

Valence-bond theory of highly disordered quantum antiferromagnets

S. ZHOU¹, J. A. HOYOS^{2(a)}, V. DOBROSAVLJEVIĆ¹ and E. MIRANDA³

¹ *National High Magnetic Field Laboratory, Florida State University - Tallahassee, FL 32310, USA*

² *Department of Physics, Duke University - Durham, NC 27708, USA*

³ *Instituto de Física Gleb Wataghin, Unicamp - C.P. 6165, Campinas, São Paulo 13083-970, Brazil*

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Abstract – We present a large- N variational approach to describe the magnetism of insulating doped semiconductors based on a disorder-generalization of the resonating-valence-bond theory for quantum antiferromagnets. This method captures all the qualitative and even quantitative predictions of the strong-disorder renormalization group approach over the entire experimentally relevant temperature range. Finally, by mapping the problem on a hard-sphere fluid, we could provide an essentially exact analytic solution without any adjustable parameters.

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The metal-insulator transition (MIT) in doped semiconductors (DS) [1] is one of the most fundamental, yet theoretically less understood problems in condensed matter physics. Even aside from their pivotal technological role, the DS have long been recognized as a bellwether system for the study of quantum criticality at the MIT. Careful transport experiments have revealed sharply defined critical behavior, although with exponents inconsistent with early theoretical predictions [2].

What are the basic physical processes that drive this transition and localize the electrons? Important clues have been provided by the thermodynamic response on the insulating side. Here, no magnetic ordering has been experimentally observed down to the lowest temperatures, while both the spin susceptibility and the specific heat display signatures of randomly interacting localized magnetic moments [1,3]. This puzzling behavior was largely explained by the Bhatt-Lee (BL) theory [4] of random singlet (RS) formation, using a strong-disorder renormalization group (SDRG) approach [5].

The remarkable success of the BL theory provides strong support to the early ideas of Mott [6], who first emphasized that strong Coulomb repulsion may localize the electrons by converting them into localized magnetic moments. According to this picture, the MIT in DS should be viewed as a disordered version of the Mott transition,

a phenomenon dominated by strong correlation effects. An appropriate theory should then be able to describe both the local moment magnetism in the insulator and the transmutation of these local moments into conduction electrons on the metallic side of the MIT. Unfortunately, the SDRG approach of BL, which was so successful in the insulator, is difficult to extend across the transition.

The essential challenge, therefore, is to develop an alternative approach to Mott localization in a strongly disordered situation, one that *at the very least* can reproduce the RS physics of Bhatt and Lee. An attractive avenue to describe strong correlations has emerged in the last twenty years from studies of various Mott systems, based on resonating-valence bond (RVB) ideas of Anderson [7] and others. At the mean field level, these theories provide variational wave functions for quasiparticle states, which become exact in appropriate large- N limits [8]. Very recent work has extended similar variational studies to disordered systems, providing a description of phenomena such as disorder-induced non-Fermi liquid behavior [9], but did not address the physics of inter-site spin correlations central to the BL paradigm.

In this letter we examine an appropriate t - J model capable of describing the Mott transition in a disordered environment. While the large- N limit of this model generally reduces to an RVB-like variational problem, here we concentrate on the localized ($t \rightarrow 0$) limit in the presence of strong positional disorder modeling the insulating DS.

^(a)E-mail: jh146@phy.duke.edu

We show that: i) the large- N formulation quantitatively reproduces all the key features of the RS regime; ii) an accurate analytic solution of the variational problem can be thus obtained, providing closed form expressions for various physical quantities; and iii) the approach can be directly extended to the metallic side, eliminating the main stumbling block in attacking the MIT in DS.

Model and large- N formulation. – We start with the large- N formulation of the two-orbital t - J model,

$$\mathcal{H} = \sum_{\mathbf{k},\sigma} (\varepsilon_{\mathbf{k}} + \varepsilon_o) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_{i \neq j, \sigma} t_{ij} \tilde{f}_{i\sigma}^\dagger \tilde{f}_{j\sigma} + \frac{1}{2N} \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \frac{V}{\sqrt{N}} \sum_{i, \mathbf{k}, \sigma} (e^{i\mathbf{k} \cdot \mathbf{r}_i} c_{\mathbf{k}\sigma}^\dagger \tilde{f}_{i\sigma} + \text{H.c.}), \quad (1)$$

under the constraint of no double occupancy on the \tilde{f} -orbital. Here each lattice site corresponds to a donor or acceptor which is randomly distributed in a periodic-boundary 3D cube of volume $V_0 = N_0/\rho_0$, where N_0 is the number of dopant sites and ρ_0 is the doping concentration. We stay at half-filling for the uncompensated DS, $\sum_{\mathbf{k},\sigma} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_{i\sigma} \tilde{f}_{i\sigma}^\dagger \tilde{f}_{i\sigma} = N_0 N/2$. The c -orbital represents the semiconductor conduction band with dispersion $\varepsilon_{\mathbf{k}}$, lying at an energy ε_o above the hydrogenic $1s$ impurity bound state (the \tilde{f} -orbital), and V is the hybridization between them. \mathbf{S}_i is the $SU(N)$ spin operator of the \tilde{f}_i -orbital. The hopping between the hydrogenic bound states [10] falls off exponentially with distance $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$, $t_{ij} = t_0 \exp(-r_{ij}/a)$ for $r_{ij} \gg a$, the Bohr radius of the bound state. Consequently, the anti-ferromagnetic super-exchange coupling

$$J_{ij} = J_0 \exp(-2r_{ij}/a), \quad (2)$$

where $J_0 \sim t_0^2$ (see footnote ¹). The projected Hilbert space of the \tilde{f} -orbital can be treated in the slave-boson formalism $\tilde{f}_{i\sigma}^\dagger = b_i f_{i\sigma}^\dagger$ enslaved to a constraint on each site $\sum_{\sigma} f_{i\sigma}^\dagger f_{i\sigma} + b_i^\dagger b_i = N/2$.

In this letter, we focus on the insulating side of the uncompensated DS $\rho_0 < \rho_c$ ($\rho_c^{1/3} a \approx 0.25$ for Si:P) where the average inter-site distance $\Lambda = \rho_0^{-1/3} \gg a$, which implies that $t_{ij} \rightarrow 0$. In this limit, the effective hybridization bV goes to zero as $b \rightarrow 0$, and the electrons become Mott localized on singly occupied \tilde{f} -orbitals. This results in an effective Heisenberg Hamiltonian for the insulating uncompensated DS, $\mathcal{H} = \frac{1}{2N} \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$. The magnetic behavior of such a disordered Heisenberg system was largely explained by Bhatt and Lee via

¹It is well known (and expected) that corrections to J_{ij} exist due to anisotropy and other effects. For instance, in $d=3$ there is an additional factor of $(r_{ij}/a)^{5/2}$ multiplying J_{ij} when $r_{ij} \gg a$ [10]. For our purposes, these corrections can be safely neglected in face of the highly disordered character of the dilute (insulating) regime. They only provide subleading (logarithmic) corrections as we confirmed numerically. Furthermore, this allows us to directly compare our results with those of the Bhatt-Lee theory [4], which also neglects them.

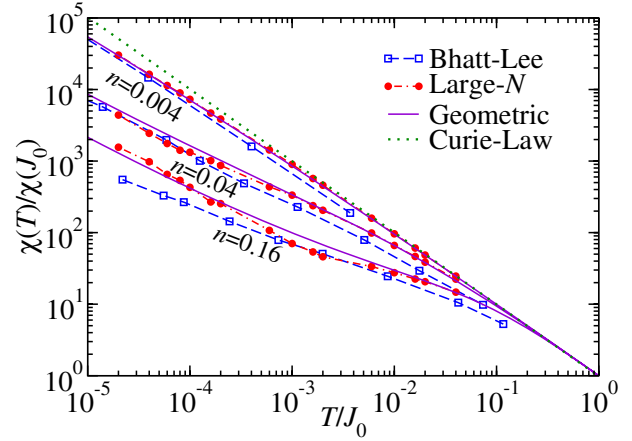


Fig. 1: (Colour on-line) Normalized magnetic susceptibility of highly disordered 3D Heisenberg magnets evaluated with the Bhatt-Lee method [4], the large- N self-consistent theory (for systems with $N_0 = 512$ spins (see footnote ²), and the geometric decimation procedure at concentrations $n = \frac{4\pi}{3} \rho_0 a^3 = 0.004, 0.04, \text{ and } 0.16$.

the SDRG method. Here we investigate the system within the large- N theory [8,11], which leads to an effective mean-field Hamiltonian through the saddle-point approximation,

$$\mathcal{H} = -\frac{N}{16} \sum_{i \neq j} J_{ij} \left(\Delta_{ij}^* \hat{\Delta}_{ij} + \text{H.c.} - |\Delta_{ij}|^2 \right),$$

with the constraint $\sum_{\sigma} f_{i\sigma}^\dagger f_{i\sigma} = N/2$ (of self-conjugate spins) implemented through the local Lagrange multiplier λ_i . Here, $\hat{\Delta}_{ij} = 2 \sum_{\sigma} f_{i\sigma}^\dagger f_{j\sigma} / N$ are valence bond (VB) operators and $\Delta_{ij} = \langle \hat{\Delta}_{ij} \rangle$ are variational parameters which minimize the free energy. They are solved self-consistently at $N \rightarrow \infty$, for a given sample realization and temperature. The results are then averaged over 20 sample realizations.

Numerical large- N solution. – At any finite temperature, our large- N solution finds two types of spins: localized and VB spins. The localized spins are those isolated from all other ones, *i.e.*, $\Delta = 0$ for all bonds connecting to them; their contribution to the magnetic susceptibility is simply a Curie term $\chi_c(T) = \mu_B^2 / k_B T$. In contrast, each VB spin forms a singlet bond ($\Delta \neq 0$) with another spin; their contribution can be neglected at low temperatures. The low- T magnetic susceptibility is, therefore, well approximated by

$$\chi(T) = \rho(T) \chi_c(T), \quad (3)$$

where $\rho(T)$ is the density of localized/free spins at temperature T . Figure 1 shows the normalized magnetic susceptibility $\chi(T)/\chi(J_0) = J_0 \rho(T) / T \rho_0$ at concentrations $n = \frac{4\pi}{3} \rho_0 a^3 = 0.004, 0.04, \text{ and } 0.16$ (see footnote ²). The susceptibility diverges at low temperatures, consistent

²We have carefully verified that all our numerical results are robust with respect to finite-size effects.

with the SDRG results of BL [4]. This divergence is usually fitted by a power law in experiments, but we shall show later that it should be viewed as a logarithmic correction to the Curie law. The higher the doping concentration, the larger this correction since couplings among spins are stronger. At extremely low concentrations, all spins are essentially free and the magnetic susceptibility follows the Curie law.

Geometric decimation procedure. – The large- N ground state at zero temperature of such a highly disordered Heisenberg system is essentially a RS state, in which most spins form inert singlets ($\Delta = 1$) with another spin and do *not* correlate with any other spin. To highlight this, we considered a simple four-spin cluster with antiferromagnetic couplings $J_{ij} > 0$, and $J_{23} \gg J_{ij}$ for all $(i, j) \neq (2, 3)$. The large- N calculation shows that for $T > J_{23}$, all bonds are zero and all four spins are free. As we lower the temperature to J_{23} , spins \mathbf{S}_2 and \mathbf{S}_3 start to form a VB singlet, $\Delta_{23} \neq 0$, and no longer contribute to $\chi(T)$. Further reducing the temperature to J_{14} , spins \mathbf{S}_1 and \mathbf{S}_4 form another VB singlet. There is *no* resonance between the (2,3) and the (1,4) VB singlets. In contrast to the Bhatt-Lee SDRG method, in which there appears a renormalized coupling between $SU(2)$ spins connected to a strong singlet pair, this effect can be shown to be of order $1/N$ between $SU(N)$ spins [12], and thus *drops out* in the large- N limit. While this simplification makes our large- N model amenable to closed form solution, we shall demonstrate that it hardly affects the quantitative predictions of the model within the experimentally relevant temperature range (as shown in fig. 1).

This also allows us to state a very simple geometric decimation procedure. We i) search for the most strongly coupled spin pair, or equivalently, the shortest one (see eq. (2)), ii) remove it from the system by coupling the spins in an inert singlet, and iii) repeat steps i) and ii) until the desired energy (temperature) scale is reached. We should emphasize that *no* other renormalizations are involved during this decimation procedure. The density of free (undecimated) spins in eq. (3) is then given by $\rho(T) = \rho_0 \int_0^T Q(J) dJ$, where $Q(J)$ is the distribution of the decimated couplings, shown in fig. 2(a) for $n = 0.16$. The distribution of nearest-neighbor couplings, $P(J)$, is also plotted for comparison. Note the dramatic difference between $P(J)$ and $Q(J)$ which stems from the fact that, during the decimation procedure, longer-distance nearest pairs are *unavoidably* generated. Therefore, $Q(J)$ will always be singular yielding the divergence of $\chi(T)$ at low temperatures. As depicted in fig. 1, this simple geometric decimation procedure captures the essential physics of the large- N theory in describing the magnetic susceptibility of strongly disordered Heisenberg spin systems.

Analytic solution. – The geometric decimation procedure will give us a long-sought analytic

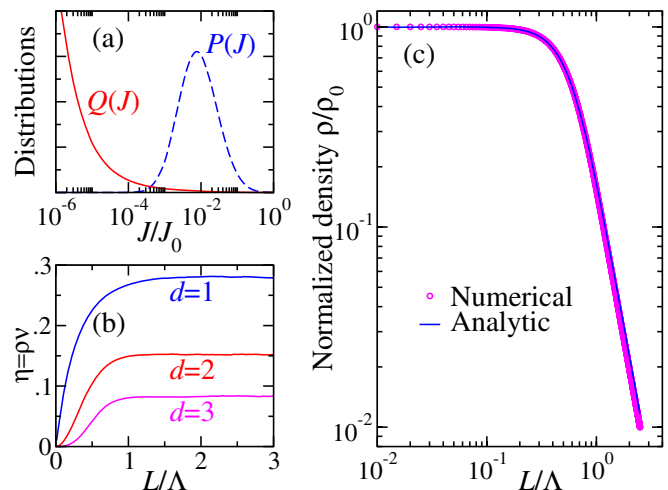


Fig. 2: (Colour on-line) (a) The distributions of the nearest-neighbor couplings $P(J)$ and of decimated couplings $Q(J)$ at concentration $n = 0.16$. (b) Numerical results for the packing fraction $\eta = \rho\nu$ as a function of decimation length L for $d = 1, 2$, and 3 ; $\Lambda = \rho_0^{-1/d}$. (c) Comparison between the numerical and analytic (eq. (5)) results for the free spin density in the geometric decimation procedure for $d = 3$. Here, systems with $N_0 = 4096$ spins averaged over 3000 samples were used (see footnote ²).

description [13,14] of the magnetic properties of insulating DS if one can keep track of ρ as a function of the energy scale $\Omega = \max\{J_{ij}\}$ (defined as the coupling to be decimated) or, equivalently, the length scale $L = \min\{r_{ij}\}$ (the distance between the spins in the pair to be decimated). Although the pair approximations [13] considerably simplify the calculations as compared to the SDRG and numerical cluster calculations, they fail to yield an analytic expression for ρ . On the other hand, the analytic formula proposed by Ponomarev *et al.* involves a tunable parameter [14]. Here we present an accurate analytic solution without any adjustable parameters for a general d -dimensional system.

Since we remove hierarchically the closest spin pair, we can imagine each spin as a hard sphere of diameter L , which naturally incorporates the constraint that no spin pair is closer than L (see footnote ³). By removing the spheres that are touching each other, we continuously increase L until the next closest pair of spins touch each other. The rate equation governing the density of free spins is given by

$$d\rho = -2^d \rho^2 g dv, \quad \text{where } g(\rho) = (1 - \alpha\rho\nu)(1 - \rho\nu)^{-d} \quad (4)$$

is the radial distribution function [15] of a hard-sphere fluid: it gives the ratio of the density of particles at distance r by the mean density, given that there is a particle at the origin. Here, α is a constant which depends only on dimensionality ($\alpha = 0, 0.436$, and 0.5 for $d = 1, 2$

³In the following, we neglect any other correlations beyond those imposed by the hard-sphere constraint.

and 3, respectively) [15], and v is the excluded volume of each hard sphere. The negative sign comes from the fact that ρ decreases as L increases, and the decrease in ρ is proportional to the density of available spins ρ times the probability that two spins (hard spheres) touch each other, *i.e.*, $2^d \rho g dv$. The 2^d factor converts the radius of the hard sphere (raised to the power d) into its diameter.

The solution of eq. (4) can be reduced to a quadrature, from which we can deduce that the packing fraction $\eta = \rho v$ increases monotonically with L , saturating at large length scales at η_c ($\simeq 0.333$, 0.182 , and 0.0968 , respectively, for $d=1, 2$ and 3). The results of a numerical solution of the decimation procedure are shown in fig. 2(b), from which we obtain $\eta_c \simeq 0.2810(5)$, $0.156(1)$, and $0.082(2)$ for $d=1, 2$ and 3 (see footnote ⁴). Since $\eta \ll 1$ throughout the decimation procedure, our hard-sphere liquid remains moderately correlated (away from the strong coupling regime in the vicinity to close packing). This provides a dramatic simplification, since we are now well justified in using the virial expansion $g^{-1} \approx 1 - (d - \alpha)\rho v$ (this linearized expression is exact [15] in $d=1$), and find a closed form solution

$$2^d \gamma \rho v = 1 - (\rho/\rho_0)^\gamma, \quad \text{with } \gamma = 1 + (d - \alpha)/2^d, \quad (5)$$

which satisfies the initial condition $\rho = \rho_0$ at $v = 0$. The magnetic susceptibility in eq. (3) is readily obtained by relating temperature and L via eq. (2), *i.e.*, $2L = a \ln(J_0/T)$. In the $L, v \rightarrow \infty$ ($T \rightarrow 0$) limit, the density decays asymptotically as $\rho \sim v^{-1} \sim L^{-d}$. Thus, the magnetic susceptibility diverges at low temperatures according to

$$\chi(T) \sim \frac{J_0}{T [\ln(J_0/T)]^d}, \quad (6)$$

which can be viewed as a logarithmic correction to the Curie law instead of the power-law divergence usually fitted to experiments. The free spin density ρ extracted from eq. (5) is plotted in fig. 2(c) as a function of L , in excellent agreement with the numerical result of the decimation procedure. Therefore, eq. (5) provides an accurate analytic solution, without any adjustable parameters, to the large- N theory of the insulating DS.

Comparison between SDRG and large- N . – It is now natural to ask how reliable the large- N theory is. To address this issue, we compare the well-known RS solution of the 1D random Heisenberg system obtained by the SDRG method [16,17] with the analytic solution, eq. (5), of the large- N theory. For randomly distributed spins, the length distribution of the nearest-neighbor bonds is a Poissonian $P(L) = \rho_0 \exp(-\rho_0 L)$, which gives rise to a power-law initial coupling constant distribution

$$P_0(J) = \theta(J)\theta(J_0 - J) \frac{\rho_0 a}{2J_0} \left(\frac{J_0}{J}\right)^{1 - \rho_0 a/2}. \quad (7)$$

⁴Small differences between the analytic and the numerical values of η_c reflect the higher-order correlations we have neglected (see footnote ³).

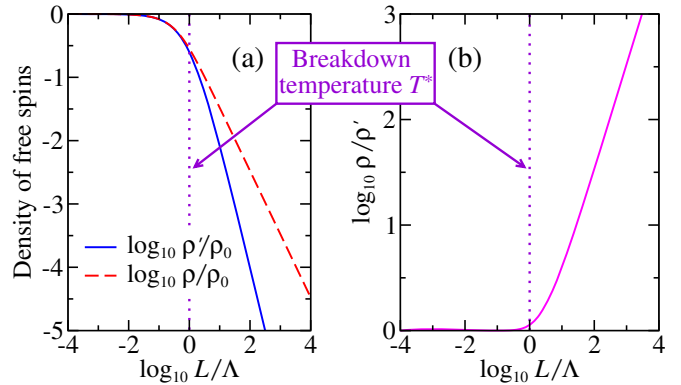


Fig. 3: (Colour on-line) Comparisons between the densities of free (undecimated) spins as functions of the length scale L obtained by the SDRG (ρ') and the large- N (ρ) methods, *i.e.*, the geometric solution, in $d=1$. The vertical dotted line highlights the breakdown temperature T^* (47 mK for $a\rho_c = a/\Lambda_c = 0.25$ (see footnote ⁵)).

In this case, the SDRG flow can be followed exactly through all energy scales, yielding [17]

$$\rho' = \rho_0 \left[1 + \frac{\rho_0 a}{2} \ln(J_0/\Omega)\right]^{-2} = \rho_0 (1 + \rho_0 L)^{-2}, \quad (8)$$

where the prime is added to distinguish this SDRG density from the large- N result in eq. (5). In the asymptotic $L \rightarrow \infty$ limit, $\rho' \sim L^{-2}$, different from the L^{-d} behavior of the large- N theory as shown in fig. 3(a). However, upon close inspection, the L -dependences of ρ and ρ' (see fig. 3(b)) reveal that the breakdown occurs only above a length scale $L^* = 1/\rho_0 = \Lambda$, corresponding to a breakdown temperature

$$T^* = J_0 \exp(-2\Lambda/a) \quad (9)$$

below which the renormalized couplings become important in the SDRG procedure. Above T^* , however, the SDRG theory can be reduced to the simple geometric decimation procedure. The smaller the concentration ρ_0 , the lower T^* is. Since T^* concerns only the energy scale at which the renormalized couplings become important, eq. (9) straightforwardly holds in higher dimensions. Interestingly, this result implies that a large class of highly disordered systems can be described by the random singlet picture above T^* even though their ground states are completely different (as they are in refs. [18]). For instance, $T^* \approx 47$ mK when $a\rho_0^{1/3} = a\rho_c^{1/3} = a/\Lambda_c = 0.25$, assuming $J_0 = 140$ K from ref. [19]⁵. Remarkably, the temperature window relevant for experiments is above the breakdown

⁵Corrections to J_{ij} in eq. (2) may be important in order to compute the precise value of T^* . If one naively inserts the factor of $(r_{ij}/a)^{5/2}$, one gets that $T^* = J_0(\Lambda/a)^{5/2} \exp(-2\Lambda/a)$ instead of eq. (9), which increases T^* by a factor of 32 at the critical concentration. However, note that $\Lambda/a = 4$ is not much greater than 1. Thus, subleading corrections of order $(r_{ij}/a)^2$ [10] become important. To our knowledge, they are not known at the present moment.

temperature (left of the dotted line in fig. 3), which also explains the success of the BL theory.

Finally, we would like to call attention to a caveat on eq. (6). As shown in fig. 3, the experiments take place in a temperature range above T^* in which both the SDRG and the geometric decimation solutions coincide *and* before their asymptotic regimes have been reached. It is thus very clear that either the numerical solution or the analytic one in eq. (5) compare well with experiments. In $d=1$ and above T^* , eq. (5) can be well approximated by eq. (8). Again, it is clear that the apparent power-law divergence of the susceptibility seen in experiments should instead be interpreted as a logarithmic correction to the Curie law.

Summary and outlook. – We have shown how a variational large- N method provides a physically transparent and quantitatively accurate description of inter-site spin correlations on the insulating side of DS. In the presence of strong positional disorder, each localized spin forms a VB singlet with a rather uniquely defined partner, allowing for a closed-form solution of the problem in the large- N limit.

Even more importantly, this approach opens a very attractive avenue to describe the behavior across the MIT. It is known that the large- N RVB approach correctly describes the high density Fermi liquid state [8]. As we established that it also works in the opposite (insulating/Bhatt-Lee) limit, then it will also provide a valid description of the transition by examining the two-orbital t - J model of eq. (1) with finite inter-site hopping t_{ij} . Each f -spin now has more than one choice: to still form a VB singlet with another localized moment, or to undergo Kondo screening by conduction electrons. Similarly as in the large- N solution of the two-impurity Kondo problem [11], we expect Kondo-screened sites to contribute to the formation of a coherent Fermi liquid, while VB singlet pairs to “drop out” from the conduction sea and remain Mott localized. Such gradual conversion of the correlated electron fluid into a localized VB solid may provide a microscopic underpinning for the phenomenological “two-fluid” model [3] —possibly the key missing link for cracking the metal-insulator transition in doped semiconductors.

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