The possibility of transverse excitation modes in liquid Ga

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Abstract
The dynamic structure factor $S(Q, \omega)$ of liquid Ga was measured at $100\,°C$ using a high
resolution inelastic x-ray scattering (IXS) spectrometer at 3-ID-C/APS. The spectra obtained
clearly demonstrate the existence of longitudinal propagating modes at small Q values, like a
previous IXS result at $42\,°C$ obtained by Scopigno *et al* and an inelastic neutron scattering
(INS) one at $47\,°C$ obtained by Bove *et al*, but unlike an INS study at $57\,°C$ by Bermejo *et al*.
The dispersion relation of the excitations deviates positively from the hydrodynamic prediction
by about 13%. There are two new findings from this experiment. Firstly, an additional lower
energy excitation is necessary to reproduce $S(Q, \omega)$ spectra in the Q range beyond $10\,nm^{-1}$, in
agreement with the result of a first-principles molecular dynamic simulation, which may
indicate a transverse acoustic mode in this peculiar liquid metal. Secondly, the quasielastic line
comprises a Gaussian contribution at Q values near the first maximum in $S(Q)$, which may
indicate the existence of short-lived covalent correlation in liquid Ga with a lifetime of 0.39 ps.

1. Introduction

Ga is a fascinating metal in several aspects. It possesses
a variety of morphological crystalline forms including both
covalent and metallic properties. The mixed covalent–metallic
character of the lowest energy phase α-Ga has been stressed in
a first-principles molecular dynamics (FPMD) study by Gong
et al [1], who showed that a low density of states (DOS) at the
Fermi energy is due to the formation of Ga_2 binary covalent
bonds, although it shows a good metallic conduction.

Upon melting, the density of Ga unusually increases by
about 3%. Ga has an extremely wide liquid range from 29.8 to
$\sim 2200\,°C$ at normal pressure, and also a very large temperature
range of the supercooled state down to about $-120\,°C$. It
can be explained that the covalent and metallic characters may
rather tend to coexist in the liquid, and successive variation of
the interatomic correlation and the local liquid structure would
induce such an unusually wide liquid range.

X-ray and neutron diffraction measurements of liquid Ga
over a wide temperature range have shown that the structure

factor, $S(Q)$, has an asymmetric first peak. It exhibits a
covalent behavior, being similar to that of group-IV elements
like Si or Ge with a shoulder at about $30\,nm^{-1}$, which lies on
the high Q side of the first peak. The coordination number,
however, ranges from 10 to 12 near the melting point, which
indicates a property similar to that in a typical densely packed
simple liquid metal, but unlike for liquid Si or Ge.

Several theoretical attempts have been carried out using
MD simulations with pseudopotential theory [2] or recent
ab initio [3, 4] techniques for a detailed and consistent
understanding of the interplay between electrons and ions
underlying the physical properties of this interesting liquid
metal. They all could reproduce qualitatively the $S(Q)$ spectra
and the corresponding pair distribution function, $g(r)$.

Hafner and Jank [2] calculated the effective pair potential
of liquid Ga at $100\,°C$ using a pseudopotential theory, and
performed an MD simulation using it to obtain the atomic
structure. Then, the electronic structure was derived using a
self-consistent linear muffin tin orbital supercell calculation,
which shows a nearly free-electron-like DOS feature. Gong
et al [3] succeeded in inquiring into the precise microscopic
nature of both the electronic and structural phenomena in liquid

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Ga using a fully FPMD calculation, although it was done only at relatively high temperature of $T \sim 1000$ K. Their findings are: (1) covalent and metallic characters coexist. The covalent aspect is reflected by the presence at each instant of time of a certain amount of very short-lived Ga_2 covalent *molecules*. (2) The 30 nm^{-1} shoulder in $S(Q)$ can be unambiguously associated with the presence of the short-lived covalent bonds. (3) In spite of the covalent aspects, the DOS at the Fermi level no longer exhibits the aforementioned minimum.

In order to obtain the dynamical structure factor, $S(Q, \omega)$, Holender *et al* [4] performed an FPMD simulation at 429 °C and 709 °C covering a long observation of 8 ps. Sound wave peaks are clearly visible in $S(Q, \omega)$ at small Q values, and the obtained results for the dispersion curve lie on the line predicted by the velocity of sound. The electronic DOS is again very close to the free-electron form.

As pointed out by the theories, the microscopic dynamics is important information for the understanding of the physical properties of liquid Ga. To our knowledge, the first $S(Q, \omega)$ was presented in a review article by Defrain [5], where a quasielastic spectrum measured by inelastic neutron scattering (INS) at $T = -60$ °C was exhibited. Bermejo *et al* performed an INS experiment on liquid Ga at 57 °C [6] using fast neutrons with a final energy of 14.8–33.9 meV to cover a broader kinematic range for exploring collective excitations. However, it was found that liquid Ga cannot sustain finite-frequency excitations. They supposed that this was caused by the interplay of the relatively high longitudinal viscosity and the existence of a frequency spectrum covering a large range in frequency space. However, in a second INS at a higher temperature of 700 °C [7], they observed well-defined (not overdamped) collective excitations. It should be noted that compared to the FPMD simulation by Holender *et al* [4], the frequencies of short-wavelength modes increase noticeably faster (>30 %) with Q than predicted by classical hydrodynamics. In addition, two excitation modes were found in the Q range beyond 12 nm^{-1}, which had not been observed in liquid metals so far.

On the other hand, Scopigno *et al* [8] measured inelastic x-ray scattering (IXS) of liquid Ga at 42 °C in the Q range 3.3–20.0 nm^{-1}, and demonstrated the existence of acoustic-like modes up to Q values larger than a half of the first maximum position in $S(Q)$, in contrast to the earlier INS result [6]. The observed *positive* dispersion is limited to be several %, and the appearance of a second collective excitation was not reported. Very recently, a new INS measurement was carried out by Bove *et al* [9] at 47 and 700 °C. The observed propagating modes exist at both the temperature up to $Q \sim 10$ nm^{-1}. The acoustic-like modes were found with a velocity slightly exceeding the bulk sound velocity, in good agreement with the IXS result [8]. In addition, the mode velocity and damping were found to be almost temperature independent.

We have again measured IXS for liquid Ga at 100 °C over a wider Q range of 3.0–28.0 nm^{-1} including the Q position of the first maximum in $S(Q)$, and using a higher resolution IXS spectrometer. In this paper, we report results of $S(Q, \omega)$, and discuss microscopic dynamics in this exotic liquid metal by comparing to the previous INS [7, 9] and IXS [8] data and

the FPMD simulation result [4]. In particular, the possibility of the existence of the low frequency excitation is argued.

2. Experimental procedure

High resolution IXS is an up-to-date technique which permits the study of the Q dependence of excitations in the meV range. Hence this method is suitable to investigate liquid dynamics, which has so far been exclusively the domain of INS. However, it is worthy noting that many limitations of neutron experiments do not appear using IXS [10]. For instance, the energy–momentum relation of the x-ray photons allows an almost unlimited energy transfer ω at any accessible momentum transfer Q, and the scattering intensity in the range of some meV is purely coherent. The difficulty of this technique for the study of liquid dynamics lies in the necessity to provide energy resolutions in the meV range, while the incident radiation is in the range of some ten keV. Such a high resolution, together with sufficient counting rates, can now be achieved at third-generation synchrotron radiation facilities using Bragg scattering from perfect single crystals in an extreme backscattering geometry.

The present experiment was carried out using a horizontal IXS spectrometer [11] at the beamline 3-ID-C of the Advanced Photon Source, Argonne National Laboratory. There, a highly resolved monochromatized x-ray beam of 3×10^8 photons s^{-1} emitted from an undulator insertion device, was achieved by two nested Si channel-cut crystals which allowed to vary the incident x-ray energy between 21.5 and 21.7 keV. The incident radiation was focused onto the sample by a toroidal mirror, producing a small spot size of 0.2×0.1 mm^2. The (18 6 0) backscattering reflection (89.975°) of a two-dimensionally curved Si analyzer with a diameter of 10 cm was used for the energy analysis of the scattered radiation, which was placed on a table, about 6 m apart from the sample position. Since its temperature was controlled within ±1 mK near room temperature, it exclusively selects x-rays at 21.657 keV. The overall energy resolution of the spectrometer was determined from the scattering of a Plexiglas sample measured at the first maximum of $S(Q)$, and a pseudo-Voigt resolution function was found comprising a width of 1.9 meV full width at half-maximum (FWHM). The Q resolution was ±0.5 nm^{-1}. Details of the IXS spectrometer are given elsewhere [11].

The Ga sample was contained in a thin-walled (0.25 mm) single-crystal sapphire cell (the so-called Tamura-type cell [12]), which is highly transparent to hard x-rays. The scattering contribution of the cell comes from only intrinsic sapphire crystal phonons. However, due to the fast sound velocity of sapphire (~11 000 m s^{-1}), these excitations lie far outside the ω–Q range of interest explored in this experiment. The high x-ray absorption by liquid Ga was handled by reducing the Ga sample thickness to about 50 μm, which roughly corresponds to an 1/e absorber. Details of the sapphire cell construction are given elsewhere [12]. The cell was located in a vessel equipped with continuous Be windows [13] capable of covering scattering angles between 0° and 25°. It was applied with 1.5 bar of high purity grade He gas. The temperature of 100 °C was achieved using an Mo resistance heater of 0.5 mm
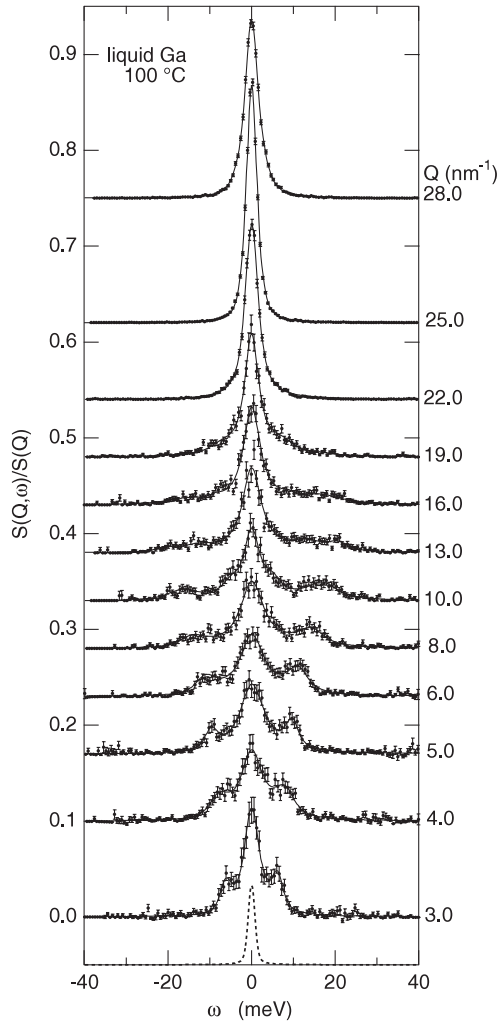
Figure 1. IXS spectra normalized to the corresponding intensity are shown by dots with error bars. The solid curves indicate the best fits of the DHO model convoluted with the resolution function (dashed curve) to the experimental data.
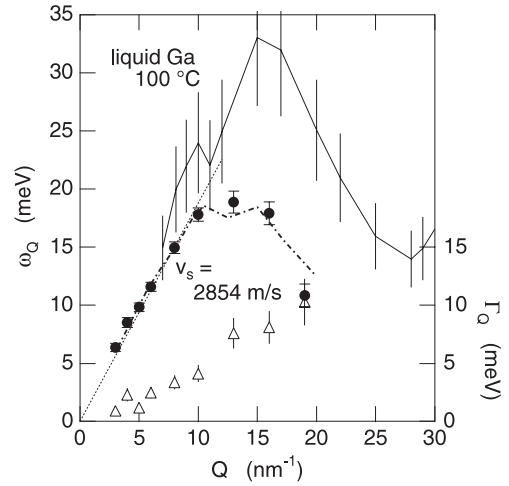

Figure 2. Dispersion relation (full circles) and width (empty triangles) of the inelastic excitations in liquid Ga at 100 °C obtained from the DHO fits with a single DHO. The dotted line represents the dispersion of hydrodynamic sound. The chain line shows the results of a previous IXS experiment [8], and the solid line with error bars are the INS results by Bermejo *et al* [7] at a higher temperature of 700 °C.

With increasing Q, the energy position of the inelastic excitation increases. This clearly demonstrates that the inelastic excitation originates from propagating modes, which was found in various liquid metals [15]. The width of the quasielastic and inelastic lines become broader with increasing Q, and eventually seems to be highly damped at $Q \sim$ 13–19 nm^{-1}.

4. Discussion

Since the resolution width is in the same range as the quasielastic and inelastic components of the spectra, a resolution correction was necessary. This was accomplished by noticing that the measured intensity is a convolution of $S(Q, \omega)$ and the experimentally determined resolution function. This method requires an appropriate model function for $S(Q, \omega)$ as an input. We approximated the central line by a Lorentzian at lower Q values below 19 nm^{-1} and as a pseudo-Voigt function at higher Q. The inelastic contribution was described by a damped harmonic oscillator (DHO) [16] given as

$$\left[\frac{1}{1 - e^{-\hbar\omega/k_B T}}\right] \frac{A_Q}{\pi} \frac{4\omega\omega_Q\Gamma_Q}{(\omega^2 - \omega_Q^2)^2 + 4\Gamma_Q^2\omega^2}. \quad (1)$$

Here, A_Q and ω_Q are the magnitude and energy of the inelastic excitations, and Γ_Q is the width close to half-width at half-maximum (HWHM).

Full circles in figure 2 indicate ω_Q obtained from the fits assuming only a single phonon mode. The dotted line represents the dispersion of hydrodynamic sound, and its slope is given by the adiabatic sound velocity [17]. As seen in the figure, the dispersion relation of ω_Q is slightly larger in energy ($\sim$13 %) than the hydrodynamic prediction. This so-called positive dispersion was already found earlier in several liquid

in diameter, and monitored with two W–5%Re/W–26%Re thermocouples.

The IXS experiments were carried out at twelve Q values between 3.0 and 28.0 nm^{-1}. A typical energy scan ranged from -40 to $+40$ meV, and took about 3.5 h. To improve statistics, 3–6 scans were performed below 13 nm^{-1}, where the scattering intensity is weak but $S(Q, \omega)$ is noticeably influenced by collective excitations. Empty cell measurements were separately carried out and used for background correction.

3. Results

Figure 1 shows the inelastic x-ray scattering spectra of liquid Ga at 100 °C [14] normalized to the respective integral intensity, which is almost identical to $S(Q, \omega)/S(Q)$ besides the resolution broadening. Also given by a dashed curve is the experimentally obtained resolution function. At low Q values, inelastic excitation modes are clearly seen as peaks or shoulders at both the sides of the central peak.
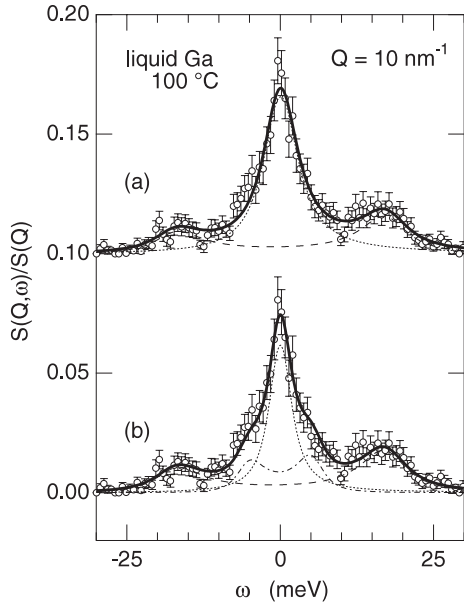
Figure 3. $S(Q, \omega)/S(Q)$ data at $Q = 10.0$ nm^{-1} (circles with error bars) fitted using (a) single and (b) double DHO functions. The thick solid curves on the data indicate the best fits, in which the dotted curves are Lorentzian approximating the quasielastic line, and dashed or chain lines the DHO components for inelastic excitations.

Figure 4. Dispersion relation (circles) of the collective modes in liquid Ga when two DHO functions are assumed for inelastic excitations. The dashed line represents the dispersion of hydrodynamic sound [17], and crosses show the dispersion assuming a single excitation (the same as full circles in figure 2). The full and empty triangles show the results of high and low energy collective excitations obtained from the FPMD simulation performed 429 °C by Holender *et al* [4]. The solid lines with error bars are the INS results by Bermejo *et al* [7] at a higher temperature of 700 °C.

metals [15]. The chain curve shows the dispersion relation obtained from a previous IXS experiment by Scopigno *et al* [8] at 42 °C. Although the measured temperatures were slightly different, the agreement between the two data sets is excellent. Good agreement is also seen in a recent INS data measured by Bove *et al* [9]. On the other hand, the solid lines in figure 2 exhibit INS results at higher temperature of 700 °C by Bermojo *et al* [7]. Since the energy positions of the modes are rather expected to decrease with increasing temperature, the dispersion relation obtained from this INS experiment appears to be considerably too high compared to the IXS results and the recent INS results. Moreover, the results of a first-principles molecular dynamics simulation by Holender *et al* [4] showed almost no positive dispersion beyond 5.8 nm^{-1} even at the higher temperatures of 429 and 709 °C, which also contradicts the INS data by Bermejo *et al* [7] but is in agreement with the other inelastic experiments.

Open triangles in figure 2 exhibit Γ_Q. With increasing Q, Γ_Q slowly increases. A practical quantity to compare the broadening of the excitations with other liquid systems is the ratio Γ_Q/ω_Q. For liquid Ga, values between 0.12 and 0.23 are found in the first quasi-Brillouin zone, i.e., Q values up to $Q_0/2$, where Q_0 is the position of the first maximum in $S(Q)$ of ~25 nm^{-1}. These values are slightly larger than those in liquid Cs [18], where the ratio ranges between 0.03 and 0.19 in the corresponding Q zone, indicating a slightly shorter lifetime of the longitudinal inelastic modes in liquid Ga with respect to those in liquid Cs.

The acoustic excitation lifetimes are shortened by heat flow and viscous drag forces. Within the thermodynamic limit [19], the width of the inelastic peaks, WQ^2, can be estimated from hydrodynamic equation,
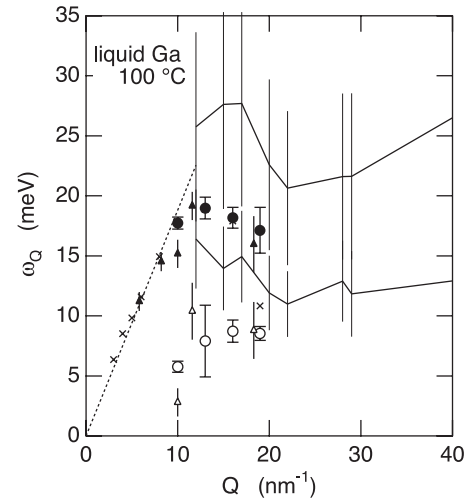
$$W = \frac{1}{2}\left[(\gamma - 1)D_T + \frac{\eta}{\rho}\right], \qquad (2)$$

where γ is the ratio of specific heats at constant pressure and constant volume, D_T the thermal diffusivity, η the longitudinal viscosity, and ρ the density. Within the hydrodynamic limit ($Q \to 0$), the Q dependence of Γ_Q is proportional to Q^2. The present IXS experimental result is, however, almost linear with Q, which indicates that the Q range of the present experiment is already outside of where the above hydrodynamic theory can be applied.

Figure 3(a) shows the $S(Q, \omega)/S(Q)$ spectrum at $Q = 10.0$ nm^{-1} (circles with error bars) together with the best fit curve using a single DHO for the excitations (solid curve). The dotted curve is a Lorentzian approximating the quasielastic line, and the dashed curve is a DHO component for inelastic excitations. As seen in this figure, the data do not perfectly lie on the model function. In particular, there is a distinct inelastic contribution visible, which is located at about 5–10 meV and appears as a weak shoulder in the quasielastic peak. It cannot be reproduced in the fitting procedure with one DHO function. Also, the fit underestimates the maximum of quasielastic peak. Similar discrepancies between the data and the fits are also seen in the previous IXS data at $Q = 12.5$ nm^{-1} in figure 2 of [8], as well as in the recent INS data at $Q = 10$ nm^{-1} in figure 8 of [9], although different model functions were used for the single excitation mode. Therefore, an additional excitation mode is needed to reproduce the $S(Q, \omega)$ spectra in this Q region. Figure 3(b) shows the fitted results using two DHOs. The dotted curve represents again a Lorentzian for the quasielastic peak, and the dashed and chain curves are the
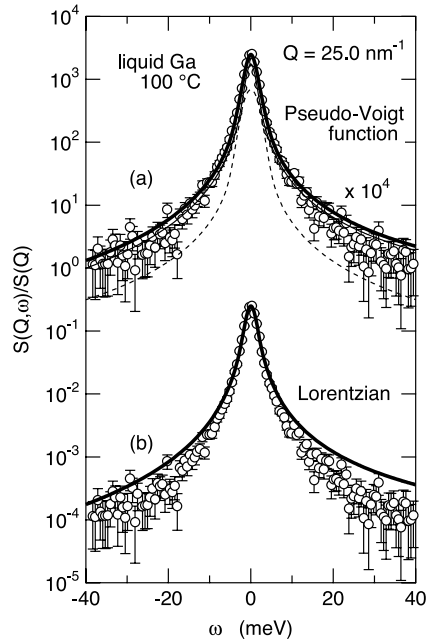
Figure 5. The thick solid curves show the best fits of (a) pseudo-Voigt function and (b) Lorentzian, respectively, to the quasielastic peak given by circles with error bars at $Q = 25.0$ nm^{-1}. Thin solid and dashed curves represent, respectively, the Lorentzian and Gaussian components of pseudo-Voigt function.

Figure 6. The Q dependence of the quasielastic line width Γ_0. With increasing Q, a Gaussian component (open circles) is needed in addition to an usual Lorentzian component (full circles) to model the central line. Crosses indicate the width of the pseudo-Voigt fits. Triangles give the Gaussian fraction, c. The arrow indicates the Q position of the first maximum in $S(Q)$, and the dotted curve is a representation of equation (4) [26].

DHO functions to model the inelastic excitation contributions. The advantage of the two DHO functions becomes apparent regarding the average value of χ^2 per degrees of freedom of 1.11 compared to $\chi^2 = 1.30$ for the single DHO fit.

Circles in figure 4 show the dispersion relation using the model assuming the double DHOs. Crosses indicate the dispersion relation assuming the single excitation, the same as the solid circles in figure 2. The higher ω_Q values indicated as closed circles almost coincide with the dispersion with the single excitations. The excitations with the lower ω_Q values appear at about a half of the energy of the higher ω_Q in a Q range of 10–20 nm^{-1}. Hence these excitations strongly resemble the transverse phonon modes in the solid state.

It should be noted that the $S(Q, \omega)$ spectra obtained from the FPMD simulation by Holender *et al* [4] also exhibit double excitation modes in the inelastic region. The full and empty triangles show the results of high and low energy collective excitations obtained from the FPMD simulation at 429 °C. Note that the lower energy excitation is not seen at 8.2 nm^{-1} and appears beyond 10 nm^{-1}. Although the temperatures of the theory and the experiment are different from each other and the energy positions are rather scattered, the coincidences are fairly good.

As mentioned before in the introductory section, liquid Ga has a covalent nature in its structural properties, which may lead to the proposed large viscosity on the microscopic scale. Thus, it is possible that the lower energy excitations originate from transverse acoustic modes as in a crystal. The existence of transverse mode was reported for a limited number of liquids with permanent bonds. For example, for covalent bonded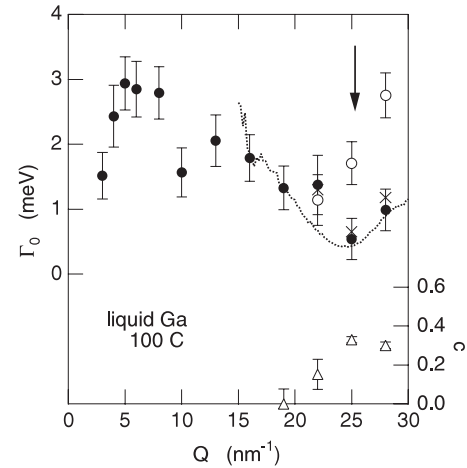 liquid SiO_2, the indications for transverse modes were drawn from a Brillouin scattering spectroscopy [20]. Similarly, a peculiarity in a phonon branch in hydrogen bonded liquid water was observed by IXS [21] and interpreted according to this view. The interpretation was supported by a subsequent MD simulation [22]. Hence it is tempting to speculate that the existence of transverse modes in a liquid is closely related to the degree of correlation between the atoms or molecules.

Bermejo *et al* [7] also pointed out the existence of two excitations beyond Q of 12 nm^{-1} at a higher temperature of 700 °C. However, since the energy positions of the excitations were much larger than the present IXS and the FPMD results, it is unclear whether they discussed the same excitation modes or not.

Usually, the quasielastic line is modeled by a Lorentzian. However, this choice is not suitable for liquid Ga beyond $Q = 20$ nm^{-1}. Instead, we used a pseudo-Voigt function in the same way as was done in the analysis of IXS data on liquid Si [23] and Sn [24]. This function is a weighted linear combination of a Lorentzian and a Gaussian, given by

$$\frac{1-c}{\pi} \frac{\Gamma_{0L}}{\Gamma_{0L}^2 + \omega^2} + \frac{c}{\Gamma_{0G}} \sqrt{\frac{\ln 2}{\pi}} \exp\left[-\ln 2 \left(\frac{\omega}{\Gamma_{0G}} \right)^2 \right]. \quad (3)$$

Here, Γ_{0L} and Γ_{0G} are respectively the widths (half-width at half-maxima, HWHM) of the Lorentzian and Gaussian contributions, and c is the mixing parameter that determines the Gaussian fraction.

Figure 5(a) shows the best fit of the pseudo-Voigt function (thick solid curve) convoluted with the resolution function at $Q = 25.0$ nm^{-1} near the $S(Q)$ maximum. The thin solid and dashed curves are respectively the Lorentzian and Gaussian components of the pseudo-Voigt function. Although the model overestimates the data in the tail region, the quality of the pseudo-Voigt fit is much better than the best fit of Lorentzian given in figure 5(b).

Full circles, empty circles, and triangles in figure 6 represent the fitted results of Γ_{0L}, Γ_{0G}, and c respectively. Crosses show the width (HWHM) of the pseudo-Voigt functions. In the low Q region, Γ_{0L} increases rapidly with increasing Q. Then, it has a maximum around $Q \sim 6$ nm^{-1}, followed by a gradual decrease and a broad minimum near the Q position at the first maximum in $S(Q)$ indicated by the arrow. The Gaussian contribution starts to get mixed into the usual Lorentzian around $Q \sim 20$ nm^{-1}, and reaches about 35% at $Q = 25$ nm^{-1} where the $S(Q)$ has a first maximum.

The relationship between the width of the quasielastic line and $S(Q)$ was derived by de Gennes [25], who pointed out that Γ_0 shows minima at Q values where $S(Q)$ comprises maxima. This is the well-known *de Gennes narrowing*. A more quantitative relation between the Γ_0 and $S(Q)$ was given by Cohen *et al* [26]. Analyzing the width of neutron spectra for several dense hard-sphere fluids within the context of a kinetic theory, they were able to obtain the following expression, which is valid in the region of the $S(Q)$ maximum;

$$\Gamma_0(Q) = \frac{D_E Q^2}{S(Q)} \frac{1}{1 - j_0(Q\sigma) + 2j_2(Q\sigma)}. \qquad (4)$$

Here, D_E is the Enskog diffusion coefficient, j_0 and j_2 the zeroth-and second-order spherical Bessel functions, respectively, and σ the hard-sphere diameter. It was demonstrated that the application of equation (4) to a variety of simple liquids gives good agreement with the corresponding experimental results [26].

The $S(Q)$ spectrum was taken from the x-ray diffraction result [27]. Using the σ value calculated from the average interatomic distance $(V/N)^{1/3} = 0.267$ nm, the D_E value could be obtained to be 1.76×10^{-9} m s^{-1}. The theoretical values of Γ_0 are described by the dotted curve in figure 6. As seen in the figure, the experimental values of the Lorentzian component Γ_{0L} coincide well with the theoretical curve over a wide Q range near the Q position of the first maximum in $S(Q)$. The successful application of the theory supports the view that the microscopic particle dynamics of liquid Ga can be essentially modeled within the framework of a simple hard-sphere pair interaction. The experimental values of the Gaussian component Γ_{0G}, however, seem to be independent of the theory. Thus, the existence of the Gaussian components in the quasielastic line cannot be understood with the context of the kinetic model.

A noteworthy question is what the origin of the Gaussian contribution in the quasielastic lineshape is. At the Q values near the $S(Q)$ maximum, where structural correlations to nearest neighbors dominate, the decay is no longer exponential-like. This finding suggests that the diffusion of the Ga atoms is not governed by a simple random walk or Fick's law. Instead, it can be concluded that subpicosecond correlations between neighboring atoms exist in liquid Ga, which may originate from cluster or molecular aggregations in this non-simple liquid metal. The lifetime of the cluster formation can be estimated from the width of the Gaussian contribution in $S(Q, \omega)$ at $Q = 25$ nm^{-1} to be $1/\Gamma_{0G}$ of 0.39 ps. Such short-lived bonds were already observed in a FPMD simulation by Gong *et al* [3], who obtained the lifetime

of the covalent bonds to be 0.05 ps at 1000 K, about one order of magnitude smaller than the present experimental result. This disagreement of the lifetime may come from the large temperature difference.

It should be noted that similar evidence for subpicosecond cluster formation was found in liquid Si [23] and liquid Sn [24]. However, the duration of the instantaneous atomic group is estimated to be only 0.08 ps in liquid Si (much shorter than that in liquid Ga), and 0.55 ps in liquid Sn (longer than that in liquid Ga). Such a difference can originate from the differences of the atomic mass, the temperature measured, and the strength of the covalent nature.

Very recently, Scopigno *et al* [28] discussed the de Gennes narrowing in liquid Ga using the aforementioned model by Cohen *et al* [26] shown in equation (4). Although liquid Ga is considered as a non-hard-sphere liquid metal, the application of this kinetic model reproduces the Q dependence of Γ_0 very well over a wide Q range of 18–70 nm^{-1} if the hard sphere diameter and the Enskog diffusion coefficient are used as free parameters. They evaluated the best fit value of $\sigma = 0.279$ nm, which is, however, larger than the average interatomic distance of 0.265 nm at 42 °C. Thus they suggested that such a large value of the effective atomic diameter could be an indication of the supra-atomic nature of the *effective particles*, which could be regarded as the mean size of the *clusters*.

The present measurement was performed in a limited Q range up to 28.0 nm^{-1}. We found that the quasielastic lineshape of $S(Q, \omega)$ near the $S(Q)$ maximum contains a Gaussian contribution, while Scopigno *et al* analyzed the data using a Lorentzian only. Thus, we could not confirm their finding. The existence of the Gaussian contribution with a broader width in the quasielastic line may cause the broadening of the Γ_0 values in their analysis, which results in the apparently larger hard-sphere diameter in their analysis. The de Gennes narrowing in liquid Ga may have a further underlying concepts. For clarifying them, experiments at different temperatures and for other systems are needed to be carried out.

5. Conclusion

The $S(Q, \omega)$ spectra of liquid Ga were measured at 100 °C using the high resolution IXS spectrometer at 3-ID-C/APS. The spectra obtained clearly demonstrate the existence of longitudinal propagating modes at small Q values, similar to a previous IXS result at 42 °C by Scopigno *et al* and an INS one at 47 °C by Bove *et al*, but unlike an INS one at 57 °C by Bermejo *et al*. The dispersion relation of the excitations deviates positively from the hydrodynamic prediction by about 13%. There are two new findings. Firstly, an additional lower energy excitation is necessary to reproduce $S(Q, \omega)$ spectra in the Q range beyond 10 nm^{-1}, in agreement with the result of a first-principles molecular dynamic simulation [4], which may indicate a transverse acoustic mode in this peculiar liquid metal similar to the crystal phase. The existence of two inelastic excitations was pointed out by Bermejo *et al*, but their excitation energies are very different. Secondly, the quasielastic line comprises a Gaussian contribution at Q

values near the first maximum in $S(Q)$, which may indicate the existence of short-lived covalent correlations in liquid Ga with a lifetime of 0.39 ps.

Acknowledgments

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