Dynamics of Long-Wavelength Fluctuations in a Fluid Layer Heated from Above

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We report an experimental study of the dynamics of thermal fluctuations in a 4.86 mm thick layer of CS_2 heated from above. Stabilizing gradients ranged from 10.3 to 61.7 K/cm. Power spectral measurements were made over the wave vector range 9 $\mathrm{cm}^{-1} < q < 100$ cm^{-1}, using a shadowgraph apparatus. Over this range, the spectra change from having a well-resolved oscillatory mode at low q to monotonic decay at higher q. For the smallest gradient, the results for the power spectra compare very well with theory, but slight deviations are seen for larger gradients.

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A horizontal fluid layer heated from above is stable in the presence of gravity and will not normally undergo convection, but fluctuations of the local temperature, and of velocity about its mean of zero, are dramatically altered by the gradient [1–16]. A remarkable prediction, made three decades ago, is that coupling of the thermal and viscous modes leads to a propagating mode in the presence of gravity [1,17]. Although this phenomenon has been observed [17], no quantitative study has been reported. In this Letter, we present measurements of the spectral power density $S(q, \omega)$, for various wave vectors q, that reveal the finite-frequency peak corresponding to the predicted mode, and are in near-quantitative agreement with recent predictions [14,18]. Some differences between theory and experiment are observed, however.

Fluctuations in nonequilibrium systems have been studied for several decades, and a recent monograph summarizes progress [19]. Theory is well advanced for single-component fluids [8,9,14,15] and binary mixtures [14–16] held in steady state by an applied temperature gradient. A gradient severely affects thermal fluctuations, making them spatially long-ranged, increasing their mean-squared amplitude, and radically altering their dynamics. Detailed predictions, with realistic treatment of the horizontal boundaries, are available [14–16,19]. However, it is *assumed* that the Langevin terms appropriate in equilibrium are also applicable in nonequilibrium steady states.

Several experiments have been carried out using fluids heated from below, with the gradient less than that required for convection. The mean-squared amplitude of temperature fluctuations in high-pressure CO_2 has been measured [20] as a function of the difference between the applied gradient ∇T, and the critical one required for convective onset, and the results were consistent with theory. Using a near-critical sample of SF_6, it has been observed [21] that sufficiently near the convective onset, the fluctuations drive the transition to convection from being continuous to discontinuous, an effect predicted by Swift and Hohenberg [22]. A study of dynamics in near-critical SF_6 has also been reported [18]. The measured decay rates did not compare well with theory, which the authors tentatively attribute to the strong dependence of fluid properties on vertical position.

The quenching of sufficiently low-q fluctuations by gravity has been measured in a near-critical binary mixture heated from above [23], but the large gradient necessary resulted in a nonlinear concentration gradient, an effect not included in the theory.

Dynamic light scattering has been used to study fluctuations in toluene heated from above [24–26], for q in the range 1500–2500 cm^{-1}, where the effects of a gradient are relatively small. Two modes having the appropriate decay times, and with amplitudes varying as $\nabla T^2/q^4$, were observed, in accord with predictions. The absolute amplitudes were initially found to be about half the predicted values; however, improved experiments resulted in excellent agreement, both for toluene [25,26] and n-hexane [27].

Similar phenomena are predicted [28] during free-diffusion. Several such experiments have been reported [28–36], but current theory makes the drastic assumption that the fluctuations are uncorrelated in the vertical direction, and is not in quantitative agreement with the data.

The most promising systems for testing theory are single-component fluids and noncritical binary mixtures with stabilizing temperature gradients. These systems are relatively insensitive to sample thickness and ∇T; however, they have small signals, and the predicted effects are large only for wave vectors below $\sim$100 cm^{-1}, where normal light scattering methods become infeasible. However, refined shadowgraph instruments can make measurements equivalent to light scattering in this wave vector regime [20,37,38]. In this Letter we present data for the dynamics of long-wavelength fluctuations in a 4.86 mm thick layer of CS_2 heated from above, obtained using such a shadowgraph.

The optical layout was similar to that described by de Bruyn *et al.* [39]. The diverging beam from an optical fiber was collimated by a lens, passed through a window into the sample, and was reflected back upon itself by a mirror that confined the sample from below. Having again passed through the lens, ~half the return beam was directed to a digital camera by a beam splitter. The optical path was evacuated. To take images sufficiently rapidly, we used a 680 nm wavelength superluminous diode coupled to a single-mode fiber as the light source. We heated the sample from above by means of a transparent layer of indium tin oxide (ITO) on the lower surface of the fused silica window, with the ITO in contact with the liquid. Heat was removed through the lower surface of a Silicon mirror by means of Peltier devices. The sample diameter was 100 mm, with the large lateral dimension chosen to avoid making measurements near the periphery. We applied temperature differences ranging from 5 to 30 K across the sample, holding its mean temperature fixed at 27.5 °C. Using a frame rate of 10 Hz, successive images were taken for about an hour for each applied gradient. The exposure time was 1 ms.

We analyzed the central 512×512 portion of the images, corresponding to a 5.16 cm square region. We used groups of $N = 256$ consecutive images, averaged the images in a set and divided each image in the set pixel by pixel by the average image to obtain a set of N ratio images per set. The ratio images were Fourier transformed to separate the fluctuations according to wave vector $\mathbf{q}$, which yielded a set of $\mathbf{q}$-space "images", $i(\mathbf{q}, n\delta t)$, ($\delta t = 0.1$ s) with $1 \leq n \leq N$ for each set. The $\mathbf{q}$-space images within a set were Fourier transformed temporally to yield $i(\mathbf{q}, p\delta\omega)$ where $-(N/2 - 1) \leq p \leq N/2$, and $\delta\omega = 2\pi/T$, with T being the measurement time for one set. The spectrum for each set was obtained, as

$$S(\mathbf{q}, p\delta\omega) \propto |i(\mathbf{q}, p\delta\omega)|^2. \tag{1}$$

We averaged the positive and negative frequency components of equal frequency magnitude, and averaged over all $\mathbf{q}$ having magnitudes q lying within a given band. The bands were chosen small enough to result in no significant distortion of the spectrum, as judged by varying their width. Finally, we averaged over the spectra obtained from each set of images. We repeated this process with no applied gradient to obtain a background noise spectrum to subtract from the spectra. The background spectra did not resolve the equilibrium fluctuations, which remain below the resolution limit of our instrument.

To compare with theory, we Fourier transformed the result given by Oh *et al.* [18] for the autocorrelation function of the fluctuations, to obtain the spectrum as the sum of two Lorentzian-like forms

$$S(q, \omega) \propto A_+\left(\frac{\Gamma_+}{\omega^2 + \Gamma_+^2}\right) + A_-\left(\frac{\Gamma_-}{\omega^2 + \Gamma_-^2}\right). \tag{2}$$

Here, Γ_+ and Γ_- are the decay rates of the coupled viscous and thermal modes, respectively, which are complex in general, and are given by Eq. 2 of Ref. [18]. In the high-q limit, the decay rate Γ_+ approaches νq^2, while Γ_- approaches κq^2, where ν and κ are the kinematic viscosity and thermal diffusivity, respectively. The amplitudes $A_\pm$ are given by Eq. 6 of [18]. These predictions result from a zeroth-order Galerkin calculation with perfectly conducting no-slip horizontal boundaries and a laterally infinite sample [14]. We used fluid parameters [40] appropriate for CS_2 to calculate $S(q, \omega)$ for our system.

Figure 1 shows data (circles) and theory (solid lines) for a 10.3 K/cm gradient, for several wave vectors. The mean wave vectors for which the theory was evaluated are averages over the data ranges, weighted by the ideal shadowgraph transfer function $\sin^2(q^2 z/2k_o)$. Here z is the viewing distance, which was 5.5 m for our apparatus, and k_o is the vacuum wave vector of the light. Changing the averaging range by a factor of 2 had little effect on the data. The amplitude of the theoretical result was adjusted to match the data, but no other parameters were adjusted, nor was any adjustment made in the subtracted baseline. As can be seen, the dynamics are definitely oscillatory for sufficiently small wave vector, as evidenced by the well-defined peaks in the upper two graphs. In general, the theory does a remarkable job with the line shapes, which vary substantially with wave vector. It is notable that although the modes are predicted to be oscillatory below 65.7 cm^{-1} for this gradient, no clearly resolved peak, which one might naively expect to be a signature of oscillatory behavior, is discernable in the data (or the theory) for q greater than ~32 cm^{-1}.

Figure 2 shows a similar set of spectra together with the theory for an applied gradient of 61.7 K/cm. Although agreement between theory and experiment is not as good for the larger gradient, it is still quite reasonable. The most obvious ways in which the theory differs from the data are revealed by Figs. 2(a) and 2(b). As shown in (a), at very low q the predicted frequency is somewhat smaller, and the predicted line width substantially smaller than observed. The main deviation revealed by (b) is that for sufficiently low q, the spectral power observed at small ω tends to be somewhat larger than predicted. This effect is also visible in Fig. 1(b) as well. These effects may be at least partially experimental in nature, and will require further testing for the reason outlined in the next paragraph.

The thermal conductivity of our upper boundary is only about 9.3 times that of the fluid, while theory assumes perfectly conducting horizontal boundaries. Non-Boussinesq effects may also play a small role; with an applied difference of 5 (30) K, the kinematic viscosity varies vertically by 2.9 (19)%, the thermal expansion coefficient α, by 1.4 (8)%, the Prandtl number by 2.6 (17)%, and the Rayleigh number by 4.5 (31)%. Because the oscillation frequency depends on α, some "dispersion broadening" of the line at low q is to be expected. We estimated this by integrating the theory vertically, assuming a linear gradient and using the temperature-dependent fluid prop-

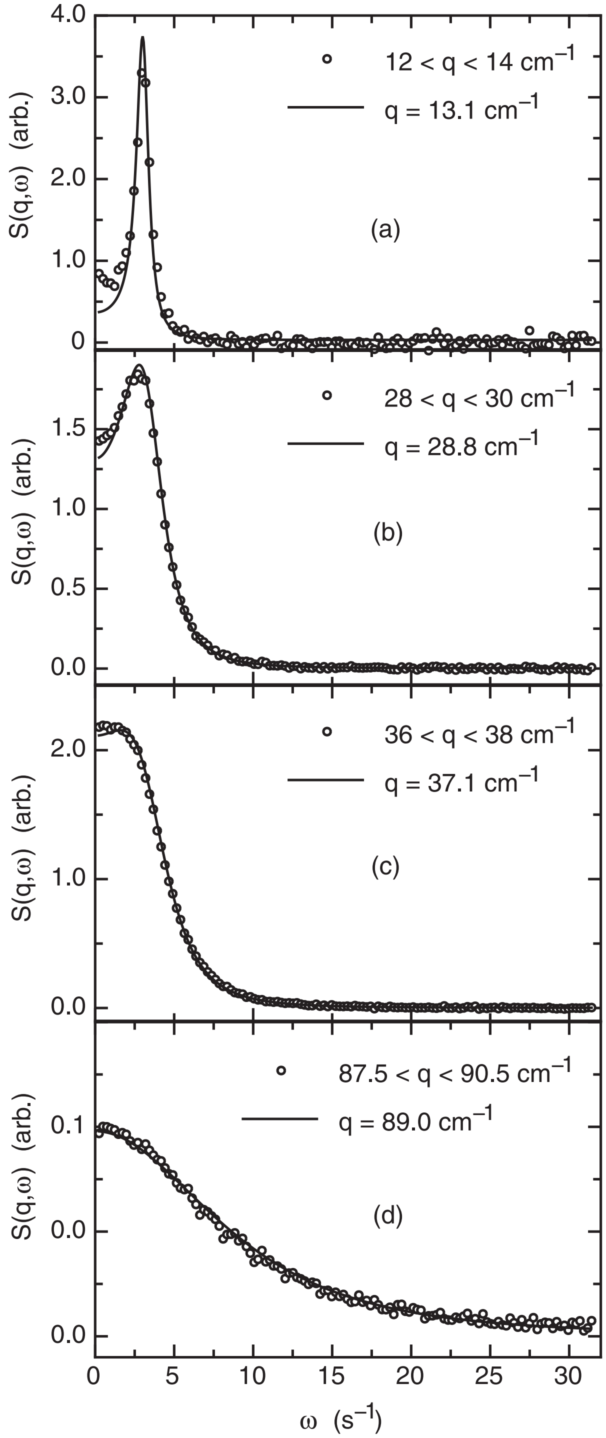

FIG. 1. Graphs of the temporal spectral power density $S(q, \omega)$ *vs*. ω for several wave vectors for a 4.86 mm thick layer of CS2 with an applied temperature difference of 5.0 K. The data are shown as circles and the theory as solid lines. The theory curves have been scaled in amplitude, but no other parameters have been adjusted.

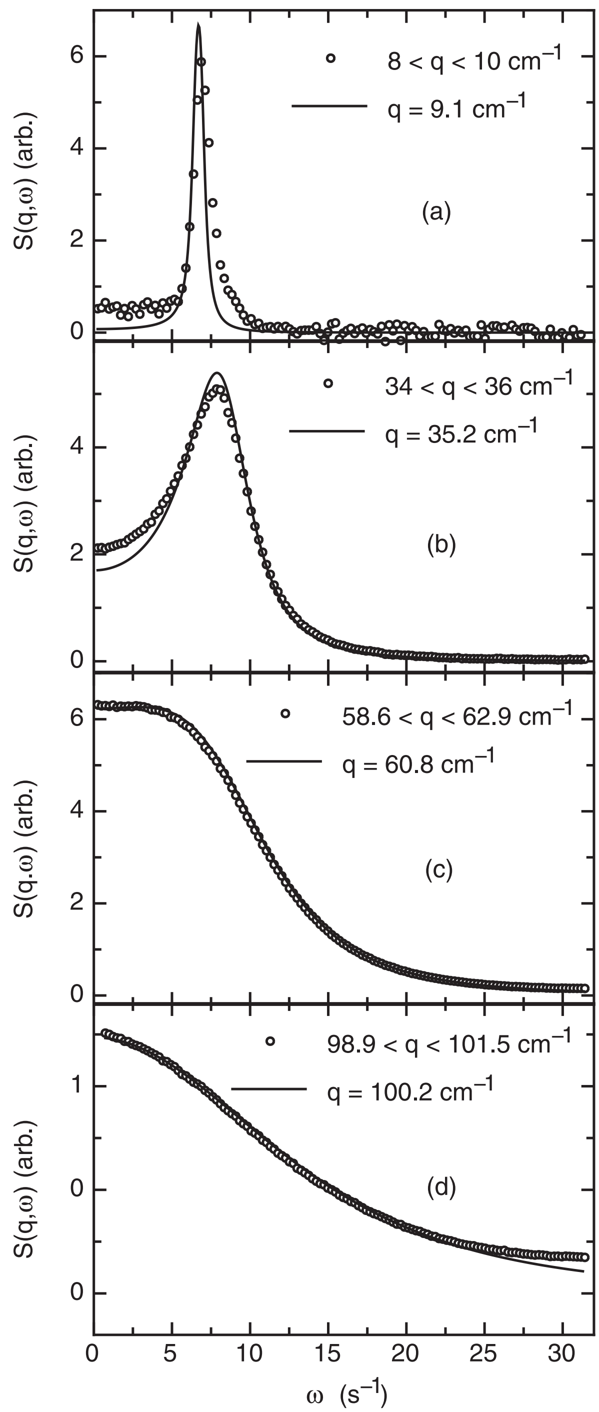

FIG. 2. Graphs similar to those of Fig. 1, but for an applied temperature difference of 30.0 K.

erties. For the conditions of Fig. 2(a), this increased the predicted line width, by $\sim$15%, which is considerably less than the observed discrepancy. The lines shown in the figures were calculated using the midplane values for all fluid parameters. One should also recall that the theory is based on a zeroth-order Galerkin approximation.

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