Quantum Phase Transition Between a Luttinger Liquid and a Gas of Cold Molecules

K. T. Law and D. E. Feldman

Department of Physics, Brown University, Providence, Rhode Island 02912, USA

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We consider cold polar molecules confined in a helical optical lattice similar to those used in holographic microfabrication. An external electric field polarizes molecules along the axis of the helix. The large-distance intermolecular dipolar interaction is attractive but the short-scale interaction is repulsive due to geometric constraints and thus prevents collapse. The interaction strength depends on the electric field. We show that a zero-temperature second-order liquid-gas transition occurs at a critical field. It can be observed under experimentally accessible conditions.

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At zero temperature most substances exist in the form of solids. The only exception is helium, which undergoes quantum melting at a critical pressure. On the other hand, zero-temperature liquid-gas transitions have never been observed. Indeed, at absolute zero any system must be in its ground state and condensed phases have lower energy than gases. Driving a quantum liquid into a zerotemperature gas state would be possible if one could control interatomic forces. At sufficiently weak interatomic interactions, a many-particle bound state (i.e., liquid or solid) would cease to exist and a gas would form instead. While this cannot be accomplished with conventional materials, recent progress in the field of cold dilute gases opens a possibility to tailor a wide range of Hamiltonians with tunable parameters. In this Letter we show that a quantum liquid-gas transition can be observed in a cold gas of polar molecules confined in an optical lattice.

Experiments with cold gases have already allowed the observation of Bose-Einstein condensation, BCS superfluidity, and Mott localization [1], and it was proposed that cold gases can serve as realizations of other analogies of electronic matter [1]. Besides, several new states of matter with different broken symmetries and/or soft modes were predicted in cold-atom systems. We address a rather different situation. Liquids and gases have the same symmetry and their only difference is the density: A gas fills all available volume, but the density of a liquid is determined by intermolecular interactions.

While cold gases do not represent true ground states of alkali metals, they are highly stable due to their low density which guarantees a low probability of multiparticle recombination processes [2]. A well-established way to control interaction in cold gases utilizes Feshbach resonances and allows changing both the strength and sign of the short-range interatomic potential which can be modeled as a delta function in space. If it is repulsive the system minimizes its energy by occupying all available volume, i.e., is a gas. Attractive interactions have different effects on fermions and bosons. A Fermi gas undergoes Cooper pair formation while a Bose gas collapses into a regime with strong many-body recombination. In our case a different interatomic potential is needed: A liquid state can be formed if the interaction is attractive at large scales, but the short-range force must be repulsive to prevent collapse.

We demonstrate that such potential can be built from the dipole interaction of polar molecules. The sign of the dipole interaction is certainly independent of the distance and depends only on the direction of the dipole moments. It was shown recently [3] that the interaction sign can be made distance dependent by driving molecules with microwave fields. However, such an approach generates many-body forces [3]. We focus on a simpler situation with only two-body interactions. It can be achieved by confining polar molecules in a helical optical lattice, i.e., a potential well of a helical shape. Hexagonal arrays of such helices [4] are among numerous periodic and aperiodic structures used in holographic microfabrication experiments. We will see below that such structures open new possibilities for tailoring cold-atom Hamiltonians which cannot be obtained with usual optical lattices in the form of sinusoidal waves.

We focus on the simplest problem of this type with the lattice in the form of a single helix [Fig. 1(a)]. It can be produced with an approach similar to Ref. [4] as discussed in the Appendix. An external electric field polarizes molecules along the axis of the helix. In zero field, molecules have zero angular momentum and hence zero average dipole moment while in strong fields the average dipole moment $p$ can reach values of the order of debyes. At large
intermolecular distances the dipole interaction $V = p^2(1 - 3\cos^2\theta)/r^3$ is attractive ($\theta = 0$). At short distances it becomes repulsive, if the angle $\gamma$ between the tangent to the helix and its axis exceeds the magic angle $\cos^{-1}(1/\sqrt{3})$. The distance dependence of the interaction may include multiple maxima and minima for $\gamma = \pi/2$. In this Letter we focus on smaller $\gamma$ so that the interaction dependence on the distance $s$ along the helix has a simpler shape shown in Fig. 1(b). The potential resembles the Lennard-Jones interaction used in models of thermal liquid-gas transitions, and we show that a transition between a liquid and a gas occurs at a critical value of the dipole moment $\rho$.

This Letter is organized as follows. We first formulate the model. Next, we find its phase diagram with a variational method. We support the variational calculation by a proof that for a class of intermolecular potentials there is a phase transition between a monoatomic gas and a liquid. Then we address the conditions for the experimental observation of the transition.

We consider $N \gg 1$ particles of mass $m$ confined in a helical potential well of radius $R$ and pitch $d - R$. We assume that the wave function is confined in a region of the width $\Delta w \ll R$ around the helix [Fig. 1(a)]. At low temperatures $T \ll \hbar^2/(m\Delta w^2)$ the problem reduces to a one-dimensional model with the Hamiltonian

$$H = -\sum_i \frac{\hbar^2}{2m} \frac{\partial^2}{\partial s_i^2} + \sum_{i \neq j} V(s_i - s_j),$$

(1)

where $s_i$ is the length of the helix between particle number $i$ and a reference point on the helix, $V(s)$ the dipole interaction [Fig. 1(b)]. Repulsive short-range interaction keeps particles apart. Hence, in 1D the particle statistics is unimportant. In what follows we focus on nonidentical particles with $s_{n+1} > s_n$ for all $n$.

As the first step, we use the self-consistent harmonic approximation (SCHA), well known in the theory of disordered classical systems [5] (for a related method, see Ref. [6]). A better approximation will be considered at the ordered classical systems [5] (for a related method, see Ref. [6]). A better approximation will be considered at the ordered classical systems [5] (for a related method, see Ref. [6]). A better approximation will be considered at the ordered classical systems [5] (for a related method, see Ref. [6]). A better approximation will be considered at the ordered classical systems [5] (for a related method, see Ref. [6]).

We consider a trial distribution function of the form $P \sim \exp(-S_0/\hbar)$, where the trial action

$$S_0 = \frac{1}{2} \int dt \left[ \sum_{k=1}^{N} m \dot{s}_k^2 + \sum_{k=1}^{N-1} K[s_{k+1}(t) - s_k(t) - \hbar^2] \right]$$

$$= \int \frac{d\omega}{2\pi} \sum_{k,l=1}^{N} G_{kl}(\omega)s_k(\omega)s_l^*(\omega)$$

$$+ \int dt K\hbar(s_1 - s_N + [N-1]\hbar/2)$$

(2)

contains the variational parameters $h$ and $K$; $h$ is related to the average length of the $N$-particle chain and $K$ describes the fluctuations of the particle positions $s_k$. We need to minimize the variational free energy $E(K, h) = \langle S - S_0 \rangle - \hbar \ln Z_0/\tau$, where $\langle \cdot \cdot \cdot \rangle_0$ denotes the average with respect to the trial distribution function $P$, the trial partition function $Z_0 = \int \Pi d\omega s \exp(-S_0/\hbar)$, and the time $\tau \to \infty$ is the size of the integration domain. SCHA provides analytic results for potentials $V(s)$ in the form of the sums of exponentials. Thus, we approximate the potential represented in Fig. 1(b) by the Morse potential [7], $V(s) = A[\exp(-2\alpha(s - s_0)) - 2\exp(-\alpha(s - s_0))]$, which has a similar shape. Since the interaction rapidly decreases at large distances, we keep only the interaction between neighboring particles ($k$ and $k + 1$).

The variational calculation includes three steps: (1) we change the variables $s_k \to s_k + \kappa$ so that $\kappa$ drops out of Eq. (2); (2) calculate $E(K, h)$, and (3) find the minimum of $E(K, h)$. Step (2) requires the calculation of the correlation function $(s_k(\omega)s_k^*(\omega)) = \hbar G_{mm}(\omega)/2$. The matrix $G_{mm}(\omega)$ differs from a tridiagonal Toeplitz matrix [8] only by the values of two matrix elements. Its inverse can be found analytically using the same method as for tridiagonal Toeplitz matrices [8]. In the large $N$ limit, step (2) yields $E(K, h)/[\bar{N}\tau] = (h/\pi) \sum K/m + A[x^2 \exp(4\alpha^2\hbar/\pi\sqrt{Km}) - 2x \exp(\alpha^2\hbar/\pi\sqrt{Km})]$, where $x = \exp(-\alpha(h - h_0))$. After the minimization with respect to $\kappa$, the energy per particle $E(K, h)/[\bar{N}\tau]$ reduces to $e(K) = (h/\pi) \sum K/m - A \exp(-2\alpha^2\hbar/\pi\sqrt{Km})$. The energy $e(K)$ has a local minimum at $K = 0$, where $e(0) = 0$. At large $A$ another minimum at $K = K_{\text{min}}$ is possible. The phase transition into the state corresponding to that minimum occurs when $e(K_{\text{min}}) = 0$. Solving the latter equation together with $de(K_{\text{min}})/dK = 0$ we find a phase transition at $\lambda_c = 2e\alpha^2/\sqrt{\pi m\Delta w^2}$. Thus, at small $A$ we get $K = 0$ and $h = \infty$; i.e., the system is a gas [9]. $K$ and $h$ are finite at large $A$. This means a finite volume at zero external pressure, i.e., a condensed state. In 1D this cannot be a solid and the calculation of the correlation function $(s_k(\omega)s_k^*(\omega)) = \hbar G_{mm}(\omega)/2$ at $k \gg 1$ shows that the particles form a Luttinger liquid [10].

Our problem is connected with the physics of atoms confined in carbon nanotubes. If one ignores the periodic potential created by carbon atoms then a model, similar to ours, emerges (certainly, one cannot tune the interaction between the atoms and obtain a phase transition in a nanotube). A variational study [6] predicted that an increasing interaction drives a monoatomic gas into a diatomic gas phase before a liquid state can be reached. This contradicts our findings. Below we sketch a proof that a liquid state has lower energy than a diatomic or multiatomic gas and hence a multiatomic gas cannot be the ground state. Qualitatively this reflects the fact that in a liquid every particle has bonds with two neighbors and only one bond in a diatomic molecule. Hence, one expects lower energy per particle in a liquid. The prediction of a diatomic gas is thus an artifact of the variational method which treats differently
different pairs of neighboring atoms (and Ref. [6] admits its unreliability).

Our proof is based on a variational upper bound for the ground state energy. We will also use that bound to improve SCHA. We will focus on models in which only neighboring particles (k and k + 1) interact. A diatomic gas was predicted in Ref. [6] for such a model and such a model is relevant for us since dipole forces rapidly decrease at large scales. We will discuss elsewhere a derivation of the statements, proven below, for systems in which all pairs of particles interact via attractive potentials with hard-core repulsion. We first demonstrate that the energy of a triatomic gas is always lower than the energy of a diatomic gas, provided that the interaction potential and the ground state wave functions are well-behaved. Then we use a similar argument to show that the energy of the liquid phase is lower than the energies of all possible multiatomic gases. In all cases we assume that the potential energy is zero at an infinite interparticle separation and impose the hard-core condition V(0) = ∞.

Consider a system of N particles in the interval −∞ < s_k < ∞. The center of mass is at rest in the ground state. Hence, the wave function can be represented as ψ_N(Δ_1, ..., Δ_{N−1}), where Δ_k = s_{k+1} − s_k. The ground state configuration can be viewed as a set of molecules. A molecule is a bond cluster of n particles k, (k + 1), ..., (k + n − 1) with finite [11] interparticle distances Δ_k, ..., Δ_{k+n−2}. The molecules are separated by infinite intervals Δ_p. Different molecules do not interact. Hence, the ground state energy is the sum of the energies of separate molecules: One can easily see that if the ground state includes k molecules with N_1, ..., N_k particles then the energy equals the sum of the ground state energies of the Hamiltonians (1) with N = N_1, ..., N_k.

We now show that the energy of a diatomic gas can always be decreased if particles rearrange into trimers. A diatomic gas may form two particles have a bound state with the energy \( E_2 < 0 \). The bound state wave function \( \psi_2(Δ) \) is a normalized eigenfunction of the Hamiltonian \( H_{12} \), given by Eq. (1) with N = 2. Since the Hamiltonian \( H_{12} = −\hbar^2/m \frac{d^2}{d\Delta_1^2} + V(Δ_1) + V(Δ_2) \) is real, we can assume that \( \psi_2 \) is real. In the diatomic gas, the energy per particle is \( ε_2/2 \). We now demonstrate that there is a three-particle state with the energy per particle \( ε' < ε_2/2 \). The three-particle Hamiltonian \( H_{123} = −\sum_{i=1}^3 \hbar^2/m \frac{d^2}{d\Delta_i^2} + V(Δ_1) + V(Δ_2) \). Let us find the average energy E of the trial wave function \( ψ_3(s_1, s_2, s_3) = ψ_2(Δ_1)ψ_2(Δ_2) \). The structure of the trial wave functions prompts the change of variables \( s_1, s_2, s_3 \rightarrow Δ_1, Δ_2, s_3 \). The Jacobian of this transformation is one. Hence,

\[
E = \int dΔ_1 dΔ_2 ψ_2(Δ_1)ψ_2(Δ_2)\left[V(Δ_1) + V(Δ_2) − \hbar^2/m \left(\frac{∂^2}{∂Δ_1^2} + \frac{∂^2}{∂Δ_2^2}\right) + \hbar^2/m \frac{∂^2}{∂Δ_1∂Δ_2}\right]ψ_2(Δ_1)ψ_2(Δ_2).
\]

The integral of the last term in the square brackets reduces to the square of the integral of a full derivative, \( \int dΔ [dψ^2/ dΔ]/2 = 0 \). The first three terms in the brackets can be represented as the sum of two two-particle Hamiltonians \( H_{12} + H_{23} \). Thus, \( E = 2ε_2 \) and the energy per particle \( E/3 < ε_2/2 \). This shows that the energy is lower in the triatomic gas than in the diatomic gas which thus cannot be the ground state.

A similar argument proves a general statement: Consider a system of N particles in the interval −∞ < s_k < ∞. Only nearest neighbors interact. Assume that a bound state exists for \( n < N \) particles. Then in the ground state the system consists of no more than \( n \) molecules and exactly one of those molecules contains more than one particle. Indeed, the ground state energy of the infinite system equals the sum of the energies of its molecules with zero center-of-mass velocities. Imagine that the ground state includes two multiatomic molecules with \( m \) and \( (k + 1) \) atoms. Let \( ψ_m(Δ_1, ..., Δ_{m−1}) \) and \( ψ_{k+1}(Δ_1, ..., Δ_k) \) be the ground states of the Hamiltonian (1) with \( N = m \) and \( N = k + 1 \) and the energies of these states be \( ε_m \) and \( ε_{k+1} \). The sum of these two energies contributes to the ground state energy \( ε_g \). We now use the argument of the previous paragraph to show that the energy decreases if we substitute those two molecules with a monoatomic molecule (whose energy is 0) and a \( (m + k) \)-atomic molecule. Indeed, the same calculation as in Eq. (3) with the variational wave function \( ψ_{m+k} = ψ_m(Δ_1, ..., Δ_{m−1})ψ_{k+1}(Δ_{m}, ..., Δ_{m+k−1}) \) shows that the ground state energy \( ε_{m+k} \) of the Hamiltonian \( H_{m+k} \) (1) with \( N = m + k \) cannot exceed \( ε_m + ε_{k+1} \). The wave function \( ψ_{m+k} \) is not an eigenfunction of \( H_{m+k} \) and hence \( ε_{m+k} < ε_m + ε_{k+1} \). Thus, there is a state whose energy \( ε_g − (ε_m + ε_{k+1}) + (0 + ε_{k+m}) \) is below the ground state energy \( ε_g \). The contradiction means that no more than one multiatomic molecule exists in the ground state. Hence, if there were more than \( n \) molecules in the ground state then at least \( n \) of them would be monoatomic. In such a situation the energy decreases if we form an additional \( n \)-atomic molecule from \( n \) free particles. This proves that there are no more than \( n \) molecules and no more than one of them is multiatomic. If \( N >> n \) then exactly one molecule contains at least \( N − n + 1 \) particles which can be described as a liquid.

The above discussion shows that only two possibilities exist for the ground state in the large-N limit: a monoatomic gas, if no bound states exist at all, and a liquid. Both possibilities take place at different interaction strengths A. At \( A = 0 \) the system must be a gas [12]. At large A, SCHA yields a negative upper bound for the energy. Hence, the monoatomic gas with its zero energy cannot be the ground state at large A and a liquid-gas transition must occur at an intermediate A. SCHA is exact at large A, where the fluctuations of \( Δ_k \) are small and it is legitimate to expand the potential energy \( V(Δ) \) up to the second order. However, SCHA is insufficient near the phase transition. This is clear
from the comparison of the variational estimate for the transition point \( A_c \) and the exact threshold \( A_d \) for the formation of diatomic molecules in the Morse potential [7]. Contrary to the above proof, the SCHA result for \( A_c \) exceeds \( A_d = \hbar^2 \alpha^2 / (4m) \). Thus, a different method is needed near the transition. We try the variational ansatz of the form \( \psi_N = \prod_{k=1}^{N-1} \psi(\Delta_k) \), where the whole function \( \psi \) is a variational parameter. From the calculation, completely analogous to Eq. (3), we find the variational energy \( E = (N - 1)E_2 \), where \( E_2 \) is the average energy of a two-particle system in the state \( \psi(s_1 - s_2) \) in the Morse potential. The lowest \( E_2 \) corresponds to \( \psi(\Delta) \) which is the ground state in the Morse potential [7]. Hence, \( E_2 = -A[1 - \sqrt{4mA/s^2}]^2 \) at \( A > A_d \) and \( E = 0 \) at \( A < A_d \). This improves the estimate for the transition point, \( A_{c,new} > A_d \). Obviously, \( E \) is lower than the SCHA variational energy \( e = 0 \) in the interval \( A_c > A > A_{c,new} \). The improved variational method leads to an unexpected prediction concerning the order of the liquid-gas transition. The size of a diatomic molecule diverges [7] as \( s \sim 1/\sqrt{E} \) at \( A \to A_d \). Hence, the size \( l \) of the \( N \)-atomic bound state \( \psi_N \) diverges according to the same law. This means that the density of the liquid \( \rho = N/1 - \sqrt{E} \to 0 \) at \( A \to A_d \). In other words, the variational method predicts a second-order liquid-gas transition. Second-order transitions between Luttinger liquid states with different densities are known [13], but in contrast to Ref. [13] the symmetry does not change at our transition. According to the Landau theory such transitions must be first order. On the other hand, examples [14] are known of second order transitions in low-dimensional systems for which the Landau theory predicts the first order. It would be interesting to find a rigorous description of the 1D liquid-gas critical point.

We see that the phase transition occurs when the characteristic kinetic and potential energies are of the same order of magnitude, \( \hbar^2 / 4m L \sim V(1/\alpha) \). For polar molecules in a helical lattice, the characteristic spatial scale \( 1/\alpha = \pi R \). Hence, near the transition the dipole energy \( p^2 / (\pi R)^3 \) must be of the order of the recoil energy \( \hbar^2 / m(\pi R)^2 \). For a realistic optical lattice with \( \pi R \sim 1 \mu \text{m} \) and the molecular mass of the order of 100 we find \( p \sim 1 \text{ D} \) at the transition. Such dipole moments are within reach. A difference between a liquid and gas can be detected in a variant of the Einstein boxes experiment. A laser beam, orthogonal to the helix, creates a potential barrier in the center of the helical lattice. A gas occupies all lattice and an approximately equal number of particles will remain on both sides of the barrier. The volume of a liquid is much smaller than the system size far from the transition. Hence, all atoms will be found on one side of the barrier.

In conclusion, a gas of polar molecules in a helical optical lattice can be driven by an electric field into a liquid state via a continuous phase transition. The gas fills all available volume while the volume of the liquid is determined by the interaction strength.

**Appendix.**—A helical lattice can be obtained from the interference of a circularly polarized wave with \( E_c = E_c(1, i, 0) \) and the wave vector \( k(0, 0, \pm 1) \) with \( n \) laser beams with wave vectors \( k_m = k(\cos \alpha \cos[2\pi m/n], \cos \alpha \sin[2\pi m/n], \sin \alpha) \) and electric fields \( e_m = E_c(- \sin[2\pi m/n], \cos[2\pi m/n], 0) \). The optical potential perceived by an atom in the \( L_c = 0 \) state is proportional to the intensity of light [2], which can be easily found analytically at large \( n \). The molecules are trapped near the intensity extrema which lay on a helix.

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