

Doped antiferromagnets in the weak-hopping limit

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(Received 26 April 1990)

We study the phase diagram of the two-dimensional t - J - V model of holes in a two-dimensional antiferromagnet in the $J \gg t$ limit. Here t is the hole hopping parameter, J is the exchange coupling between nearest-neighbor spins, and V is the nearest-neighbor electron-electron repulsion. We also include the possibility of Heisenberg-Ising anisotropy, $J_\perp \neq J_z$. We obtain a complete description for $J_\perp \ll J_z$ and approximate results for $J_\perp = J_z$. There is no region of the zero-temperature phase diagram that supports a dilute gas of holes in an antiferromagnet. Rather, the system always phase separates into hole-rich and no-hole phases, a result we believe to be more general than the model we have considered. We find various hole-crystal phases and, at high hole density, we find a “Fermi-liquid” phase and at least two superfluid phases, one with a charge $2e$ and one with a charge $4e$ order parameter.

I. INTRODUCTION

The discovery of the intimate proximity of superconductivity and quantum antiferromagnetism in the phase diagram of the cuprate perovskites has led to an intense effort to understand the effects of dilute holes in a spin- $\frac{1}{2}$ Heisenberg antiferromagnet. The strategy adopted in many of these studies is to study the apparently simpler problem of a gas of holes of vanishing density in a background that is therefore at least locally antiferromagnetic far from any of the holes. One question that has not been addressed sufficiently carefully in these studies is whether holes can, in fact, dissolve in an antiferromagnet. In other words, does there exist a zero-temperature phase that can be described as a hole gas in an antiferromagnet? In nature, there are many examples of systems that form self-bound solid or liquid phases at low temperatures, but very few that remain gases. A notable exception to this general rule is found in ^3He - ^4He mixtures, where the “gas” phase consists of up to 6% ^3He in ^4He . (We return to this example below.) We recently have shown¹ that in the t - J model, the holes in an antiferromagnet are always self-bound. In other words, if the average hole concentration is held fixed and sufficiently small, then in thermal equilibrium the system will phase separate into a “hole-rich” region with finite hole concentration (determined by the short-ranged interactions between the holes) and a pure antiferromagnetic region. We also conjectured¹ that self-binding is a more general feature of holes in an antiferromagnet in the absence of long-ranged Coulomb interactions. The intuitive reasons for this conjecture are summarized below. In the present paper, we explore the phase diagram of a simple, short-range model of holes in an antiferromagnet, the t -(J_\perp, J_z)- V model, in the limit $T \ll J_z$ where we can obtain asymptotically exact results:

$$\begin{aligned}
 H = & \sum_{\langle \mathbf{R}, \mathbf{R}' \rangle} [J_z S_{\mathbf{R}}^z S_{\mathbf{R}'}^z + J_\perp (S_{\mathbf{R}}^x S_{\mathbf{R}'}^x + S_{\mathbf{R}}^y S_{\mathbf{R}'}^y)] \\
 & - \sum_{\langle \mathbf{R}, \mathbf{R}' \rangle} t (c_{\mathbf{R},s}^\dagger c_{\mathbf{R}',s} + \text{H.c.}) \\
 & + \sum_{\langle \mathbf{R}, \mathbf{R}' \rangle} V n_{\mathbf{R}} n_{\mathbf{R}'} + \sum_{\mathbf{R}} \mu n_{\mathbf{R}} ,
 \end{aligned} \tag{1}$$

where μ is the chemical potential, $c_{\mathbf{R},s}^\dagger$ creates an electron of spin s on site \mathbf{R} ,

$$S_{\mathbf{R}}^a \equiv \frac{1}{2} \sum_{s,s'} c_{\mathbf{R},s}^\dagger \sigma_{s,s'}^a c_{\mathbf{R},s'} , \tag{2a}$$

$$n_{\mathbf{R}} \equiv \sum_s c_{\mathbf{R},s}^\dagger c_{\mathbf{R},s} , \tag{2b}$$

and the Hamiltonian is supplemented by the constraint that there be no doubly occupied sites, $n_{\mathbf{R}} = 0$ or 1 for all \mathbf{R} . The creation operators $c_{\mathbf{R},s}^\dagger$ obey the usual fermion anticommutation relations. We will talk about empty sites with $n_{\mathbf{R}} = 0$ as being occupied by holes. The concentration of holes x , is given by

$$x = 1 - \frac{1}{N} \sum_{\mathbf{R}} n_{\mathbf{R}} , \tag{3}$$

where N is the number of sites. We will explicitly consider the case of a two-dimensional square lattice, although many of our results are trivially generalizable to three dimensions. We have chosen to define our Hamiltonian so that J_z and J_\perp positive corresponds to an antiferromagnetic exchange interaction; at the end of the paper we will briefly consider the “ferromagnetic” case which is obtained by taking J_\perp negative. For $J_z = J_\perp = J$ and $V = -J/4$, this model is commonly referred to as the t - J model. In the limit $t \gg J$, the t - J model is a piece of the low-energy effective theory of the large- U Hubbard mod-

el; the omitted pieces are the pair-hopping terms which may have important effects.² Independent of its microscopic origins, the $t-(J_z J_\perp)-V$ model is a simple model of holes in an antiferromagnet, and we learn much that is generic about the model by studying it in the more theoretically tractable small- t limit.

Our most important finding is that there is no region of the zero-temperature phase diagram in which a dilute gas of holes dissolved in an antiferromagnetic is stable. At low-hole concentration the system always phase separates into a hole-rich and an undoped phase. Depending on the relative magnitudes of V and J , the hole-rich phase is either an all-hole phase, with one hole per site, or a finite density hole crystal. While phase separation is most likely in the small- t limit, we have found¹ that it occurs even for large t/J .

The results of our calculations are given in tables, and are best summarized in the phase diagrams shown later in the figures. At zero temperature and small V we find complete phase separation; the hole-rich phase has hole density $x=1$, one hole per site. For larger V , the lowest hole-density phase is a crystal with $x=\frac{1}{5}$ and a $\sqrt{5}\times\sqrt{5}$ structure. At hole density $x=\frac{1}{2}$ there are at least three crystalline phases that are stable for different ranges of V . Finally, for $0 < x < \frac{1}{2}$, and moderate V there are at least two superfluid phases, one with a charge $2e$ and the other with a charge $4e$ order parameter. The charge $2e$ phase is describable in terms of a hard-core quantum dimer model.³ For large V and $x < \frac{1}{2}$, we find a Fermi-liquid phase. We also study the finite temperature phase diagram and make some inferences concerning how the phase diagram changes as a function of T and t .

II. THE $t=0, J_\perp=0$ MODEL: ($V < \frac{1}{4}J_z$)

To warm up, we consider the Ising limit $J_\perp=0$, with static holes, $t=0$. Because $t=0$, the statistics of the electrons are irrelevant; the model is classical and we can rewrite it as an effective spin-1 Ising model

$$H = \frac{J_z}{4} \sum_{\langle R, R' \rangle} S_R^z S_{R'}^z + V \sum_{\langle R, R' \rangle} (S_R^z S_{R'}^z)^2 + \mu \sum_R (S_R^z)^2, \quad (4)$$

where S_R is a spin-1 operator. We associate $S_R^z = \pm 1$ with the states in which site R is occupied by a spin-up or spin-down electron respectively, and $S_R^z = 0$ with the state in which site R is occupied by a hole. This is a familiar model which has been widely studied in the context of $\text{He}^3\text{-He}^4$ mixtures.⁴ We restrict our attention to the region of the phase diagram where the model is well behaved, $V < J_z/4$. (For $V > J_z/4$ the model has finite zero-temperature entropy.) The phase diagram as a function of x and temperature T computed in mean-field theory in Ref. 4 is shown in Fig. 1 for $V=0$. Similar phase diagrams result for nonzero $V < J_z/4$. The $T=0$ result can be verified by noting that the ground state flips from $(S_R^z)^2=0$ to $(S_R^z)^2=1$ as μ decreases past $\mu_c = J_z/2 - 2V$. In this regime, the phase diagram does not depend qualitatively on V ; there is complete phase

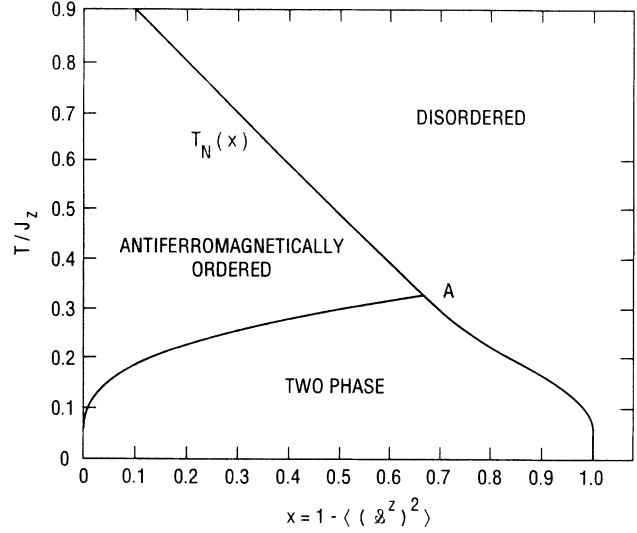


FIG. 1. Schematic phase diagram for the $t=V=J_\perp=0$ model. x is the hole concentration per site and T_N is the Néel temperature at zero doping (see Sec. II). As discussed in Sec. III, we expect this phase diagram to be little different if $J_\perp \neq 0$ so long as $|J_\perp| \ll J_z$. The actual phase diagram reproduced here was computed in mean-field theory as described in Ref. 4. Since the undoped system is simply the two-dimensional Ising model, we know that an exact solution would yield $T_N=0.57J_z$.

separation in the $T \rightarrow 0$ limit and the phase boundary for low T goes like

$$x_c \sim \frac{1}{2} \exp[(4V - J_z)/2k_B T].$$

Near the $x=0$ axis there is a Néel ordered phase which ends in a tricritical point on the coexistence curve.

III. THE $t \& J_\perp \ll J_z$ MODEL ($V > \frac{1}{4}J_z$)

We now consider the effect of small admixtures of quantum processes (nonzero J_\perp and t) on the phase diagram of the model. For temperatures T greater than both t and J_\perp , or for $V < \frac{1}{4}J_z$ (where there is a unique ground state and a gap in the excitation spectrum), these couplings have little effect. Thus, the phase diagram for $V < \frac{1}{4}J_z$ is changed little by the addition of nonzero J_\perp and t .

We therefore focus our attention on low temperatures and $V > J_z/4$ where the degeneracy of the classical ground state is lifted by quantum fluctuations. We keep terms computed in straightforward degenerate perturbation theory. The results of this calculation are summarized in Table I for dilute holes, $x \ll 1$, and in Table II for dilute electrons, $1-x \ll 1$. As mentioned above, for $1 > x > 0$ and $V > J_z/4$ there is a large zeroth order degeneracy of the ground state. Any configuration of holes in the otherwise perfectly Néel ordered background such that there are no present nearest-neighbor pairs of holes is degenerate with any other. We use degenerate perturbation theory to construct an effective Hamiltonian which connects the zeroth order degenerate states. For low hole concentrations, $x \ll 1$, it suffices to keep one-

TABLE I. Dilute holes to order $J_1^2/J_z, t^2/J_z, J_1 t^2/J_z$ [See Eq. (5)]. All other one- and two-body interactions are 0 to this order. $\hat{\mathbf{e}}_x$ and $\hat{\mathbf{e}}_y$ are the basic lattice vectors.

ε	$= J_z - 4V$	$+ \left[\frac{2}{15} \right] \frac{J_1^2}{J_z}$	$- \left[\frac{8}{3} \right] \frac{t^2}{J_z}$
$\Phi(\hat{\mathbf{e}}_x)$	$= - \left[\frac{1}{4} \right] J_z + V$		$+ \left[\frac{8}{3} \right] \left[\left[\frac{8V - J_z}{4V - 5J_z} \right] \right] \frac{t^2}{J_z}$
$\Phi(\hat{\mathbf{e}}_x + \hat{\mathbf{e}}_y) = 2\Phi(2\hat{\mathbf{e}}_x)$	$=$	$+ \left[\frac{1}{30} \right] \frac{J_1^2}{J_z}$	$+ \left[\frac{8}{3} \right] \left[\left[\frac{4V - J_z}{5J_z + 4V} \right] \right] \frac{t^2}{J_z}$
$\Phi(2\hat{\mathbf{e}}_x + \hat{\mathbf{e}}_y) = 3\Phi(3\hat{\mathbf{e}}_x)$	$=$	$- \left[\frac{1}{40} \right] \frac{J_1^2}{J_z}$	
$\tau_2(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y, \hat{\mathbf{e}}_x) = \tau_2(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_x, \hat{\mathbf{e}}_x)$	$=$		$+ \left[\frac{4J_z}{4V - 5J_z} \right] t^2$
$\tau(\hat{\mathbf{e}}_x + \hat{\mathbf{e}}_y) = \tau(2\hat{\mathbf{e}}_x)$	$=$		$+ \left[\frac{8}{15} \right] \left[\left[\frac{J_1}{J_z} \right] \left[\frac{t^2}{J_z} \right] \right]$

and two-hole terms in the effective Hamiltonian. The result is

$$\begin{aligned}
 H^{\text{eff}} = & \sum_R \varepsilon n_R + \sum_{|R, R'\rangle} \Phi(\mathbf{R} - \mathbf{R}') n_R n_{R'} \\
 & - \sum_{|R, R'\rangle} \tau(\mathbf{R} - \mathbf{R}') (c_R^\dagger c_{R'} + \text{H.c.}) \\
 & + \sum_{R_1, R_2, R'_1, R'_2} \tau_2(\mathbf{R}_1 - \mathbf{R}'_1, \mathbf{R}_2 - \mathbf{R}'_2, \mathbf{R}_1 - \mathbf{R}_2) \\
 & \quad \times c_{R_1}^\dagger c_{R'_1} c_{R_2}^\dagger c_{R'_2} + \dots, \quad (5)
 \end{aligned}$$

where c_R^\dagger is a spinless fermion creation operator that creates a vacancy on site R and $n_R = c_R^\dagger c_R$ is the vacancy number operator. The first term is the hole self-energy. The second term is a hole potential energy. The third term is a hole hopping and the fourth term is a hole pair hopping. The \dots signifies three and more hole interactions which can be ignored for $x \ll 1$ and $V > J_z/4$. To avoid double counting, we define

$$\Phi(0) = \tau(0) = \tau_2(R, 0, 0) = 0,$$

where the last equality holds for any R . All of the

TABLE II. Small clusters of electrons. Here $\tau = t^2 / (\frac{1}{4}J_z - V)$, and the terms labeled ε_a are the delocalization energy as a function of wave number to this order. (See especially Sec. IV.) $\varepsilon_0(k) = 2\tau[\cos(k_x) + \cos(k_y)]$, $\varepsilon_1(k) = 4\tau[\cos(k_x) + \cos(k_y)] + 2\tau$ and $2\tau[\cos(k_x) + \cos(k_y)] - 2\tau$ for the two different bands.

Picture	Perturbative Energy per electron	Heisenberg Energy per electron
	$0 - \varepsilon_0(k)$	0
	$\frac{1}{2}(V - \frac{1}{4}J_z) - \frac{1}{4}J_1 - \tau - \varepsilon_1(\mathbf{k})$	$\frac{1}{2}(V - \frac{3}{4}J)$
	$(V - \frac{1}{4}J_z) - \frac{1}{4} \left[\frac{J_1^2}{J_z} \right] - \tau$	$V - \frac{1}{2}J$
	$\frac{7}{6}(V - \frac{1}{4}J_z) - \frac{31}{144} \left[\frac{J_1^2}{J_z} \right] - \frac{7}{9}\tau$	$\frac{7}{6}V - 0.5216J$
	$\frac{5}{4}(V - \frac{1}{4}J_z) - \frac{5}{24} \left[\frac{J_1^2}{J_z} \right] - \frac{2}{3}\tau$	$\frac{5}{4}V - 0.5366J$
	$\frac{17}{12}(V - \frac{1}{4}J_z) - \frac{17}{90} \left[\frac{J_1^2}{J_z} \right] - \frac{3}{8}\tau$	$\frac{17}{12}V - 0.5326J$
	$\frac{3}{2}(V - \frac{1}{4}J_z) - \frac{89}{480} \left[\frac{J_1^2}{J_z} \right] - \frac{5}{12}\tau$	$\frac{3}{2}V - 0.5756J$
Infinite lattice	$2(V - \frac{1}{4}J_z) - \frac{1}{6} \left[\frac{J_1^2}{J_z} \right]$	$2V - 0.668J$

effective interactions must respect the point-group symmetries of the lattice. The values of all the effective interactions (up to symmetry transformations) are listed in Table I. These expressions are valid for arbitrary values of V/J_z not too close to $V = \pm(\frac{5}{4})J_z$. However, we are most interested in the results for $V > J_z/4$. Here, the potential energy terms strongly dominate the hopping terms; t is higher order in perturbation theory than the potential terms and the pair-hopping term, τ_2 , only operates on nearest-neighbor pairs which are suppressed in zeroth order by the large nearest-neighbor repulsion between holes. It is easy to see that at $T=0$ in this limit, the holes self-bind to form the $\sqrt{5} \times \sqrt{5}$ crystal structure shown in Fig. 2. We expect that the crystal will melt at a temperature $T \sim E_b = (J_\perp)^2/30J_z$, the binding energy of the crystal.

Even though the effective Hamiltonian has rather uninteresting consequences in thermal equilibrium due to the fact that the holes self-bind into a crystal, it may be interesting to use it to study the properties of nonequilibrium holes injected into an antiferromagnet, or to extrapolate to parameter regions in which well-controlled perturbative results cannot be obtained. With this in mind we note that to all orders in perturbation theory in J_\perp and t , $\tau(R-R')$ is nonzero only if R and R' are on the same sublattice; there are two distinct types of holes which do not mix.⁵ (This follows from the fact that we are doing degenerate perturbation theory, so only effective interactions which connect states in the zeroth-order degenerate manifold are produced.) Moreover, it is clear from Table I that the potential interactions produced by spin fluctua-

tions are attractive between holes on opposite sublattices and repulsive between holes on the same sublattice. The same sort of oscillatory interactions are produced by the exchange of spin waves in the Heisenberg limit, since the spin waves carry wave vector $Q = (\pi, \pi)$. Thus, some of the properties of H^{eff} may be more general features of holes in an antiferromagnet.

For hole densities $x > \frac{1}{2}$, the zeroth-order ground state is degenerate but of wholly different character; all states in which no two nearest-neighbor sites are occupied by *electrons* are degenerate. For nonzero t , we recognize this phase as a liquid of Fermions with short-range repulsions, i.e., a normal Fermi liquid. (We ignore the possibility of some more exotic phase transition in the Fermi liquid at extremely low temperature.)

Density $x = \frac{1}{2}$ is special even for $V > J_z/4$. Here, in zeroth order, the ground state is the $\sqrt{2} \times \sqrt{2}$ hole crystal shown in Fig. 2. This state has a macroscopic spin degeneracy but only a twofold translational degeneracy due to the ordering of the holes. For nonzero t , the spin degeneracy for $x = \frac{1}{2}$ is lifted by terms of order t^2 . Note, this is not superexchange, which in any case would be of order t^4 . The effect occurs due to the Ising interactions between nearest-neighbor spins in the intermediate states. The spins are governed by an effective Hamiltonian with Ising symmetry of the form

$$H^{\text{eff}} = \sum_R (\mathcal{J}_0 + \mathcal{J}_1 X_R^2 + \mathcal{J}_2 X_R^4), \quad (6)$$

where \sum' runs over the empty sites R (sites occupied by holes) in the $\sqrt{2} \times \sqrt{2}$ crystal structure, X_R is the total z component of the spin of the electrons on the four nearest-neighbor sites of the hole at R , and the values of \mathcal{J}_a are listed in Table III. Whereas this Hamiltonian is formally site diagonal, X_R from neighboring hole sites are not independent; they have spins in common. The ground state is still highly degenerate; any state in which $X_R = 0$ for all R is a ground state. The perfectly antiferromagnetically (Néel) ordered state on the electron lattice is such a state, but so is any state related to the antiferromagnetic state by reversing the direction of all the spins in any row or rows. This results in a ground-state entropy that is not extensive, but which grows like the square root of the size of the system. Presumably, this remaining ground-state degeneracy is lifted by terms of order t^4 ; we have not completed the analysis of these terms but it seems clear that they will stabilize the Néel state. Thus, the magnetic order should vanish at a "Néel" temperature of order \mathcal{J}_a , while the crystalline lattice will melt at much higher temperature of order $(V - J_z/4)$.

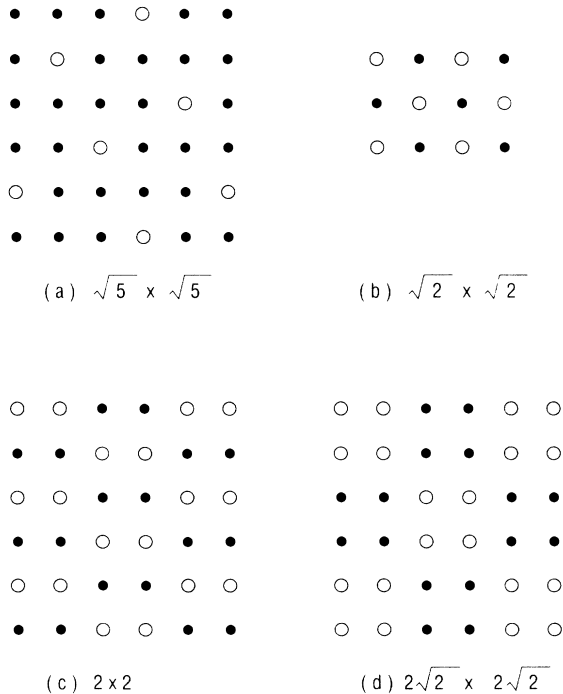


FIG. 2. Hole crystal structures referred to in the text: (a) is the $\sqrt{5} \times \sqrt{5}$ hole crystal, (b) is the $\sqrt{2} \times \sqrt{2}$ structure, (c) is the 2×2 dimer crystal, and (d) is the 2×2 square crystal structure.

TABLE III. Effective interactions in Eq. (6).

$$\begin{aligned} \mathcal{J}_0 &= -16 \left[\frac{t^2}{12V - J_z} \right] \\ \mathcal{J}_1 &= \frac{16t^2 J_z (12V + 7J_z)}{3(12V - J_z)(12V + J_z)(4V + J_z)} \\ \mathcal{J}_2 &= \frac{8t^2 J_z (12V - 5J_z)}{3(12V - J_z)(12V + J_z)(4V + J_z)} \end{aligned}$$

IV. THE $J_z = J_1 = J \gg t$ MODEL

The results in this section are summarized in the zero-temperature phase diagram shown in Fig. 3. The results for $x \geq \frac{1}{2}$ are exact; the results for $x < \frac{1}{2}$ are approximate.

For $x > \frac{1}{2}$, the system can best be thought of as consisting of a dilute collection of electrons. For $t=0$ and large to moderate V , these electrons do not tend to form large clusters, but they do form little "molecules" consisting of one, two, four, or more electrons. The optimal size and shape of the molecule is a function of V/J . We have calculated the energies of various small clusters and the infinite lattice as summarized in the third column of Table II. [The second column lists perturbative results for the same energies to second order in J_1/J_z and $t/(4V-J_z)$.] The results on clusters of size four or less are all analytic. The results on larger finite size clusters were obtained by exact numerical diagonalization. We did not obtain numerical results for molecules larger than 16 sites. The energy per electron of an infinite Heisenberg model has been calculated accurately from series expansion and Green's function Monte Carlo calculations.⁶ By directly comparing the exact energies it is easy to see that the optimal molecule is a monomer for $V/J > \frac{3}{4}$, a dimer for $\frac{3}{4} > V/J > \frac{1}{2}$, and a four-electron square for $\frac{1}{2} > V/J > 0.168$. We find that the energy is minimized by the infinite cluster (i.e., complete phase separation) when $V/J < 0.168$. We believe that these represent all the stable molecular phases of the model although we have not ruled out the possibility that there are interven-

ing phases where molecules of size larger than 16 and less than infinity are optimal. One should also note that there is nothing fundamental about this particular hierarchy of phases; by turning on longer-range potential interactions we can surely stabilize phases within which arbitrary size molecules are optimal.

To zeroth order in t , the ground state for $x > \frac{1}{2}$ and $V/J > 0.168$ is highly degenerate; all configurations such that no two molecules come within one lattice constant of each other have the same energy. This degeneracy is lifted by a small but finite t .

For $V/J > \frac{3}{4}$, the molecules are simply electrons with strong short-range repulsions between them. Thus, for finite t , the system is a Fermi liquid (again, ignoring the possibility of a further, ultra-low-temperature phase transition into a more exotic phase).

For $\frac{3}{4} > V/J > \frac{1}{2}$ and $x > \frac{1}{2}$, the system is a liquid of dimers. Each dimer consists of a pair of singlet paired electrons and hence behaves as a hard-core boson. If we simply do degenerate perturbation theory, the hard-core includes the nearest-neighbor sites of the dimer. [Equivalently, we could organize the perturbation theory such that the repulsion between two dimers which have exactly one pair of nearest-neighbor sites is $V - (2\sqrt{3} - 3)J/4$, while the repulsion between two dimers on the same plaquette is $2(V - J/2)$. It makes little difference if we treat these repulsions relative to the dimer hopping τ , defined below.] Thus, the low-energy physics in this region of parameter space is described by the quantum-hard-core dimer gas³ with second quantized Hamiltonian given pictorially by

$$\begin{aligned}
 H &= -\tau \sum_3 (\begin{array}{|c|c|c|} \hline \text{---} & \cdot & \text{---} \\ \hline \end{array} \begin{array}{|c|c|c|} \hline \cdot & \text{---} & \cdot \\ \hline \end{array} + \text{H.c.}) \\
 &= \tau \sum_{3'} (\begin{array}{|c|c|} \hline \cdot & \text{---} \\ \hline \end{array} \begin{array}{|c|c|} \hline \text{---} & \cdot \\ \hline \end{array} + \text{H.c.}) \\
 &\quad - 2\tau \sum_4 (\begin{array}{|c|c|c|c|} \hline \text{---} & \cdot & \text{---} & \cdot \\ \hline \end{array} \begin{array}{|c|c|c|c|} \hline \cdot & \text{---} & \cdot & \text{---} \\ \hline \end{array} + \text{H.c.}),
 \end{aligned} \tag{7}$$

where a bar represents a singlet pair of electrons and a dot represents an empty site (i.e., occupied by a hole), the sum over 3 runs over all triples of nearest-neighbor sites that form a right triangle, the sum over $3'$ runs over all triples of collinear nearest-neighbor sites, and the sum over 4 runs over all quadruples of sites around a plaquette. The effective dimer hopping matrix is easily seen to be second order in t ,

$$\tau = 8t^2 / (3J - 4V). \tag{8}$$

In addition to the terms in Eq. (7), there are corrections to the dimer self-energy, and somewhat longer-range dimer-dimer repulsions of order τ induced to this order of perturbation theory. Without going into details we note that since Eq. (7) describes a liquid of bosons with short-range repulsions, it should condense into a superfluid phase with charge $2e$ quasi-long-range order below a Kosterlitz-Thouless transition temperature $T_{KT} = f(x)\tau$, where $f(x) \sim (1-x)$ as $x \rightarrow 1$.

In similar fashion, for $x > \frac{1}{2}$ and $\frac{1}{2} > V/J > 0.168$, the

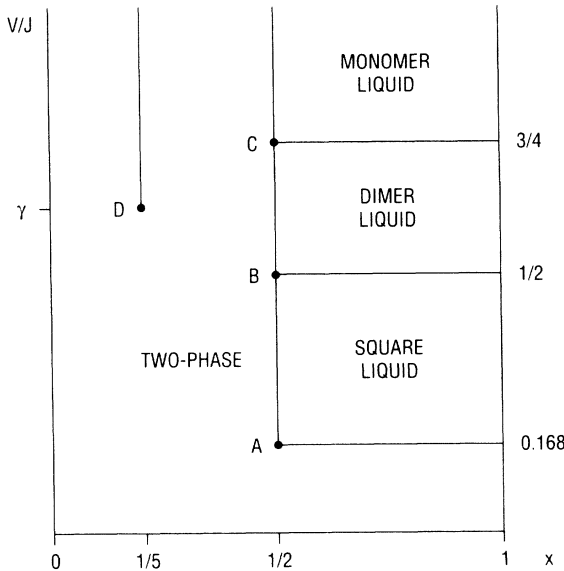


FIG. 3. Zero-temperature phase diagram for the t - J - V model ($J_z = J_1$) for $t \ll J$. On the line A - B is the square crystal shown in Fig. 2(d); on BC is the dimer crystal shown in Fig. 2(c), on the line going up from point C is the monomer crystal shown in Fig. 2(b), and on the line going up from D is the hole crystal shown in Fig. 2(a). As discussed in the Appendix, the numerical value of γ , which marks the endpoint of the hole crystal phase, has only been very crudely estimated.

low-energy effective theory is a theory of hard-core bosonic molecules with charge $4e$. The effective hopping-matrix elements for these molecules is of order t^4 , so the superfluid transition temperature is very low. Nonetheless, we feel that the existence of a superfluid phase with a charge $4e$ order parameter is quite interesting.

For $x < \frac{1}{2}$ we cannot obtain a formally well-controlled systematic solution to the model. However, we believe that the results are qualitatively the same as in the $J_{\perp} \ll J_z$ limit where there is an attractive interaction of order J between static holes on opposite sublattices. Since, by assumption, $J \gg t$, this immediately implies phase separation into a hole crystal phase. This feature certainly persists in the Heisenberg limit. Moreover, since the hole crystal is always fairly dense (see phase diagram in Fig. 3), the presence or absence of long-range magnetic order at zero temperature has almost no effect on this result; it is the attractive interactions between holes no more than a few lattice sites apart which drives the phase separation. One can estimate the magnitude of the interactions for the t - J - V model by simply taking the perturbative results in Table II and evaluating them for $J_{\perp} = J_z = J$. The results are shown in Table IV. Alternatively, one can attempt to extract results from exact numerical diagonalization of finite size systems with static holes. We have done this for 4×4 systems with periodic boundary conditions and have analyzed the results as described in the Appendix. As is clear from the table, there is good agreement between the two methods with regard to the sign of the interactions, at least at short distances. The numerical results consistently yield larger magnitudes for the attractive part of the interactions. On the basis of these approximate results we infer that the doped Heisenberg antiferromagnetic forms an $x = \frac{1}{5}$ hole crystal for $V > \gamma J$ where from perturbation theory we estimate $\gamma = \frac{13}{60} \approx 0.2$, and from the numerics we estimate $\gamma \approx 1$. For $V < 0.168 J$ we find complete phase separation into a perfect antiferromagnet and an $x = 1$ all hole region.

TABLE IV. Interactions between static holes in the Heisenberg model. The Φ notation is the same as in Table I. The second column is the obtained by substituting $J_z = J_{\perp} = J$ in the expressions from Table I. The estimates in the third column were obtained from numerical solution of the model on a 4×4 lattice with periodic boundary conditions as described in the Appendix.

Picture	Extrapolated P.T.	Numerics
$\Phi(\hat{e}_x)$	$V - 0.25J$	$V - 0.52J$
$\Phi(\hat{e}_x + \hat{e}_y)$	$0.033J$	$0.028J$
$\Phi(2\hat{e}_x)$	$0.017J$	$0.028J$
$\Phi(2\hat{e}_x + \hat{e}_y)$	$-0.025J$	$-0.024J$
$\Phi(3\hat{e}_x)$	$-0.008J$	
ϵ	$-4V + 1.133J$	$-4V + 1.283J$
E_{AF}	$2V - 0.667J$	$2V - 0.668J$

V. THE FERROMAGNETIC MODEL,

$-J_z > -J_{\perp} \gg t$ (ANALOGY WITH He MIXTURES)

So long as the ground state is completely ferromagnetically aligned, we can compute the low-energy properties of the system exactly; there are no perturbative corrections in J_{\perp}/J_z . The ferromagnetic state will be the ground state if $|J_{\perp}| < (-J_x)$ and $t \ll (-J_z) - |J_{\perp}|$. Thus, the low-energy hole motion is described by the effective Hamiltonian

$$H = \epsilon \sum_R a_R^\dagger a_R = t \sum_{\langle R, R' \rangle} (a_R^\dagger a_{R'} + \text{H.c.}) + \sum_{\langle R, R' \rangle} v a_R^\dagger a_R a_{R'}^\dagger a_{R'}, \quad (9)$$

where a^\dagger is the hole creation operator, $\epsilon = -(4V + J_z)$, and $v = (V + J_z/4)$. Note that the hole bandwidth is entirely unrenormalized. For $V > -J_z/4$, the hole-hole interactions are purely repulsive. Thus, we expect the hole gas to be stable at all concentrations. In addition to the excitations of the hole gas, the system possesses spin excitations above a gap energy that vanishes as $J_z \rightarrow J_{\perp}$, the massive magnon. The ferromagnetic order persists up to a finite temperature which is roughly of order J_z , although it depends on the hole concentration and vanishes as $x \rightarrow 1$ or as $J_{\perp} \rightarrow J_z$. For $V < -J_z/4$, the interactions are attractive and the system may phase separate.

Note that the results we have obtained apply equally well to the case in which J_{\perp} is positive. In this case the magnetic ground state is still ferromagnetically ordered and so the hole kinetic energy is unfrustrated. From the point of view of magnetism this is curious since for $J_z < 0$ and $J_{\perp} > 0$, the magnetic part of the Hamiltonian can be transformed into a purely antiferromagnetic Ising-Heisenberg model by a canonical transformation. If we are dealing with magnetism alone it is conventional to call this model "antiferromagnetic" even though the actual spin order is ferromagnetic since all its thermodynamic functions are exactly equal to those of the pure antiferromagnetic model. However, in the presence of mobile holes, the canonical transformation cannot be performed without at the same time transforming the hole hopping term. Thus, the holes distinguish between a true antiferromagnet, where the hole kinetic energy is frustrated, and a model with a ferromagnetic order which is formally related to the true antiferromagnet but in which the hole kinetic energy is unfrustrated.

VI. ^3He - ^4He MIXTURES ($0 > J_x > J_{\perp}$)

The ferromagnetic t - (J_z, J_{\perp}) - V model in the XY limit incorporates many of the important features of ^3He - ^4He mixtures. Here we imagine a lattice-gas model of the He liquid and we incorporate the hard cores by imposing the constraint that no two He atoms can occupy the same lattice site. We also ignore the nuclear spin of the ^3He atom. (Since, in any case, two ^3He atoms cannot occupy the same site due to the hard core, the existence of two types of ^3He atoms, spin up and spin down, which would in principle allow double occupancy of a site, should not

have a very important effect on the physics at low ^3He densities.) Thus, we are left with a lattice-gas model of hard-core bosons and spinless fermions. Such a model is exactly of the same form as Eq. (1), where a spin-up site corresponds to a site which is occupied by a ^4He atom, a site occupied by a hole corresponds to a site occupied by a ^3He atom, and a spin-down site corresponds to an empty site. Thus $-J_\perp$ is the ^4He hopping matrix element, t is the ^3He hopping matrix, J_z is the nearest-neighbor ^4He - ^4He repulsion, V is the ^3He - ^3He repulsion, μ is the ^3He chemical potential. The ^4He chemical potential has been set equal to zero. Another value of the ^4He chemical potential would correspond to an external magnetic field in the z direction in the spin language. ^4He superfluid order corresponds to the XY order in the spin language, which is why the analogy holds only in this limit. (Ising order in the spin model would correspond to crystalline order in the ^4He .) From the experimental fact that ^3He is immiscible in ^4He beyond 6% concentration, we infer that, at least in three dimensions, the holes in a ferromagnetic XY -Heisenberg model will phase separate for concentrations larger than some small, but nonzero hole concentration.

For $0 > J_z > J_\perp$ we cannot obtain similarly exact results as in the Ising limit. However, the only differences we expect in the qualitative behavior in this region of parameter space are the following: (1) The ferromagnetic order parameter will lie in the XY plane rather than along the z axis. (2) There will be a massless spin-wave mode due to Goldstone's theorem, as opposed to the massive magnon we observe in the Ising limit. (3) The ferromagnetic long-range order will be destroyed by thermal fluctuations at any finite temperature. Instead, we expect quasi-long-range ferromagnetic order below a Kosterlitz-Thouless temperature, which is roughly of order J_\perp but which vanishes as $J_z \rightarrow J_\perp$ or as $x \rightarrow 1$. (4) There are long-range effective interactions induced between the holes by the exchange of ferromagnetic spin waves. It is not completely clear what the effect these interactions have; as suggested below they could lead to phase separation beyond a critical hole density.

VII. CONCLUSIONS AND SUMMARY

We have studied the t -(J_z, J_\perp)- V model in the small- t limit. In this limit we find that wherever the ground state is antiferromagnetically ordered (whether the purely magnetic part of the Hamiltonian is related by a unitary transformation to an antiferromagnet or to a ferromagnet), there is no stable zero-temperature phase with vanishing hole density; holes introduced into a pure antiferromagnet are always self-bound and so there is a minimum stable hole concentration necessary for a homogeneous phase. In addition to various hole crystal phases, we also found that at relatively high hole concentrations there exist charge $2e$ and charge $4e$ superfluid phases.

Based on the intuition we have developed in this study, we have made several *speculations* concerning the behavior of more realistic models of doped antiferromagnets. (1) We believe that some of the features of the phase dia-

gram we have derived in the small- t limit survive even when t is larger than J . In particular, we feel that the self-binding of holes at low temperature and low-hole concentration is a general feature of holes in an antiferromagnet. Of course long-range Coulomb interactions must drastically change this behavior. Consider the behavior of the system at fixed low concentration of holes (in the forbidden region of the phase diagram) and imagine adding to the model a long-range Coulomb interaction of the form $V(R) = e^2/\epsilon R$. For ϵ large enough, the long-range Coulomb interactions will only affect the nature of the ground state over very long distances. Thus, the holes will form droplets of the preferred hole density and those droplets will in turn form a crystal of some sort. In three dimensions some sort of staging in which hole-rich and undoped layers alternate in a commensurate pattern should occur. On the other hand, as we reduce ϵ , we expect that the Coulomb interactions will destroy even the short-ranged hole order. Finally, for small enough ϵ , the holes will form a Wigner crystal in the antiferromagnetic background. (2) We expect that at sufficiently high dopant concentration, there exists a charge $2e$ s -wave superconducting state of electrons. (Note it is possible that at dilute hole concentrations and large t/J , a superconducting state, if it exists, may be d wave.) It is interesting to ask whether the charge $4e$ superconducting state survives to large t .

Finally, we note the present results reinforce the idea⁷ that data on the lightly doped perovskite superconductors should be examined to determine whether, in fact, they are intrinsically inhomogeneous. It is possible that, even if phase separation of the holes occurs, it does so for purely extraneous reasons, such as preferred packing structures of the oxygens. However, we would also like to suggest that in lightly doped antiferromagnetic materials there is an intrinsic tendency to form an inhomogeneous state. In fact, there is strong evidence⁸ that phase separation occurs in oxygen doped La_2CuO_4 . Moreover, in Sr doped La_2CuO_4 , muon-spin-relaxation experiments⁹ have revealed the existence of a low-temperature transition to a state with frozen moments, even in superconducting samples, with a transition temperature that extrapolates to zero at 15% at. Sr concentration. A natural interpretation of this is that the critical hole concentration in these materials is $x = 0.15$, and that all samples with lower concentration of holes are intrinsically inhomogeneous. This would further imply that the superconducting transition as a function of x at $x = 0.05$, is a percolative transition.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge constructive criticism by S. Chakravarty. We would like to acknowledge the hospitality of the Institute for Theoretical Physics at the University of California, Santa Barbara (UCSB) where this work was complete and S.K. would like to acknowledge the hospitality of Brookhaven National Laboratory. This work was supported in part by the National Science Foundation (NSF) Grant Nos. DMR 87-06250 [at the State University of New York (SUNY)] and PHY82-17853 (at UCSB) supplemented by funds from NASA at

UCSB and by the U.S. Department of Energy under Contract No. DE-AC02-76CH00016.

APPENDIX; ESTIMATING THE HOLE-HOLE INTERACTIONS FROM THE NUMERICS

We have estimated the interactions between static holes from the results on the 4×4 system in various ways, all of which produce roughly equivalent results. The method we finally used was as follows. (1) The energy per bond of the 4×4 lattice with no holes is $-0.702J$. The best estimate⁶ of the same energy for the infinite system is $-0.669J$. Since at a crude level, the energy to add a hole to an antiferromagnet is simply the energy to break a certain number of bonds, we have rescaled all energies computed for the 4×4 system by an overall multiplicative factor of $0.669/0.702=0.953$. (2) The energy to add one static hole to an antiferromagnet, ϵ , was computed by taking the difference of the ground-state energy of fifteen spins and one vacancy and the 16 spin ground-state energy, and rescaling the energy as above. (3) To es-

timate the two-hole energy we computed the ground-state energy for two vacancies in the 4×4 system. We chose to estimate the energy of two noninteracting holes not by doubling the result in (2), but rather by taking the energy for two vacancies as far apart as possible (in this case, that means at a distance of $2\sqrt{2}$ lattice constants apart) to be the energy of two independent holes. The difference between this energy and the ground-state energy of two vacancies at smaller separation, rescaled as above, was then taken to be the interaction energy between the two holes.

One further aspect of the calculation should be noted. For the 16 spin system with no holes, the ground state has spin 0. For the 15 spin system with one hole, the ground state has spin $\frac{1}{2}$. For 14 spins and two holes, one on the red sublattice and one on the black sublattice, the ground state has spin zero, while for both holes on the same sublattice it has spin 1. This is in keeping with the extrapolated results of perturbation theory in J_{\perp}/J_z , in which the z component of the spin in the ground state has the same dependence on hole number and location.

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