

(1961).

¹⁰B. Ya'akobi, E. V. George, G. Bekefi, and R. J. Hawryluk, *J. Phys. B* **5**, 1017 (1972).

¹¹Why this quantity should be used for $\langle E_{1f}^2 \rangle$ of Eq. (1) is a nontrivial question. We do not preclude the possibility of a Stark effect stronger than that treated by Baranger and Mozer. In any case, neither the ion-wave field itself nor the equivalent electric field of the beat-frequency ponderomotive force is large enough to cause

the observed effect.

¹²The inhomogeneous-plasma threshold is higher in this case. Thresholds and their meanings have been summarized by F. F. Chen, in *Laser Interaction and Related Plasma Phenomena*, edited by H. J. Schwarz and H. Hora (Plenum, New York, 1974), Vol. 3A, p. 291.

¹³G. Bekefi, *Radiation Processes in Plasmas* (Wiley, New York, 1966), p. 258.

Brownian Motion near Hydrodynamic-Regime Transitions

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The diffusion coefficient of suspended Brownian particles diverges near a hydrodynamic-regime transition point.

Pretransitional phenomena in nonequilibrium systems evolving towards an instability point bear a striking resemblance to those occurring near equilibrium phase transitions.¹ One of the most interesting analogies is the amplification of the thermal fluctuations near the transition point. In particular the enhancement of the fluid velocity fluctuations induces an increase of the diffusion coefficient of Brownian particles suspended in the fluid. This was shown for the particular case of the convective instability² and is seen here to be a general feature of hydrodynamic-regime transitions.

The system considered is infinite in the z direction and finite in the x and y directions; then the fluctuating fluid velocity field $\tilde{v}(\vec{r}, t)$ can be written as

$$v_j(\vec{r}, t) = \sum_n \int dk v_j(n, k, t) e^{ikx} f_n(x, y), \quad j = x, y, z, \quad (1)$$

where the expansion functions $f_n(x, y)$ are assumed to form a complete set and satisfy the boundary conditions. Near a hydrodynamic instability point, there is a branch of modes—say those with $n = 1$ —which become unstable; their corresponding damping factor, $\lambda(1, k)$, can in general be written as³

$$\lambda(1, k) = \alpha\epsilon + \beta(k - k_c)^2. \quad (2)$$

Here α and β are merely expansion coefficients and $\epsilon = (R_c - R)/R_c$. R is the parameter characterizing the nonequilibrium constraints on the system and R_c denotes its critical value,⁴ i.e., when $\epsilon \rightarrow 0$, the modes with wave number k_c become unstable. Furthermore, in view of the analogy between regime transitions and phase transitions, $\lambda(1, k)$ can be cast into the form of an Ornstein-Zernike-type expression by rewriting Eq. (2) as

$$\lambda(1, k) = \alpha\epsilon [1 + \xi^2(k - k_c)^2], \quad (3)$$

where ξ denotes the correlation length, with $\xi \propto d\epsilon^{-1/2}$, d being the characteristic dimension of the system. On the other hand, from the stochastic linear hydrodynamic equations describing the system, the correlation function of the fluid velocity fluctuations of the unstable branch is given by⁵

$$\begin{aligned} \langle v_j^*(1, k, 0) v_j(1, k', t) \rangle \\ = \frac{C_j(k)}{\lambda(1, k)} e^{-\lambda(1, k)t} \delta(k - k'), \end{aligned} \quad (4)$$

where the explicit form of the quantity $C_j(k)$ depends on the specific problem considered.⁶ It is clear from Eqs. (3) and (4) that when $\epsilon \rightarrow 0$ the fluctuations of the modes with $n = 1$ and with wave number $k \simeq k_c$ are amplified and decay very slow-

ly in time (critical slowing down).

Now it follows from Faxén's theorem⁷ that the diffusion coefficient of a spherical Brownian particle can be written in the hydrodynamic limit as²

$$D_j = \int_0^\infty dt \langle \bar{v}_j^s(0) \bar{v}_j^s(t) \rangle, \quad (5)$$

where \bar{v}^s denotes the average of the fluctuating fluid velocity field over the surface of the Brownian particle. Here we are interested in the effect of the enhancement of the fluid velocity fluctuations on the diffusion coefficient. However, only the fluctuations of modes with $n=1$ are amplified. Therefore restriction will be made to the analysis of the contribution of these modes to the integrand of Eq. (5); this contribution will be labeled D^{CRIT} . In general the radius a of the Brownian particle is much smaller than d , and the function $f_1(x, y)$ varies slowly over the surface of the sphere. Therefore D_j^{CRIT} , as obtained from Eqs. (1) and (5), reads to a good approximation

$$D_j^{\text{CRIT}} \simeq f_1^2(X, Y) \int_0^\infty dt \int dk \int dk' \left(\frac{\sin ka}{ka} \right) \left(\frac{\sin k'a}{k'a} \right) \langle v_j^*(1, k, 0) v_j(1, k', t) \rangle, \quad (6)$$

where X and Y are the coordinates of the center of mass of the Brownian particle in the (x, y) plane. Substitution of (4) into (6) yields

$$D_j^{\text{CRIT}} \propto \int dk \left(\frac{\sin ka}{ka} \right)^2 \frac{C_j(k)}{\lambda^2(1, k)}. \quad (7)$$

Near the instability point where ϵ becomes small, it follows from Eq. (3) that the integrand on the right-hand side of Eq. (7) is sharply peaked around $k=k_c$. Consequently, to a good approximation, we may set the functions $(\sin ka)/ka$ and $C_j(k)$ equal to their values at $k=k_c$. In addition, since k_c is of the order of d^{-1} , and because $a \ll d$, one has $(\sin k_c a)/k_c a \simeq 1$. Introducing the scaled variable $\kappa = \xi k$, it then follows from Eqs. (3) and (7) that

$$D_j^{\text{CRIT}} \propto \frac{C_j(k_c)}{\alpha^2 \epsilon^2 \xi} \int_{-\infty}^{+\infty} d\kappa [1 + (\kappa - \kappa_c)^2]^{-2}. \quad (8)$$

Now using the fluctuation-dissipation theorem, one can show⁵ that $C_j(k)/\alpha^2 \propto k_B T/\gamma$, where γ is a transport coefficient.⁸ It then follows straightforwardly from Eq. (8) that

$$D_j^{\text{CRIT}} \propto \frac{k_B T}{\gamma d} \epsilon^{-3/2}. \quad (9)$$

This result shows that D^{CRIT} diverges as $\epsilon^{-3/2}$ when the system approaches the transition point. The other modes (with $n \geq 2$) are so little affected by the nonequilibrium constraints that their contribution yields essentially the regular Stokes-Einstein diffusion coefficient.

In conclusion we have presented a calculation of the bare diffusion coefficient (i.e., as valid in the strict hydrodynamic limit: $\omega \rightarrow 0$, $k \rightarrow 0$) for a suspension of Brownian particles in a system near a hydrodynamic-regime transition. We have shown that this diffusion coefficient contains a critical contribution which within the frame-

work of linear hydrodynamic theory diverges at the transition point. This divergence arises from the amplification of the fluid velocity fluctuations. Note that these fluctuations lead, at finite k and ω , to a renormalization of the diffusion coefficient.⁹ This effect was not considered in the present work and will be treated in a subsequent paper.

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¹See, e.g., the recent review by H. Haken, *Rev. Mod. Phys.* **47**, 67 (1975), and the references contained therein.

²H. N. W. Lekkerkerker, *Physica (Utrecht)* **80A**, 415 (1975).

³H. Haken, *Z. Phys.* **B21**, 105 (1975).

⁴For instance, in the Bénard problem, R is the Rayleigh number; for Couette flow, it is the Taylor number; for Poiseuille flow, R is the Reynolds number.

⁵H. N. W. Lekkerkerker and J. P. Boon, in *Fluctuations, Instabilities, and Phase Transitions*, edited by T. Riste (Plenum, New York, 1975), p. 205.

⁶E.g., in the case of the convective instability, $C_j(k) \propto k_B T \rho^{-1} \nu k^2$.

⁷R. Zwanzig, *J. Res. Nat. Bur. Stand., Sect. B* **68**, 143 (1964); D. Bedeaux and P. Mazur, *Physica (Utrecht)* **76**, 247 (1974).

⁸In the case of the Bénard instability γ is merely the shear viscosity. However in general γ can be a more complicated linear combination of transport coefficients.

⁹R. Zwanzig, in *Statistical Mechanics: New Concepts, New Problems, New Applications*, edited by S. A. Rice, K. F. Freed, and J. C. Light (The Univ. of Chicago Press, Chicago, 1972); D. Bedeaux and P. Mazur, *Physica (Utrecht)* **73**, 431 (1974).