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Dynamics of Normal and Superfluid Fogs Using Diffusing-Wave Spectroscopy

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Abstract
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Disciplines
Physical Sciences and Mathematics | Physics

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Dynamics of normal and superfluid fogs using diffusing-wave spectroscopy

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The dynamics of normal and superfluid fogs are studied using the technique of diffusing-wave spectroscopy. For a water fog generated with a 1.75 MHz piezoelectric driver below the liquid surface, the 7 μm diameter droplets are found to have diffusive dynamics for correlation times long compared to the viscous time. For a fog of 10 μm diameter superfluid helium droplets in helium vapor at 1.5 K the motion appears to be ballistic for correlation times short compared to the viscous time. The velocity correlations between the helium droplets are found to depend on the initial velocity with which the droplets are injected from the helium surface into the fog.

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In a previous experiment [1] it was possible to create a dense fog of superfluid helium droplets by driving the capillary waves on a liquid helium surface unstable with an intense ultrasonic beam from a transducer under the surface. The fog droplets were found to have a diameter that is about one wavelength of the surface waves created by the transducer, and were ejected into the vapor with velocities of order 1 m/s, the maximum velocity of the oscillating surface. A puzzle in these experiments was the question of how the fog could be formed at heights up to 5–6 cm from the surface. A calculation of the viscous drag on a droplet from the helium vapor appeared to limit the droplets to a maximum height of only a few millimeters, if the vapor was assumed to be stationary. It was speculated in Ref. [1] that the vapor must in fact be moving along with the injected droplets to produce the observed fogs. The dynamics of the creation of a steady-state fog appears to be quite complex, and to check whether the injected droplets are indeed still moving rapidly centimeters above the surface requires a probe that is sensitive to the individual droplet motion. In this paper we report the first use of diffusing-wave spectroscopy (DWS) to investigate the droplet dynamics of fogs.

The technique of diffusing-wave spectroscopy has been used extensively to study the dynamics of small particles immersed in fluids. DWS utilizes a laser beam passing through the sample, and the temporal intensity fluctuations in the speckle field of the light that is multiply scattered from the particles allows a determination of their motion on small length scales [2,3]. This has been used to characterize the microscopic dynamics of systems such as fluid suspensions of micron-sized beads [4] and of flowing granular materials [5]. Although light beams have generally been employed in these studies, it was recently shown that the scattering of an acoustic beam can also be used in a similar fashion to determine the velocity fluctuations in a fluidized suspension of particles [6].

To compare with the superfluid fog we have also applied the DWS light-scattering method to a water fog in air. Although water fogs have been extensively studied over the years with various techniques [7], the microscopic dynamics of the droplets has not been well characterized. We find that the dynamics of the water droplets take the form of diffusive motion through the air, for particle correlation times longer than the viscous relaxation time. The dynamics of the helium droplets, on the other hand, appears to be ballistic for correlation times short compared to the viscous time of the helium vapor. From an analysis of the helium fog data the average relative velocity between the droplets is found to be linearly proportional to the initial velocity that they are ejected from the helium surface. This provides a picture of the steady-state dynamics of the fog, where a small number of high-velocity droplets emerging from the surface appear to move upwards relatively freely, while the bulk of the droplets fall back under gravity.

For the water fog experiment shown in Fig. 1, a commercial transducer unit ("Mist-Maker") is immersed 2 cm under the surface of distilled water at room temperature (22°C) to generate the fog. This utilizes two spring-mounted piezos operated at 1.75 MHz, and the average diameter of the water droplets which are produced is 7 μm, as measured with a long-distance microscope. This corresponds well with the capillary wavelength of the water surface at this frequency [1]. The diameters of the droplets appear to be fairly uniform in the microscope photographs, though we are limited by the 4 μm resolution of the microscope. Due to the spring mounts, the piezoelectric unit also produces much larger drops up to about 0.5 cm diameter, which are ejected up to 15–25 cm above the surface. Since these drops still carry the strong ultrasonic field (the reflection coefficient at the surface is practically unity) they also generate small droplets at their surface, and this greatly increases the height and the density

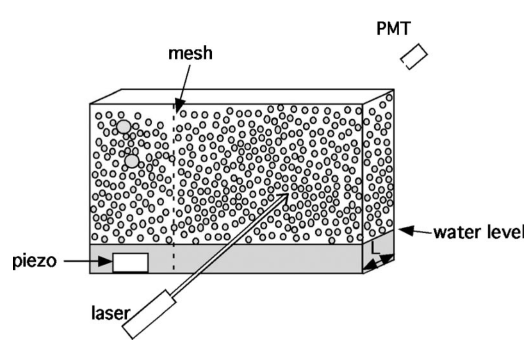

FIG. 1. Apparatus for diffusing-wave spectroscopy with water fogs.

of the fog. To avoid the disruption of the large drops, the transducer is placed at the far end of the rectangular glass cell that is 7 cm width × 28 cm length × 25 cm height, and a mesh screen is placed in the vapor to prevent large drops from spraying across the cell. The small drops diffuse through the mesh and produce a fairly uniform steady-state thick fog in the other side of the cell to a height of 12–15 cm, and the measuring laser is at a height of 6 cm. In steady-state conditions there are no observable fluctuations in the transmitted laser intensity at low frequency. As the drops fall under gravity they reach a terminal velocity of a few mm/s; it takes about 60 s for the fog to clear after the transducer is turned off. An antifog surfactant is sprayed on the inside glass surfaces that prevents drop formation on the walls and allows the impinging droplets to drain uniformly. A 100 mW argon-ion laser at 514.5 nm, whose coherence length is much longer than the average photon mean free path through the fog, is focused on the small-drop side of the sample cell. The speckle-pattern fluctuations at a small angle from the exiting diffuse light are viewed with a photomultiplier through a 200 μm pinhole. The autocorrelation function data were collected for 10 min while holding the conditions as steady as possible.

The density of the water fog was determined from the attenuation of the laser beam with and without the fog present, as measured with a photodiode at the beam exit point. The intensity of the laser beam is exponentially attenuated by scattering from the drops through a fog of thickness L as

$$I = I_0 \exp\left(-\frac{L}{l_s}\right), \tag{1}$$

where l_s is the average distance between scattering events, $l_s = 1/nQ_{ext}\pi R^2$, with n the density of droplets, R the droplet radius, and Q_{ext} the extinction efficiency [8]. The photon transport mean free path, $l^* = l_s/\langle 1 - \cos\theta\rangle$, is the distance over which the propagation direction is randomized, and is defined such that the photon diffusion constant is $l^*c/3$, with c the speed of light. The average cosine of the scattering angle and extinction efficiency of the water droplet for 514.5 nm wavelength are 0.845 and 2.14, respectively, which are calculated from Mie scattering theory for our droplet size. In the steady state the density was stable at $n = 5 \times 10^6/\text{cm}^3$ over the measuring time, a volume fraction for the droplets of 9.8×10^{-4}. We find $L/l^* = 4.7 \pm 1$, where $L = 7$ cm is the cell thickness, and the main uncertainty is from the droplet size.

The characteristic viscous time scale $\tau_v = R^2\rho_v/\eta_v$, where ρ_v and η_v are the density and viscosity of the vapor, determines the droplet dynamics [4]. For times long compared to τ_v the motion will be diffusive, with the mean-square displacement of a droplet after time t given by

$$\langle \Delta r^2 \rangle = 6Dt, \tag{2}$$

where

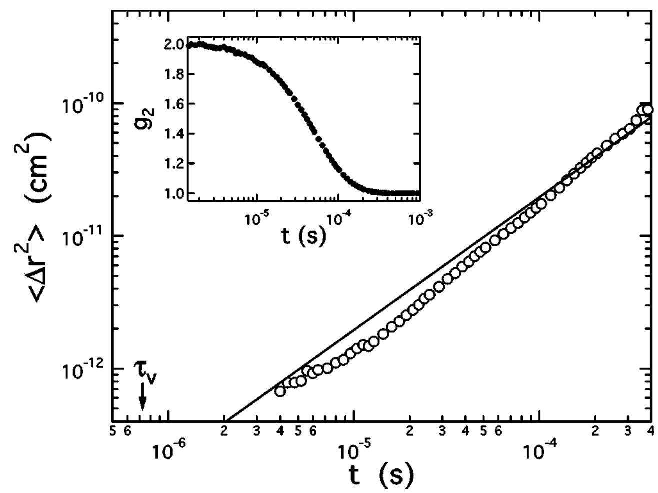

FIG. 2. The mean-square drop displacement as a function of the correlation time for water fog. The line represents the theoretical prediction $6Dt$ of Eqs. (2) and (3). The inset shows the autocorrelation data.

$$D = \frac{k_B T}{6\pi\eta_v R} \tag{3}$$

is the self-diffusion coefficient given by the Stokes-Einstein formula. This is the asymptotic value of the diffusion coefficient, reached after the velocity autocorrelation function has decayed but before a drop has moved far enough to interact with its neighboring droplets. The factor of 6π in Eq. (3) is appropriate for a water drop where the viscosity is high compared to the vapor viscosity, but for superfluid helium drops where the normal fluid viscosity is much lower the factor is closer to 5.2π [9].

The measured autocorrelation function [10] for the water fog, g_2, is shown in the inset to Fig. 2, which as expected, varies between the limiting values of two at short times and one at long times. We can extract the mean-square displacement of the droplets from the autocorrelation by using Eq. (7) of Ref. [10], and the result is shown as the main curve in Fig. 2. It shows approximately a power-law increase, with an exponent close to the value of one predicted by Eq. (2), which should be valid since the viscous time scale for the water fog is $\tau_v = 8.7 \times 10^{-7}$ s, considerably shorter than these correlation times. With values appropriate for ambient atmospheric air in Eq. (3), the diffusion coefficient is $3.3 \times 10^{-8}\ \text{cm}^2/\text{s}$, and the solid line in Fig. 2 is the prediction of Eq. (2) using this value. The agreement is quite good; the dynamics of a water fog droplet in air appears to be well approximated by Brownian motion for correlation times exceeding the viscous time.

The experimental apparatus to generate the superfluid helium droplets is shown in Fig. 3. A thin composite brass/piezoelectric disk transducer is used to drive the helium surface, where the piezo is 2 cm diameter and the brass disk 4 cm in diameter. The sealed cell 6 cm in diameter is surrounded by superfluid helium held at 1.5 K, and is mounted in an optical dewar. The cell is partially filled by condensing purified helium gas into it. The vapor pressure in the cell is

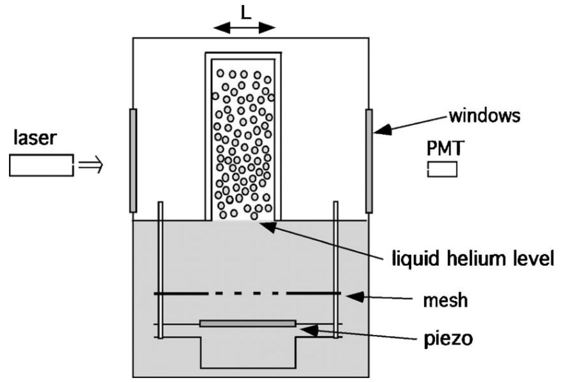

FIG. 3. Apparatus for diffusing-wave spectroscopy with superfluid helium fog at 1.5 K.

monitored with a capacitance pressure gauge to ensure that the temperature of the droplets and the liquid in the cell does not rise more than 0.02 K above the bath temperature from the piezoelectric dissipation. The average droplet size ranges from about 100 μm at a drive frequency of 1 kHz to 10 μm at 120 kHz. For the DWS experiment a Nd:YAG laser producing 150 mW at 532 nm and droplets with an average diameter of 10 μm at 124 kHz drive frequency [11] were employed. A Plexiglass rectangle closed at the top and having dimensions 2.5 cm width $\times$ 5 cm length $\times$ 7 cm height defines the fog region and keeps the density uniform over the 2.5 cm path of the laser. The maximum fog height above the liquid helium depends somewhat on the drive level, but is typically 4–6 cm, and the laser beam traverses it at about one-half the maximum height. The terminal velocity of the helium droplets is a few cm/s, and it typically takes 4–8 s for the fog to clear after turning off the drive. Evaporation of the drops is not significant over this time; we calculate it would take 150 s for these drops to completely evaporate.

The density of the helium fog increases and then saturates at a value of 5×10^{7} cm^{-3} as the drive voltage increases [1], a volume fraction of 0.026, and the experiment is carried out at this saturated density. Mie scattering theory shows that for 10 μm helium droplets and a 532 nm wavelength the extinction efficiency is 1.84 and the average cosine of the scattering angle is 0.979, since the small dielectric constant of liquid helium results in strong forward scattering. The corresponding value of L/l^* is 3.6 ± 1 for these experimental conditions.

For the superfluid helium fogs the characteristic viscous time is $\tau_v=9.5\times10^{-6}$ s, longer than that of the water fogs, due to the smaller helium vapor viscosity ($\eta_v=4\mu P$) compared with air. The autocorrelation function was taken at a number of different drive amplitudes of the piezoelectric transducer where sufficiently high densities of fog could be produced to keep $L/l^*>3$. As with the water fog experiment, we can extract the mean-square displacement from the autocorrelation data [10], shown in the inset of Fig. 4. The main curve in Fig. 4 shows the mean-square displacement at a drive amplitude of 57 V. At short times the displacement is proportional to t^2, showing that the droplets initially move in ballistic trajectories, since for that case one expects

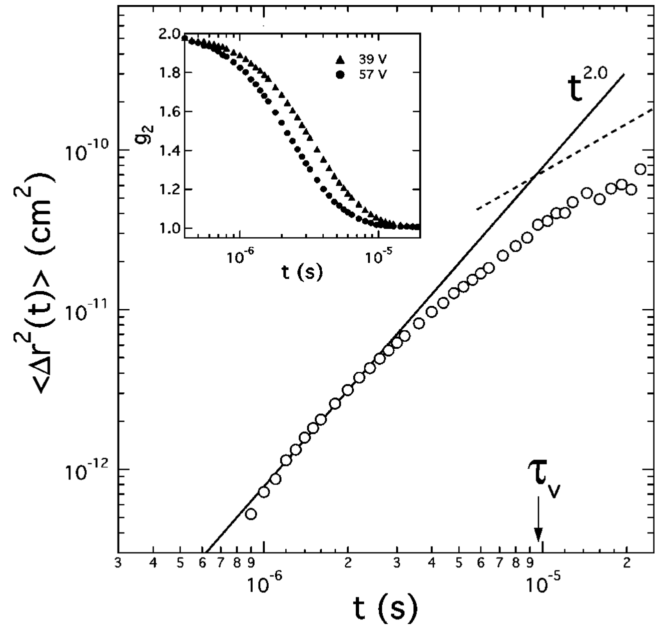

FIG. 4. The mean-square displacement as a function of the correlation time for a helium fog (57 V drive on the piezo, where the critical drive voltage to produce droplets was 15 V). The solid line indicates the t^2 behavior that would be expected for ballistic dynamics, while the dashed curve shows the linear in t behavior of diffusive dynamics. The inset shows the autocorrelation data for two drive voltages, 39 and 57 V.

$$\langle\Delta r^2\rangle=\langle\Delta v_{rel}^2\rangle t^2, \tag{4}$$

where Δv_{rel} is a randomly directed relative velocity between two drops. At longer times there is a deviation from the t^2 behavior, with a slower increase in the mean-square displacement, which begins near the viscous time. This may be the crossover to the diffusive regime which is expected for times greater than τ_v. The dashed line is the diffusive prediction of Eq. (2) for helium parameters. Unfortunately, our data at times longer than 10^{-5} s become increasingly uncertain since the correlation function is rapidly approaching its limiting value of one where no further information on the relative displacement can be extracted.

We repeated these helium fog measurements for several different piezoelectric drive amplitudes, which increases the initial velocity of the droplets being emitted from the helium surface, but has almost no effect on the fog density. This initial velocity was measured previously [1] using a pulsed drive and time-of-flight techniques, and was found to be directly proportional to the velocity of the oscillating helium surface. The relative velocities are found from the data of the mean-square displacement versus the correlation time (as in Fig. 4) by fitting to Eq. (4). In Fig. 5 the measured relative velocities $\delta v=\sqrt{\langle\Delta v_{rel}^2\rangle}$ from the DWS data at different drive amplitudes are shown as a function of the initial velocity of the emitted droplets, calibrated from the onset drive voltage as in Ref. [1]. From Fig. 5 it is apparent that the relative velocity is proportional to the initial droplet velocity, but two orders of magnitude smaller. We believe that this magnitude can be understood from the steady-state dynamics of the fog.

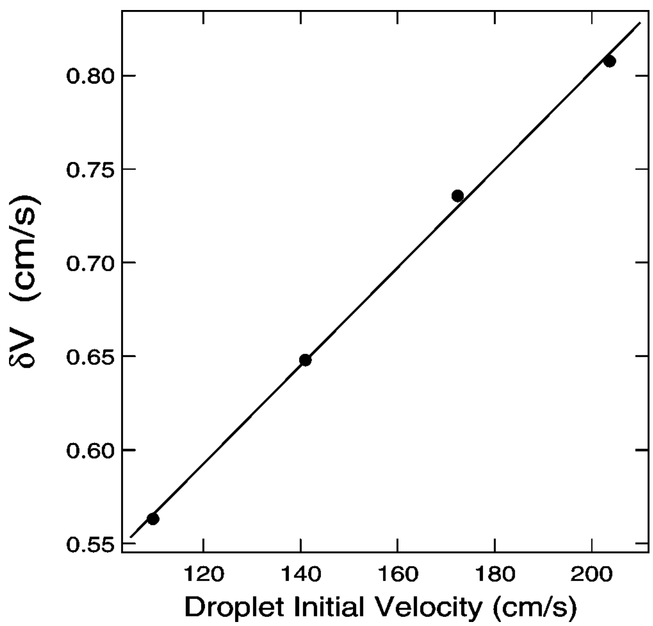

FIG. 5. The relative velocity of helium droplets vs the velocity with which the helium droplets are being ejected from the helium surface. These are for drive voltages of 39, 45, 51, and 57 V, where the onset voltage for drop generation was 15 V. The line shows a linear fit, with the relative velocity linearly proportional to the initial droplet velocity.

It is likely that the fog is made up of essentially two classes of drops: the small fraction emitted from the surface on every cycle with velocities of order 1 m/s that increase with the drive, and the vast majority falling back to the surface with terminal velocities of a few cm/s that will be independent of the drive. A photon will occasionally scatter both from a fast droplet and a slow one, giving a large relative velocity, but then when the mean-square average is taken over those which only scatter from slow drops the net result is a greatly reduced value, but one which does increase with the drive amplitude. Of course a disadvantage of the DWS method is that the beam samples the entire width of the fog and gives no information on the spatial dependence of the droplet velocities. It is possible that there could be a central region where the fast droplets and vapor move together to heights of several centimeters, surrounded by an outer region where the droplets fall back. Further experiments varying the scattering geometry and possibly employing fiber-optic probes will be necessary to check if this is the case.

This experiment is the initial application of diffusing-wave spectroscopy to study the dynamics of normal and superfluid fogs. The results are in agreement with expectations: the motion of a water fog in air appears to be diffusive, with the measured diffusion constant being reasonably close to the theoretical value for motion at times long compared to the viscous time. The motion of helium droplets, on the other hand, is found to be ballistic for time scales short compared to the viscous time. We have not seen any evidence that the superfluidity of the helium droplets plays a role in the dynamics. We also find that the relative velocity between the helium droplets increases proportional to the velocity that the droplets are being ejected from the helium surface, but with a small value that appears to be an average of the high-speed droplets being injected from the surface and the much larger number of droplets drifting down under gravity.

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[1] H. Kim, K. Seo, B. Tabbert, and G. A. Williams, Europhys. Lett. **58**, 395 (2002); J. Low Temp. Phys. **121**, 621 (2000).
[2] G. Maret and P. E. Wolf, Z. Phys. B: Condens. Matter **65**, 409 (1987).
[3] D. J. Pine, D. A. Weitz, P. M. Chaikin, and E. Herbolzheimer, Phys. Rev. Lett. **60**, 1134 (1988).
[4] J. X. Zhu, D. J. Durian, J. Muller, D. A. Weitz, and D. J. Pine, Phys. Rev. Lett. **68**, 2559 (1992).
[5] N. Menon and D. J. Durian, Science **275**, 1920 (1997).
[6] M. L. Cowan, J. H. Page, and D. A. Weitz, Phys. Rev. Lett. **85**, 453 (2000).
[7] R. Ferrara, G. Fioccho, and G. Tonna, Appl. Opt. **9**, 2517 (1970); A. Deepak and O. Vaughan, *ibid.* **17**, 374 (1978); G. Gimmestad, L. Winchester, W. Choi, and S. Lee, Opt. Lett. **7**, 471 (1982); R. Wei, Y. Tian, and Q. Lu, J. Acoust. Soc. Am. **81**, 1350 (1987); R. Sutherland, Y. Yee, G. Fernandez, and J. Millard, Atmos. Res. **41**, 299 (1996).
[8] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1983).
[9] L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon Press, Oxford, 1959), p. 69. We now believe that the value of 4π quoted in Ref. [1] should actually be 5.2π to account for the finite normal fluid viscosity.
[10] P.-A. Lemieux, M. U. Vera, and D. J. Durian, Phys. Rev. E **57**, 4498 (1998).
[11] The droplet size observed in this experiment is somewhat larger than that at the same frequency in Ref. [1] , where an average size of about 7 μm was measured. The reason for this difference is unknown, but in the present cell the droplet-emitting helium surface is confined to the area of the rectangular box rather than extending to the edges of the cylindrical cell, and also the drive levels (and hence the fog density) are considerably higher than in the earlier experiment. The drop diameters appear to be fairly uniform in the microscope photographs, with no more than a 5–10% variation in size. However, as already noted, we are limited by the 4 μm resolution of the microscope.