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LETTER TO THE EDITOR

Observation of fast sound in liquid Li_4Tl and liquid Li_4Pb by inelastic neutron scattering

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Abstract. We performed inelastic neutron scattering experiments on liquid Li_4Pb and Li_4Tl. We obtained the first experimental evidence for the existence of fast sound in a molten binary alloy of elements of strongly different weight. This phenomenon was observed for the first time ten years ago in a computer molecular dynamics (CMD) simulation. The results are compared with the results of the CMD simulation and with experimental results for the sound dispersion of pure lithium.

Jacucci *et al* [1], [2] found in a computer molecular dynamics (CMD) simulation of liquid Li_4Pb indications for a mode propagating at a velocity faster than (macroscopic) sound. Bosse *et al* [3] named this phenomenon fast sound. The fast-sound mode was interpreted as a propagating density fluctuation of the Li atoms only in a background of heavy lead ions which do not participate in this high-frequency motion [3]. However, the actual fast-sound velocity in the simulation (7500 $m\,s^{-1}$) was even higher than the experimental sound velocity in liquid lithium (4500 $m\,s^{-1}$), which does not seem in complete agreement with this interpretation. The fast-sound mode is visible as an inelastic peak in the Li–Li partial dynamic structure factor $S_{LiLi}(\kappa, \omega)$.

Due to the large difference in mass (the ratio is 30) in Li–Pb the Pb–Pb partial dynamic structure factor $S_{PbPb}(\kappa, \omega)$ is expected to be much narrower than the Li–Li partial, $S_{LiLi}(\kappa, \omega)$. Moreover, the sound velocity of pure lithium v_{Li} (4500 $m\,s^{-1}$) is much larger than the normal sound velocity in Li_4Pb (2100 $m\,s^{-1}$) and the sound velocity in pure Pb. Hence the inelastic peak in $S_{LiLi}(\kappa, \omega)$ should be well separated from the other contributions, and the fast-sound mode should be clearly distinguishable from the ordinary sound mode.

Experimental evidence for the occurrence of fast sound in gases was found by inelastic neutron scattering (INS) on a mixture of He–Ne [4] and by inelastic light scattering on a mixture of H_2–Ar [5]. In order to investigate the occurrence of fast sound in a molten binary alloy we performed INS experiments on liquid Li_4Pb and liquid Li_4Tl. The latter alloy was chosen in order to study the effect (if any) of the potential.

The total dynamic structure factor $S(\kappa, \omega)$ of these alloys is given by

$$S(\kappa, \omega) = 4\pi\langle b^2\rangle[c_2S_{11} - 2\sqrt{c_1c_2}S_{12} + c_1S_{22}] + c_1\sigma_1^{inc}S_1^{inc}(\kappa, \omega) + c_2\sigma_2^{inc}S_2^{inc}(\kappa, \omega) \qquad (1)$$

with $\langle b^2\rangle = c_1b_1^2 + c_2b_2^2$, where c_j and b_j denote the concentration and coherent scattering length of species j (1: Li, 2: Pb or Tl) respectively, $S_{ij}(\kappa, \omega)$ denotes the Ashcroft–Langreth

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partial dynamic structure factor, σ_j^{inc} and $S_j^{inc}(\kappa, \omega)$ denote the incoherent scattering cross-section and the incoherent dynamic structure factor of species j respectively. The relevant scattering lengths and cross-sections are listed in table 1.

Table 1. Relevant scattering lengths and cross-sections (from [12]). σ_i denotes the incoherent scattering cross-section of the pure elements or the alloys, b and $\langle b^2 \rangle$ denote the coherent scattering length of the pure elements and the mean squared coherent scattering length of the alloys, respectively.

	^{7}Li	Pb	Tl	$^7Li_4Pb$	$^7Li_4Tl$
b/fm	−2.22	9.41	8.78		
σ_i/b	0.78	0.00	0.21	0.624	0.666
$4\pi\langle b^2\rangle$/b				2.72	2.43

The total $S(\kappa, \omega)$ was measured with the spectrometer MARI at the neutron spallation source ISIS. The experiment was performed with an incident energy of 496 meV (wavelength $\lambda_0 = 0.0406$ nm, neutron velocity is 9750 m s^{-1}) and 548 detectors positioned between 3.86° and 135°. In the present report we use only the data from the detectors at the eight smallest scattering angles ($\varphi \leqslant 6.86°$). The samples were held in sealed niobium cylinders with a wall thickness of 0.2 mm and an outer diameter of 25 mm.

The measurement on liquid Li_4Pb at 1020 K had to be terminated after two hours because the high temperature of the sample caused a vacuum leak in the sample chamber of MARI. The measurement on liquid Li_4Tl at 753 K went well for 34 hours. We performed additional measurements on an empty furnace, on a thin-walled vanadium cell filled with vanadium turnings for normalization and on a single niobium cell filled with ^{3}He gas. The latter two samples had the same transmission as the Li_4Tl sample.

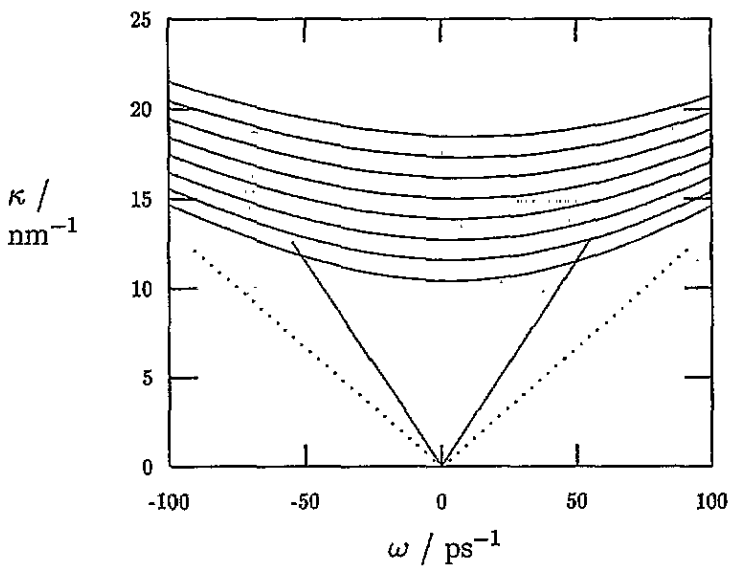

Figure 1. The kinematically accessible κ–ω plane for an incident energy of 496 meV. The curves drawn indicate detectors and correspond to scattering angles of 3.86°, 4.29°, 4.71°, 5.14°, 5.57°, 6.00°, 6.43° and 6.86°. The solid line denotes the sound velocity of pure lithium at 753 K; the dotted line denotes v_F (7500 m s^{-1}) from the CMD simulation.

In figure 1 we show the kinematically accessible κ–ω plane. The velocity of the fast-sound mode v_F from the CMD simulation (7500 m s^{-1}) is much larger than the sound velocity of pure lithium (4372 m s^{-1} at 753 K and 4209 m s^{-1} at 1023 K). Calculations of Campa and Cohen [6] show that v_F is given by the sound velocity of the pure light component.

We have corrected the data for background, multiple scattering and detector efficiency. In figure 2 we show $S(\kappa, w)$ for few scattering angles. We omit the interpolation of the data to a rectangular κ–ω grid in order to avoid introducing an additional uncertainty due to the interpolation. We have determined the resolution of the spectrometer using the measurement on the vanadium sample. We have applied the same corrections as mentioned above to the vanadium spectra and we have fitted a Gaussian to the vanadium data. It appears that a single Gaussian with standard deviation σ_r (10.2 ps^{-1}) gives a good description of the experimental resolution. The fitted σ_r is in good agreement with the value found by de Jong *et al* [7].

The INS experiment of Soltwisch *et al* [8] on Li_4Pb at 1023 K with a low incident energy shows that the central peak of $S(\kappa, \omega)$ is well described by a sum of two Lorentzians. The widths of these two contributions (3 ps^{-1}) are much narrower than the width of the resolution (20 ps^{-1}) in the present experiment. The width of $S^{inc}_{Tl}(\kappa, \omega)$ is also expected to be much narrower than the width of the resolution [2]. We have therefore used the following model to fit the experimental data:

$$S(\kappa, \omega) = A_0 \mathcal{G}(0, \sigma_r) + A_s[\mathcal{G}(\omega_s, \sigma_s) + \mathcal{G}(-\omega_s, \sigma_s)] \tag{2}$$

with A_0 and A_s the areas of the central peak and the shifted Gaussians respectively, where σ_r is 10.2 ps^{-1}, ω_s and σ_s denote the position and the standard deviation of the shifted Gaussians respectively. $\mathcal{G}(a, b)$ denotes a Gaussian normalized to unity with standard deviation b centred at a. We employ this simple model because the energy-gain and energy-loss branches of a detector through the κ–ω plane are nearly mirror images of each other at small angles for an incident energy of 496 meV.

The results of the weighted least-squares fits are shown in figure 2. We have omitted the Li_4Pb data at the first two scattering angles due to poor statistics. The mean square error of the fit averaged over the available angles is 2.1 for Li_4Tl and 2.5 for Li_4Pb. In figure 3 we show the position and the full width at half maximum ($2\sqrt{2\ln(2)}\sigma_s$) of the shifted Gaussians as a function of $\kappa_e = 4\pi \sin(\varphi/2)/\lambda_0$. In this figure we also show the results of the CMD simulation for liquid Li_4Pb [3], the experimental results for the sound dispersion of pure lithium [7] [9], and the expected positions of the fast-sound modes for sound velocities of 4372 m s^{-1} (pure lithium at 753 K), 7500 m s^{-1} (from the CMD simulation) and 2100 m s^{-1} (hydrodynamic sound in Li_4Pb). We observe a collective mode at a frequency which is much larger than the expected position of the hydrodynamic sound mode (2100 m s^{-1}) in Li_4Pb. Moreover, the dispersion relation is in good agreement with the sound dispersion of pure lithium which was determined by INS [7] and by inelastic x-ray scattering [9]. The data are furthermore in qualitative agreement with the results of the CMD simulation. The deviation of v_F from the CMD simulation with v_{Li} is most likely due to the model potential used [10]. Consequently, we ascribe the side peaks in $S(\kappa, \omega)$ of Li_4Tl and Li_4Pb to the occurrence of fast sound in these binary liquid mixtures.

This is the first experimental verification of fast sound in liquid Li_4Pb and Li_4Tl, nearly ten years after the CMD results were published. We conclude with a comment. We have not been able to observe fast sound for values of κ less than 11 nm^{-1} due to the limited κ range. Hence an experimental determination of v_F is still lacking. Of particular interest is the behaviour of the fast-sound mode for $\kappa \to 0$: does the fast-sound merge with the hydrodynamic sound mode or does it disappear as is suggested by theory [11]? Apparently, a

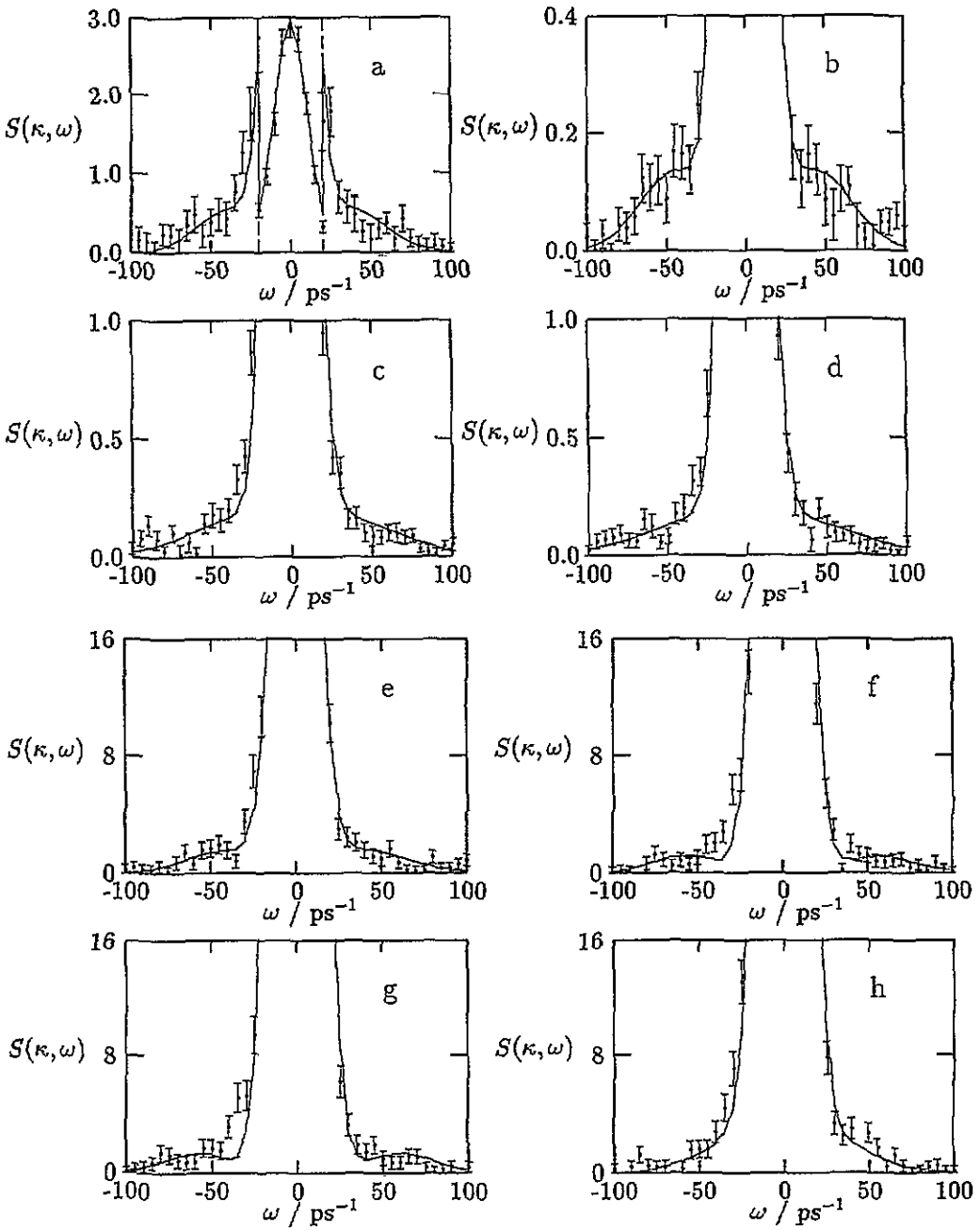

Figure 2. $S(\kappa, \omega)$ for liquid Li_4Tl (a)–(d) and liquid Li_4Pb (e)–(h) for a few scattering angles. Dots: experimental data; solid line: fit. The error bars correspond to one standard deviation up and down. The intensities for $|\omega| \geqslant 20$ ps^{-1} in (a) have been multiplied by 5 for clarity. a: $\varphi = 3.86°$ (κ_e =10.4 nm^{-1}), b: $\varphi = 4.71°$ (12.7 nm^{-1}), c: $\varphi = 5.57°$ (15.0 nm^{-1}), d: $\varphi = 6.43°$ (17.4 nm^{-1}), e: $\varphi = 4.71°$ (12.7 nm^{-1}), f: $\varphi = 5.15°$ (13.9 nm^{-1}), g: $\varphi = 5.57°$ (15.0 nm^{-1}) and h: $\varphi = 6.43°$ (17.4 nm^{-1}).

spectrometer with neutrons of high energy (such as MARI), with very good energy resolution

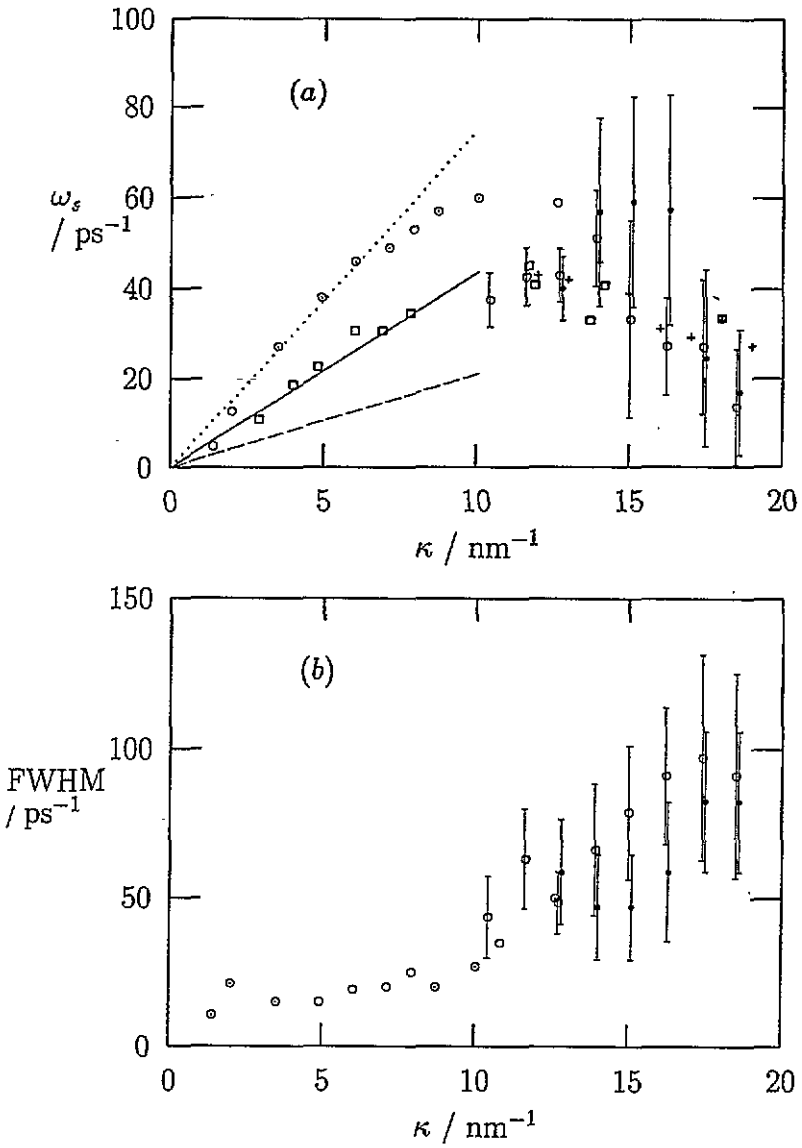

Figure 3. Position (a) and full width at half maximum (FWHM) (b) of the shifted Gaussians. Circles: results for Li_4Tl; dots: result for Li_4Pb (shifted by +0.1 nm^{-1}); circles with dots: results from the CMD simulation of liquid Li_4Pb [3]; squares in (a): inelastic x-ray data for liquid lithium at 600 K [9]; plusses in (a): inelastic neutron data for liquid ^7Li at 470 K [7]; solid line in (a): sound velocity in pure lithium at 753 K (4372 m s^{-1}); dotted line in (a): sound velocity in Li_4Pb from the CMD simulation [3]; and broken line in (a): hydrodynamic sound velocity in liquid Li_4Pb.

and angular resolution, and with a small-angle facility is needed.

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