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A viscoelastic analysis of IXS spectra from He/Ne mixtures

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A generalization of the viscoelastic model to binary systems has been applied to analyze Inelastic X-ray Scattering spectra from $He_{0.8}Ne_{0.2}$ mixture. Experiments have been carried out at temperature $T = 82$ K and number density $n = 18.5$ nm⁻³. In order to test the reliability of such a generalization, we measured a few representative Inelastic X-ray Scattering spectra. The model exhaustively describes the experimental data and provides a determination of the spectral densities of the separate mixture components as well as the ones of both density and concentrations fluctuations. These results can be considered as a first test for further developments.

Keywords: fluid binary mixtures; viscoelastic model; Inelastic X-ray Scattering

1. Introduction

It is very well assessed that, in the hydrodynamic limit, a binary mixture behaves as a one-component fluid, its spectrum being characterized by only one inelastic mode whose propagation speed, c_s , results from the weighted average of the adiabatic sound velocities of the separate components. In the 1986 a molecular dynamics simulation [1] demonstrated that if the two components have very disparate masses a new faster sound mode can be observed at sufficiently short wavelengths (λ) and high frequencies (ω). Moreover, a second excitation dispersing with a group velocity lower than c_s has been observed by light scattering experiments [2, 3] and ascribed to density fluctuations separately propagating through the heavier component only.

The whole body of results seems somehow supportive of the occurrence of a bifurcation of the hydrodynamic excitation, which splits into two separate modes whenever the exchanged wave-vector (Q), overcome some threshold value.

Theoretical studies accounting for such phenomenology have been developed either in the frame of the Revisited Enskog theory (RET) [4–6] or within the so called "two-temperature" approach [7]. Recently, a new approach has been proposed on the Generalized Collective Modes (GCM) description [8, 9]. Even if all these theories provide a consistent account of the observed phenomenology, they strongly differ in quantitative prediction of transport parameters along the crossover region (from hydrodynamic to sound-decoupled regime) and also suggest different physical interpretations of the observed dispersive behavior.

Experimental evidences on the occurrence of two coexisting collective modes mainly come from inelastic scattering experiments performed either in nobles gas

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mixtures [2, 10–12] or in molten alloys, such as Li_4Pb [13–15]. The experimental insight on the dynamics of binary mixture is, however, deeply hampered by the scarce hope of properly resolving the two excitations in the spectra [10] and also by the rather limited Q -range accessible by most of instruments. Actually these limitations have so far prevented an unambiguous validation of any of the mentioned theoretical predictions [11, 12].

These difficulties are even worsened by the lack of a well assessed analytical model providing an accurate approximation of the spectrum of mixtures beyond the hydrodynamic limit. In order to amend to this lack, we test here the soundness of a new line-shape description obtained as a generalization of the memory function formalism to the case of binary mixtures [16]. Interestingly enough, such formalism has been extensively employed for single component (both atomic and molecular) fluids in the mesoscopic range [17–19], yet its "two-components" extension has never been considered to describe experimental spectra of binary mixture.

2. Experimental method

The experiment was performed at the ID28 beamline of the European Synchrotron Radiation Facility (Grenoble, France). The spectrometer was operated using the Si(11,11,11) reflection from both monochromator and analyzer, which yields an overall energy resolution bandwidth of about 1.5 meV . The present configuration of ID28 spectrometer allows to simultaneously measure Inelastic X-ray Scattering (IXS) spectra at nine different Q -values mutually separated by a constant offset of about 1.5 nm^{-1} . The chosen $\hbar\omega$ -range was $\pm 40 \text{ meV}$ with steps of 0.3 meV . The acquisition time for each $\hbar\omega$ point was $\approx 100 \div 200 \text{ sec}$ in order to obtain high statistical quality data. The actual Q -values of IXS spectra were established by varying the angle of the spectrometer arm in between 5° and 20° interval. The explored Q -range was $6 - 14$ and $24 - 30 \text{ nm}^{-1}$. The Q indetermination (ΔQ) was determined by the aperture of the analyzer slits and set to be better than 0.3 nm^{-1} at the lowest Q 's. At the highest Q 's a more relaxed ΔQ ($< 1 \text{ nm}^{-1}$) was instead tolerable and this allowed us to keep the slit wide open thus increasing the count rate and compensating the strong intensity decrement in this dynamic region.

The high purity $He_{0.8}Ne_{0.2}$ sample (purchased from Gas Liquide) was embedded in a high pressure hydrostatic cell made of Inconel 625 with 1 mm thick crystal diamond windows. A 12 mm , sample length combined with a 2.3 mm window diameter, allowed a 30° angular acceptance, necessary to probe the highest exchanged momenta. The cell was connected to an external high pressure generator through a capillary. The pressure stability was monitored by a pressure gauge and found to be better than $\pm 5 \text{ bar}$, within the typical acquisition time ($12 \div 18 \text{ hours}$) of a single set of IXS spectra. A closed-cycle He cryostat and a resistive heater in thermal contact with the sample cell were used to steadily keep the sample at the desired temperature. The actual sample temperature was measured by two thermocouples and electronically controlled by a PID device. The overall temperature stability was better than 0.2 K . All the experimental setup was kept under high vacuum ($\sim 10^{-8} \text{ mbar}$) in order to avoid thermal gradients and spurious scattering from the air surrounding the cell. The IXS spectra of $He_{0.8}Ne_{0.2}$ were collected at a temperature $T = 82 \text{ K}$ and $P = 320 \text{ bar}$, corresponding to a sample number density of 18.5 nm^{-3} .

Ancillary measurements of the energy resolutions of each of the nine analyzers have been performed through the acquisition of the nearly elastic scattering of a Plexiglas sample at low temperature (5 K) and at the Q of its first diffraction peak. It has been found that both the shape and the width (about $1.4 - 1.7 \text{ meV}$) of the

energy resolution function, $R(\omega)$ are quite insensitive to the specific experimental conditions (i.e., to the angle of the spectrometer arm and slit aperture). Empty cell measurements were also performed for comparison. The measured scattering was found more than an order of magnitude weaker than the signal from the sample, even at the higher Q 's, thus it has been neglected in the analysis to be discussed.

Fig.1 displays two selected IXS spectra at the indicated Q -values along with the measured shape of $R(\omega)$. The spectral profiles $I(Q, \omega)$, were approximated by the following model function:

$$I(Q, \omega) = I(Q)[n(\omega, T)S^{ex}(Q, \omega) \otimes R(\omega)] + c(Q). \quad (1)$$

Here the symbol $\otimes$ represents the numerical convolution operator, $I(Q)$ is an overall intensity factor, $c(Q)$ is a background term taking into account both the electronic background of the detector and the environmental one. The factor $n(\omega, T)$ is a frequency dependent factor accounting for the detailed balance:

$$n(\omega, T) = \frac{\hbar\omega/k_B T}{1 - e^{-\hbar\omega/k_B T}}, \quad (2)$$

where k_B is the Boltzmann's constant and T the temperature. The "experimental" $S^{ex}(Q, \omega)$ is related to the partial dynamic structure factors through weighting coefficients, $I^{\alpha\beta}$, which depend on the specific probe and measurement conditions. In the Faber-Ziman representation [20]:

$$S^{ex}(Q, \omega) = I^{11}S^{11}(Q, \omega) + I^{12}S^{12}(Q, \omega) + I^{22}S^{22}(Q, \omega), \quad (3)$$

where the apices 1 and 2 refer to the two specie components. By comparing eq.1 with the IXS cross section for binary mixtures, one obtains:

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \omega_f} \propto \sum_{\alpha, \beta=1,2} (x_\alpha x_\beta)^{1/2} f^\alpha(Q) f^\beta(Q) S^{\alpha\beta}(Q, \omega), \quad (4)$$

from which it follows that $I^{\alpha\beta} = (x_\alpha x_\beta)^{1/2} f^\alpha(Q) f^\beta(Q)$, being x_α and $f^\alpha(Q)$ the concentration and the atomic form factor of particles of specie $\alpha = 1, 2$. Here $S^{\alpha\beta}(Q, \omega)$ is the partial dynamic structure factor, defined as

$$S^{\alpha\beta}(\mathbf{Q}, \omega) = \frac{1}{(x_\alpha x_\beta)^{1/2}} \int_V d\mathbf{r} \int_{-\infty}^{\infty} dt \langle \delta n_\alpha \delta n_\beta \rangle e^{i(\mathbf{Q} \cdot \mathbf{r} - \omega t)}, \quad (5)$$

being V the volume occupied by the system and δn_α the fluctuations of partial number density of specie α . $S^{\alpha\beta}(\mathbf{Q}, \omega)$ are calculated using a generalization of the viscoelastic model to binary systems as described in the next section.

The data analysis consists of a fitting procedure based on a standard minimization of a χ^2 function, defined as:

$$\chi^2 = \sum_{i=1}^N \frac{(I(Q, \omega_i) - y_i)^2}{\sigma_i}, \quad (6)$$

where N is the number of data points in the IXS spectra, y_i are the measured counts at the energy transfer $\hbar\omega_i$ and σ_i is their relative standard deviation. A Poisson statistics is assumed, so that $\sigma_i = \sqrt{y_i}$. The minimization was performed

with the software MATLAB, employing two routines: one exploiting the Nelder-Mead Simplex algorithm [21] and, subsequently, a second one based on a large-scale algorithm, which uses the method of preconditioned conjugate gradients (PCG) [22]. Finally, the errors were estimated through the calculation of the covariance matrix.

3. Data analysis

For a one-component system the dynamic structure factor $S(Q, \omega)$ is given by the expression:

$$S(Q, \omega) = \frac{1}{\pi} \Re(\tilde{F}(Q, -i\omega)), \quad (7)$$

where the intermediate scattering function, $\tilde{F}(Q, -i\omega)$ can be evaluated, in the memory function frame, through the continue fraction expansion, truncated to the third order:

$$\tilde{F}(Q, -i\omega) = S(Q) \left(-i\omega + \frac{M(Q)}{-i\omega + \frac{N(Q)}{-i\omega + K(Q)}} \right)^{-1}. \quad (8)$$

$S(Q)$, $M(Q)$, $N(Q)$ and $K(Q)$ are the zero, first, second and third order memory function calculated at $t = 0$, respectively. $S(Q)$ can be identified with the static structure factor, while $M(Q)$, $N(Q)$ and $K(Q)$ can be associated to the zero and infinite sound frequencies, $\omega_0(Q)$ and $\omega_\infty(Q)$, and to the compliance relaxation time, $\tau_c(Q)$, through the following relations:

$$\omega_\infty^2(Q) = N(Q) + M(Q) \quad (9)$$

$$\omega_0^2(Q) = M(Q) = \frac{k_B T}{MS(Q)} \quad (10)$$

$$\tau_c = K^{-1}(Q) \frac{\omega_\infty^2}{\omega_0^2}, \quad (11)$$

being M the mass of the particles. By imposing that in the $Q \rightarrow \infty$ limit the expected short time behavior is reached, it is possible to link $K(Q)$ and $N(Q)$:

$$K(Q)^{-1} = \frac{\sqrt{\pi}}{2\sqrt{N(Q)}}, \quad (12)$$

also known as Lovesey relation [23].

As the memory function formalism holds for a generic set of dynamical variables, ν , the generalization of the viscoelastic model to binary systems is, in principle, quite straightforward. The physical problem is focused on the choice of the suitable set of dynamical variables describing the dynamics of the system. By choosing $\nu = (\delta n_1(\mathbf{r}, t), \delta n_2(\mathbf{r}, t))$ (i.e. exploiting the Faber-Ziman representation) the dynamic structure factor result to be a 2×2 matrix, $\mathbf{S}(Q, \omega)$, whose generic elements

are $S^{\alpha\beta}(Q, \omega)$. $\mathbf{S}(Q, \omega)$ can be obtained from the intermediate scattering matrix, $\tilde{\mathbf{F}}(Q, -i\omega)$, assuring it to be hermitian, i.e.:

$$\mathbf{S}(Q, \omega) = \frac{1}{2\pi} [\tilde{\mathbf{F}}^+(Q, -i\omega) + \tilde{\mathbf{F}}(Q, -i\omega)], \quad (13)$$

where $\tilde{\mathbf{F}}^+$ is the adjoint of $\tilde{\mathbf{F}}$. The matrix $\tilde{\mathbf{F}}(Q, -i\omega)$ is, in the viscoelastic model, given by an expression formally equal to eq.8, although in this case $S(Q)$, $M(Q)$, $N(Q)$ and $K(Q)$ are matrixes expressed in the vectors basis $(\delta n_1(\mathbf{r}, t), 0)$ and $(0, \delta n_2(\mathbf{r}, t))$. In order to reduce the number of free parameters in the fitting routine, the Lovesey relation in Eq.12 is used as generalized to the case of binary systems. Explicit calculations of the involved matrix elements $K^{\alpha\beta}(Q)$ as a function of $N^{\alpha\beta}(Q)$ can be found in [16]. The matrix $\mathbf{M}(Q)$ is, moreover, related to $\mathbf{S}(Q)$ through the squared single components isothermal velocities, $Q^2 k_B T / M_{1(2)}$ [16]. The identities $N^{21} = N^{12}(M^{22}/M^{11})$ and $S^{12} = S^{21}$ follow from symmetry requirements.

The parameters determined by the fitting procedure are, therefore: $S^{11}(Q)$, $S^{12}(Q)$, $S^{22}(Q)$, $N^{11}(Q)$, $N^{12}(Q)$, $N^{22}(Q)$, $c(Q)$ and $I(Q)$, while T was fixed to the measured value. $f^{1(2)}(Q)$ were calculated [24] and kept fix during the minimization routine.

As clearly emerges from Fig.1 the agreement between the model function and the raw IXS data is satisfactory for both the reported Q values. As a matter of fact, the χ^2 normalized to the number of degrees of freedom turns out to be rather close to 1. The logarithmic scale emphasizes the agreement also in the high frequency spectral tails.

As will be discussed in the next section, the present data analysis allowed to extrapolate $S^{\alpha\beta}(Q, \omega)$ from a single IXS spectra of the mixture. Matrix expressions equivalent to Eqs.9-11 can be, moreover, defined so that the zero and infinite frequency as well as the compliance relaxation time matrix remain defined. In the chosen representation (i.e. assuming $\nu = (\delta n_{He}(\mathbf{r}, t), \delta n_{Ne}(\mathbf{r}, t))$), the ω_0 , ω_∞ , and τ_c values associated with He and Ne subsystems can be identified with the diagonal elements of the respective 2×2 matrix.

4. Results and discussion

$\mathbf{S}(Q, \omega)$ can be also expressed in the basis defined by the vectors $(\delta n(\mathbf{r}, t), 0)$ and $(0, \delta c(\mathbf{r}, t))$, being $\delta n(\mathbf{r}, t)$ and $\delta c(\mathbf{r}, t)$ the fluctuations of the total density and concentration, respectively. These are related to $\delta n_1(\mathbf{r}, t)$ and $\delta n_2(\mathbf{r}, t)$ by linear combinations. Consequently, the partial dynamic structure factors $S^{nn}(Q, \omega)$ and $S^{cc}(Q, \omega)$ can be obtained by $S^{11}(Q, \omega)$, $S^{22}(Q, \omega)$ and $S^{12}(Q, \omega)$ by means of a change of vectors basis. For each partial dynamic structure factor the respective longitudinal current can be finally obtained as:

$$J(Q, \omega) = \omega^2 S(Q, \omega). \quad (14)$$

In the left panel of Fig.1-(a) the longitudinal partial current $J^{HeHe}(Q, \omega)$, $J^{NeNe}(Q, \omega)$, $J^{HeNe}(Q, \omega)$ (upper frame) are reported as directly extrapolated by the fitting procedure for the IXS spectra at $Q = 9.4 \text{ nm}^{-1}$, while $J^{nn}(Q, \omega)$ and $J^{cc}(Q, \omega)$ are obtained from the former. A low-frequency peak ($\omega_1 \cong 1.9 \text{ meV}$) is observed in $J^{HeHe}(Q, \omega)$, $J^{NeNe}(Q, \omega)$, $J^{nn}(Q, \omega)$ and $J^{cc}(Q, \omega)$. This feature dominates $J^{NeNe}(Q, \omega)$ and $J^{cc}(Q, \omega)$. It is located in a frequency range where the cross

current, $J^{HeNe}(Q, \omega)$ touches negative values, which reveal out-of-phase motion of particles of different specie. The peak centroid is, finally, around $D_{12}Q^2 \cong 1.6 \text{ meV}$, which leads to estimate an inter-diffusion coefficient D_{12} consistent to the one derived from Generalized Enskog theory [25]. These trends suggest that this low-frequency feature can be related to interspecies diffusion dynamics. A further higher frequency peak is observable in $J^{HeHe}(Q, \omega)$ and $J^{nn}(Q, \omega)$ at $\omega_2 \cong 4.6 \text{ meV}$. The phase velocity associated to such mode is $c_2 = \omega_2/Q \cong 750 \text{ m/s}$, a value similar to the speed of sound of pure *He* at the same temperature and equivalent density. Moreover, the peak is located in a frequency range corresponding to positive value of the cross correlation. It can be, thus, related to an acoustic-like collective mode mainly supported by the *He* particles. This scenario strongly suggests a dynamic decoupling of the two components. Similar behavior is observed also in others *He/Ne* mixtures [12]. The partial static structure factors obtained after the minimization routine are $S^{HeHe}(Q) = 1.3 \pm 0.5$, $S^{HeNe}(Q) = -0.23 \pm 0.06$, $S^{NeNe}(Q) = 0.61 \pm 0.05$. A calculation made by using the hypernetted chain (HNC) closure relations in the Ornstein-Zernike (OZ) equation, solved with the Gillan method extended to binary systems, shows consistency with these experimental results [26]. The zero and infinite sound frequencies are $\omega_0^{HeHe} = 2.3 \pm 0.5 \text{ meV}$, $\omega_\infty^{HeHe} = 5.2 \pm 0.3 \text{ meV}$ and $\omega_0^{NeNe} = 1.5 \pm 0.7 \text{ meV}$, $\omega_\infty^{NeNe} = 4.5 \pm 0.2 \text{ meV}$, i.e. $\omega_0^{HeHe(NeNe)}/\omega_\infty^{HeHe(NeNe)} \sim 0.3 \div 0.4$ as usually found for one-component fluids [17, 19, 27]. The compliance relaxation times associated to *HeHe* and *NeNe* correlations have been as well calculated. It is found $\tau_c^{HeHe} = 0.8 \pm 0.4 \text{ psec}$ and $\tau_c^{NeNe} = 1.5 \pm 0.3 \text{ psec}$. These values are, respectively, three and for times bigger than the single specie collision times: $\tau_{coll}^{He} \cong 0.27 \text{ psec}$, $\tau_{coll}^{Ne} \cong 0.36 \text{ psec}$, respectively.

Fig.1-(b) reports the same profiles as in Fig.1-(a), but for $Q = 27.6 \text{ nm}^{-1}$. The first relevant observation concerns the cross current $J^{HeNe}(Q, \omega)$. Its amplitude is much smaller than these of $J^{HeHe}(Q, \omega)$ and $J^{NeNe}(Q, \omega)$. $S^{HeNe}(Q)$ is, as well, nearly vanishing (i.e., $S^{HeNe}(Q) = -0.009 \pm 0.008$). This is consistent with the expectation that the dynamical variables $\delta n_1(\mathbf{r}, t)$ and $\delta n_2(\mathbf{r}, t)$ became statistically independent in the high Q limit, where the struck atoms basically act as free particles [28]. Single peaks are present in $J^{HeHe}(Q, \omega)$ and $J^{NeNe}(Q, \omega)$ at $\omega_1 \cong 11.1 \text{ meV}$ and $\omega_2 \cong 4.9 \text{ meV}$, respectively. The $\omega_{1(2)}$ values are rather close to $(2Q^2 k_B T / M_{1(2)})^{0.5}$, being $(2Q^2 k_B T / M_{He})^{0.5} \cong 10.6 \text{ meV}$ and $(2Q^2 k_B T / M_{Ne})^{0.5} \cong 4.7 \text{ meV}$, which is the expression for $J(Q, \omega)$ maxima corresponding to a Gaussian function of variance $\sigma = Q^2 k_B T / M_{1(2)}$ for $S(Q, \omega)$. This suggests that the spectral density is gradually approaching its single particle limit. According to this finding, moreover, the values of the compliance relaxation times ($\tau_c^{HeHe} = 0.21 \pm 0.08 \text{ psec}$ and $\tau_c^{NeNe} = 0.4 \pm 0.1 \text{ psec}$) are consistent, within the errors, with the single components collision times.

In conclusion, IXS spectra from $He_{0.8}Ne_{0.2}$ mixture at $T = 82 \text{ K}$ and $n = 18.5 \text{ nm}^{-3}$ in the mesoscopic region have been analyzed through a generalization to binary systems of the viscoelastic model [16]. So far, this model has been largely used to successfully describe the dynamic structure factor of one-component systems in this regime. We applied it for the first time to analyze experimental data of a two-component system. These preliminary results are encouraging, yet clearly worth further tests hopefully with the support of parallel molecular dynamics techniques. Moreover, its natural evolution consists in a full characterization of the parameters describing collective dynamics in a binary mixture as function of the thermodynamic conditions.

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Figure 1. IXS spectrum of $He_{0.8}Ne_{0.2}$ mixture at $Q = 9.4 \text{ nm}^{-1}$ (panel (a), left) and $Q = 27.6$ (panel (b), left). The corresponding best fit curve as obtained by the viscoelastic model for binary systems and the instrumental resolution curve are represented by solid red line and dotted black line, respectively. The longitudinal currents extrapolated by the fitting procedure are reported in the right panels.

(a)

(b)