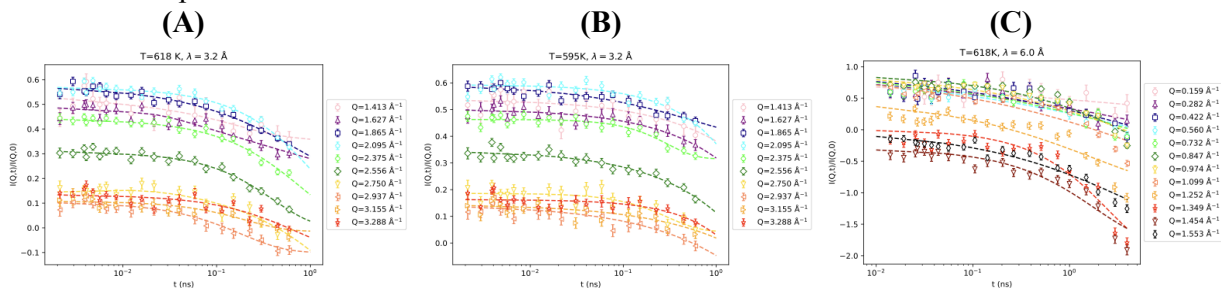


Figure 1: NSE spectrum for ZnCl_2 at (A) 618 K with $\lambda=3.2$ Å neutrons, (B) 595 K with $\lambda=3.2$ Å neutrons, and (C) 618 K with $\lambda=6.0$ Å neutrons. Dashed lines indicate fits to the data with a Kohlrausch-Williams-Watts (KWW) function with background: $KWW(t) = fe^{-(t/\tau)^\beta} + bkg$

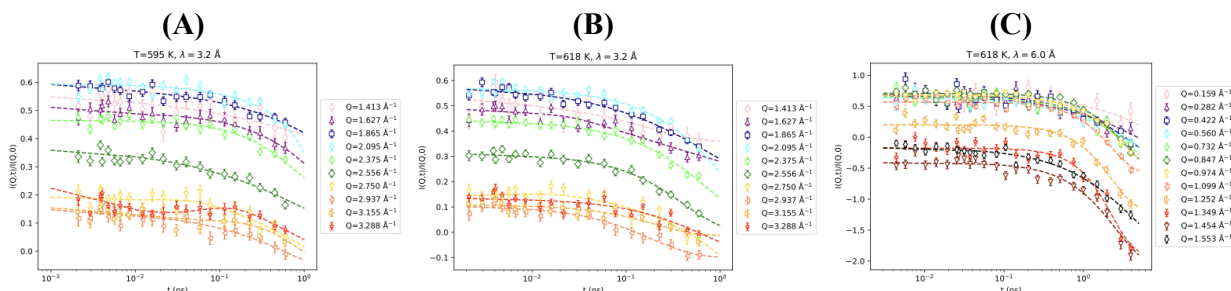


Figure 2: NSE spectrum for ZnCl_2 at (A) 618 K with $\lambda=3.2$ Å neutrons, (B) 595 K with $\lambda=3.2$ Å neutrons, and (C) 618 K with $\lambda=6.0$ Å neutrons. Dashed lines indicate fits to the data with a two step relaxation with background: $TwoStep(t) = (1 - A)e^{-t/\tau_f} + Ae^{-(t/\tau_s)^\beta} + bkg$

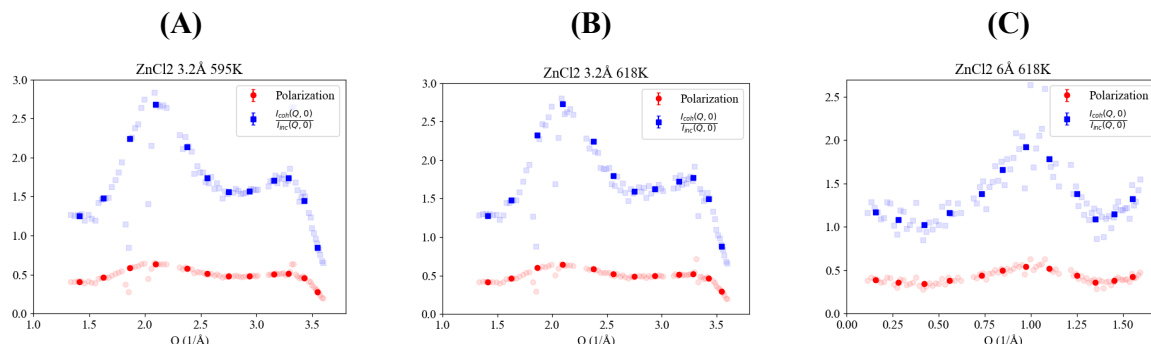


Figure 3: Polarization and coherent/incoherent ratio for ZnCl_2 at (A) 618 K with $\lambda=3.2$ Å neutrons, (B) 595 K with $\lambda=3.2$ Å neutrons, and (C) 618 K with $\lambda=6.0$ Å neutrons. Faded symbols indicate binning for each of the 90 collected Q values, whereas vibrant symbols indicate data with the same binning as $I(Q, t)$ data. Polarization is normalized by total count.