Neutron Brillouin Scattering Study of Collective Dynamics in a Dense He-Ne Gaseous Mixture

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The dynamic structure factor $S(k, \omega)$ of a 77% He and 23% Ne gaseous mixture at $T = 39.3$ K and total number density $n = 15.8$ nm^{-3} has been measured by inelastic neutron scattering at small angles. In the range of wave vectors studied, $0.7 < k/\text{nm}^{-1} < 1.8$, we find clear side peaks at frequencies following the hydrodynamic sound dispersion $c\ k$ of the binary mixture (with c the adiabatic sound speed in the mixture). Surprisingly, neither fast modes nor a transition from normal to fast sound are observed in the above low-k region, indicating that hydrodynamic behavior persists up to k values which are larger than those predicted by theories and computer simulations.

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After the discovery of the so-called fast sound modes in the molecular dynamics simulation of a Li_4Pb liquid alloy [1], and the prediction of the existence of fast kinetic modes in binary rare-gas mixtures [2], theoretical calculations [3,4], computer simulations [5–8], and experiments, both by light [9,10] and neutron [4,7,11–13] scattering, have been devoted to investigations of the existence and behavior of fast and/or slow sound modes in the dynamics of two-component fluids outside the hydrodynamic regime. These studies show that, for wave vectors $k > k_H$, with k_H denoting the upper limit of the hydrodynamic region, there are two propagating kinetic modes, with velocities c_1 and c_2, which are, respectively, higher and lower than the hydrodynamic sound velocity c of the mixture. Differently from hydrodynamic sound modes, related to in-phase atomic motions of both components, the fast mode, with speed c_1, is attributed to oscillations of the light species which are too fast for the heavy particles to follow, while the slow sound, with speed c_2, is interpreted to arise from density fluctuations of the heavy component (in this Letter, subscripts 1 and 2 refer to the lighter and heavier component, respectively).

Experimental evidence of modes propagating faster than the ordinary sound has been obtained for various simple systems, such as H_2-Ar, H_2-Xe, and He-Xe dilute mixtures by light scattering [9,10], and He-Ne and He-Ar dense mixtures by neutron scattering [4,7,11]. The identification of fast sound with the dynamics of the light component alone is supported, on the one hand, by the observation that the corresponding propagation speed turns out to be very close to that of the hydrodynamic sound in the fluid composed of atoms of species 1 only (at the same total density), and, on the other hand, by the fact that, when the partial contributions to the dynamic structure factor can be isolated, as in the case of molecular dynamics simulations, the fast-sound inelastic peaks are visible only in the correlations between the light atoms [1,5–8]. Analogous indications, obtained by light scattering experiments [9,10], induce to relate the slow sound to the heavy component dynamics, since the propagation takes place with a velocity of the order of the adiabatic sound velocity of the equivalent "heavy" fluid.

However, so far, theoretical and simulation results provided contrasting pictures on the way in which the above kinetic modes are connected to the hydrodynamic mode with decreasing k towards k_H. In particular, these suggest either that fast and slow sound merge into the hydrodynamic sound [6,7], or that the fast sound disappears, its frequency dropping to zero at $k \approx k_H$, while the slow sound goes over into the normal sound [4], where the predicted value of k_H for dense He-Ar and He-Ne mixtures is around 0.7 nm^{-1} [4,6,7]. The coexistence of all three modes in a limited k range between 1.5 and 2.5 nm^{-1} has also been reported in one case [6], though not confirmed in another work [7].

It is also worth mentioning that in very recent neutron scattering [12] and simulation studies [13] of liquid Li_4Pb it is demonstrated that the high-frequency mode, supported by the Li atoms only, behaves quite differently from the acoustic mode in pure liquid Li and is rather localized. Here too, molecular dynamics shows that deviations from hydrodynamics appear to set on at about $k = 0.7$ nm^{-1}.

In previous neutron experiments on dense $He_{0.65}Ne_{0.35}$ [4,11] and $He_{0.75}Ar_{0.25}$ [7] gas mixtures, minimum k values of 4 and 2 nm^{-1}, respectively, were reached. Therefore, the region around $k_H \approx 0.7$ nm^{-1} (a value predicted either [4] by extrapolating to lower k a kinetic model that agreed with the neutron data, or [7] by use of the revised Enskog theory) has never been experimentally probed. In both quoted experiments a linear dispersion in good agreement with the sound speed of pure He was found, proving that the transition from hydrodynamic to fast sound takes place at k values lower than those investigated there.

In Ref. [11] it was suggested that the extension of the measurements on dense He-Ne mixtures by means of neutron Brillouin scattering, i.e., at wave vectors about 1 order of magnitude lower than in conventional inelastic neutron scattering, might provide important information on this transition. Indeed, neutron Brillouin experiments on one-component fluids [14,15] have shown that the region where deviations from hydrodynamic behavior take place can be effectively investigated.

Here we report the main results of such an inelastic neutron scattering measurement of the dynamic structure factor $S(k,\omega)$ of a He-Ne mixture in the range $0.7 < k/\mathrm{nm}^{-1} < 1.8$, at thermodynamic conditions similar to those of Refs. [4,11], i.e., the same temperature $T = 39.3$ K and partial He number density $n_1 = 12.1\ \mathrm{nm}^{-3}$ [16], with a slightly lower total pressure ($p = 105$ bars). The partial Ne number density was, according to the van der Waals equation [17], $n_2 = 3.7\ \mathrm{nm}^{-3}$, thus leading to a 77% He and 23% Ne mixture.

The experiment was performed with the IN5 time-of-flight (TOF) spectrometer of the Institut Laue Langevin in Grenoble, with the small-angle multidetector covering the range $2.0° < \theta < 7.9°$, using an incident neutron wavelength $\lambda = 0.475$ nm. The corresponding neutron velocity of 832.8 $\mathrm{m\,s^{-1}}$ ($E_0 = 3.63$ meV) was sufficiently high to enable interaction between neutrons and the fast mode, whose propagation speed c_1 was expected to be close to that of pure He at the same temperature and total density, namely, 544 $\mathrm{m\,s^{-1}}$ [16]. The instrument energy resolution (HWHM of 0.09 ps^{-1}, or 60 μeV) was also appropriate for resolving a fast as well as a hydrodynamic mode near the predicted k_H, since the sound velocity c of the mixture is 374 $\mathrm{m\,s^{-1}}$, as estimated from the van der Waals equation. The sample was contained in a cylindrical aluminum container with an inner diameter of 16 mm and a wall thickness of 0.4 mm.

The total dynamic structure factor $S(k,\omega)$ measured in neutron scattering experiments can be expressed as

$$S(k,\omega) = x_1\tilde{b}_1^2 S_{11}(k,\omega) + x_2\tilde{b}_2^2 S_{22}(k,\omega) + 2\sqrt{x_1x_2}\,\tilde{b}_1\tilde{b}_2 S_{12}(k,\omega), \quad (1)$$

where the partial structure factors $S_{ij}(k,\omega)$ are the Fourier transforms of the time correlation functions of density fluctuations, with wave vector k, of species i and j; $x_i = n_i/n$ and $\tilde{b}_i = b_i/\sqrt{x_1b_1^2 + x_2b_2^2}$ are, respectively, the concentration and the normalized scattering length of species i, with $b_1 = 3.26$ fm and $b_2 = 4.566$ fm denoting the bound scattering lengths of natural He and Ne. The incoherent contribution from Ne is negligible. With our experimental values for the He and Ne concentrations, the coefficients of the partial structure factors in Eq. (1) are $x_1\tilde{b}_1^2 = 0.63$, $x_2\tilde{b}_2^2 = 0.37$, and $2\sqrt{x_1x_2}\,\tilde{b}_1\tilde{b}_2 = 0.97$, which imply a rather high sensitivity of the total $S(k,\omega)$ to the light component dynamics, due to the considerable contribution of $S_{11}(k,\omega)$.

In order to derive the experimental $S(k,\omega)$, the TOF spectra, collected at 20 scattering angles, were corrected for background, attenuation, and detector efficiency. Multiple scattering was considered to be negligible because of the small scattering power of our sample (3.2% in the forward direction) and because multiple scattering is much broader (virtually flat) than the single scattering. Also, no important frame overlap effect was detected in the data. The data taken at different angles were normalized to consistent, though arbitrary, units by means of a vanadium measurement, also used for determining the experimental resolution function.

$S(k,\omega)$ was then obtained by constant-ω and constant-k interpolation of the spectra at all angles and symmetrized for comparison with classical models. An absolute normalization of the dynamic structure factor was not necessary for the physical problem under consideration. Figure 1 shows the experimental $S(k,\omega)$ of the He-Ne mixture as a function of ω at three values of k.

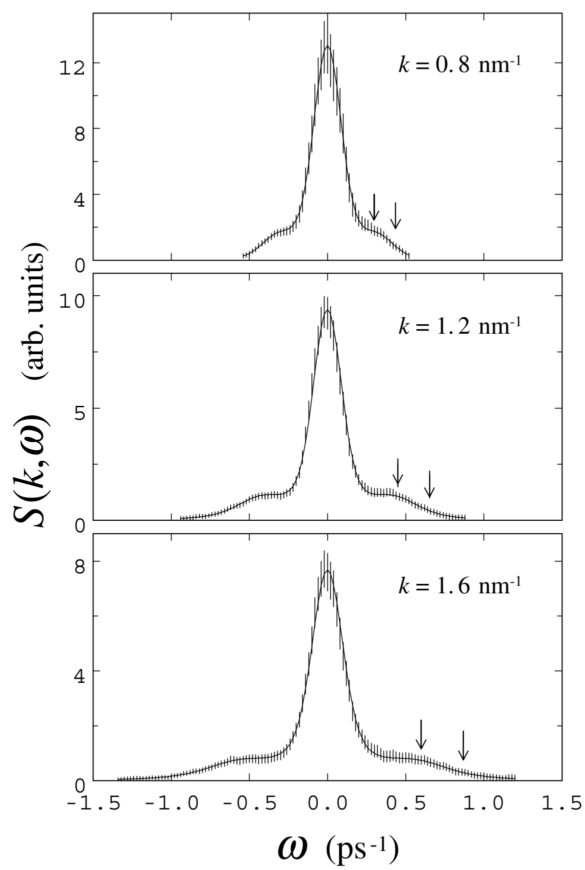

FIG. 1. Experimental $S(k,\omega)$ (error bars) at three k values after symmetrization. The line is the best fit obtained with Eq. (2). The expected positions of hydrodynamic and fast-sound modes calculated as $\omega = c\,k$ with $c = 374$ and 544 $\mathrm{m\,s^{-1}}$, respectively, are shown by the arrows.

In Ref. [4] it was found that at $k > 4$ nm^{-1} the measured $S(k, \omega)$ could be described properly by a sum of four Lorentzian lines, all located at nonzero frequencies, but not by a model including one quasielastic and two inelastic peaks. Here we tried to analyze our data in both the above ways, by fitting, at each k, two different models to our experimental $S(k, \omega)$. The first model is

$$S(k,\omega) = \frac{I_0}{\pi}\left[a_0 \frac{z_0}{z_0^2 + \omega^2} + a_s \frac{z_s + b_s(\omega + \omega_s)}{z_s^2 + (\omega + \omega_s)^2} + a_s \frac{z_s - b_s(\omega - \omega_s)}{z_s^2 + (\omega - \omega_s)^2}\right], \qquad (2)$$

i.e., the usual three-Lorentzian spectrum that is appropriate for extended hydrodynamics in a pure system [15], with one central line and two inelastic peaks located at frequencies $\pm\omega_s$. A physical constraint was imposed on the parameters by forcing $S(k, \omega)$ to have integrated intensity I_0, thus leaving six parameters free. The second model is the same four-Lorentzian spectrum as used in Ref. [4], with two lines centered at frequencies $\pm\omega_s^{(1)}$, and two at $\pm\omega_s^{(2)}$, with $\omega_s^{(1)} > \omega_s^{(2)}$ (in Ref. [4] a kinetic model consisting of one central Lorentzian and two pairs of inelastic peaks was also fitted to the experimental data. The central line turned out to be very weak, which explains why the four-Lorentzian fit worked equally well). In this case the number of free parameters is eight. Both model functions were convoluted with the measured resolution function before being fitted to the experimental data.

The convoluted line shapes obtained from the best fit with either model agree very well with the experimental points and turn out to be practically indistinguishable from each other. In particular, $\omega_s(k)$ obtained from the three-Lorentzian fit agrees well, in the whole k range, with $\omega_s^{(1)}(k)$ obtained from the four-Lorentzian one. On the other hand, $\omega_s^{(2)}(k)$ from the four-line fit turns out to be very close to zero frequency (<0.02 ps^{-1}) at all k values. It is worth noting that such value is about 1 order of magnitude lower than all theoretical predictions for the frequency of the slow mode in He-Ne [4,6]. We take this as an indication that no slow sound is present, and that the central peak in the experimental $S(k, \omega)$ corresponds actually to a single nonpropagating quasielastic mode (heat mode). Indeed, the model given in Eq. (2) is sufficient to accurately describe the experimental spectra (see Fig. 1) and we have no convincing reason to use models with more than one pair of inelastic (propagating) modes.

$\omega_s(k)$ is shown in Fig. 2, together with the hydrodynamic sound dispersion $\omega = ck$ of the mixture, and the equivalent pure-He dispersion curve. From Fig. 2 we note that in the whole investigated k range the fitted inelastic peak frequency depends linearly on k, with a slope coincident with the estimated hydrodynamic sound velocity of the mixture. Therefore, our data appear to be incompatible with both fast and slow propagating sound modes, which, if present, should be detectable with the available energy resolution and energy window, and with the present width of the central line ($z_0 < 0.06$ ps^{-1} at all k). Conversely, they show the persistence of a collective mode propagating at the speed of the ordinary hydrodynamic sound of the mixture up to $k = 1.8$ nm^{-1}.

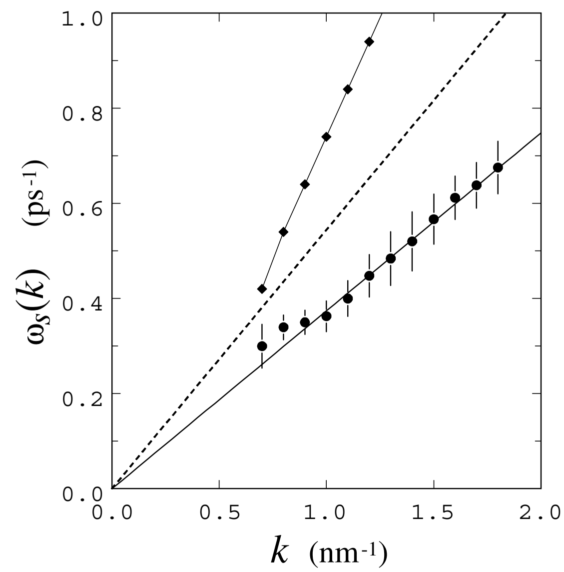

FIG. 2. The Brillouin peak position ω_s (dots with error bars) obtained from the three-Lorentzian fit [Eq. (2)] as a function of k. The solid and dashed straight lines are, respectively, the hydrodynamic sound dispersion of the mixture ($\omega = ck$ with $c = 374$ m s^{-1}), and the hydrodynamic dispersion for pure He at the same total density ($\omega = ck$ with $c = 544$ m s^{-1}). The diamonds, connected by a line to guide the eye, give the maximum frequency in each experimental $S(k, \omega)$.

It has been repeatedly stated in the literature that the relevant parameter for discriminating between different microscopic dynamical regimes is the product of the wave vector k and a length scale of the fluid identified with an (effective) mean free path l. Following Ref. [17], we use an "averaged mean free path" of the mixture defined as the Enskog mean free path $l = [\sqrt{2}\,\pi n \sigma^2 g(\sigma)]^{-1}$ of an equivalent monatomic fluid of hard spheres with density $n = n_1 + n_2$ and diameter $\sigma = (x_1\sigma_1^3 + x_2\sigma_2^3)^{1/3}$, with $g(\sigma)$ the radial distribution function at contact. For the previous neutron experiments, where only fast sound was detected, we find $kl \geq 0.42$ for $k \geq 4$ nm^{-1} in He-Ne (σ_1 and σ_2 from Ref. [4]) and $kl \geq 0.34$ for $k \geq 2$ nm^{-1} in He-Ar (σ_1 and σ_2 from Ref. [7]). For the present data, where instead only hydrodynamic dispersion is found, we have $0.10 \leq kl \leq 0.26$ (with the same σ_1 and σ_2 as in Ref. [4]). Therefore, all the available neutron results indicate that a rather sharp transition between the hydrodynamic and the kinetic regimes should occur around $kl = 0.3$. Although some difference between the high- and the low-density situations is possible, in the light scattering case, where k is much lower and l is much higher than in neutron experiments, similar transition kl values are again found. In fact, with the above definition of l, the onset of the reported fast sound in H_2-Ar is located around $kl \approx 0.25$ [9]. However, the result $k_H = 0.7\ \text{nm}^{-1}$ obtained in theoretical and simulation studies of dense He-Ne corresponds to $k_H l = 0.10$.

The absence in the present experimental data of the fast sound excitations found at higher k in previous experiments might, however, also have another explanation, namely that with decreasing k this mode behaves more like an optical phonon branch, so that high energy excitations occur even in the present k range, but outside the available energy window.

In conclusion, by using neutron Brillouin scattering, we have shown that, in a dense He-Ne mixture very similar to that previously studied [4,11], the acoustic excitations behave hydrodynamically at least up to $k = 1.8\ \text{nm}^{-1}$, in contrast with the predicted upper limit $k_H = 0.7\ \text{nm}^{-1}$. Experimental data at $1.8 \le k/\text{nm}^{-1} \le 4$ (i.e., $kl \approx 0.3$) and possibly also at higher frequencies are needed for a deeper investigation of the transition to fast sound and the behavior of the dispersion curves, although the conflicting requirements in terms of neutron energy and resolution power may be difficult to fulfill, especially if the fast sound frequency turns out to go to zero at finite k with a high slope.

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