>1 over most of this temperature range. However, except at very small angles of incidence large k implies large K since $k > k_M$, and therefore, through the integral of $G(K)$, phonons with $k\xi < 1$ are emphasized.

In conclusion, I have shown the following: (1) When surface roughness is taken into account the acoustic mismatch theory is in excellent agreement with Kapitza conductance measurements on Cu-^{3}He, Cu-^{4}He, and NaF-^{4}He, at temperatures $T > 0.2$ K. (2) For large impedance mismatch the calculation is only weakly dependent upon the properties of the liquid. This explains the weak pressure dependence observed for ^{4}He and ^{3}He interfaces as well as the similarity of experimental results for ^{4}He, ^{3}He, and other low-impedance materials. (3) The two channels invoked by Wyatt and Page[14] are just h_K^{AM} and h_K^s. (4) From Eq. (5) and what followed, the frequency distribution of the scattered radiation is angle dependent and generally not the same as that of the input. This "filtering" effect can partially explain the apparent inelastic scattering observed in some heat-pulse experiments.

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Frustrated Spin-Gas Model for Doubly Reentrant Liquid Crystals

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A mechanism, based on the competition between short-range positional order and long-range antiferroelectric order, is presented for reentrant liquid crystals. Antiferroelectric frustration is inherent to the triangular close packing of layers normal to the molecular axis, but is lifted at intermediate temperatures by positional disorder. Phase diagrams evaluated by using prefacing transformations include the doubly reentrant case exhibiting, as temperature is lowered, nematic, bilayer smectic, reentrant nematic, and monolayer smectic phases.

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The reappearance[1-3] in certain liquid crystals of a nematic (less ordered) phase as temperature is lowered from a smectic (more ordered) phase is an *a priori* unexpected phenomenon which requires microscopic explanation. Although the importance of dipolar interactions in such systems has been empirically established and stressed,[1] a microscopic mechanism explaining the occurrence or nonoccurrence of a macroscopic phase through many-body correlations, and making possible quantitative calculations of phase diagrams and other observable properties, has been lacking. We propose such a mechanism based on the competition between short-range positional order and long-range antiferroelectric order, and on the mismatch between the optimal dipolar permeation length and the atomic length scale. Starting from a particulate picture with microscopic potentials, calculations are performed by using a special prefacing[4] transformation which could

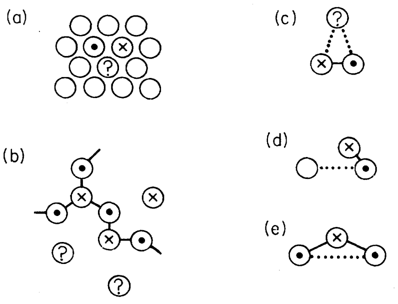

FIG. 1. Positional configurations of a layer normal to the molecule axes. Molecular dipoles which are pointing into or out of the figure, or which are frustrated, are respectively indicated by crosses, closed circles, or question marks. Strong (weak) bonds are shown with full (dotted) lines. Permeation fluctuations, not seen here, similarly relieve frustration.

also be useful for other fluid or glassy systems where local distortions are important. Our calculated phase diagrams include reentrant as well as doubly reentrant cases.

We begin with an intuitive explanation of the mechanism, to be made quantitative further on. Consider the two-dimensional array of molecules intersected by a plane normal to the average molecular axis, in either the smectic or the nematic phase. As in the experimental systems with reentrant phase diagrams, the molecules are taken to have an aliphatic tail and a polar head along the molecular axis. First, for the sake of argument, consider the limit of complete positional order [Fig. 1(a)]. The close-packing arrangement in two dimensions is triangular. The interaction between the dipoles is antiferroelectric, but an antiferroelectric long-range order cannot be supported[5] because each elementary triangle of the array is frustrated.[6] However, we are actually dealing with a liquid, with positional disorder. If the local distribution is like Fig. 1(e) with one weak and two strong bonds, frustration will be lifted, and two-dimensional antiferroelectric order can propagate across the unit. On the other hand, if the elementary triangle is like Fig. 1(c), with one strong and two weak bonds, it will still be frustrated, and at most a one-dimensional antiferroelectric correlation can propagate across, which is not sufficient for antiferroelectric ordering. Actually, in any given snapshot of the layer, each elementary triangle is between these two cases, with one strong, one intermediate, and one weak bond [Fig. 1(d)]. Whether the intermediate bond is closer to the strong one or to the weak one determines the ordering character of the unit. [This is made quantitative in Eq. (2).] If there are enough of the former type of units, they will percolate across the system and form an infinite network ("polymer") of positionally disordered, but antiferroelectrically ordered molecules [Fig. 1(b)]. Each layer, consecutively along the z direction, will have its own network. These networks will not pass through each other because this would involve disrupting infinitely many strong bonds. The result is the density modulation along the z axis, namely the smectic phase. Note first that, even in the presence of the networks (which indeed should have zero weight close to the nematic phase), many antiferroelectrically ordered but finite clusters ("n-mers") slide up and down the z axis and give the constant background to the smectic modulation. Second, although the percolating network must be sustained for the smectic modulation, individual molecules join it and leave it as time progresses. Finally, as the system is further cooled, it is not unreasonable that local positional order will set in, turning on frustration, eventually destroying the network, and reentering the nematic phase.

Calculations are performed with molecules interacting via the potential

$$V(\vec{r}_1,\hat{s}_1,\vec{r}_2,\hat{s}_2) = [A\hat{s}_1\cdot\hat{s}_2 - 3B(\hat{s}_1\cdot\hat{r}_{12})(\hat{s}_2\cdot\hat{r}_{12})]/|\vec{r}_{12}|^3, \qquad (1)$$

where $\vec{r}_i$ is the position of molecule i, $\hat{s}_i = \pm 1$ is the orientation of its dipole along the z axis, $\vec{r}_{12} \equiv \vec{r}_1 - \vec{r}_2$, and $\hat{r}_{12} \equiv \vec{r}_{12}/|\vec{r}_{12}|$. For purely dipolar interaction, $B/A = 1$. Tail-tail interactions are simply taken into account by letting this ratio be less or greater than one, respectively, corresponding to tail-tail interaction dominated by steric hindrance or van der Waals attraction. More complicated potentials can and have been used within our method, without qualitatively affecting the results.

The simplest treatment of (1) is by a prefacing transformation, which restructures a given system onto another more readily solvable one via partially carrying out the partition sum.[4] First, we discuss the model onto which this mapping is done. For an Ising ($s_i = \pm 1$) antiferromagnet on a uniformly distorted triangular lattice, e.g., with couplings $-\beta J_\alpha = K_1$, K_2, and K_3 along the three directions, Houtappel[7] showed that

$$H[K_\alpha] \equiv \sinh 2\bar{K}_1 \sinh 2\bar{K}_2 + \sinh 2\bar{K}_2 \sinh 2\bar{K}_3 + \sinh 2\bar{K}_3 \sinh 2\bar{K}_1 = 1 \qquad (2)$$

is the criticality condition, where $\overline{K}_\alpha$ are obtained by pairwise inverting signs of K_α (i.e., relabeling $s_i \to -s_i$ every other row of spins) until the two largest magnitude couplings are positive. The ordered (disordered) phase occurs at $H[\overline{K}_\alpha] > 1$ (< 1). Further, the exact position-space renormalization group of Hilhorst, Schick, and van Leeuwen[8] shows that, when the Houtappel condition is locally satisfied, $H[K_\alpha(\vec{r})] = 1$, it remains so under rescaling. The latter is, to our knowledge, the first exact result of its kind for a nontrivial system with quenched bond disorder. It requires a quenched bond distribution whose continuum limit can be taken, namely $K_\alpha(\vec{r})$ varying smoothly with $\vec{r}$ on the scale of lattice spacing. We claim that the phase boundary of such a system is a very good approximation for that of a system with annealed bond disorder, as in the liquid-crystal problem. Thus, when the Houtappel condition is satisfied, on the average, by the K_α of the elementary triangles discussed earlier, a good approximation to the phase boundary should be obtained.

The prefacing transformation is effected in a finite-cluster approximation,[9] by considering three molecules of the layer defined above and summing over the positional degrees of freedom to obtain effective spin interactions:

$$C \exp(K_S s_1 s_2 + K_I s_2 s_3 + K_W s_3 s_1) = \int (dr) \exp[-\beta V_S(\vec{r}_1, s_1, \vec{r}_2, s_2) - \beta V_I(\vec{r}_2, s_2, \vec{r}_3, s_3) - \beta V_W(\vec{r}_3, s_3, \vec{r}_1, s_1)]. \quad (3)$$

The positional degrees of freedom summed above should include lateral displacements from the average positions within the layer and permeation displacements out of the layer (in the z direction). The latter degrees of freedom are much softer and therefore much more important. In fact, we have performed the calculation including just the lateral displacements, a boundary condition of a fixed cage of nine other molecules, and a Lennard-Jones potential in addition to (1). The qualitative shape of the resulting phase diagram did not match the experiments, as could be expected. Thus, all results reported below include only the important permeation fluctuations. Phase diagrams which are not qualitatively different are obtained by including both types of fluctuations, at the expense of increased computational burden.

From the definition of a layer, the permeation displacements span $\pm l/2$, where l is the molecular length. Because of the unsmooth molecular architecture, there are n preferred notches along this length, separated by l/n, where n is of the order of the number of carbons in the tail. At each notch, a permeation of $\pm \delta \ll l/n$ accounts for molecular libration.

Most importantly, we have already noted that the instantaneous distortions from equilateral triangular symmetry cause the long-range antiferroelectric order. These distortions are projected in (3) by always labeling s_1 and s_2 the spins coupled with the strongest antiferroelectric interaction, itself denoted by the subscript S. The spins s_2 and s_3 are those coupled with the intermediate antiferroelectric interaction, with subscript I. The spins s_3 and s_1 are coupled with the weakest antiferroelectric interaction, W. Thus, the positions of the spins within the partial sum determine their labeling. This new prefacing transformation in fact yields the average strong-

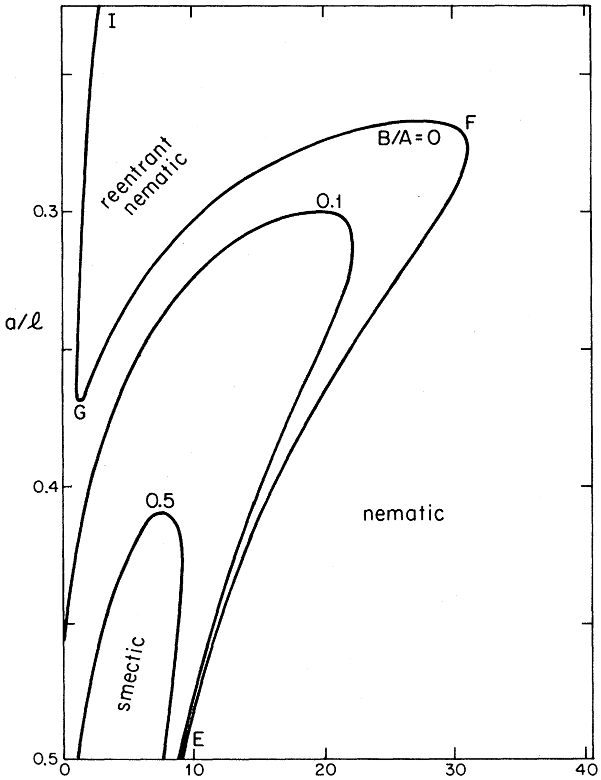

FIG. 2. Reentrant phase diagrams calculated with $n = 5$. The librational length is $\delta = 10^{-2} l/n$ for the outermost curve, but, for clarity, zero for the other curves.

est, average intermediate, and average weakest coupling, thereby reflecting the variance.

The phase diagrams are thus obtained by submitting the outcome of the prefacing transformation (3) to the Houtappel condition (2). Figure 2 shows cases with $B/A = 0$, 0.1, and 0.5. The ratio of the average lateral separation to the fixed molecular length, a/l, should monotonically decrease with increasing pressure. Along the boundary from E to F, as a decreases the transition temperature increases, because the couplings get stronger by a factor of a^{-3}. But frustration takes over at F. From the underlying calculation, reentrant boundaries such as FG are seen to be due to frustration created by neighboring molecules falling into the same notch. (The boundaries such as GI are due to frustration being relieved via molecular libration, with neighboring molecules still in the same notch. We believe that this very-low temperature, high-pressure transition is preempted by crystallization.) For experimental comparison, the compounds 8OCB and 6OCB exhibit, respectively, a reentrant phase diagram and no smectic phase at all.[2] They have a/l of about 0.26. This is consistent with Fig. 2, indicating that these compounds are in the steric hindrance regime of tail-tail interactions, with this effect being less pronounced in 6OCB. This is of course sensible, since 6OCB has a shorter (thus less bulky through floppiness) tail.

Because of the antiferroelectric spin ordering, the smectic phase in Fig. 2 is composed of interdigitated bilayers, with a z-axis repeat distance of approximately two tail lengths plus one head length, as seen experimentally.[1] However, the interactions V_α and K_α in (3) can become ferroelectric, for $B/A \gtrsim 1$ and $l/n \gtrsim a$. Thus, when $H[K_\alpha] > 1$ is satisfied with $K_W \geqslant K_I > |K_S|$, a smectic phase composed of monolayers obtains, with z-axis repeat distance of approximately one tail length plus one head length (whereas, for the bilayer phase, at least one of the two largest-magnitude K_α must be antiferroelectric). This situation is shown in Fig. 3 for $B/A = 2$. This double-reentrance phenomenon has recently been reported[10] for the compound "T_8." Our analysis thus suggests that this compound is in the attractive tail-tail interaction regime ($B/A > 1$). Finally, our approach indicates that there will always be a nematic phase between the two types of smectics, be it very narrow at low temperatures (with a zero-temperature bicritical point).

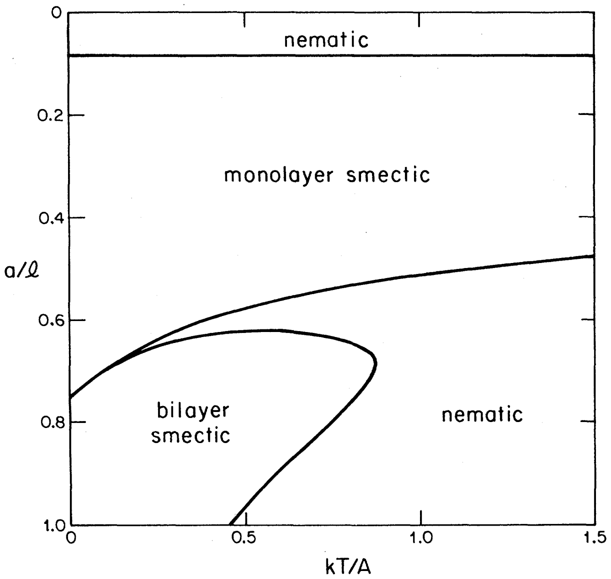

FIG. 3. Doubly reentrant phase diagram, with the zero-temperature bicritical point ($n = 7$, $B/A = 2$, $\delta = 0$).

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