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Strong-Coupling Hubbard Chain[*]

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An exact expression is obtained for the partition function of the arbitrary-electron-density Hubbard chain in the infinite-coupling ($U \to \infty$) limit. It is shown that the magnetic susceptibility obeys a Curie law, while the orbital contribution to the specific heat corresponds to that of a noninteracting band of spinless fermions. The electronic mobilities in this limit are shown to be infinite.

I. INTRODUCTION

One-dimensional systems[1] have received considerable theoretical attention for several reasons. Often problems exactly solvable in one dimension may only be approximately treated in higher dimensionalities. Thus one expects that the insight gained in the exact solution may lead to a better understanding of the underlying physics applicable in other situations. On the other hand, there exist special features of one-dimensional problems which often lead to drastically different results from similar problems, say, in three dimensions. A well-known example is the absence of long-range order in one dimension. Over recent years the discovery of a variety of compounds possessing quasi-one-dimensional structures has lent further impetus to the theoretical investigation of lower-dimensionality models. Some examples of such systems include (1) magnetic insulators such as (TMMC) tetramethylmanganese chloride[2] and various hydrated salts[3]; (2) one-dimensional "metallic" compounds such as the square planar platinum salts[4] and the organic charge-transfer salts based on tetracyanoquinodimethan (TCNQ).[4] It is this latter situation which interests us here. The salt *N*-methylphenizinium (NMP)-TCNQ has been studied in detail by the University of Pennsylvania group[5] and its conducting-insulating transition analyzed in terms of a half-filled-band Hubbard model. Other TCNQ salts may be described as more or less than half-filled bands, e.g., in quinolinium (Q) TCNQ_2, there presumably exists one electron per two TCNQ molecules and thus a one-quarter-filled band. Therefore the one-dimensional Hubbard model with variable electron density is of interest.

The one-dimensional Hubbard-model[6] Hamiltonian which is to be discussed here is

$$\mathcal{H} = -t \sum_{i,\sigma} \left(c^\dagger_{(i+1)\sigma} c_{i\sigma} + c^\dagger_{i\sigma} c_{(i+1)\sigma}\right) + U \sum_i n_{i\uparrow} n_{i\downarrow} \quad , \tag{1.1}$$

where t is the hopping integral which is the matrix element for the transfer of an electron to a nearest-neighbor site; $c_{i\sigma}$, $c^\dagger_{i\sigma}$ are, respectively, annihilation and creation operators for an electron with spin σ at the ith site; U is the local Coulomb repulsion which operates when two electrons occupy the same orbital; $n_{i\sigma} = c^\dagger_{i\sigma} c_{i\sigma}$ is the number operator for electrons on the ith site. We consider only one nondegenerate orbital at each site. Lieb and Wu[7] have solved for the ground state exactly for arbitrary t, U, and electron density ρ. Takahashi[8] has calculated the magnetic susceptibility at $T = 0\,°\mathrm{K}$ for the half-filled band ($\rho = 1$) and Shiba[9] has extended this result to arbitrary density. Ovchinnikov[10] has computed elementary excitations (at $T = 0\,°\mathrm{K}$) of both magnon and single-particle character; Coll[11] has extended his results to arbitrary density. Thus we have a fairly complete description of the $T = 0\,°\mathrm{K}$ properties for all values of the parameters. At finite temperatures, there exist fewer results. For finite chains, of up to six sites with $\rho = 1$, the partition function and thermodynamic properties (specific heat, spin susceptibility) have been computed[12] in both the canonical and grand canonical ensembles for arbitrary U and t. For infinite chains, the statistical mechanics of the noninteracting limit ($U \to 0$) is of course simply a problem of Fermi statistics on a tight-binding chain. The purely local limit ($t \to 0$) is also completely solvable.[13]

The purpose of this paper is to discuss another situation which leads to exact thermodynamics and for which correlation functions associated with dynamical response functions can be calculated. This is the strong-coupling limit, i.e., t and ρ arbitrary but $U \to \infty$. In this case, Sokoloff[14] has already derived an expression for the partition function and shown that the susceptibility obeys a Curie law. By a rather different method, we rederive these results showing that there is a complete decoupling of the orbital and spin degrees of freedom, i.e., (1) the spin susceptibility is a Curie law corresponding to an electron density of $(2 - \rho)$ (for $\rho > 1$) and ρ (for $\rho < 1$); and (2) the specific heat is that for a noninteracting tight-binding band of spinless fermions of density $|\rho - 1|$. Finally we demonstrate that correlation functions associated with dynamical properties can be calculated. As an example, we explicitly compute the velocity-velocity correlation function and show that the mobility is infinite.

In Sec. II, we consider the case of a half-filled band with one extra electron (or hole). This situation has previously been considered by Brinkman and Rice[15] who showed that the mobility is infinite. We shall reconsider their calculation in detail and then generalize the result to arbitrary densities in Sec. III.

II. HALF-FILLED BAND PLUS ONE CARRIER

In this section, we consider a uniform-Hubbard-model chain described by the Hamiltonian (1.1) in the limit $U\rightarrow\infty$ containing one electron per site plus one additional carrier. In particular, we take the extra carrier to be an electron, but by electron-hole symmetry, this is equivalent to adding a hole (i.e., subtracting one electron from the half-filled band). The infinite local repulsion described by $U\rightarrow\infty$ ensures that states of the chain with more than one doubly occupied site are excluded. By projecting out such states explicitly, Brinkman and Rice[15] point out that the resulting Hamiltonian commutes (in one dimension only) with the velocity operator,

$$v=(ita/\hbar)\sum_{i,\sigma}\left(c^\dagger_{(i+1)\sigma}c_{i\sigma}-c^\dagger_{i\sigma}c_{(i+1)\sigma}\right)\ , \tag{2.1}$$

where a is the lattice constant. Thus, the total velocity is a constant of the motion and the mobility is infinite. These authors also explicitly computed the velocity correlation function

$$\phi(\tau)=\langle v(\tau)v(0)+v(0)v(\tau)\rangle\ , \tag{2.2}$$

where $v(\tau)$ is the velocity at time τ and the brackets indicate an ensemble average, and showed that indeed the correlation function $\phi(\tau)$ is independent of time, τ. In this section, we shall reconsider this problem in a slightly different formalism and derive the partition function, yielding the thermodynamic properties, as well as the velocity correlation function. In the following section, the method is easily extended to arbitrary density.

Since we are interested in the strong-coupling situation, it is appropriate to solve the interaction term in the Hamiltonian (proportional to U) exactly and treat the transfer term (proportional to t) by perturbation theory. For $t=0$, each site is singly occupied except for the one doubly occupied site. The energy of the system is then U, which we now take as our zero of energy. Each singly occupied site has spin $\frac{1}{2}$ and thus the application of a magnetic field removes the up-down degeneracy by $\pm\mu_B H\equiv\pm h$ (μ_B is the Bohr magneton and H the external field), where + and − refer, respectively, to spin parallel and antiparallel to the field. For the zero-transfer case, each site is independent and the partition function is then simply

$$Z_0=(2\cosh\beta h)^{N-1}\ , \tag{2.3}$$

where N is the number of sites on the chain. The doubly occupied site is necessarily in a singlet state involving no spin degeneracy and thus does not contribute to the partition function; hence, the exponent $N-1$. The extreme simplicity of Z_0 is a direct consequence of the fact that we need not take into account multiply-occupied sites because $U=\infty$. Denoting the transfer term [the first term of (1.1)] by V, standard formal thermodynamic perturbation theory[16] for the thermodynamic potential Ω gives

$$e^{-\beta(\Omega-\Omega_0)}=1+\sum_{n=1}^{\infty}(-1)^n\int_0^\beta\cdots\int_0^{x_{n-2}}\int_0^{x_{n-1}}dx_n\ldots dx_1 \times\langle V(x_1)V(x_2)\cdots V(x_n)\rangle_0\ , \tag{2.4}$$

where

$$V(x)=e^{x\mathcal{H}_0}Ve^{-x\mathcal{H}_0} \tag{2.5}$$

and $\mathcal{H}_0$ is the unperturbed Hamiltonian, in this case the interaction term [second term of (1.1)]; Ω_0 is the unperturbed thermodynamic potential ($\Omega_0=-\beta^{-1}\ln Z_0$); and the ensemble average in (2.4), $\langle\ \rangle_0$, is with respect to the unperturbed density matrix. The $U=\infty$ limit ensures that the transfer term V has nonvanishing matrix elements only when operating on one of the electrons of the doubly occupied site. Thus the nth-order term of the sum in (2.4) involves the doubly occupied site moving n steps, and because we are calculating a trace the final step must be a return to the initial site. Such a round trip must be independent of the spin configuration of all singly occupied sites because (1) no matter whether a given singly occupied site has spin up or down an electron of the appropriate spin may move onto it, and (2) in one dimension all paths contributing to a diagonal matrix element must be retraced leaving the original spin configuration unaltered. Furthermore, since any motion of the doubly occupied site leaves the interaction energy (or $\mathcal{H}_0$) unchanged, $V(x)$ is independent of x. Only the even terms in V contribute to the sum; the $(2n)$th term is then simply

$$[(\beta t)^{2n}/(2n)!]\,p(2n)\ , \tag{2.6}$$

where $p(2n)$ is the number of distinct paths by which the doubly occupied site may move and return to its original site in $2n$ steps. This must involve n steps to the right and n steps to the left; thus $p(2n)$ is the number of ways one may make n right steps out of $2n$ steps, i.e.,

$$p(2n)=(2n)!/(n!)^2\ . \tag{2.7}$$

Substituting, (2.6) and (2.7) into (2.4), we obtain an exact expression for the partition function,

$$e^{-\beta(\Omega-\Omega_0)}=1+\sum_{n=1}^{\infty}(\beta t)^{2n}/(n!)^2=I_0(2\beta t)\ , \tag{2.8}$$

where $I_0(x)$ is the hyperbolic Bessel function of zero order.[17] The thermodynamic properties are now easily computed from the thermodynamic potential Ω. In particular, since $\Omega - \Omega_0$ is independent of magnetic field, the spin susceptibility χ is completely determined by Ω_0 and is easily shown to be a Curie law corresponding to $(N-1)$ noninteracting spins,

$$\chi = (N-1)\mu_B^2 \beta \ . \tag{2.9}$$

In a finite magnetic field, there exist two contributions to the specific heat: (1) the Schottky-type anomaly associated with the $N-1$ localized spins whose Zeeman levels are split by the field; (2) the kinetic heat capacity arising from the motion of doubly occupied site. The Schottky term is well known and we shall not discuss it further. The kinetic contribution is given by

$$C_v = -\frac{d^2\Omega}{dT^2} = k_B x^2 \frac{d}{dx}\frac{I_1(x)}{I_0(x)} \ , \tag{2.10}$$

where $x = 2\beta t$. For $k_B T \gg t$, $C_v \approx \frac{1}{2} k_B x^2$ and for $k_B T \ll t$, $C_v \approx \frac{1}{2} k_B$. The low-temperature result is just the familiar Dulong–Petit value of $\frac{1}{2} k_B$ per degree of freedom (which is one for a free particle in one dimension). This result obtains because at low temperature the band-structure effects on a single carrier are negligible; the zone boundaries play no role.

Let us now turn our attention to the velocity correlation function $\phi(\tau)$ which should be explicitly independent of time, according to Brinkman and Rice.[15] This quantity is explicitly

$$\begin{aligned} \phi(\tau) &= \langle v(\tau)v(0) + v(0)v(\tau)\rangle \\ &= Z^{-1}\,\mathrm{Tr}\{e^{-\beta\mathcal{H}}[e^{i\mathcal{H}(\tau/\hbar)} v e^{-i\mathcal{H}(\tau/\hbar)} v \\ &\qquad + v e^{i\mathcal{H}(\tau/\hbar)} v e^{-i\mathcal{H}(\tau/\hbar)}]\} \ , \end{aligned} \tag{2.11}$$

where the velocity operator v is given by (2.1). Again rewriting the Hubbard Hamiltonian (1.1) as $H_0 + V$, where the transfer term V is treated by a perturbation expansion to all orders, the correlation function becomes

$$\begin{aligned} \phi(\tau) = Z^{-1}\,\mathrm{Tr}\{&Q(\beta, i\tau/\hbar) v Q(i\tau/\hbar, 0) v \\ &+ Q(\beta + i\tau/\hbar, 0) v Q(0, i\tau/\hbar) v\} \ , \end{aligned} \tag{2.12}$$

where

$$Q(x,y) = \sum_{n=0}^{\infty} Q_n(x,y) \tag{2.13}$$

and

$$\begin{aligned} Q_n(x,y) = (-1)^n \int_y^x dx_1 \int_y^{x_1} dx_2 \cdots \int_y^{x_{n-1}} dx_n \\ \times \langle V(x_1)V(x_2)\cdots V(x_{n-1})\rangle \ . \end{aligned} \tag{2.14}$$

The expression (2.12) for $\phi(\tau)$ can be easily interpreted in the following way: Suppose that the carrier (i.e., the doubly occupied site) is initially at some site which we may take as the origin. The velocity operator causes the carrier to move one step either to the right or left, i.e., $0 \to \pm 1$. The final result must be independent of the sign of the initial step, so we can arbitrarily choose to consider the first step to be to the right $(0 \to 1)$. The development operator Q, because it contains an arbitrary number of transfer operations, allows the carrier to move an arbitrary number of steps to some site q, i.e., $1 \to q$. The velocity operator again generates a shift by one site, i.e., $q \to q \pm 1$. Finally the operator Q must return the carrier to its original location, i.e., $q \pm 1 \to 0$. Thus in the site representation,

$$\begin{aligned} \phi(\tau) = 2Z^{-1} \sum_{q=\infty}^{\infty} \sum_{\eta=\pm 1} [&\langle 0|Q(\beta, i\tau/\hbar)|q+\eta\rangle\langle q+\eta|v|q\rangle \\ &\times \langle q|Q(i\tau/\hbar, 0)|1\rangle\langle 1|v|0\rangle \\ &+ \langle 0|Q(\beta + i\tau/\hbar, 0)|q+\eta\rangle\langle q+\eta|v|q\rangle \\ &\times \langle q|Q(0, i\tau/\hbar)|1\rangle\langle 1|v|0\rangle] \ , \end{aligned} \tag{2.15}$$

where the factor of 2 arises from the fact that the first step may be either to the right or left, the $U = \infty$ condition again forces $V(x)$ in the Q integrals to be independent of x; also the second term in the square brackets of (2.15) is easily seen to be the complex conjugate of the first. Then performing the integrals in (2.14) and using the velocity matrix elements,

$$\langle q+\eta|v|q\rangle = (ita\eta/\hbar) \ , \quad \eta = \pm 1 \tag{2.16}$$

leads us to

$$\begin{aligned} \phi(\tau) = 4\left(\frac{ta}{\hbar}\right)^2 \left(\frac{Z_0}{Z}\right) \mathrm{Re}\Bigg\{ \sum_{q=-\infty}^{\infty} \left[\sum_{n=0}^{\infty} \left(\frac{-it\tau/\hbar}{n!}\right)^n P_n(1,q) \right] \\ \times \left[\sum_{m=0}^{\infty} \frac{[t(i\tau/\hbar - \beta)]^m}{m!} [P_m(q-1,0) - P_m(q+1,0)] \right] \Bigg\} \ , \end{aligned} \tag{2.17}$$

where $P_n(a,b)$ is the number of ways one can move from site b to site a in n steps; the factor Z_0 appears because the motion of the carrier is independent of the over-all spin configuration of the chain. The combinatorial function $P_n(a,b)$ is

$$P_n(a,b) = \frac{n!}{[\frac{1}{2}(n+b-a)]!\,[\frac{1}{2}(n+a-b)]!} \tag{2.18}$$

for $n > |b-a|$ and zero otherwise. Using (2.18), the typical series that occurs in (2.17) is easily summed[17]:

$$\sum_{n=0}^{\infty} \frac{Z^n}{n!} P_n(a,b) = J_{|b-a|}(2iZ)\, i^{|b-a|} \ , \tag{2.19}$$

where $J_\alpha(x)$ is the Bessel function of order α. Inserting (2.19) into (2.17), and after some manipulations,

$$\phi(\tau) = 4\left(\frac{Z_0}{Z}\right)\left(\frac{ta}{\hbar}\right)^2 \mathrm{Re}\Bigg\{ \sum_{q-0}^{\infty} (-1)^q \alpha_q J_q(-x)$$

$$\times[J_{q+2}(y)+2J_q(y)+J_{q-2}(y)]\Big\} \quad , \qquad (2.20)$$

where $\alpha_q = \frac{1}{2}$ for $q=0$, 1 otherwise, and where

$$x = 2t\tau/\hbar \ , \quad y = x + 2it\beta \quad . \qquad (2.21)$$

Finally, the q sum is performed with the aid of the Bessel-function-addition theorem,[18] which yields the result that $\phi(\tau)$ is only a function of $y-x$; i.e., independent of the time τ. Using the partition function, (2.8), the explicit expression for the correlation function is

$$\phi(\tau) = 4(ta/\hbar)^2\{1-[I_2(2t\beta)/I_0(2t\beta)]\} \quad . \qquad (2.22)$$

Thus we indeed find that the mobility is infinite in agreement with Brinkman and Rice.[14] The function ϕ is then simply $2\langle v^2\rangle$, i.e., twice the average square velocity of the carrier which according to (2.22) tends to zero as $T\to 0$ and for $\beta t \ll 1$, tends to $4(ta/\hbar)^2$; i.e., $\langle v^2\rangle \to 2(ta/\hbar)^2$. In the next section we generalize the experience gained in this problem to the case of arbitrary density.

III. ARBITRARY DENSITY

Let us now consider the $U\to\infty$ limit in the situation where we add an arbitrary number of electrons $(2>\rho>1)$ to a half-filled band. Electron-hole symmetry dictates that the identical results should obtain for the physical quantities of interest when $1>\rho>0$. Sokoloff[14] derived an expression for the partition function for this situation by generalizing the method of the Lieb-Wu[7] calculation of the ground-state energy to finite temperature. Our method is to generalize the technique utilized in the preceding section. In particular, we begin with the general expression for the partition function (2.4). In that case where there existed only one carrier (doubly occupied site), the nth term in the perturbation expansion was determined by summing up the contributions from all possible excursions of the carrier such that it returned to its initial site in exactly n steps. For the present situation, we must carry out the same type of analysis with the additional constraint imposed by the Pauli principle that two doubly occupied sites cannot pass through one another. This clearly limits the number of available excursions and greatly complicates the combinatorial problem. However, the path counting can be avoided by noting that it is identical to an already solved problem. Consider a noninteracting tight-binding chain $(U=0)$ which contains a density Γ of electrons with all spins parallel. In principle, we may compute the partition function for this problem by using (2.4) and summing to all orders. Notice that Fermi statistics imposes the same paths to count for this problem as for the $U=\infty$ problem with a density $\rho = 1+\Gamma$. Thus, we are immediately led to the result

$$Z = Z_0 Z' \quad , \qquad (3.1)$$

where Z_0 is the partition function for the singly occupied sites:

$$Z_0 = \left|2\cosh(\beta h)\right|^{N(2-\rho)} \qquad (3.2)$$

and Z' is the partition function for $n=N(\rho-1)$ spinless fermions in a tight-binding band. The magnetic field dependence of the partition function is entirely in Z_0 leading immediately to the Curie susceptibility

$$\chi = N(2-\rho)\mu_B^2\beta \quad . \qquad (3.3)$$

Note that $\chi\to\infty$ as $T\to 0$ in agreement with Shiba's[9] calculation that $\chi(0)\to\infty$ as $U\to\infty$. On the other hand, the properties based on the orbital motion are completely associated with the spinless doubly occupied sites. The fermion partition function in the grand canonical ensemble is given by

$$Z' = \mathrm{Tr}\, e^{-\beta(\mathcal{H}-\mu\mathfrak{N})} \quad , \qquad (3.4)$$

where the chemical potential μ is determined by

$$\mathfrak{N} = N(\rho-1) = \sum_k f(\epsilon_k) \quad . \qquad (3.5)$$

Here $f(x)$ is the usual fermion function

$$f(x) = 1/(e^{\beta(x-\mu)}+1) \qquad (3.6)$$

and ϵ_k denotes the energy spectrum of the noninteracting tight-binding band,

$$\epsilon_k = 2t\cos k \ , \quad -\pi < k < \pi \quad . \qquad (3.7)$$

The internal energy W is calculated in the usual way,

$$W = \sum_k \epsilon_k f(\epsilon_k) \quad , \qquad (3.8)$$

which using (3.6) and (3.7) is

$$W = \frac{2tN}{\pi}\int_{-1}^{1} \frac{yf(y)}{(1-y^2)^{1/2}}\, dy \quad . \qquad (3.9)$$

The specific heat $C_v = \partial W/\partial T$ is easily computed numerically and is shown in Fig. 1 for several densities. Let us compute the ground-state energy W_g explicitly. First the Fermi momentum k_F at $T = 0$ °K is found from (3.5),

$$\mathfrak{N} = \sum_k f(\epsilon_k) = \frac{N}{\pi}\int_0^{k_F} dk = \frac{Nk_F}{\pi} \qquad (3.10)$$

or $k_F = \pi(\rho-1)$. The ground-state energy is then

$$W_g = \sum_k \epsilon_k f(\epsilon_k) = -(2tN/\pi)\int_0^{k_f} \cos k\, dk$$

$$= -(2tN/\pi)\sin k_F = (2tN/\pi)\sin\pi\rho \quad , \qquad (3.11)$$

which may be written as $W_g = -(2tN/\pi)|\sin\pi\rho|$ and is identical to the $U=\infty$ limit of Shiba's calculation.[9] At infinite temperature, $f(\epsilon_k)$ must be in-

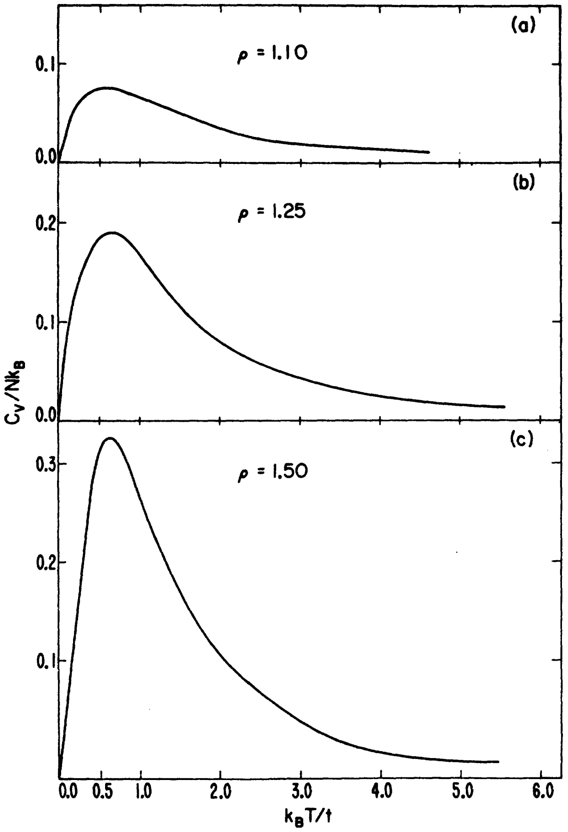

FIG. 1. The orbital contribution to the specific heat in zero external field for various values of the electron density ρ.

dependent of k, leading to $W(\infty)=0$.

As in the case of one carrier considered previously, the mobility is infinite. This follows directly from the argument given by Brinkman and Rice[15] that the velocity operator commutes with the Hamiltonian when $U=\infty$. The mean square velocity is

$$\langle v^2\rangle=\left(\frac{a}{\hbar}\right)^2\sum_k\left(\frac{\partial\epsilon_k}{\partial k}\right)^2 f(\epsilon_k)$$

$$=\left(\frac{2ta}{\hbar}\right)^2\sum_k \sin^2 k f(\epsilon_k)\quad . \tag{3.12}$$

At absolute zero, using the Fermi momentum given in (3.10),

$$\langle v^2\rangle_0=(2ta/\hbar)^2(N/2)[\rho-1-(2\pi)^{-1}\sin 2\pi\rho]\quad , \tag{3.13}$$

which for $\rho=1+N^{-1}$ gives zero, in agreement with the result of Sec. II for one extra carrier. In the infinite-temperature limit all $f(\epsilon_k)$'s are equal to $\rho-1$, leading to

$$\langle v^2\rangle_\infty=(2ta/\hbar)^2(N/2)(\rho-1)\quad , \tag{3.14}$$

which for $\rho-1+N^{-1}$ again agrees with the result of Sec. II.

As we have seen the $U=\infty$ limit of the arbitrary-density Hubbard chain is exactly soluble, at least for its thermodynamic quantities. The basic result is that the magnetic and orbital properties are completely decoupled. The magnetic behavior is that of a set of localized spins while the kinetic effects correspond to the appropriate density of spinless noninteracting fermions. Future studies will consider the use of these exact results as a guide to obtain good approximations for finite but large U situations.

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