Dilution Effects in Two-Dimensional Quantum Orbital Systems

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Interacting orbital degrees of freedom in a Mott insulator are essentially directional and frustrated. In this Letter, the effect of dilution in a quantum-orbital system with this kind of interaction is studied by analyzing a minimal orbital model which we call the two-dimensional quantum compass model. We find that the decrease of the ordering temperature due to dilution is stronger than that in spin models, but it is also much weaker than that of the classical model. The difference between the classical and the quantum-orbital systems arises from the enhancement of the effective dimensionality due to quantum fluctuations.

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Orbital degree of freedom (ODF) in transition-metal (TM) compounds has attracted much interest for more than a decade [1,2]. ODF in a TM ion, denoted by M, corresponds to a direction of the electronic cloud. In a single M-M bond, ODF is usually quenched so as to gain bond energy. On the other hand, in a solid, equivalent M-M bonds with different directions can coexist. When orbitals are arranged to gain bond energy for one direction, this configuration is not fully favorable for other bonds. This causes a type of frustration in a system with ODF. This frustration is often lifted by the order-by-disorder mechanism due to thermal fluctuations, and a long-range order (LRO) appears at finite temperature (T). The e_g orbital systems in the perovskite lattice, such as $LaMnO_3$ and $KCuF_3$, may correspond to this situation.

Recently, much attention has been focused on quantum effects in the orbital systems. ODF is described by a quantum operator, e.g., in the case of the e_g orbital, pseudospins (PS) $\mathbf{T}_{\mathbf{r}_i}$ with an amplitude of $1/2$ where large quantum fluctuations are expected. The quantum fluctuations often release frustration even at $T = 0$ by the quantum order-by-disorder mechanism, and induces LRO. "Orbital liquid states," proposed in a ferromagnetic $La_{1-x}Sr_xMnO_3$ [3] and in a Mott insulating $LaTiO_3$ [4], are other examples of the quantum-orbital states. Here, LRO is prevented due to reduction of an effective dimensionality and large quantum fluctuations. This arises from an interplay between the directional and frustrated interactions and quantum effects.

Interesting aspects are expected when disorder is introduced into the quantum-orbital systems. Disorder of our interest implies that ions with ODF are replaced randomly by other ions without ODF. Disorder effects in the quantum spin systems have been studied intensively and extensively. Doping of nonmagnetic impurities into the gapped spin liquids is one of the examples, where quantum spin fluctuations are suppressed and a classical antiferromagnetic (AF) LRO emerges [5,6]. In the same way, an impurity without ODF may destroy the quantum resonance of orbitals and stabilize LRO. In a different way from the spin systems, it is expected that quantum fluctuation weakens the anisotropic and directional character of the classical LRO. This may be crucial for dilution effects and the percolation scheme. Thus, doping of disorder is a good way to reveal the interplay of the directional and frustrated interactions and the quantum-orbital fluctuations.

In this Letter, we examine disorder effects in a quantum-orbital model, termed the two-dimensional orbital-compass model (OCM) [7–9]. This is a minimal quantum-orbital model involving the characteristics in the orbital system, i.e., the directional and frustrated interaction. A long-range correlation for one of the PS components appears along the vertical or horizontal directions in a square lattice, termed the directional order (DO). We focus on quantum effects in the orbital dilution by using the quantum Monte Carlo (QMC) method. We have found that quantum fluctuations from the classical DO state enhance the effective dimensionality of the system and makes DO robust against dilution.

An interacting orbital system in a Mott insulator is generally described by the following model [10,11]

$$\mathcal{H} = -2J\sum_{i,\ell}(\mathbf{T}_{\mathbf{r}_i}\cdot\hat{m}^{\ell})(\mathbf{T}_{\mathbf{r}_i+\hat{\ell}}\cdot\hat{m}^{\ell}). \tag{1}$$

PS for ODF is defined by $\mathbf{T}_{\mathbf{r}_i} = \frac{1}{2}\sum_{\gamma s} d^{\dagger}_{\mathbf{r}_i\gamma s}\sigma_{\gamma\gamma'}d_{\mathbf{r}_i\gamma' s}$, where $d_{\mathbf{r}_i\gamma s}$ is the electron annihilation operator with spin s ($=\uparrow,\downarrow$) orbital γ ($=\alpha,\beta$) at site $\mathbf{r}_i$. The subscript ℓ indicates the bond direction in a crystal, and the factor $\hat{m}^{\ell}$ is the unit vector in the PS space. This interaction explicitly depends on the bond direction ℓ [1,10]. When we take $\hat{m}^{\ell} = [\sin(\frac{2n_{\ell}}{3}\pi), 0, \cos(\frac{2n_{\ell}}{3}\pi)]$ with $(n_x, n_y, n_z) = (2, 1, 0)$, Eq. (1) describes the three-dimensional e_g-orbital system [10,12]. OCM in a two-dimensional square lattice [11] of the present interest corresponds to another choice of $\hat{m}^{\ell} = \hat{e}_{\ell}$ with the unit vector $\hat{e}_{\ell}$. The explicit form of OCM associated with the orbital-less impurities is given as

$$\mathcal{H} = -2J\sum_{i,\ell=(x,z)} T^{\ell}_{\mathbf{r}_i}T^{\ell}_{\mathbf{r}_i+\hat{\ell}}\varepsilon_{\mathbf{r}_i}\varepsilon_{\mathbf{r}_i+\hat{\ell}}. \tag{2}$$

Here, $\varepsilon_{\mathbf{r}_i}$ takes one for an orbital ion and zero for an impurity. Since the sign of the interaction J is gauged away by rotating $\mathbf{T}_{\mathbf{r}_i}$ with respect to the y axis, we take it

to be positive. This Hamiltonian without dilution ($\varepsilon_{\mathbf{r}_i} = 1$ for $\forall\ i$) is invariant under the following two-symmetry operations [8,13]. (i) The global fourfold symmetry: PS's at all sites and the crystal lattice are rotated by $\pi/2$, simultaneously, with respect to the y axis. (ii) The local symmetry at each column and row: the z (x) component of all PS's at each column (row) along the z (x) axis are flipped, i.e., $T^{z(x)}_{r_x,r_z} \rightarrow -T^{z(x)}_{r_x,r_z}$ for each $r_x(r_z)$. In addition, in the classical uniform PS state, there is the following continuous symmetry: (iii) the global PS direction is arbitrary in the T^x-T^z plane.

Let us explain in more detail the symmetry (ii) which is unique in OCM. This symmetry operation at a row r_z is done by the operator $P_{r_z}(\equiv \prod_{r_x} \sigma^z_{r_x,r_z})$, with the Pauli matrices $\sigma_{\mathbf{r}}$, as $P^{-1}_{r_z}\sigma^x_{r_x,r_z}P_{r_z} = -\sigma^x_{r_x,r_z}$ and $P^{-1}_{r_z}\sigma^z_{r_x,r_z}P_{r_z} = \sigma^z_{r_x,r_z}$. On an equal footing, the symmetry operation at a column r_x is done by the operator $Q_{r_x} = \prod_{r_z} \sigma^x_{r_x,r_z}$. The operators P_{r_z} and Q_{r_x} commute with $\mathcal{H}$, and P_{r_z}'s commute with each other as well as Q_{r_x}'s, but $[Q_{r_x}, P_{r_z}] \neq 0$. The energy eigenvalues are characterized by the eigenvalue set of P_{r_z}'s $(p_1, \ldots, p_L)$ or that of Q_{r_x}'s $(q_1, \ldots, q_L)$ in a $L \times L(= N)$ square lattice. That is, at least, there are L conserved quantities. Since $P^2_{r_i} = 1$ and $P^{\dagger}_{r_i} = P_{r_i}$, $p_i = \pm 1$. It is easily shown that the conventional site-diagonal order parameter $N^{-1}\sum_{\mathbf{r}}\langle \mathbf{T}_{\mathbf{r}}\rangle$ is not invariant under this local-symmetry operation. The Elitzur's theorem states that the only operators with a nonzero expectation value are locally gauge invariant. According to this theorem, it is shown that the site-diagonal LRO does not occur [14]. Instead, a kind of LRO, termed the directional order, appears. This order corresponds to a breaking of the global fourfold symmetry (i) explained above. This nematiclike order has been studied by using the correlation function $C^{zz}_{r_x} \equiv \lim_{r_z\rightarrow\infty}\langle T^z_{r_x 0} T^z_{r_x r_z}\rangle$ at $T = 0$ [13] and $q = N^{-1}\sum_{\mathbf{r}}(T^{z2}_{\mathbf{r}} - T^{x2}_{\mathbf{r}})$ in the classical OCM [7]. We introduce D, as an order parameter of DO, given by $D = \frac{1}{N(1-x)}\sum_{\mathbf{r}}(T^z_{\mathbf{r}} T^z_{\mathbf{r}+\hat{\mathbf{z}}}\varepsilon_{\mathbf{r}}\varepsilon_{\mathbf{r}+\hat{\mathbf{z}}} - T^x_{\mathbf{r}} T^x_{\mathbf{r}+\hat{\mathbf{x}}}\varepsilon_{\mathbf{r}}\varepsilon_{\mathbf{r}+\hat{\mathbf{x}}})$. This represents the correlation of T^z along the z axis or that of T^x along x. Since D commutes with P_{r_z} and Q_{r_x}, being different from $\mathbf{T}_{\mathbf{r}}$, it is possible for $\langle D\rangle$ to be finite.

To examine OCM numerically with and without vacancies, we adopt QMC method for a finite-size cluster. There is no negative-sign problem in OCM. The QMC calculations have been performed on a square lattice of $L \times L$ sites ($L = 14$–18) with periodic-boundary conditions. The Suzuki-Trotter formula is applied to decomposition of the density matrix, $e^{-\beta\mathcal{H}} = \lim_{n\rightarrow\infty}(\Pi_i e^{-\beta\mathcal{H}_i/n})^n$, where we assume $\mathcal{H}$ is decomposed into the local part as $\mathcal{H} = \sum_i \mathcal{H}_i$. The Trotter number n is chosen to be from 12 to 22 and the extrapolation of the results to $n = \infty$ is performed. At low temperatures for $T < 0.04J$, we perform the simulation with $n = 60$–72. We take, at maximum, 10 000 MC steps for measurement after 3000 MC steps for thermalization. Physical quantities are averaged over 20–60 MC samples at each parameter set. We have checked by MC calculations with 10^6 MC steps, which is much longer than the integrated autocorrelation time determined by the binning analyses, that the order parameter values in the present calculation are reliable within numerical errors. The ordering temperature (T_{DO}) of DO is estimated from the finite-size scaling analyses of the fourth-order cumulant of the order parameter. This is termed the Binder cumulant defined by $Q = 1 - \frac{\langle D^4\rangle}{3\langle D^2\rangle^2}$ [7,15]. It is known that Q-T curves for several system size L cross at the critical temperature T_{DO}.

First we show the numerical results in OCM without vacancies. Temperature dependence of $\sqrt{\langle D^2\rangle}$ [$\equiv \bar{D}(T)$] for several L is presented in Fig. 1(a). Around $T/J = 0.17$, $\bar{D}(T)$ grows with decreasing T and is saturated to about 0.13 below $T/J = 0.12$. With increasing L, this dependence becomes steep, but the saturated value of $\bar{D}(T)$ at low T does not change much. Below $T/J = 0.07$, $\bar{D}(T)$ seems to decrease. We have performed careful numerical calculations with the larger Trotter numbers $n = 60$–72. As shown in the inset of Fig. 1(a), $\bar{D}(T)$ takes about 0.125 down to $T/J = 0.015$. Therefore, the reduction of $\bar{D}(T)$ in low T is an artifact due to the small n. Temperature dependence of the Binder cumulant Q is presented in Fig. 2. From the definition, we expect that, in the limit of $L = \infty$, $Q = 0$ for $T > T_{\mathrm{DO}}$ and $Q = 2/3$ for $T < T_{\mathrm{DO}}$. The Q-T curves for the several system sizes cross around $T/J = 0.15$ corresponding to the temperature where $\bar{D}(T)$ grows. We conclude that DO is realized in the quantum OCM and $T_{\mathrm{DO}} = 0.150 \pm 0.003$. The saturated value of $\bar{D}(T)$ at low T is about half of the classical value of $1/4$. This reduction is much stronger than that in the quantum AF Heisenberg model with $S = 1/2$ in a square lattice [16]. That is, DO in quantum OCM is not similar to the simple classical picture. This is due to large quantum fluctuations which induce correlations between columns or rows. We note that the site-diagonal order parameter $M \equiv \sqrt{\langle (N^{-1}\sum_{\mathbf{r}}\mathbf{T}_{\mathbf{r}})^2\rangle}$ de-

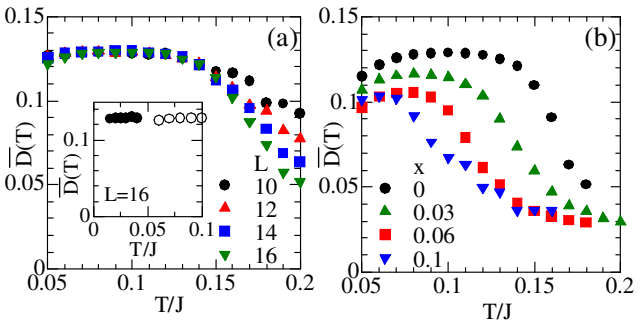

FIG. 1 (color online). (a) Temperature dependence of the directional order parameter $\bar{D}(T)$ at $x = 0$ for various system sizes L. The inset shows $\bar{D}(T)$ at low temperatures obtained in the calculation with the large Trotter numbers (●). ○ are the same data with those in the main panel. (b) $\bar{D}(T)$ for various impurity concentrations.

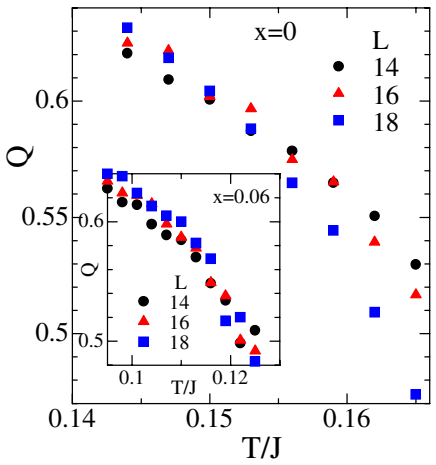

FIG. 2 (color online). Temperature dependence of the Binder cumulant Q at $x = 0$ for various system sizes L. Inset shows temperature dependence of Q at $x = 0.06$.

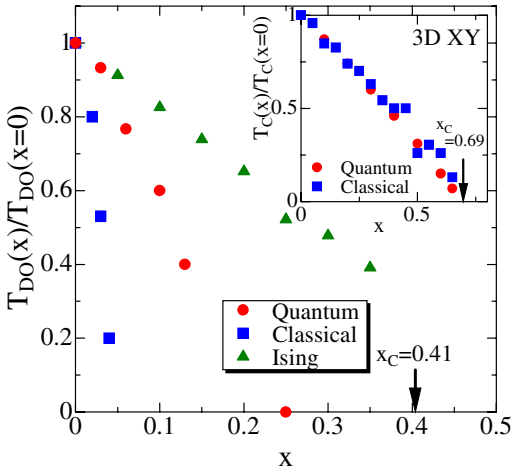

FIG. 3 (color online). Impurity concentration x dependence of the ordering temperature. ● are for T_{DO} in the quantum OCM, and ■ are for T_{CL} in the classical OCM. For comparison, the ordering temperature in the Ising model (▲) are also plotted. Bold arrow indicates the percolation threshold x_c in a square lattice. The inset shows the ordering temperature T_C in the quantum XY model with $S = 1/2$ (●) and that in the classical one (■) in a three-dimensional cubic lattice. System size is chosen to be $N = 32^3$ at maximum. The continuous imaginary-time algorithm is utilized.

creases with increasing L. This is in contrast to the e_g-orbital model [8,12,17].

Now we present the dilution effects in OCM. The results on $\bar{D}(T)$ for several x are shown in Fig. 1(b). We observe that $\bar{D}(T)$ gradually increases with decreasing T and saturates to about 0.1 at $x = 0.06$. The temperature where $\bar{D}(T)$ rises is reduced with doping. The saturated value of $\bar{D}(T)$ becomes small by doping in spite of the factor $(1 - x)^{-1}$ in the definition of D. The analyses for Q work well at $x = 0.06$, as shown in the inset of Fig. 2, where T_{DO}/J is identified to be 0.113 ± 0.005. We have checked that, at $x = 0.25$, the Q-T curves for several L do not cross each other down to $T = 0.01J$.

The main result of the present work, the x dependence of T_{DO} in the quantum OCM, is shown in Fig. 3. For comparison, we also plot the ordering temperature T_{CL} of DO in the classical OCM, and the Curie temperature T_{Ising} on the $S = 1/2$ Ising model on a square lattice. We observe that T_{DO} decreases monotonically with x and seems to take zero around $x = 0.15$ ($\equiv x_c$). As well as the diluted e_g orbital systems [12,18], the critical concentration x_c is much smaller than the percolation threshold 0.41 on the square lattice [19]. It is worth noting that the decrease of T_{DO} is much stronger than that in T_{Ising}, but is weaker than T_{CL}. In other words, quantum effects make DO robust against dilution.

To understand the dilution effects in the quantum OCM, we present a rigorous treatment of the dilute OCM in two-coupled chains along the z axis, that is, a two-leg ladder OCM. As mentioned previously, the energy eigenstates are classified by the eigenvalue set $(p_1, \ldots, p_L)$ of the operator $P_{r_z} = \sigma^z_{r_L r_z}\sigma^z_{r_R r_z}$, where $r_R(r_L)$ indicates the x coordinates of the right (left) chain in the ladder. We consider one of the doubly degenerate ground states: the state characterized by $p_1 = \cdots = p_L = 1$ which is degenerate with the state by $p_1 = \cdots = p_L = -1$. In a row r_z of the ladder, the eigenstates are $|T^z_{r_L r_z} T^z_{r_R r_z}\rangle = |\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$, or their linear combinations $|\pm\rangle \equiv \frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle \pm |\downarrow\downarrow\rangle)$. There are relations $(T^x_{r_L r_z} T^x_{r_R r_z})|\pm\rangle = \pm\frac{1}{4}|\pm\rangle$, $T^z_{r_L r_z}|\pm\rangle = \frac{1}{2}|\mp\rangle$, and $T^z_{r_R r_z}|\pm\rangle = \frac{1}{2}|\mp\rangle$. Therefore, within the subspace of $p_1 = \cdots = p_L = 1$, we introduce a new PS operator $\mathbf{S}_{r_z}$, and show correspondences $(T^x_{r_L r_z} T^x_{r_R r_z}) \to \frac{1}{2} S^z_{r_z}$ and $T^z_{r_L(r_R) r_z} \to S^x_{r_z}$. The state $|+\rangle$ ($|-\rangle$) is the up- (down-) eigenstate of $S^z_{r_z}$. As a result, OCM in this subspace is mapped onto the transverse-Ising model in a single chain [20]: $\mathcal{H}_{\mathrm{eff}} = -4J\sum_{r_z} S^x_{r_z} S^x_{r_z+1} - J\sum_{r_z} S^z_{r_z}$. Now, one vacancy is introduced at a site (r_L, r^0_z). Since, in the row r^0_z, we have $P_{r^0_z} = \sigma^z_{r_R r^0_z}$, the energy eigenstates in this row, $|-\uparrow\rangle$ and $|-\downarrow\rangle$, belong to the different subspaces, and $T_{r_R r^0_z}$ is replaced by a C number in each subspace. Thus, OCM in the two-leg ladder with one vacancy is denoted by the following model: $\mathcal{H}_{\mathrm{eff}} = -4J{\sum_{r_z}}' S^x_{r_z} S^x_{r_z+1} - J\sum_{r_z \neq r^0_z} S^z_{r_z} \pm J(S^x_{r^0_z+1} + S^x_{r^0_z-1})$, where ${\sum_{r_z}}'$ indicates a sum of r_z except for r^0_z and $r^0_z - 1$. The sign $+$ ($-$) in the third term corresponds to the state $|-\downarrow\rangle$ ($|-\uparrow\rangle$) at the site (r_R, r^0_z). This corresponds to the model of the two transverse-Ising chains where the external field $\pm J$ along the S^x axis is applied at edges of the chains, i.e., the $r^0_z + 1$ and $r^0_z - 1$ sites. In the original language, this field acts on $T^z_{r_R(r_L), r^0_z+1}$ and $T^z_{r_R(r_L), r^0_z-1}$, and it enhances the correlation $\langle T^z_{\mathbf{r}_i} T^z_{\mathbf{r}_j}\rangle$ around the vacancy in OCM. This situation does not depend on the sign of the third term in $\mathcal{H}_{\mathrm{eff}}$, since $T^z = 1/2$

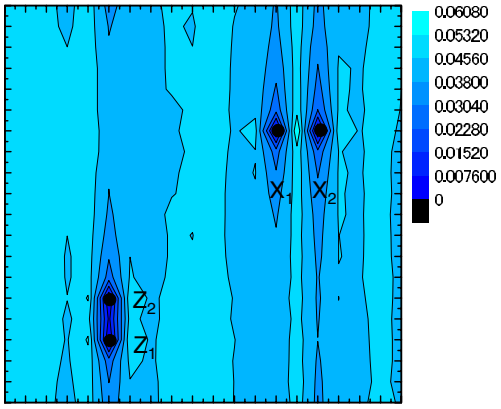

FIG. 4 (color online). Contour map of local static correlation function $C^{zz}(\mathbf{r}_i)$ in a 20×20 site cluster. Four vacancies $X_{1(2)}$ and $Z_{1(2)}$ are represented by ●. Temperature is chosen to be $T/T_{\mathrm{DO}}(x=0) = 1.27$.

$(-1/2)$ is favored in the case of $-J$ $(+J)$, and the correlation is enhanced in both of the cases.

Based on the above picture, we discuss first the difference between T_{DO} and T_{Ising} in Fig. 3. Consider a situation in a two-dimensional square lattice where two vacancies are located closely in a same row. As suggested above, the correlation of T^z is enhanced around the vacancies. On an equal footing, the correlation of T^x is enhanced around the vacancies located closely in the same column. A certain number of domains, where T^z or T^x correlation is enhanced, coexist in a system, and competition between them reduces $\bar{D}(T)$ and T_{DO}. To confirm this picture, we calculate the local PS correlation function around the impurity defined by $C^{zz}(\mathbf{r}_i) = \frac{1}{N(1-x)}\sum_j \langle T^z_{\mathbf{r}_i} T^z_{\mathbf{r}_j}\rangle$ (Fig. 4). Two pairs of vacancies introduced in a 20×20 site cluster are denoted by $X_{1(2)}$ and $Z_{1(2)}$ in the figure. It is clearly shown that $C^{zz}(\mathbf{r}_i)$ is enhanced between X_1 and X_2, and almost vanishes between Z_1 and Z_2. These are not seen in dilute spin models.

Now we focus on quantum effects in dilution, i.e., discrepancy between T_{DO} and T_{CL}. In the classical DO state at $T = 0$, the system is decoupled into the independent Ising chains along the x or z axis, although DO is the two-dimensional order where the fourfold symmetry is broken. The rapid decrease of T_{CL} by dilution reflects the quasi-one-dimensional nature of DO, and at finite T, weak two dimensionality is recovered due to the thermal fluctuations. The quantum fluctuations from the classical DO state bring about a coupling between the independent chains and increases the effective dimensionality of DO even at low T. This causes a type of resonant states between the independent chains, and thus DO becomes robust against dilution. This remarkable quantum effect is unique in the orbital system; we compare the x dependence of the Curie temperatures T_C in the quantum and classical XY models in a cubic lattice (the inset of Fig. 3). It is seen that the difference between the two are much smaller than that in OCM.

In summary, we have studied the dilution effects in the quantum-orbital system where the interaction is directional and frustrated. As a minimal quantum-orbital model, the two-dimensional OCM is analyzed by using the QMC method. The magnitude of the DO parameter is largely reduced from its classical value. That is, a classical picture of DO, i.e., a vertical (horizontal) alignment of T^z (T^x), is considerably modified by quantum fluctuations. The reduction of the ordering temperature due to dilution is stronger than that in the spin model, but is much weaker than that of the classical OCM. Quantum fluctuations increase the effective dimensionality of the system and make the order robust against dilution. The present results are in contrast to the conventional sense that quantum fluctuations destroy LRO.

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