in the experiment with ethyl alcohol ($t_p/\tau = 2.0$) is drawn through the experimental points. The calculated curve fits our data quite well at $t_D = 0$, at the position of the maximum, and also for larger values of t_D. The comparison with theory confirms the dephasing time of $\tau = 4$ psec. It is interesting to compare this value of τ with measurements of the spontaneous Raman linewidth. Earlier data suggested a value of $\Delta\omega/c \cong 10\ \mathrm{cm}^{-1}$. More recently the isotope structure of the Raman emission in CCl_4 has been resolved and $\Delta\omega/c \cong 1.5\ \mathrm{cm}^{-1}$ was reported for the most prominent Raman line.[13] Quite obviously our experimental dephasing time of $\tau = 4.0 \pm 0.5$ psec corresponds to the width of the individual isotope components of the Raman line.

Finally, we wish to emphasize once more the importance of knowing the shape (the wings) of the laser pulse. The determination of a dephasing time of $\tau = 4$ psec with a probe pulse width of $t_p = 8$ psec requires rapidly rising (Gaussian) pulses. Calculations show, for example, that with a Lorentzian pulse of the same duration ($t_p = 8$ psec), only τ values larger than $\simeq 80$ psec can be measured.

Investigations of the type discussed here are readily extended to other media, to compressed gases and especially to solids where TO phonons (e.g., in diamond[14]) and internal vibrations (e.g.,[9] in $CaCO_3$) have been excited by stimulated Raman scattering. Using two incident pumping beams with the proper frequency difference, it will be possible to excite molecular and lattice vibrations with smaller gain factors.

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Lattice Gas with Short-Range Pair Interactions and a Singular Coexistence-Curve Diameter*

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A simple lattice gas, of the familiar kind with only short-range pair interactions, is proved to have a coexistence curve whose diameter is singular at the critical point. Particle-hole symmetry is violated only because the range of interaction energies a molecule can experience depends on which of two nonequivalent sets its lattice site belongs to.

Theoretical analysis has cast doubt on the validity of the law of the rectilinear diameter, which asserts that $d\rho_d(T)/dT$ is asymptotically constant at the liquid-vapor critical point $\{\rho_d(T) = \frac{1}{2}[\rho_V(T) + \rho_L(T)]\}$. Three models have now been described with diameters reflecting the constant-volume specific-heat singularity[1-3]:

$$d\rho_d(T)/dT \sim \mathrm{const} \times c_v, \quad T \to T_c. \tag{1}$$

One can argue that each of these models is atypical. That of Hemmer and Stell[1] is one dimensional, requires forces of infinite range, and yields the singular diameter only for special values of the interaction parameters that result in the coalescence of two classical critical points. The penetrable-sphere model[2] has a short-range interaction which requires, however, 2-, 3-, $\cdots$, N-body interactions for its representation. The bar model[3] interaction is a sum of short-range four-body potentials.

Although models with short-range *pair* interactions are perhaps as much hallowed by tradition as required by physical reality, the presence of a singular diameter in such a model would throw into sharper focus the problems raised by the existence of such singularities. It is therefore important to note that there are, in fact, many lattice-gas models, interacting only through perfectly ordinary *short-range pair* potentials, whose diameters carry the specific-heat singularity, as in Eq. (1). We describe here an especially simple case.

Take an ordinary two-dimensional lattice gas on a square lattice, with only nearest-neighbor attractive coupling of strength $\eta\epsilon$. (We use the dimensionless parameter η to measure the strength of the coupling.) Number the rows and columns of lattice sites $1, 2, 3, \cdots$. We now *remove* (or forbid occupation of) those sites whose row and column numbers are both even (even-even sites). Finally, it is instructive (but not necessary—λ may be zero) to introduce an additional attractive coupling of strength $\lambda\epsilon$ between molecules occupying nearest-neighbor sites in the sublattice of odd-odd sites.

Note that this lattice gas does not have particle-hole symmetry, because it has two kinds of sites: Odd-odd sites have coordination number 4, and odd-even sites have coordination number 2. Only when $\eta = -2\lambda$ does a particle-hole transformation map the model onto itself; when $\eta \neq -2\lambda$, the particle-hole transformation leads to an additional nonuniform external field.

The model can also be described as one in which the ordinary lattice gas with attractive nearest-neighbor coupling $\lambda\epsilon$ that occupies the odd-odd sites has been augmented by a set of sites on nearest-neighbor bonds (the odd-even sites), and by the addition of an attractive interaction of strength $\eta\epsilon$ between a molecule on an odd-even site and either of its two neighbors on odd-odd sites. From this point of view, the model is nothing more than a lattice-gas version of the decorated Ising models discussed by Fisher over ten years ago.[4] Following Fisher's analysis, we can easily express the grand partition function of the *decorated* lattice gas, $\Xi(\zeta, K)$, in terms of the grand partition function $\overline{\Xi}(\overline{\zeta}, \overline{K})$ of the *ordinary* nearest-neighbor square lattice gas, with attractive nearest-neighbor coupling of strength ϵ (we use dimensionless variables $\zeta = \mu/k_B T$, $\overline{\zeta} = \overline{\mu}/k_B\overline{T}$, $K = \epsilon/k_B T$, $\overline{K} = -\epsilon/k_B\overline{T}$):

$$\Xi(\zeta, K) = (1 + e^{\zeta})^{2\overline{N}}\overline{\Xi}(\overline{\zeta}, \overline{K}), \tag{2}$$

where $\overline{N}$ is the number of sites in the ordinary lattice gas (so that the decorated lattice gas has $3\overline{N}$ sites) and

$$\overline{\zeta}(\zeta, K) = \zeta + 4\ln\left[\frac{1 + e^{\zeta + \eta K}}{1 + e^{\zeta}}\right],$$
$$\overline{K}(\zeta, K) = \lambda K + \ln\left[\frac{(1 + e^{\zeta})(1 + e^{\zeta + 2\eta K})}{(1 + e^{\zeta + \eta K})^2}\right]. \tag{3}$$

Below the critical temperature, the ordinary lattice-gas density is discontinuous across the coexistence line $\overline{\zeta}(\overline{K}) = -2\overline{K}$. The coexistence line for the decorated lattice gas is therefore given by

$$\zeta(K) + 2\lambda K + 2\ln\left[\frac{1 + e^{\zeta(K) + 2\eta K}}{1 + e^{\zeta(K)}}\right] = 0. \tag{4}$$

By calculating from (2) the mean occupation number per site in the decorated lattice gas, on opposite sides of the coexistence line, one finds that

$$\rho_d(K) = \tfrac{2}{3}[1 + e^{-\zeta(K)}]^{-1} + \tfrac{1}{3}\overline{\rho}_d(\overline{K}(K))[(\partial\overline{\zeta}/\partial\zeta)_K]_{\zeta=\zeta(K)} + \tfrac{1}{3}\overline{w}_d(\overline{K}(K))[(\partial\overline{K}/\partial\zeta)_K]_{\zeta=\zeta(K)}, \tag{5}$$

where $\overline{K}(K)$ is defined by

$$\overline{K}(K) = \overline{K}(\zeta(K), K) \tag{6}$$

and $-\epsilon\overline{w}_d = \frac{1}{2}(\overline{u}_L + \overline{u}_V)$, where $\overline{u}$ is the mean energy per site of the ordinary lattice gas.

The particle-hole symmetry of the ordinary lattice gas establishes that $\overline{\rho}_d = \frac{1}{2}$ and $\overline{w}_d(\overline{K}) = \overline{w}(\overline{\rho}_c, \overline{K})$. The $\overline{K}$ derivative of $\overline{w}_d$ therefore diverges logarithmically at the critical point, and so will the K derivative of ρ_d provided that this is the only divergence that differentiation with respect to K introduces on the right-hand side of Eq. (5), and provided that $(\partial\overline{K}/\partial\zeta)_K$ does not vanish at the critical point.

The first proviso is readily verified (i) from the fact that the first and second derivatives of the mappings (3) are all bounded, (ii) from the boundedness of $d\zeta(K)/dK$ that (4) implies when the coupling is attractive, and (iii) from the boundedness of $d\bar{K}(K)/dK$ that follows from (i), (ii), and Eq. (6). That $(\partial\bar{K}/\partial\zeta)_K$ does not vanish at the critical point follows from the mapping (3) and the form of the coexistence line (4) *except* in the case $\eta=0$ (where the odd-even sites are decoupled from the odd-odd), *and* in the case $\eta=-2\lambda$ (where particle-hole symmetry is restored).[5]

Thus in the decorated lattice gas, the *lack* of a singular diameter clearly emerges as an accidental consequence of particle-hole symmetry, which causes the coefficient of the singularity to vanish. This lends considerable support to the conjecture[1,2] that the rectilinear diameter of the conventional lattice gas is, in fact, the atypical case, found only in models with the artificial particle-hole symmetry. To test the conjecture further, it would be interesting to know, for example, whether a singular diameter would emerge if, instead of prohibiting occupation of even-even sites, we had merely changed the strength of their coupling (thereby destroying particle-hole symmetry). Or, would a lattice gas in which the horizontal coupling constant alternated in value from row to row have a singular diameter? These are hard questions to answer, but it may well be that the remaining peculiarities of models known to have singular diameters reflect only our inability to answer more than a few hard questions, rather than any lingering shreds of the law of the rectilinear diameter.

Indeed, the somewhat stronger conjecture that (1) holds whenever there is no symmetry to prevent it has considerable explanatory power. Thus continuum systems (which *ipso facto* lack particle-hole symmetry), with classical critical points, have finite heat capacities at T_c and, *as implied by (1), non*singular diameters. The singular diameter discovered in Ref. 1 has its peculiar setting because it occurs in a model with infinite-range forces and a classical critical point. Hemmer and Stell must therefore make the short-range part of their interaction complex enough to yield a new kind of classical critical point, possessing a *singular* specific heat, before (1) implies a *singular* diameter.

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