nonlinear event occurs. The slopes of the dashed lines drawn through the data are again exactly inversely proportional to the square of the H_0 field. There is a slight dependence on the H_1 field of the frequency at which the large jumps occur. The larger H_1, the higher the frequency at which the jump occurs.

The role that the gradient in magnetic field plays in these nonlinear phenomena cannot be unambiguously determined from the measurements presented here. However, an estimate of the importance of the gradient field may be made by comparing the experiments reported here with those of Ref. 1 in which the gradient was nearly 100 times smaller. Assuming that the nonlinear events depend primarily upon the magnitude of $\Delta M_B/M_B$ as in the ferromagnetic resonance case, then the two experiments can be compared by using the standard relationship[12] $\Delta M_B/M_B = \gamma^2 H_1^2 \times T_1 T_2$. I find, using the value for T_1 and T_2 appropriate for the two examples of Ref. 1 in 623 Oe at a reduced temperature $T/T_c = 0.60$, that no nonlinear phenomena were observed for $\Delta M_B/M_B \approx 5\times 10^{-5}$ while for $\Delta M_B/M_B \approx 2\times 10^{-2}$ the first nonlinear event was observed. In the present work, at the lowest temperature and highest field employed, the onset of nonlinear phenomena began at $\Delta M_B/M_B = 1.1\times 10^{-2}$. Although this comparison is based on only two events, it does suggest that the only major effect of the gradient field is to change the values of T_1 and T_2 and it does not play a dominant role in the physics of these interesting and unexplained nonlinear phenomena.

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Ornstein-Zernike Theory of Classical Fluids at Low Density

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We show rigorously that for dilute classical systems with finite-range interactions the pair-correlation function has the form predicted by Ornstein and Zernike.

The Ornstein-Zernike theory of pair-correlation functions is a cornerstone, albeit a heuristic one, in discussion of fluids and lattice gases outside the critical region.[1,2] It predicts that the truncated pair-correlation function for a fluid $u(\vec{r},\rho)$ should behave asymptotically as

$$u(\vec{r},\rho) = A \exp(-k_1|\vec{r}|)\cos k_2|\vec{r}|/|\vec{r}|^{(d-1)/2}, \qquad (1)$$

where A, k_1, and k_2 are functions of density and temperature and d is the dimensionality. In this Letter we establish the theory rigorously both for continuum systems with finite-range potentials and for lattice systems under the same conditions, essentially for low densities and high temperatures. The latter extends results obtained by transfer-matrix techniques in this region to non-nearest-neighbor interactions in the transfer direction. Paes-Leme and Shor have obtained similar results for lattice gases independently.[3] Prior to this work, *bounds* had been obtained on the spatial behavior of correlation functions.[4] Our results should prove useful in a field-theoretic context,[5] and more generally in statistical mechanics.[6]

The outline of our approach is as follows: We use the direct correlation function $c(\vec{r},\rho)$ first defined by OZ as a solution of

$$\rho^2 c(\vec{r},\rho) = u(\vec{r},\rho) - \rho(c*u)(\vec{r},\rho). \tag{2}$$

OZ *assumed* that $c(\vec{r},\rho)$ has essentially the range, here assumed finite, of the potential. In this Letter, *rigorous* exponential bounds on the decay of $c(\vec{r},\rho)$ will be obtained by working with the graphical expansion of $c(\vec{r},\rho)$ in powers of ρ; $c(\vec{r},\rho)$ decays at least twice as fast as the bound on $u(\vec{r},\rho)$ mentioned in the previous paragraph.[4] This provides a meromorphic extension in k of $\hat{u}(k,\rho)$ through the Fourier transforms of Eq. (2). The nearest singularities are then simple poles at $\pm k_2 \pm ik_1$ [see Eq. (1)]. The essential ingredients here are that we have analyticity in ρ and expansion in terms of rooted, *connected* graphs, thus conferring a strictly finite range $r(G)$ on a graph G with n vertices; in fact, $r(G) \sim n$. It does not appear to have been noticed heretofore that combining this with analyticity enables one to control the decay of $c(\vec{r},\rho)$, and to *derive* Eq. (2) in terms of a $c(\vec{r},\rho)$ defined *a priori.*

The truncated correlation function between two particles at $\vec{0}$ and $\vec{r}$ in R^d for the canonical ensemble is given by

$$u(\vec{r},\rho) = \sum_{n=0}^{\infty} u_{n+2}(\vec{r})\rho^{n+2}, \tag{3}$$

where ρ is the density and the $u_n(\vec{r})$ are defined for a pair potential $\varphi(\vec{r})$ by

$$u_n(\vec{r}) = \sum_G \frac{1}{S_G}\int \prod_{e\in G} f(\vec{r}_e), \tag{4}$$

where G are connected simple graphs with n vertices, none being articulation points,[7,8] rooted at $\vec{0}$ and $\vec{r}$ having edges of length $\vec{r}_e$, with weight $f(\vec{r}_e)$ defined by

$$f(\vec{r}) = \exp[-\beta\varphi(\vec{r})] - 1, \tag{5}$$

with $\beta = 1/k_B T$ for temperature T. S_G is the symmetry number of the graph G.[7] The integral is performed over the coordinates of all the unrooted vertices.

The series (3) converges[9] in $\mathfrak{D} = \{\rho : |\rho| < \rho_0\}$ where

$$\rho_0 = (3 - 2^{3/2})/C(\beta)\exp(2\beta B + 1), \tag{6}$$

with

$$C(\beta) = \int |f(\vec{r})|\, d^3r, \tag{7}$$

and $B \geq 0$ is such that

$$\varphi(\vec{r}) \geq -2B. \tag{8}$$

Let functions $c_n(\vec{r})$ be defined by

$$c_n(\vec{r}) = \sum_G \frac{1}{S_G}\int \prod_{e\in G} f(\vec{r}_e), \tag{9}$$

where the graphs G have the additional restriction over those in (4) that they shall be *nodeless*.[8] Note that $c_2(\vec{r}) = u_2(\vec{r})$ and that for $n \geq 1$ (note that $c_0 = c_1 = 0$),

$$u_{n+2} = c_{n+2} + \sum_{l=1}^{n} c_{l+1} * u_{n+2-l}, \tag{10}$$

where $*$ denotes the convolution in R^d. The essential feature of the $c_n(\vec{r})$ is that they have *half the range* of the $u_n(\vec{r})$.

Lemma 1: Let the potential $\varphi(\vec{r})$ be spherically symmetrical and have range b. Then

$$u_n(\vec{r}) = 0, \quad |\vec{r}| > nb; \tag{11}$$

but

$$c_n(\vec{r}) = 0, \quad |\vec{r}| > nb/2.$$

The idea of the proof[10] is that the longest-range graph in Eq. (9) has two vertex-disjoint chains, between the roots at $\vec{0}$ and $\vec{r}$ (each having an equal number of vertices). [Note that $f(r) = 0$ if $|r| > b$.] We shall use this lemma to make assertions about the domain of analyticity in $\vec{k}$ of the Fourier transforms $\hat{u}(k,\rho)$ and $\hat{c}(k,\rho)$ where for any $f \in L^2(R^d)$ we have

$$\hat{f}(\vec{k}) = \int e^{i\vec{k}\cdot\vec{r}} f(r)\, d^3r. \tag{12}$$

Then we obtain the following result:

$$\hat{u}(\vec{k},\rho) = \sum_{n=0}^{\infty} \rho^{n+2}\hat{u}_{n+2}(\vec{k}), \tag{13}$$

and both $\hat{u}_n$ and $\hat{u}$ lie in $L_2 \cap L_\infty$. The connection between the $c_n(\vec{r})$ and a direct correlation function is established as follows: Let $\vec{k} \in R^d$ and *define* $\hat{c}$ by

$$\hat{c}(\vec{k},\rho) = \hat{u}(\vec{k},\rho)/[\rho^2 + \rho\hat{u}(\vec{k},\rho)]. \tag{14}$$

This function is analytic in ρ in the intersection of $\mathfrak{D}$ and

$$\{\rho : \rho \neq -\hat{u}(\vec{k},\rho)\ \forall \vec{k} \in R^d\}.$$

This set is, in fact, $\mathfrak{D}$ itself. Furthermore, $\hat{c}$ lies in $L_2 \cap L_\infty$ for ρ in an compact subset of $\mathfrak{D}$. It has the expansion

$$\hat{c}(\vec{k},\rho) = \sum_{n=0}^{\infty} \rho^n \hat{c}_{n+2}(\vec{k}), \tag{15}$$

where $\hat{c}_n(\vec{k})$ are the Fourier transforms of the $c_n(\vec{r})$ in Eq. (9); this converges for ρ in $\mathfrak{D}$ *uniformly* in $\vec{k} \in R^d$. Finally, we recapture the usual definition, given by Eq. (2), of the direct correlation function.[11]

For spherically symmetrical potentials, $\hat{u}(\vec{k})$ and $\hat{c}(\vec{k})$ depend only on $k=|\vec{k}|$. From Lemma 1, the convergence of the fugacity expansion, and Cauchy inequalities, we obtain the following.

Lemma 2: Let $\rho \in R \cap \mathfrak{D}$; then $\hat{u}(k,\rho)$ is analytic in $|\mathrm{Im}k| < k_0$ whereas $\hat{c}(k,\rho)$ is analytic in $|\mathrm{Im}k| < 2k_0$ with

$$k_0 = b^{-1}\ln(\rho_0/\rho). \tag{16}$$

Lemma 3: The equation

$$\hat{u}(\vec{k},\rho) = \rho^2\hat{c}(\vec{k},\rho)/[1-\rho\hat{c}(\vec{k},\rho)] \tag{17}$$

supplies a *mermorphic extension of* $\hat{u}$ which has (i) poles in $k_0 < |\mathrm{Im}k| < 2k_0$ (as well as $|\mathrm{Im}k| > 2k_0$) at zeros of $1-\rho\hat{c}(k,\rho)$; and (ii) branch points (*ceteris paribus*) at the branch points (if any) of $\hat{c}(k,\rho)$ in $|\mathrm{Im}k| \geq 2k_0$. Singularities at the poles of $\hat{c}(k,\rho)$, if any, are removable. From Eqs. (5), (9), (12), and (15) we have

$$\hat{c}(k,\rho) = \hat{f}(k) + \hat{d}(k,\rho), \tag{18}$$

where $\hat{d}(k,\rho)$ has a uniform bound for k in any compact subset of the strip $|\mathrm{Im}k| < 2k_0$. Given any zero of $1-\rho\hat{f}(k)$ at k_1, say, we can construct a box B with k_1 in its interior such that on its boundary $|\rho\hat{d}(k,\rho)/[1-\rho\hat{f}(k)]| < 1$. Then by Rouché's theorem $1-\rho\hat{c}(k,\rho)$ also has a zero inside B. Finally, as $\rho \to 0$, B can be made arbitrarily small, but the precise details of its shape depend on the particular potential chosen.

In the remainder of this Letter, we summarize the results found in special cases, for $d=3$.

Results for gases with hard cores. 1. *Hard core*.—Here we have $f(\vec{r}) = -1$ if $|\vec{r}| < b$; $f(\vec{r}) = 0$ if $|r| > b$; and $C(\beta) = 4\pi b^3/3$. By means of a Rouché argument, there are simple poles of $\hat{u}$ close to the points given by

$$kb/2 = i(z - a_n^{\pm}), \tag{19}$$

where

$$a_n^{\pm} = -\ln\alpha + (2n \pm \tfrac{1}{2})\pi i, \quad \alpha = 2^{3/2}\ln(3/4\pi\rho b^3), \tag{20}$$

and

$$\exp z = a_n^{\pm} - z \tag{21}$$

with the condition that $0 < \mathrm{Im}z < \pi$ if $\mathrm{Im}a_n^{\pm} > 0$ and that $-\pi < |\mathrm{Im}z| < 0$ if $\mathrm{Im}a_n^{\pm} < 0$. This result is valid asymptotically for large α. Then the nearest poles to the real axis are

$$kb = \pm\pi[1 + O((\ln\alpha)^{-1})] \pm [\ln\alpha + O(\ln(\ln\alpha))], \tag{22}$$

which gives an oscillatory $u(\vec{r})$ of the form (1) with $k_1 b = \ln\alpha + O(\ln(\ln\alpha))$ and $k_2 b = \pi[1 + O((\ln\alpha)^{-1})]$.

2. *Hard core plus attractive well*.—In this case we show, within the domain of validity of the theory, that for T small enough, the decay of $u_2(\vec{r})$ is *asymptotically monotonic* and of Ornstein-Zernike type, as $|\vec{r}| \to \infty$, but that for T large enough we recapture the oscillatory decay characteristic of the hard core. This should be compared with the exact results[12] for one-dimensional systems.

3. *Lattice gases*.—The whole of our analysis can be carried through for lattice gases; sums over Z^d replace integrals over space. Then

$$u(\vec{r},\rho) = \int_0^{2\pi} \cdots \int d(\omega)_d\, e^{i\vec{\omega}\cdot\vec{r}}\rho^2\hat{c}(\omega,\rho)/[1-\rho\hat{c}(\omega,\rho)] \tag{23}$$

with $(\omega)_d \equiv (\omega_1, \ldots, \omega_d)$ is valid for ρ in a domain similar to (4). This expression may be placed in a more transparent form by performing a single contour integral over ω_d, say. The singularities of the integrand in ω_d, when ω_i (with $i=1,\ldots,d-1$) are all real, are localized as before in terms of those of $1-\rho f(\vec{k})=0$ by means of a Rouché argument. A typical result of this procedure in the cubic Ising ferromagnet[13] with nearest-neighbor interactions, for which

$$u(r,\rho) = \int_0^{2\pi} \cdots \int d(\omega)_{d-1} \exp[-|r_d|\gamma((\omega)_{d-1}) + i\vec{\omega}\cdot\vec{r}]/\sinh[\gamma((\omega)_{d-1})] + O(\exp[-2|r_d|\gamma(0)]), \tag{24}$$

where

$$\cosh[\gamma((\omega)_d)] = (1+\rho)/\rho(e^{2k}-1) - \sum_{i=1}^{d}\cos\omega_i. \tag{25}$$

Equation (25) is a typical consequence of "one-particle" states in transfer-matrix problems.[2] Its form should be compared particularly with the exact results for the $d=2$ Ising ferromagnet.[14]

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[8]See Ref. 7, p. II-204. We repeat the definitions for the reader's convenience here. *Articulation point:* Let G be rooted. A point $\alpha \in V(G)$ is an articulation point $\Longleftrightarrow$ if α is removed, then the component of G of which it is part separates into at least two parts, at least one g which contains no roots. *Node:* a vertex γ is a node of a graph G $\Longleftrightarrow G$ has at least two roots, γ is not a root, and any root-connecting chain must pass through γ.

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Buoyancy-Driven Instability in a Dilute Solution of Macromolecules

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We have performed optical measurements on a hydrodynamic instability occurring in a dilute polymer solution characterized by a negative value of the thermal diffusion ratio and heated from above. We determine the threshold condition and the behavior of the convected mass flow above threshold. Qualitative evidence of critical slowing down is also reported together with a discussion on the convective-mode wavelength.

As is well known, a horizontal layer of a single-component fluid heated from below will remain stationary as long as the temperature gradient across it does not exceed the critical value corresponding to $R = 1708$, where R is the Rayleigh number. When this value is exceeded, a convective instability sets in, and both temperature and fluid velocity become space-dependent variables according to a well-defined spatial structure. This is the so called Rayleigh-Bénard instability (RBI).[1] Recently a new type of convective instability occurring in two-component systems has been described in the literature [Soret-driven instability (SDI)[2,3]]. The instability can be generated both by heating from below or from above depending on the sign of the thermal diffu-