High-frequency dynamics in a molten binary alloy

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The nature of the finite wavelength collective excitations in liquid binary mixtures composed of atoms of very different masses has been of interest for more than a decade. The most prominent fact is the high frequencies at which they appear, well above those expected for a continuation to large wave vector of hydrodynamic sound. To better understand the microscopic dynamics of such systems, an inelastic neutron scattering experiment was performed on the molten alloy Li_4Pb. We present the high-frequency excitations of molten Li_4Pb which indeed show features substantially deviating from those expected for the propagation of an acoustic mode.

The presence of overdamped high-frequency collective excitations within the microscopic regime in liquid binary mixtures composed of atoms of disparate masses has been observed by scattering experiments and computer simulations, in different systems, such as molten salts [1], simple liquid mixtures [2], molecular liquids [3,4] and metallic alloys [5]. Kinetic-theory predictions portray such excitations as being supported by the light component only so that they apparently travel with phase velocities close to those characteristic of the lighter pure component, which are well above those given by the elastic constants of the mixture. From the exploration of the phase velocity trend within the low-Q region of such systems, the presence of a mode of acoustic nature (where the atoms execute in-phase displacements) propagating with a velocity well above that corresponding to hydrodynamic sound has been inferred. However, some aspects are still the subject of controversy in regard to, on the one hand, the adequacy of describing the nature of excitations appearing at relatively large-Q in terms of constructs which only retain full sense within the realm of hydrodynamics (i.e. a sound mode), and on the other hand, the assignment of the observed frequencies to a definite underlying microscopic mechanism in the absence of further information.

In order to get a coherent understanding of the high-frequency dynamics in these mixtures, we chose to study a system which has been a flagship for tests of the above mentioned phenomena, namely molten Li_4Pb (at 1075 K). The measurements were carried out on the three-axis spectrometer IN1.

The full accessible dynamical range at each wavenumber of the experimental dynamic structure-factors, $S(Q,\omega)$, is shown in Fig. 1. These data reveal heavily damped excitations, i.e. a broad but relatively intense inelastic signal. The total $S(Q,\omega)$ was fitted using two main components, the central quasielastic part plus the inelastic contribution. The model used for the latter was the *damped harmonic oscillator* [6] which is defined in terms of three parameters, I_Q, excitation strength, ω_Q, the frequency and Γ_Q ($\propto \tau_Q^{-1} Q^2$), the linewidth.

The experiment covered a kinematic range ($0.6 \leq Q \leq 2.4$ Å^{-1} and $\hbar\omega \leq 60$ meV) which enabled an unambiguous characte-

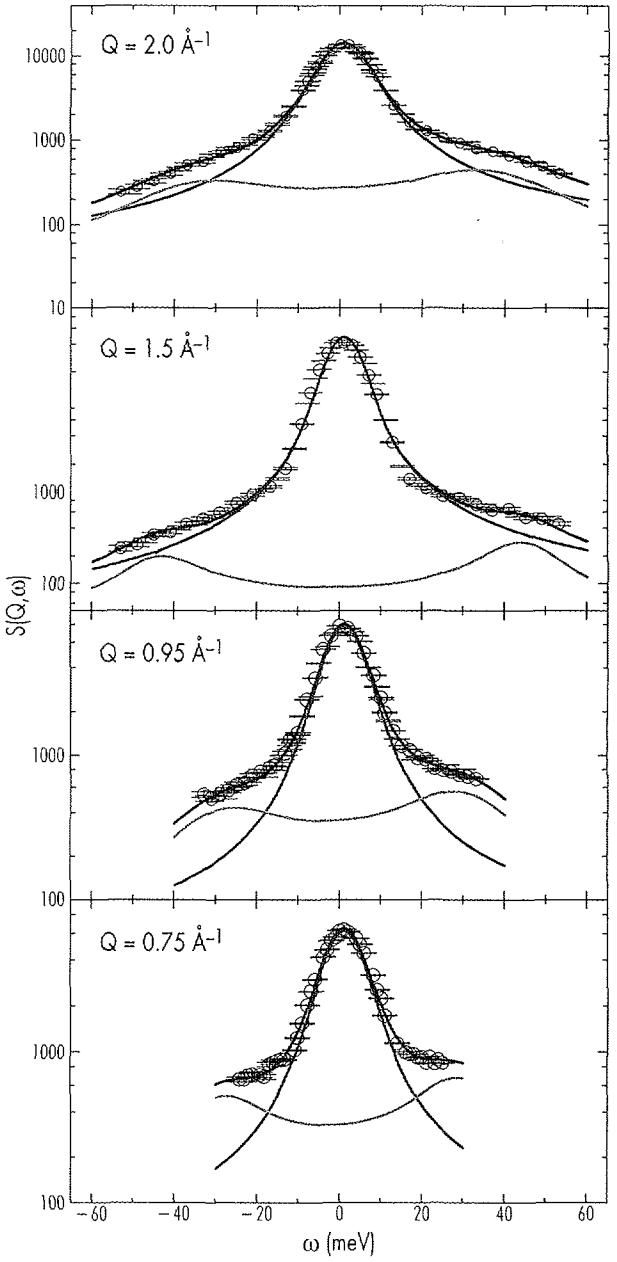

Figure 1: *The experimental data are shown by symbols, the fitted model by blue lines, and the quasielastic and inelastic components by red and green lines, respectively.*

risation of such excitations by studying the wavevector dependence of their frequencies, lifetimes and signal amplitudes. The fitted parameters vs. Q are all plotted in Fig. 2. The Q dependence of the inelastic integrated intensity, I_Q (Fig. 2a), does not follow the form of the static structure factor of the mixture (S(Q), blue circles), as the quasielastic integrated intensity does. This fact indicates that the collective mode does not propagate through the whole network of atoms, or in other words, it is a signature of a non-acoustic behaviour.

On the other hand, the dispersion relation, ω_Q vs. Q, is displayed in Fig. 2b. It shows a maximum at about $Q_m/2$, where Q_m is the position of the first peak of S(Q) for pure Li, which confirms that these excitations are supported by the lighter component of the alloy. Moreover, the phase velocity, ω_Q/Q, approaches the sound propagation velocity in pure Li (blue line). The Q dependence of Γ_Q is also plotted in Fig. 2c. This provides an estimate of the lifetime of the collective excitations being sampled, $\tau_Q \approx 0.02$ ps, which implies that these excitations are of a localised nature. In summary, the Q-dependence of the inelastic intensity is considered here on the same footing as that shown by the excitation frequencies and linewidths, since it provides a direct access to the relative phases of the motion of atoms taking place at frequencies well above hydrodynamic sound. The results confirm the presence of overdamped excitations in a molten alloy composed of particles with a disparate mass ratio. In agreement with predictions from kinetic theories, such motions are shown to be supported by the light component only. However, the experiment provides for the first time clear indications that these motions are mostly out-of-phase and very much like the 'optic' modes in binary crystals.

The situation is highly reminiscent of liquid water and ice polycrystals, where a similar effect ('fast sound') has repeatedly been reported [e.g. 4,7]. Thus, our measurements provide new evidence pointing towards a microscopic origin of the steep dispersion. This gives an alternative view to that based on the interpretation of the deceptively simple 'dispersion' in terms of a unique acoustic excitation which travels at speeds several times higher than hydrodynamic sound. The results also show the unique capabilities of IN1, the three-axis spectrometer for liquids and amorphous materials research at ILL, when dealing with liquid mixtures composed of atoms of very different masses and when steep dispersions come into play.

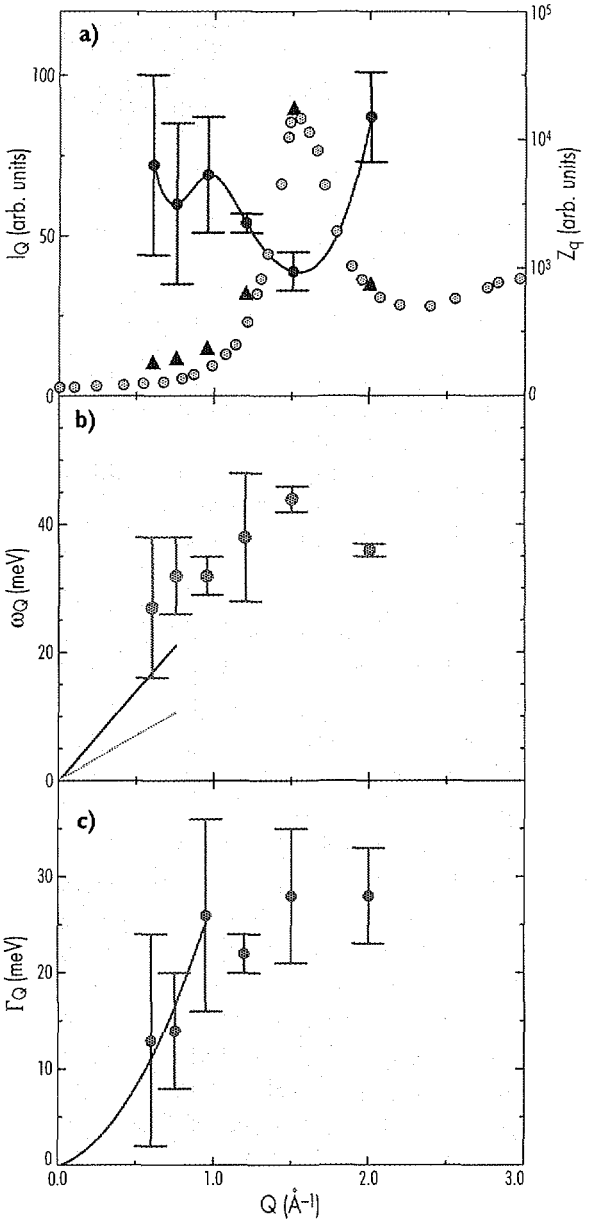

Figure 2: *Wave-vector dependence of the damped harmonic oscillator parameters. The Q dependence of both the integrated intensities of the quasi-elastic component (Z_q, ▲) and the inelastic part (I_Q, ●), together with the static structure factor of the mixture (S(Q), ●), is shown in frame* ***a)****. The dispersion relation is displayed in frame* ***b)****, together with the expected positions at high Q of the hydrodynamic sound modes corresponding to pure liquid Li (blue line) and to liquid Li_4Pb (pink line). The wavevector dependence of the inelastic linewidth Γ_Q is also plotted in frame* ***c)****; the line merely shows the Q^2dependence at low Q values, from which the lifetime, τ_Q, can be estimated (see text).*

References

■ [1] R.L. McGreevy, et al., J. Phys. C17 (1984) 775 ■ [2] W. Montfrooij, et al., Phys. Rev. Lett. 63 (1989) 544; E. Enciso et al., Phys. Rev. Lett. 74 (1995) 4233 ■ [3] A. Criado, et al., Phys. Rev. B 47 (1993) 3516, F.J. Bermejo, M. Alvarez, S.M. Bennington, and R. Vallauri, Phys. Rev. E 51 (1995) 2250 ■ [4] F. Sette, G. Ruoco, M. Krisch, C. Masciovecchio, R. Verbeni and U. Bergmann, Phys. Rev. Lett. 77 (1996) 83 ■ [5] P.H.K. de Jong, P. Verkerk, C.F. de Vroege, L.A. de Graaf, W.S. Howells, and S.M. Bennington, J. Phys.: Condens. Matter 6, (1994) L681; M. Alvarez, F.J. Bermejo, P. Verkerk, and B. Roessli, Phys. Rev. Lett. 80 (1998) 2141 ■ [6] The damped harmonic oscillator model is discussed in Appendix B in 'Theory of Neutron Scattering from Condensed Matter', by S.W. Lovesey (Oxford University, Oxford, 1984), and in 'Excitations in Liquid and Solid Helium', by H.R. Glyde (Clarendom, Oxford, 1994), p. 184, 185 ■ [7] J. Teixeira, M.C. Bellissent-Funel, S.H. Chen, and B. Dorner, Phys. Rev. Lett. 54 (1985) 2681.