PHYSICAL REVIEW E VOLUME 47, NUMBER 4 APRIL 1993

Zero-sound-like modes in simple liquid metals

Chr. Morkel, T. Bodensteiner, and H. Gemperlein
Physik Department, Technische Universität München, D-8046 Garching, Germany

(Received 2 November 1992)

Coherent neutron-scattering experiments on liquid cesium and rubidium near the melting point reveal an anomalous dispersion of collective modes for frequencies $\omega\tau_M \gg 1$ with τ_M the Maxwell relaxation time for shear stress in the liquid. Within the framework of generalized hydrodynamics, these modes are shown to propagate similar to zero sound in a restricted range of wave numbers around $Q = 0.4$ Å^{-1}. Such a behavior is known from glass-forming systems, but here simple liquid metals near the melting point are shown to exhibit such modes too. The feature of the dispersion relation in this Q region yields quantities known from the theory of elasticity of solids such as the shear modulus G_∞ and the Poisson ratio ν. Some evidence is found for the existence of small clusters in the liquid on the picosecond time scale.

PACS number(s): 61.25.Mv, 61.12.Ex, 67.40.Fd

I. INTRODUCTION

Short-wavelength collective modes are an experimentally well-studied feature of both liquid metals [1–4] and rare-gas fluids [5–9]. Among these two classes of liquids the former is known to show distinct propagating collective modes extending to wavelengths as short as the interparticle distance. This property has been traced back in molecular-dynamic (MD) simulations [10,11] and theoretical considerations to the more pronounced harmonicity of the interparticle potential and smaller compressibility of the metallic systems [12,13]. Thus, a simple metal such as cesium at the melting point is a favorable system for studying collective modes in the liquid state. These excitations are observed experimentally in the dynamic structure factor $S(Q,\omega)$ by inelastic coherent neutron scattering as so-called Brillouin lines, using here the common extension of this term into the region beyond the hydrodynamic limit. In hydrodynamics collective modes are well understood as propagating pressure fluctuations (first sound) with the rate of collisions restoring thermal equilibrium being large compared to the frequency of the fluctuation. At increasing frequencies in the region beyond hydrodynamics, collisions become less and less important for sound propagation, which then depends mainly on the existence of local order in a liquid at the melting point. This enables the system to support collective excitations via an effective interaction potential between the particles as in a solid. Such short-wavelength high-frequency collective modes in a liquid are therefore called "zero-sound-like" because of the analogy with collisionless sound, first revealed in liquid helium [14]. In the numerous literature on collective modes a zero-sound-like propagation in simple liquids has long been discussed [15–19], with only one experimental attempt to verify such behavior in liquid argon [19–21].

The paper is organized in the following way. After a short description of the experiment on liquid cesium in the next section we discuss the observed dispersion relation of longitudinal collective modes within the framework of generalized hydrodynamics and viscoelastic theory of liquids [22–25], including a brief reference to solid cesium [26]. The data are then compared with recent molecular-dynamic results for liquid rubidium [27] and corresponding experimental data from our own laboratory [28], measured with an improved resolution of about twice that of Copley and Rowe's Rb experiment [2]. This supplement shows clearly that the revealed positive dispersion is a general feature of liquid metallic systems.

II. EXPERIMENT

The present neutron-scattering measurements on liquid cesium at 308 K (melting temperature $T_m = 301.6$ K) were performed at the Forschungsreaktor München [$E_f = 22.5$ meV, $\Delta E = 0.45$ meV full width at half maximum (FWHM)] and the High Flux Reactor, Grenoble ($E_f = 14.66$ meV, $\Delta E = 0.68$ meV FWHM) using the triple-axis spectrometers DAS and IN8, respectively. For the reason of correct normalization of the data a constant k_f constant Q mode was chosen, with k_f the final wave vector and $\hbar Q$ the momentum transfer in the scattering process. Covering the Q range from 0.20 to 2.55 Å^{-1} with the maximum of the structure factor $S(Q)$ at $Q_0 = 1.42$ Å^{-1}, spectra were taken for energy transfers $\hbar\omega$ from -1.5 to $+10$ meV. The excitation energies of the collective modes varied from 1.3 meV at the lowest Q to about 6 meV at $Q_0/2 \approx 0.7$ Å^{-1}, the maximum of the dispersion relation (see Fig. 1). It is this low Q range that is discussed further in this paper. The scattering data are corrected for all relevant effects (background, container scattering, self-shielding, second-order contamination, incoherent intensity, resolution broadening, and multiple scattering). A more detailed description of the experiment and the data evaluation procedure is given elsewhere [29,30].

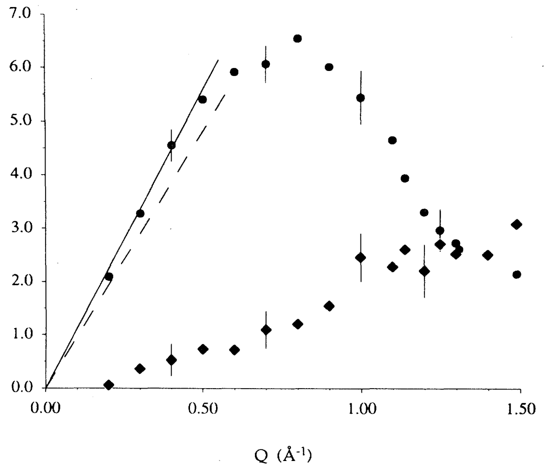

FIG. 1. Dispersion relation of liquid cesium. Dots, peak position $\omega^m(Q)$ of $J_l(Q,\omega)$; dashed line, $c_s(0)=965$ m/s; full line $c_\infty=1120$ m/s; rhombs, width (HWHM) of $J_l(Q,\omega)$.

III. RESULTS AND DISCUSSION

Generalized hydrodynamics describe the scattering law $S(Q,\omega)$ via the wave number and frequency-dependent density response function $\chi(Q,z)$ as

$$S(Q,\omega)=\frac{1}{\pi\omega}\chi''(Q,\omega)\ , \tag{1}$$

with $\chi''(Q,z=\omega+i0)$ the imaginary part of the generalized susceptibility

$$\chi(Q,z)=-[z^2-\omega_0^2(Q)+izQ^2D(Q,z)]^{-1}\ . \tag{2}$$

Here $\omega_0^2(Q)=k_BTQ^2[MS(Q)]^{-1}$ and $D(Q,z)$ is the so-called damping function. Although there are attempts nowadays to extract the complex quantity $D(Q,z)$ from experimental scattering data [31] we use the following prescription of the memory function $D(Q,z)$

$$Q^2D(Q,z)=\frac{(\gamma-1)\omega_0^2(Q)\tau_h}{1-iz\tau_h}+\frac{[\omega_1^2(Q)-\gamma\omega_0^2(Q)]\tau_v}{1-iz\tau_v}\ . \tag{3}$$

Here $\gamma=c_p/c_v$ is the specific-heat ratio, $\omega_1^2(Q)$ is the well-known normalized fourth moment of $S(Q,\omega)$. The time constants τ_h and τ_v characterize the thermal and viscoelastic decay of a density fluctuation in a simple liquid at low Q. Whereas there are recent more sophisticated mode-coupling concepts to derive memory functions for supercooled liquids [32], Eq. (3) seems to be the simplest working ansatz to describe the experimental findings in a liquid at the melting point. Such a form has been used earlier, especially in Chung and Yip's [20] and Mountain's [33] work on the collective dynamics in liquid argon. In contrast to the case of argon Eq. (3) can be simplified further in the case of liquid cesium. Because of the condition $|z\tau_h|\gg 1$ being fulfilled rather well in the alkali metal for collective excitations in the ps^{-1} region [29], the first term of Eq. (3) can be approximated accurately by $i(\gamma-1)\omega_0^2/z$. Moreover the factor $(\gamma-1)$ reduces the weight of the thermal decay channel in the case of an alkali metal, where $(\gamma-1)$ is of the order of 0.1 [34]. Thus one is left with Eq. (4) for $\chi(Q,z)$

$$\chi(Q,z)=-\left[z^2-\gamma\omega_0^2(Q)+i\frac{[\omega_1^2(Q)-\gamma\omega_0^2(Q)]z\tau}{1-iz\tau}\right]^{-1}\ . \tag{4}$$

Here and below the time constant τ stands for the viscoelastic relaxation time introduced by Maxwell [35].

Looking for collective excitations the poles of $\chi(Q,z)$ can be determined and lead to the dispersion relation

$$\omega^2(Q)=\frac{1}{2\tau^2}\{(\omega_1^2\tau^2-1)+[(\omega_1^2\tau^2-1)^2+4\gamma\omega_0^2\tau^2]^{1/2}\}\ . \tag{5}$$

In the limit $(\omega\tau)^2\ll 1$ this yields the well-known adiabatic sound velocity $c_s=v_0[\gamma/S(Q)]^{1/2}$ with the thermal velocity $v_0=(k_BT/M)^{1/2}$. As can be seen from Fig. 1 this limit is not reached properly in our present experiment. The real hydrodynamic limit $\omega\tau\ll 1$ for collective excitations is only accessible by means of the neutron Brillouin scattering technique [7], which has made some progress recently [36].

More interesting here is the limit $(\omega\tau)^2\gg 1$. From Eq. (4) it is seen that the viscoelastic part of the memory function starts to contribute noticeably at $\omega\tau\approx 1$. This affects the poles of $\chi(Q,z)$ leading to a shift of the excitation frequency towards higher frequencies. This positive dispersion beyond the hydrodynamic limit is clearly seen in Fig. 1. In the case $\omega\tau\gg 1$ the dispersion relation [Eq. (5)] has the limit

$$\omega(Q)=c_\infty(Q)Q\ , \tag{6}$$

where $c_\infty(Q)$ is the high-frequency sound velocity, which is no longer determined by the system's compressibility alone as in the case of the adiabatic sound velocity. Instead c_∞ is ruled by elastic moduli as in a solid. According to Schofield c_∞ is given as

$$c_\infty(Q)=\left[\frac{B_\infty(Q)+\frac{4}{3}G_\infty(Q)}{\rho}\right]^{1/2}\ , \tag{7}$$

where ρ is the mass density and B_∞ and G_∞ are the high-frequency bulk and shear modulus of the liquid, with the elastic constant $c_{11}(Q)=B_\infty(Q)+\frac{4}{3}G_\infty(Q)$ in the limit $Q\to 0$ [18]. It is seen from this formula that both compression and shear are involved in a high-frequency sound mode in contrast to hydrodynamic sound with vanishing shear modulus $G(\omega\tau\ll 1)$. In the low Q limit and $\omega\tau\gg 1$ the two elastic moduli are related by the generalized Cauchy relation $B_\infty=\frac{5}{3}G_\infty+2(p-\rho k_BT)$ derived by Zwanzig and Mountain [23] for an isotropic system of particles interacting via two-body central forces. As in a dense liquid near the melting point the contribution $(p-\rho k_BT)$ is negligibly small, the high-frequency sound velocity can be finally expressed as

$$c_\infty(Q)=\left[\frac{3G_\infty(Q)}{\rho}\right]^{1/2} \quad (Q\rightarrow 0)\ , \tag{8}$$

where $G_\infty(Q)$ corresponds to the elastic constant $c_{44}(Q)$ in a disordered system [18] and yields for vanishing Q,

$$G_\infty(Q\rightarrow 0)=\rho\left[\frac{k_BT}{M}+\frac{\omega_E^2\sigma^2}{10}\right]\ . \tag{9}$$

In a liquid metal at the melting point the kinetic contribution k_BT/M in Eq. (9) is of the order of 5×10^{-2} compared to the solidlike potential part, where the Einstein frequency ω_E and the effective core diameter σ simply characterize the interparticle potential ($\omega_E=4.13\times10^{12}$ s^{-1}, $\sigma=4.8$ Å here [29]). Thus high-frequency sound ($\omega\tau\gg1$) is propagated mainly via the strong coupling between neighboring particles as in a disordered solid.

In addition we show in Fig. 1 the width [half width at half maximum (HWHM)] of the collective modes (rhombs). Whereas a damping varying with Q^2 is known from hydrodynamics, we find a much weaker increase beyond the hydrodynamic region. Such a nearly linear increase with Q can be viewed as another feature of zero-sound-like excitations [14].

The approach of the sound velocity to c_∞ is shown in Fig. 2, where the phase velocity $c_l(Q)=\omega_{\max}(Q)/Q$ is plotted. Whereas the adiabatic sound velocity is 965 m/s in the hydrodynamic limit (dashed line [34]), the sound velocity increases to $c_\infty(Q)$ beyond $Q=0.2$ Å^{-1}, reaching a maximum at about $Q=0.4$ Å^{-1}. At this Q value a length scale of two to three particle diameters σ is resolved in the neutron-scattering experiment. Obviously, on this scale, which covers the well-defined first to second coordination shell, the sound propagation is solidlike. From the peak value of the sound velocity in Fig. 2 $c_\infty(0)=1120$ m/s the shear modulus G_∞ has been extrapolated to $G_\infty(0)=(7.67\pm0.95)\times10^9$ g/cm s^2 [Eq. (8)], which agrees well with the calculated value $G_\infty=7.55\times10^9$ g/cm s^2 from Eq. (9). The value in the liquid reaches nearly two-thirds of that in solid cesium, where G_∞ is 1.2×10^{10} g/cm s^2 at room temperature [26]. The shear modulus of the liquid now known, the calculation of the Maxwell relaxation time

$$\tau_M=\eta_s/G_\infty \tag{10}$$

yields $\tau_M=0.86\times10^{-12}$ s with $\eta_s=6.61\times10^{-3}$ g/cm s the shear viscosity at 308 K [34].

It is the quantity τ_M that sets the time scale for the transition from first sound ($\omega\tau_M\ll1$) to zero sound ($\omega\tau_M\gg1$). In Fig. 3 the measured quantity $\Delta=[c^2(Q)-c_s^2(Q)]/[c_\infty^2(Q)-c_s^2(Q)]$, which represents the contribution of $\mathrm{Im}[zD(Q,z)]$ to sound dispersion [Eq. (2)], clearly shows the transition to the zero sound regime. This quantity can simply be expressed as

$$\Delta=\frac{[\omega(Q)\tau_M]^2}{1+[\omega(Q)\tau_M]^2} \tag{11}$$

(full curve in Fig. 3 with $\tau_M=0.86$ ps).

A weak Q dependence of τ_M in the limited wave-vector region ($Q\leq0.8$ Å^{-1}) has been ignored here for the sake of simplicity. It is seen again that the hydrodynamic limit $\omega\tau_M\leq1$ is not covered in our present experiment. From $\omega\tau_M\leq1$ and $\omega=c_s\times Q$ for the low Q dispersion a limiting Q value of $Q_{hd}=(c_s\tau_M)^{-1}=0.12$ Å^{-1} can be estimated for the true hydrodynamic regime. Beyond this limit, the dispersive part of the memory function $zQ^2D(Q,z)$ attains about 95% of its high-frequency saturation value where sound propagation is ruled by elastic properties [Eq. (7)]. Hence it is obvious that the liquid approaches the limit of solidlike elastic response. Such a transition to solidlike sound propagation is known from glass-forming supercooled liquids [32,33,37]. It is re-

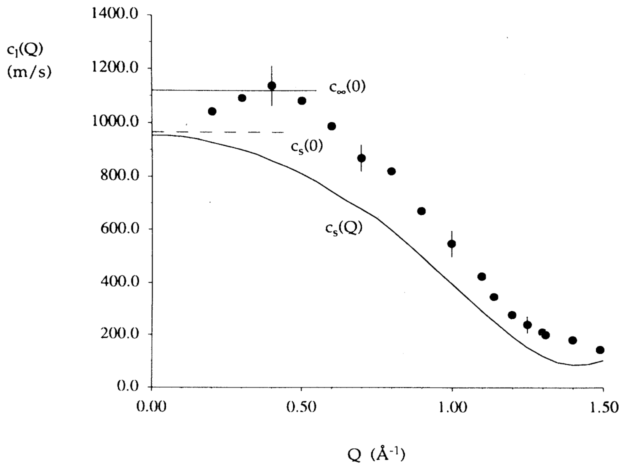

FIG. 2. Anomalous sound dispersion in liquid cesium. Dots, $c(Q)=\omega^m(Q)/Q$; full line, $c_s(Q)=v_0[\gamma/S(Q)]^{1/2}$.

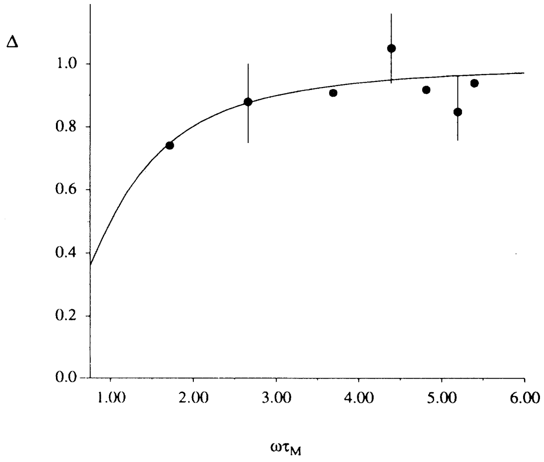

FIG. 3. $\Delta=(c^2-c_s^2)(c_\infty^2-c_s^2)^{-1}$ (dots) vs scaled frequency $\omega\tau_M$ indicating the increasing contribution of the term $\mathrm{Im}[zQ^2D(Q,z)]$ [see Eq. (2)] in the zero sound regime. Full line, Eq. (11) with $\tau_M=0.86$ ps.

markable that dense simple liquids show such modes, albeit in a restricted Q region around $Q = 0.4$ Å^{-1}, too.

The solidlike high-frequency response of the liquid now established, a quantity known from the theory of elastic continua—the Poisson ratio ν—seems to be useful in the context of simple liquids. The ratio $\nu = (\Delta r/r)/(\Delta l/l)$ describes the relative change $\Delta r/r$ of the lateral dimension of a volume element in the case of an applied longitudinal strain $\Delta l/l$ in the theory of elasticity [38]. Extending ν to nonzero wave numbers yields a generalized Q-dependent Poisson ratio

$$\nu(Q) = \frac{1 - \{c(Q)/[\sqrt{3}c_s(Q)]\}^2}{1 + \{c(Q)/[\sqrt{3}c_s(Q)]\}^2}, \quad (\omega\tau \gg 1) . \tag{12}$$

This quantity is easily evaluated from the experimentally measured sound velocities in the region where $\omega\tau \gg 1$ holds, i.e., for $Q \geq 0.4$ Å^{-1} (Fig. 4). Below this limit $\nu(Q)$ is no longer determined by $c(Q)/c_s(Q)$ as $\omega\tau \gg 1$ is not valid. The ratio can then be shown to approach $\nu(Q = 0) = 0.25$, the well-known bulk value for the Poisson rate of an isotropic solid [23]. In a solid the Poisson ratio is defined as $\nu = (c_{12} + p)/(c_{11} + c_{12})$ [39], with c_{11} and c_{12} the elastic constants and p the thermodynamic pressure. Inserting Schofield's generalizations of the elastic constants for a liquid [18] the approximation

$$\nu(Q) = \frac{\omega_t^2(Q)}{\omega_l^2(Q) + \omega_t^2(Q)} \tag{13}$$

can be derived, where $\omega_t^2(Q)$ and $\omega_l^2(Q)$ are the known second moments of the transverse and longitudinal current correlation function, respectively [22].

In Fig. 4 the experimental values [Eq. (12)] are compared with the above theoretical result [Eq. (13)], showing an increase of $\nu(Q)$ which indicates more solidlike behavior of the liquid at shorter distances. The value of ν for solid Cs ($\nu = 0.38$) is indicated for comparison [26].

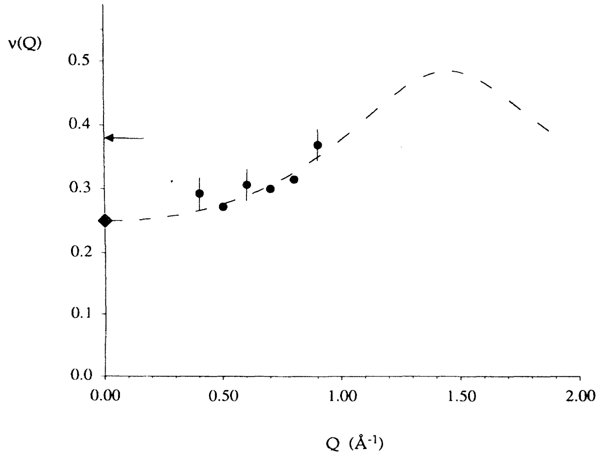

FIG. 4. Wave-vector dependent Poisson ratio $\nu(Q)$ showing a maximum at $Q_0 = 1.4$ Å^{-1}. Dots, experimental values, Eq. (12); dashed line, Eq. (13); rhomb, isotropic bulk value $\nu(0) = 0.25$; arrow, $\nu = 0.38$ for solid cesium [26].

A Poisson ratio deviating significantly from the isotropic limit $\nu = 0.25$ indicates a certain extent of a short-time anisotropy in a simple liquid at nonvanishing wave numbers. It is remarkable that the calculated Poisson ratio [Eq. (13)] reaches a maximum value of about 0.5 at Q_0, the position of the principal peak of $S(Q)$. In this region again the condition $\omega\tau \gg 1$ is not valid and hence we could not evaluate experimental values for the Poisson ratio. The anisotropy seems to be most pronounced at Q_0, corresponding to the next-neighbor distance $d \approx 2\pi/Q_0$. This may indicate again the close resemblance of a melting point liquid with a solid on this length scale.

Returning to the results in the Q region around $Q = 0.4$ Å^{-1}, our findings give some evidence for the existence of small clusters of about 15-Å size, as this length scale is resolved around 0.4 Å^{-1}. Such a volume contains about 12 to 15 atoms in liquid cesium. Thus for a short time there seems an ensemble of about 15 highly correlated particles to be formed, which shows shear relaxation on the picosecond scale. There is an interesting argument that gives evidence for the formation of clusters in a liquid [44]. In a simple calculation long ago, Frank has shown that an icosahedral cluster of 13 particles (12 around a central one) interacting via a Lennard-Jones (LJ) potential has a binding energy that is 8.4% greater than that with the corresponding crystalline structure, which is fcc in the case of LJ potentials. Indeed, for these energetic reasons, LJ liquids do show some tendency towards icosahedral order, especially in the supercooled state, as has been shown in molecular-dynamical simulations [45]. Carrying out an analogous calculation for a liquid metal—the cesium potential for $T = 308$ K of Kambayashi and Kahl has been used [46]—we find an energetic preference of 14% for the formation of such a cluster in the liquid state. Hence it can be concluded that the occurrence of clusters is even more pronounced in liquid metals than in LJ fluids. It is our conjecture that the picosecond cluster dynamics in the liquid is the beginning of a process, which is known as so-called structural relaxation in the theory for the glass transition of disordered media [32].

IV. COMPARISON WITH OTHER SYSTEMS

A short comparison of sound dispersion in other liquid metals shows the general feature of the above-described phenomena. Whereas from Copley and Rowe's rubidium data (resolution FWHM ≈ 1 meV) a zero-sound-like dispersion can only be conjectured [2], the effect clearly shows up in our experimental data for rubidium at 313 K ($T_M = 312.6$ K). These data (Fig. 5) have been obtained at the FRM Munich with an improved resolution (FWHM) of 0.54 meV [28] and are in good agreement with a recent MD simulation study of Balucani, Torcini, and Vallauri [27], showing the characteristic sound velocity maximum beyond the hydrodynamic limit.

An explanation based on a hydrodynamic mode-coupling theory as proposed for anomalous dispersion in liquid argon [9] definitely fails for a liquid metal: Calculating the value of a_s—the mode-coupling coefficient of

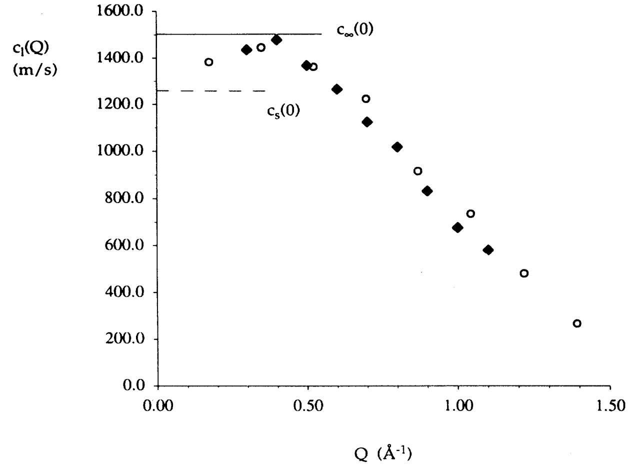

FIG. 5. Comparison with the sound velocity $c(Q)$ for liquid rubidium. Circles, Rb at 335 K (MD simulation [27]); rhombs, Rb at 313 K (scattering experiment [28]); full line, $c_\infty = 1503$ m/s; dashed line, $c_s(0) = 1260$ m/s.

the anomalous contribution in the dispersion law $\omega = c_s Q + a_s Q^{5/2}$ [9]—yields a value of $a_s = 4.3 \times 10^{10}$ Å$^{5/2}$ s^{-1} for liquid cesium at 308 K, which is two orders of magnitude smaller than for liquid argon ($a_s = 6.4 \times 10^{12}$ Å$^{5/2}$ s^{-1}, $T = 120$ K, $p = 20$ bar [9]). This is mainly due to the fact that the coefficient a_s is essentially proportional to $(\gamma - 1)^2$ [9], which is of order unity in LJ systems and 10^{-2} in a liquid metal. It is the factor $(\gamma - 1)$ that is crucial for the different dynamics in liquid metals ($\gamma \approx 1.10$) and LJ systems ($\gamma \approx 2$). The former are characterized by solid features like elastic response from collective excitations, whereas the latter show thermal relaxation. Moreover the high-frequency limit $\omega\tau \gg 1$ is not reached properly in these liquids, except for the case of strongly pressurized argon (850 bar) [10], where Verkerk, van Well, and de Schepper [10] interestingly state a glasslike dispersion of sound modes. A more recent demonstration of thermal relaxation in a LJ system is the systematic study of Montfrooij for dense helium gas [40], thus representing a physical system contrary to a liquid metal. Also, from the extensive study of hard-sphere systems at various densities both experimentally [41] and theoretically [42,43] can be deduced that solidlike sound propagation only occurs at high densities near the solidification. Hence, again a zero-sound-like dispersion can be viewed as a precursor to the liquid-solid transition, which recently has found wide interest as glass transition in the case of disordered systems [32].

V. CONCLUSIONS

In conclusion, we report high-resolution thermal neutron-scattering results on liquid cesium and liquid rubidium near the melting point. In these liquid metals short-wavelength collective excitations with anomalous dispersion are observed for wave numbers beyond the hydrodynamic limit. These propagating modes are identified as zero sound in the region around $Q = 0.4$ Å^{-1}. First, a renormalization of the known adiabatic sound velocity c_s, leading to an enhanced high-frequency sound velocity c_∞, is found. Second, a reduced damping of the zero sound modes is observed, varying only linearly with wave number. Furthermore the elastic shear modulus G_∞ of liquid cesium could be determined experimentally. From the solidlike features of sound dispersion a quantity known from the theory of elasticity of solids—the Poisson ratio ν—is introduced into the discussion of liquids, showing an increase over the isotropic value $\nu = 0.25$ at $Q = 0$, indicating a local anisotropy at nonvanishing wave numbers in a simple liquid. The existence of short-time clusters of 12 to 15 particles has been discussed and corroborated by an estimation of the binding energy of an icosahedrally ordered cluster in the liquid metal.

An interesting issue for further experiments would be to follow the collective modes down to pure adiabatic sound, which has been estimated above to occur at $Q = 0.1$ Å^{-1}. A Brillouin scattering experiment on a liquid metal could shed some light upon this question and is yet an outstanding task. Second, it might be rather interesting to unravel the microscopic solidlike features of a simple liquid further in molecular-dynamics simulations, which moreover could substantiate the existence of small clusters on the picosecond time scale in a simple liquid metal.

ACKNOWLEDGMENTS

The help of the FRM Munich workshop and the staff of the ILL Grenoble with the performance of the experiments is gratefully acknowledged. We are indebted to U. Balucani for providing us with molecular-dynamics simulation data for liquid rubidium and thank G. Kahl for leaving to us the cesium potential in tabulated form. Encouraging discussions with B. Alder, P. Egelstaff, and W. Götze are gratefully acknowledged. This work was supported by the German Bundesministerium für Forschung und Technologie.

---

[1] S. J. Cocking and P. A. Egelstaff, J. Phys. C **2**, 507 (1968).
[2] J. R. D. Copley and J. M. Rowe, Phys. Rev. A **9**, 1656 (1974); Phys. Rev. Lett. **32**, 49 (1974).
[3] O. Söderström, Phys. Rev. A **23**, 785 (1981).
[4] T. Bodensteiner, Chr. Morkel, P. Müller, and W. Gläser, J. Non-Cryst. Solids **117&118**, 116 (1990); Chr. Morkel and T. Bodensteiner, J. Phys. Condens. Matter **2**, SA251 (1990).
[5] K. Sköld, J. M. Rowe, G. Ostrowski, and P. D. Randolph, Phys. Rev. A **6**, 1107 (1972).

[6] A. A. van Well, P. Verkerk, L. A. de Graaf, J. B. Suck, and J. R. D. Copley, Phys. Rev. A **31**, 3391 (1985).

[7] H. Bell, H. Moeller-Wenghoffer, A. Kollmar, R. Stockmeyer, T. Springer, and H. Stiller, Phys. Rev. A **11**, 316 (1975).

[8] P. A. Egelstaff, W. Gläser, D. Litchinski, E. Schneider, and J. B. Suck, Phys. Rev. A **27**, 1106 (1983).

[9] I. M. de Schepper, P. Verkerk, A. A. van Well, and L. A. de Graaf, Phys. Lett. **104A**, 29 (1984).

[10] P. Verkerk, A. A. van Well, and J. M. de Schepper, J. Phys. C **20**, L979 (1987).

[11] J. W. Lewis and S. W. Lovesey, J. Phys. C **10**, 3221 (1977).

[12] J. Bosse, W. Götze, and M. Lücke, Phys. Rev. A **18**, 1176 (1978).

[13] L. Sjögren, Phys. Rev. A **22**, 2883 (1980).

[14] D. Pines and Ph. Nozières, *Theory of Quantum Liquids* (Benjamin, New York, 1966), Vol. 1.

[15] S. W. Lovesey, *Theory of Neutron Scattering from Condensed Matter* (Oxford University Press, Oxford, 1986), Vol. 1.

[16] J. P. Hansen and J. R. McDonald, *Theory of Simple Liquids,* 2nd ed. (Academic, London, 1986).

[17] P. A. Egelstaff, *An Introduction to the Liquid State* (Academic, London, 1967).

[18] P. Schofield, Proc. Phys. Soc. London **88**, 149 (1966).

[19] S. Yip, in *New Directions in Physical Acoustics,* edited by D. Sette (North-Holland, Amsterdam, 1976), p. 55.

[20] C. H. Chung and S. Yip, Phys. Lett. **50A**, 175 (1974).

[21] S. W. Lovesey, Z. Phys. B **58**, 79 (1985).

[22] J. P. Boon and S. Yip, *Molecular Hydrodynamics* (McGraw-Hill, New York, 1980).

[23] R. Zwanzig and R. D. Mountain, J. Chem. Phys. **43**, 4464 (1965).

[24] S. W. Lovesey, J. Phys. C **4**, 3057 (1971).

[25] J. Jäckle, Z. Phys. B **64**, 41 (1986).

[26] N. Nücker and U. Buchenau, Phys. Rev. B **31**, 5479 (1985).

[27] U. Balucani (private communication). For a similar comparison with cesium, see U. Balucani, A. Torcini, and R. Vallauri, Phys. Rev. A **46**, 2159 (1992).

[28] H. Gemperlein, Diplom-Arbeit, Technische Universität München, 1984.

[29] T. Bodensteiner, Ph.D. thesis, Technische Universität München, 1990.

[30] T. Bodensteiner, *et al.*, Phys. Rev. A **45**, 5709 (1992).

[31] P. A. Egelstaff and W. Gläser, Phys. Rev. A **31**, 3802 (1985).

[32] W. Götze and L. Sjögren, Rep. Prog. Phys. **55**, 241 (1992), and references therein.

[33] R. D. Mountain, Rev. Mod. Phys. **38**, 205 (1966).

[34] W. Ohse, *Handbook of Thermodynamic and Transport Properties of Alkali Metals* (Blackwell, Oxford, 1985).

[35] J. Frenkel, *Kinetic Theory of Liquids* (Dover, New York, 1955).

[36] P. A. Egelstaff, G. Kearley, J. B. Suck, and J. P. A. Youden, Europhys. Lett. **10**, 37 (1989).

[37] M. Soltwisch, J. Sukmanowski, and D. Quitmann, J. Chem. Phys. **86**, 3207 (1987).

[38] R. T. Beyer and S. V. Letcher, *Physical Ultrasonics* (Academic, New York, 1969).

[39] D. Frenkel and A. Ladd, Phys. Rev. Lett. **59**, 1169 (1987).

[40] W. Montfrooij, Ph.D. thesis, Technical University of Delft, 1990.

[41] W. E. Alley and B. J. Alder, Phys. Rev. A **27**, 3158 (1983); **27**, 3174 (1983).

[42] E. Leutheusser, J. Phys. C **15**, 2801 (1982); **15**, 2827 (1982).

[43] U. Bengtzelius and L. Sjögren, J. Chem. Phys. **84**, 1744 (1986).

[44] F. C. Frank, Proc. R. Soc. London Ser. A **215**, 43 (1952).

[45] P. Steinhardt, D. Nelson, and M. Ronchetti, Phys. Rev. B **28**, 784 (1983).

[46] S. Kambayashi and G. Kahl, Phys. Rev. A **46**, 3255 (1992).