Experimental measurements of the temperature-dependent Van Hove function in a Zr$_{80}$Pt$_{20}$ liquid - Confirmation of a structural origin for the dynamics at high temperature

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Even though viscosity is one of the most fundamental properties of liquids, determining its atomic origin has proven elusive. By combining inelastic neutron scattering with the electro-static levitation technique, the viscosity crossover temperature has been determined for liquid Zr$_{80}$Pt$_{20}$. We show that the decay-time of the first peak of the Van Hove function is directly related to the Maxwell relaxation time of the liquid, which is proportional to shear viscosity. This result proves that the local dynamics of atomic bond breaking and forming determines the viscosity, lending support to predictions from molecular dynamics simulations.

The viscosity, $\eta$, of liquids shows common behavior among various disparate groups of liquids [1]. At high temperatures it follows an Arrhenius temperature dependence with a constant activation energy. But below a certain temperature, the viscosity crossover temperature, $T_A$, it becomes super-Arrhenius. Kivelson [2] first showed that the viscosity of various liquids can be scaled into one curve as a function of $T/T_A$ ($T_A = T^*$ in their work). $T_A$ is also the temperature below which the mode-coupling becomes appreciable [3, 4]. The fundamental time-scale for viscosity is the Maxwell relaxation time, $\tau_M = \eta/G_\infty$, where $G_\infty$ is the infinite-frequency shear modulus. Recent molecular dynamics (MD) studies of metallic liquids suggest that for $T > T_A$, $\tau_M$ is approximately equal to $\tau_{LC}$, the lifetime of the atomic bond [4, 5].

To evaluate this relationship inelastic neutron scattering measurements were made on liquid Zr$_{80}$Pt$_{20}$. To access the supercooled state and to avoid contamination all measurements were made with the liquid held in a containerless environment in high vacuum using the Neutron Electrostatic Levitation (NEL) facility [7] located at the Spallation Neutron Source (SNS). The results were converted into the time-dependent pair-distribution function, i.e. the Van Hove function $G(r,t)$, which allowed a study of the spatial and temporal correlations of the atoms. Due to experimental difficulties, studies of the Van Hove function in the past have been largely limited to computer simulations. Only in a few cases have measurements of $G(r,t)$ been made in metallic liquids at the melting temperature [6], and for water by inelastic x-ray scattering [10, 11]. While $\tau_{LC}$ cannot be measured directly from experiment, new MD results discussed here show that it can be related to the decay time of the first peak area in the distinct part of the Van Hove function, $G_d(r,t)$. A comparison of the activation energies of $\tau_{VH}$ and $\tau_M$ confirms the prediction that both have an Arrhenius temperature dependence and the same activation energy. To our knowledge, this is the first significant experimental evidence indicating that local structural rearrangements underlie the dynamical behavior of high temperature metallic liquids.

Measurements of the high temperature properties of liquid metals such as Zr are often plagued by sample reactivity and oxygen contamination. These are minimized by processing the liquids without a container in a high vacuum environment using the technique of electro-static levitation [12]. The viscosity measurements were made with the Washington University Beamline Electro-static Levitation (WU-BESL) facility; the experimental methods are discussed elsewhere [14, 15]. Inelastic neutron scattering measurements were made at Oak Ridge National Laboratory (ORNL) on the wide angular-range chopper spectrometer [16] (ARCS) beamline at the SNS. The samples were processed in high vacuum using the NESL facility, which is optimized for both elastic and inelastic time-of-flight (TOF) neutron scattering studies [3]. The TOF inelastic neutron diffraction measurements on the levitated liquid samples were made with incident energies $E_i = 20, 50$, and 80meV. Since the energy resolution worsens with increasing incident energy, only the 20meV data are reported here. Due to the kinematic restrictions inherent to inelastic neutron scattering experiments, however, the maximum $q$-range for this incident energy is restricted to $q < 6\AA^{-1}$. Though this restricted $q$-range can introduce termination ripples in the spatial Fourier transform to obtain the Van Hove correlation function, the increased energy resolution was deemed to be more important for the data needed. The procedure for analyzing the data is only briefly described here; a more detailed description will be given in a future publication.

The MD simulations were performed with the LAMMPS software [17] using the Zr-Pt embedded
atom \textsuperscript{18} (EAM) potential developed by H. Sheng \textsuperscript{19}. The Zr\textsubscript{80}Pt\textsubscript{20} system was simulated with 15000 atoms under the NPT ensemble (P=0) with periodic boundary conditions. The Nosé-Hoover thermostat \textsuperscript{20, 21} was used to equilibrate the system at a target temperature for 15ns before data collection. The Maxwell time was calculated from the atomic level stress using the Green-Kubo formula (see \textsuperscript{22} for example). The method for computing the Van Hove time is the same as for the experimental data which is discussed later. Additional details of the MD simulation are provided in the supplemental material.

The steps to obtain the dynamic structure factor $S(q, E)$ include the conversion of the TOF data to energy and momentum transfer, a physical normalization factor, the assumption of detailed balance and a correction for the resolution of the spectrometer. The initial conversion used a standard reduction routine employing the MANTID \textsuperscript{23} software. The required source beam parameters \textsuperscript{24} for this reduction were obtained from previous calibration experiments, since they could not be measured in the NESL studies due to the presence of an internal beam-stop. For normalization, the condition that $S(q) \rightarrow 1$ as $q \rightarrow \infty$ was enforced, using the $S(q)$ obtained from the intermediate scattering function, $F(q, t)$. This normalization was checked by comparing the $S(q)$ obtained here with one obtained from earlier neutron and x-ray diffraction data. Detailed balance was used to extend the negative energy transfer data into regions that are inaccessible in the positive energy transfer region. A typical $S(q, E)$ obtained after these corrections is shown in Fig. 1a. Since $F'(q, t) = F(q, t)R(t)$, where $F'$ is the true and $F$ is the measured intermediate scattering functions and $R$ is the resolution function, the true intermediate scattering function can be obtained by dividing the measured signal with the resolution function. The resolution function was obtained from inelastic scattering measurements from vanadium at room temperature.

The intermediate scattering function, $F(q, t)$, is obtained by a Fourier transform of $S(q, \omega)$

$$F(q, t) = \int_{-\infty}^{\infty} S(q, \omega)e^{i\omega t}d\omega \quad (1)$$

The self ($F_s(q, t)$) and distinct ($F_d(q, t)$) parts of the intermediate scattering function, which describe single particle and collective density fluctuations respectively, are extracted by assuming that the self-part has a Gaussian form, i.e. $F_s(q, t) = A\exp(-wq^2)$ \textsuperscript{25}, where the decay rate, $w$, and the amplitude, $A$ are fitting parameters. The Gaussian approximation comes from expressions for the self-part of the Van Hove correlation function, $G_s(r, t)$, which has a Gaussian dependence in $r$ in both the hydrodynamic and free-particle limits \textsuperscript{22, 26}. For intermediate times, which are of interest here, the Gaussian approximation should still be a good approximation \textsuperscript{24, 27}. The distinct Van Hove correlation function is obtained from the Fourier transform of $F_d(q, t)$

$$G_d(r, t) - 1 = \frac{1}{2\pi \rho} \int_{-\infty}^{\infty} F_d(q, t)q^2 \sin(qr) dq \quad (2)$$

where $\rho$ is the number density for the sample. A representative example of $G_d(r, t)$ obtained from the data is shown in Fig. 1b. The integrated peak intensity is computed for each temperature from

$$N(t) = \int_D 4\pi r^2 \rho (G_d(r, t) - 1) dr \quad (3)$$

where $D$ is the positive region of the first intense peak of the integrand. $N(t)$ is proportional to the dynamic coordination number, and reflects the average decorrelation time for atoms located near the first peak of $G_d(r, t)$.
The decorrelation time is the time for an atom initially located near the central atom to begin to diffuse away. This time is a function of the distance that an atom is from the central atoms initial position. It also depends on the local structure, which can be quite varied. Since the exponential decay time for each atom is different, the overall decay of \( N(t) \) could be described by a Kohlrausch-Williams-Watts (KWW) stretched exponential function,

\[
y(t) = \exp\left(-\left(t/\tau\right)^\beta\right)
\]

where \( \tau \) is the time constant and \( \beta \) is the stretching factor. From MD simulations and a recent study on water, \( N(t) \) is expected to have two decay rates; one is due to ballistic motion and another one describes the changes in the configuration of the nearest-neighbors. However, due to the limited energy range of these experiments for \( \text{Zr}_{80}\text{Pt}_{20} \) it was not possible to determine the decay rate in the ballistic region.

The normalized peak intensity \( N(t)/N(0) \) is shown in Fig. 2. The initial decay in \( N(t)/N(0) \), \( t < 0.1 \text{ps} \), is due to ballistic motion and is only weakly dependent on temperature. As \( t \to \infty \) it is expected that \( N(t)/N(0) \to 0 \) as the correlations between the initial position are lost. The data show plateaus at longer times; these are artifacts from the resolution function correction (see supplemental material) and are not fit to the KWW expression. The ballistic region is not well described by the single KWW expression. The dashed lines in Fig. 2 are fits to the KWW expression, which describes well the data beyond 0.1ps.

As mentioned earlier, the local configuration time, \( \tau_{LC} \), cannot be obtained directly from the experimental data. However, our MD simulations show that \( \tau_{LC} \) is related to a measurable quantity called here the Van Hove time, \( \tau_{VH} \), which is the long decay time corresponding to the configuration of nearest-neighbors in the first peak in \( G_{d}(r, t) \). The experimental value of the Van Hove time was obtained from the mean relaxation time of the KWW function fit to the data, \( \langle \tau \rangle = \frac{\beta}{\Gamma(\frac{\beta}{4})} \equiv \tau_{VH} \). The results from the MD simulations shown in Fig. 3 indicate that \( \tau_{LC} \approx \tau_{VH}/3.6 \) for \( T > T_{A} \). Since the ratio is approximately constant for \( T/T_{A} > 1.2 \) the activation energy for \( \tau_{VH} \) will be the same as for \( \tau_{LC} \). For water \( \tau_{LC} \) was approximately equal to \( \tau_{VH} \) (\( \tau_{2} \) in their work). Since metallic liquids have more nearest neighbors than does water (\( \sim 13 \) for metallic liquids and 4 for water), the ratio \( \tau_{VH}/\tau_{LC} \) should reflect this difference.

As shown in Fig. 3, \( \tau_{VH} \) obtained from the scattering data shows an Arrhenius temperature dependence for \( T_{A}/T > 1 \), as indicated by the previous MD simulations for the related local configuration time, \( \tau_{LC} \). The results from the MD studies indicate that \( \tau_{LC} \) remains Arrhenius far below \( T_{A} \). Based on the results in Fig. 3, \( \tau_{VH} \) should become super-Arrhenius below this temperature, as suggested by the data in Fig. 4. The activation energy for \( \tau_{VH} \) (and from Fig. 3 for \( \tau_{LC} \)) above \( T_{A} \) is 750±90meV. As shown in Fig. 4, the activation energy for the measured viscosity above \( T_{A} \) is 731±4meV. Within experimental error, these activation energies are equal, indicating that the energy barrier is the same for each process.
from the viscosity (not shown) and comparing to \( \tau_{LC} \) we find that \( \tau_M \approx \tau_{LC} \). This provides experimental evidence that the MD predictions \[3, 6\] are correct, i.e. showing that the atomic rearrangements that determine \( \tau_{V_H} \) (and \( \tau_{LC} \)) are controlling the viscosity at high temperatures.

In summary, the time dependent pair distribution function (distinct Van Hove function, \( G_d(r, t) \)) was obtained as a function of temperature from inelastic neutron scattering data for equilibrium and supercooled ZrPt\(_20\) liquids made in a containerless environment. Molecular dynamics simulations showed that the relaxation time of the of the positive peak area in \( G_d(r, t) \) (defined as the Van Hove time, \( \tau_{V_H} \)) is related to the local configuration time, \( \tau_{LC} \), and has the same temperature dependence above the crossover temperature, \( T_A \). A comparison of the experimental neutron scattering and viscosity data show that the activation energy of \( \tau_{V_H} \) and that of the Maxwell time, \( \tau_M \) are equal to within experimental error, strongly suggesting that they are governed by the same process. To our knowledge this is the first experimental evidence for a key prediction from recent MD studies for metallic liquids, which indicate that local structural excitations underlie the viscosity at high temperature.

FIG. 4. (Color online) (a) Van Hove time and (b) viscosity data for liquid Zr\(_{50}\)Pt\(_{20}\) versus inverse temperature normalized to the Arrhenius crossover temperature determined from the viscosity (\( T_A = 1450K \)). The best fit lines for the activation energy are shown; the slopes give an activation energy of 731meV for the viscosity and 750meV for \( \tau_{V_H} \). The error bars shown for \( \tau_{V_H} \) are 3\( \sigma \) and are calculated from the error in the fit parameters from Eq. [4].

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Supplemental Material: Experimental measurements of the temperature-dependent Van Hove function in a Zr$_{80}$Pt$_{20}$ liquid - Confirmation of a structural origin for the dynamics at high temperature

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EXPERIMENTAL INFORMATION

Master ingots of Zr$_{80}$Pt$_{20}$(1-2g, using purity Zr (99.97%) and Pt (99.997%)) were first prepared by arc-melting on a water-cooled hearth in a high purity (99.999%) Ar atmosphere. To further reduce the oxygen concentration in the atmosphere, a Ti-Zr getter was melted prior to arc-melting the ingots. Both the ingot and the getter were held molten for 60 sec. This procedure was repeated three times, flipping the samples between melting to further increase mixing. The ingots were subsequently crushed and portions were used to create smaller spherical samples for viscosity (45 mg) and inelastic neutron scattering measurements (350 mg).

The inelastic neutron scattering samples were levitated in the NESL and heated to the desired temperatures using two fiber-coupled diode lasers (980nm, 110W continuous maximum power output) that were focused on opposite sides of the samples to reduce the temperature gradient. The sample temperature was measured using a single Process Sensors Metis MQ22 two-color ratio pyrometer. To obtain sufficient statistics, the samples were held for 1.5-2hrs. at each temperature while inelastic scattering measurements were made. Occasionally it was not possible to maintain levitation of the same sample for the full time duration, in which case a different sample was used to obtain the remaining data and the two scattering data sets were combined. The temperature data were corrected after processing by using the solidus temperature as a point of reference. A more detailed discussion of this technique can be found elsewhere [1].

MOLECULAR DYNAMICS SIMULATION

The MD simulations reported in this publication were made using the Washington University in St. Louis Physics department high performance computing cluster. The atoms in the simulation were initialized to random locations. The system was then allowed to relax to remove overlapping atoms and evolved for 0.5ns to obtain a structure more like that of the liquid. The temperature in the simulation was decreased at a rate of $7 \times 10^{11}$ K/s and they system was subsequently equilibrated at each target temperature for 15ns. After these initial steps the Van Hove correlation function ($G(r, t)$), and the viscosity ($\eta$) were obtained.

Van Hove Function

The distinct Van Hove correlation function is given by:

$$G_d(r, t) = \sum_{\alpha, \beta} \left( \sum_{\gamma} G_{\alpha\beta}^d(r, t) \right)$$

where $c_\alpha$ is the concentration and $b_\alpha$ is the scattering length of element $\alpha$. The sum $\alpha\beta$ ranges over all atomic pairs. $G_{\alpha\beta}^d(r, t)$ is the partial distinct Van Hove correlation function given by

$$G_{\alpha\beta}^d(r, t) = \frac{N}{\rho N_\alpha N_\beta} \sum_i \sum_{i \neq j} \langle \delta(r - r_i(0) + r_j(t)) \rangle,$$
where $N_\alpha$ is the number of atoms of $\alpha$ and $\rho$ is the number density. Structural data used to calculate $G_d(r, t)$ was collected for 10ps. A typical $G_d(r, t)$ calculated from MD is shown in Fig. 1.

**Viscosity Calculation**

As mentioned in the publication the viscosity was calculated using the Green-Kubo formula and the method derived by Daivis and Evans [2]:

$$\eta = \frac{V}{10k_BT} \sum_{i,j} \langle P_{ij}(t)P_{ij}(0) \rangle \, dt \tag{3}$$

where $V$ is the volume, $T$ is the temperature in absolute units, and $P_{ij}$ is given by

$$P_{ij} = \frac{\sigma_{ij} + \sigma_{ji}}{2} \cdot \frac{1}{3} \sum_k \sigma_{kk} \tag{4}$$

where $\sigma_{ij}$ is the $ij$-th element of the atomic stress tensor, with $ij$ spanning all values of the tensor. The atomic stress tensor was recorded for 4 ns. at each temperature. The autocorrelation function for $P_{ij}$ was computed using fast Fourier transforms according to the Weiner-Khinchin theorem:

$$\langle P_{ij}(t)P_{ij}(0) \rangle = \text{IFFT}[\text{FFT}[P_{ij}][\text{FFT}[P_{ij}^*]] \tag{5}$$

where IFFT and FFT are the inverse and forward fast Fourier transforms and * indicates complex conjugation.

From the viscosity the Maxwell time, $\tau_M$, was computed using

$$\tau_M = \frac{\eta}{G_\infty}$$

where $G_\infty$ is the instantaneous shear modulus given by

$$G_\infty = \frac{V}{k_BT} \langle P_{ij}^2(0) \rangle \tag{6}$$

**Local Configuration Time**

The local configuration time, $\tau_{LC}$, is the time it takes for an atom to leave the local cluster (i.e. the coordination number to increase or decrease by 1). The local configuration time was calculated following the methods set forth in [2] and [4].

**DATA ANALYSIS**

Figure 2 gives a schematic diagram of the data reduction procedure some of which is outlined in the paper. All analysis apart from the standard instrument reduction, performed using the MANTID software, is performed using an in-house Python script. For the current analysis the resolution function (discussed later) is applied to the total intermediate scattering function $F(q, t)$ but could be applied to the Van Hove correlation function $G(r, t)$ without significant changes.

As mentioned in the publication the resolution function is obtained from the scattering profile of a polycrystalline sample of vanadium. It is assumed that the vanadium scatters completely incoherently so that the result is the neutron beam profile convoluted with the resolution of the detectors (i.e. $S'(q, E) = S(q, E) * R(q, E)$). It is also assumed that the resolution function is independent

![FIG. 2. A flowchart which describes a simplified data reduction method for inelastic neutron scattering measurements. Curved boxes indicate the functions while square boxes correspond to an analysis technique applied to the function or used to obtain the function.](image)

![FIG. 3. The resolution function calculated from the Fourier transform of the vanadium sample normalized to its value at $t = 0$ps. The value and error are computed from the mean and standard deviation, respectively, assuming that $F(q, t)$ is $q$-independent.](image)
of $q$ which should be approximately true for small energy transfer about $|E| < 10\text{meV}$ (see Fig.4 for an example vanadium scattering profile) or for a sufficiently small $q$-range. To approximate the resolution function only the restricted section of the vanadium data $1.0 < q < 2.5\text{Å}^{-1}$ was used. The calculated resolution function given by the Fourier transform of the $I(q, E)$ data is shown in Fig.4.

Figure 4 shows a comparison between the data for $N(t)/N(0)$ obtained from the inelastic neutron scattering experiments both with the resolution function correction and without. A noticeable effect coming from the resolution function correction is an elongation of the long time tail of the decay (i.e. the plateauing mentioned in the paper). This is an artifact, arising from an incomplete knowledge of the true resolution function for the beam line. To account for this effect the decay data is only fit with the KWW function out to 2ps corresponding to when the resolution function decays to approximately $1/e$. The minimum time which can be considered in this experiment, set by the maximum energy transfer 20meV, is $\sim 0.2\text{ps}$.

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