Spin-fluctuation-mediated $d+id'$ pairing mechanism in doped β-MNCl (M=Hf,Zr) superconductors

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We study two types of models for the superconducting layered nitride β-MNCl (M=Hf,Zr); a single-band model on a triangular lattice, and a two-band model on a honeycomb lattice. We find that the former model does not suffice as an effective model while the latter one can be a good candidate. We propose from the study on the two-band model a possibility of spin-fluctuation-mediated $d+id'$-wave superconductivity in the doped β-MNCl. We show that the relatively high T_c obtained in the doped band insulator is a characteristic feature of the spin-fluctuation-mediated superconductivity on a honeycomb lattice. We also find that the gap anisotropy on the Fermi surface strongly increases upon increasing the doping concentration, and the interlayer hopping suppresses superconductivity. These results are in qualitative agreement with the experimental findings.

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I. INTRODUCTION

Layered nitride β-MNCl (Ref. 1) (M=Hf,Zr) doped with carriers is one of the most interesting group of superconductors. The mother compound β-MNCl is composed of alternate stacking of honeycomb (HC) MN bilayer and Cl bilayer.[2] This is a band insulator and becomes a superconductor upon doping electrons by Na or Li intercalation. They exhibit relatively high T_c up to $\sim$25 K for M=Hf and $\sim$15 K for M=Zr. The bilayer HC lattice structure consisting of M and N is considered to be playing the main role in the occurrence of superconductivity, and the two-dimensional nature of the superconductivity has been revealed by nuclear magnetic resonance (NMR) (Ref. 3) and muon spin relaxation (μSR) studies.[4,5]

Despite the relatively high T_c, experimental as well as theoretical studies indicate extremely low density of states (DOS) at the Fermi level.[6–8] In fact, they have the highest T_c among materials with the specific-heat coefficient γ as small as $\sim$1 mJ/mol K^2. The electron-phonon coupling is also estimated to be weak,[6,8–10] and the isotope effect is found to be small.[11,12] These experiments suggest that some kind of unconventional pairing mechanism may be at work, but on the other hand, NMR knight shift measurement suggests spin-singlet pairing,[11] and the tunneling spectroscopy[13] and specific-heat[7] experiments find a fully open, seemingly s-wave-like gap. However, regarding this fully open gap, recent experiments show that the anisotropy of the gap increases with doping. Namely, when the doping concentration is small, the specific-heat coefficient increases linearly as a function of the magnetic field, suggesting an isotropic gap,[14] while a steep increase in the coefficient at low magnetic field is observed for higher doping.[7,14] Also, $2\Delta/(k_BT_c)$ determined from the specific-heat measurement ranges from $\sim$5 in the lightly doped regime to less than $\sim$3 in the heavily doped regime,[14] which may be an indication that the maximum and minimum values of the gap have a substantial difference when heavily doped, and the specific heat is mainly governed by the minimum value. Moreover, a recent superfluid density measurement by μSR shows that the gap is nearly isotropic when the doping concentration is small, while the anisotropy increases for large doping.[15] Very recently, absence of coherence peak in the spin-lattice-relaxation rate has been found in an NMR experiment,[16] again suggesting unconventional pairing, most probably with some kind of sign change in the superconducting gap. Furthermore, for Li_xHfNCl, an intercalation of organic molecules tetrahydrofuran (THF) between the layers is found to enhance T_c.[17]

Given these experimental circumstances, here we study two types of models for the doped β-MNCl, a two-band model on a HC lattice, and a single-band model where the nitrogen site is effectively integrated out. While the latter model does not seem to suffice for explaining the relatively high T_c in this series of material, the former one provides a possibility of spin-fluctuation-mediated d-wave superconductivity in the doped β-MNCl.[18] Spin-fluctuation-mediated pairing in a doped *band insulator* may sound surprising and odd at first sight since one cannot expect strong spin fluctuations. Nevertheless, we show that the relatively high T_c despite the low DOS and the temperature-independent spin susceptibility is a characteristic feature of the spin-fluctuation-mediated superconductivity of a doped band insulator on the HC lattice. The most probable pairing state below T_c is the $d+id'$ state, whose gap anisotropy on the Fermi surface is found to be strongly enhanced upon increasing the carrier concentration. We also study the effect of the three dimensionality, where the interlayer hopping suppresses superconductivity.

II. FORMULATION

We first obtain the effective model for β-MNCl. Although the material consists of bilayer HC lattice, here we consider models on a single-layer lattice, and try to reproduce the bands that lie close to the Fermi level in the first-principles band calculations. The simplest one is a single-band model on a triangular lattice, in which the nitrogen site degrees of freedom is effectively integrated out, so that there is only one site per unit cell. This is a model that corresponds to the single-band Hubbard model for the cuprates, where the oxygen $2p_x$, $2p_y$ degrees of freedom is integrated out from the

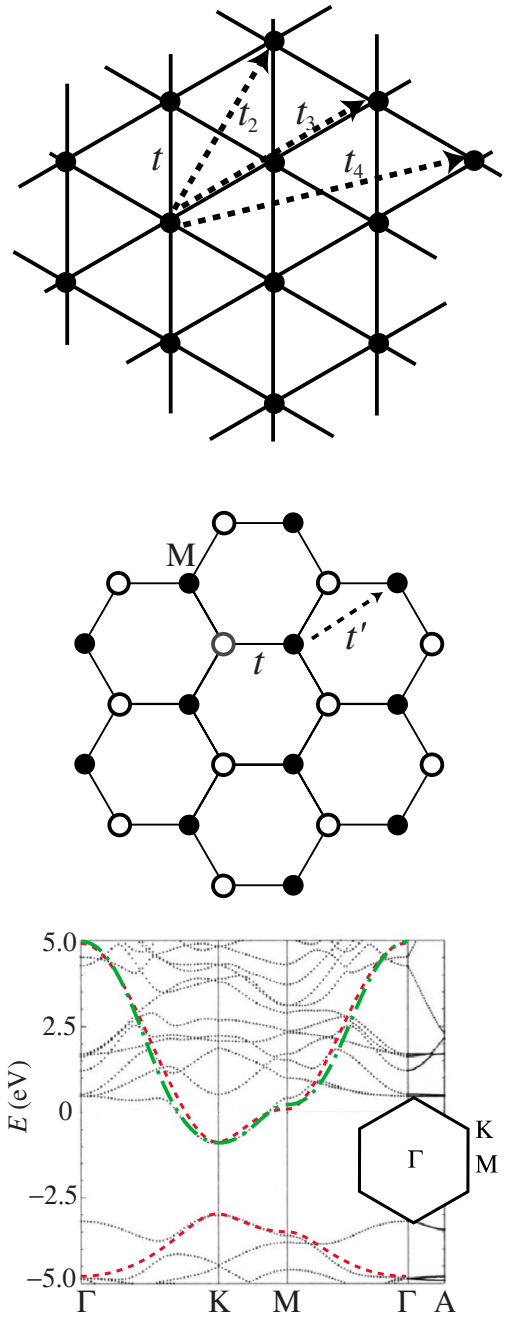

FIG. 1. (Color online) Upper: the single-band model on a triangular lattice. Middle: the two-band model on a honeycomb lattice. Lower: the first-principles band calculation taken from Ref. 8 and the band dispersion of the single-band model (dash-dotted green) and the two-band model (dashed red).

original three-band d-p model. By considering hopping integrals up to fourth nearest neighbors (Fig. 1, upper panel), we can reproduce the single band that crosses the Fermi level in the first-principles band calculations.[8,10,19,20] The band dispersion of the single-band model with $t=0.65$ eV, $t_2/t=-0.03$, $t_3/t=0.06$, and $t_4/t=-0.025$ is shown by the green dash-dotted line in Fig. 1.

To go a step further, we also consider a model on a single-layer HC lattice consisting of alternating "M" and "N" orbitals with a level offset, which roughly reproduces the conduction and valence bands closest to the Fermi level, as shown by the dashed lines in the middle panel of Fig. 1. Here we take the nearest-neighbor (M-N) hopping $t=1.2$ eV, the level offset $\Delta/t=2.7$, and the next-nearest-neighbor (M-M) hopping $t'/t=0.35$.

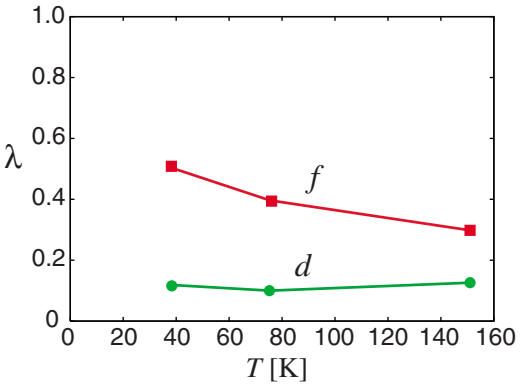

FIG. 2. (Color online) Eigenvalue of the Eliashberg equation as functions of temperature for f-wave and d-wave pairings in the single-band model.

The on-site interaction $U/t=6$ is considered in the single-band model, and the on-site interaction $U/t=6$ is introduced on both M and N orbitals in the two orbital model.[21] The band filling n is defined as the number of electrons/number of sites. For the single-band model, $n=2x$, where x is the Li or Na content, since there are two M sites per unit cell, which results in bonding and antibonding bands, and the electrons are doped only in the bonding band. For the two-band model, $n=1$ corresponds to the nondoped case, and x and n is related by $n=1+x$. We use fluctuation exchange (FLEX) method,[22–25] which is kind of a self-consistent random-phase approximation, to obtain the Green's function. Then we solve the linearized Eliashberg equation,

$$\lambda\phi_{lm}(k) = -\frac{T}{N}\sum_{k'}\sum_{l',m'} V_{lm}(k-k')G_{ll'}(k')$$
$$\times G_{mm'}(-k')\phi_{l'm'}(k').$$

Here, G is the Green's function matrix (with $l,m,\ldots$ labeling sites in a unit cell) obtained by FLEX. V is the spin-singlet pairing-interaction matrix given by $V=\frac{3}{2}U^2\chi_{\rm sp}-\frac{1}{2}U^2\chi_{\rm ch}$ with the spin- and charge-susceptibility matrices $\chi_{\rm sp(ch)}(q)=\chi_{\rm irr}(q)[1-(+)U\chi_{\rm irr}(q)]^{-1}$, where $\chi_{\rm irr}(q)$ is the irreducible susceptibility matrix $\chi_{\rm irr}(q)=-\frac{1}{N}\Sigma_k G(k+q)G(k)$ (N is the number of k-point meshes). In the following, the maximum eigenvalue of the spin-susceptibility matrix will be referred to as the spin susceptibility. We take up to 128×128 k-point meshes and up to 16384 Matsubara frequencies. The eigenvalue of the Eliashberg equation λ increases upon lowering the temperature and reaches unity at $T=T_c$.

III. SINGLE-BAND MODEL: CALCULATION RESULTS

We first present calculation results for the single-band model. In Fig. 2, the eigenvalue of the Eliashberg equation is shown as functions of temperature for spin-singlet and

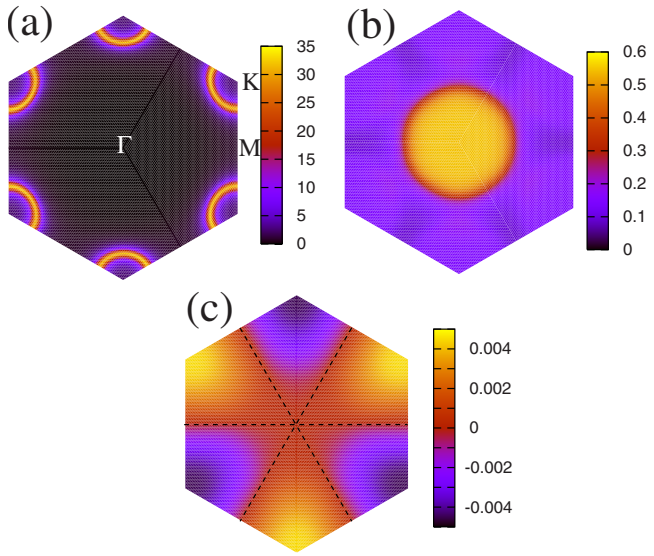

FIG. 3. (Color online) The contour plots of the FLEX result at the lowest Matsubara frequency for the single-band model in the hexagonal Brillouin zone with $n=0.2$ and $T=0.02t$. (a) The Green's function of the upper band squared, (b) the spin susceptibility, and (c) the superconducting gap.

-triplet pairings for $n=0.2$. It can be seen that the triplet pairing strongly dominates over singlet pairing. The gap of the spin-triplet pairing has an f-wave form, where the nodes of the gap do not intersect the Fermi surface (the ridge of the Green's function squared) as shown in Fig. 3. On the other hand, the singlet pairing has a d-wave gap (not shown). The possibility of this type of spin-triplet f-wave pairing was, in fact, proposed on a canonical triangular lattice in Ref. 26. The triplet pairing dominates over singlet pairing because ferromagnetic spin fluctuations arise due to the dilute band filling as shown in Fig. 3(b), and also because the nodes of the f-wave gap do not intersect the Fermi surface.

Nonetheless, as can be seen from Fig. 2, the temperature at which the f-wave eigenvalue reaches unity, if any, seems to be very low. We have also tried a model that considers the nearest-neighbor off-site interaction V in addition to U. In this case, strong charge fluctuations arise, which makes the triplet vs singlet competition more subtle, but in any case, the eigenvalue of the Eliashberg equation remained small even when the magnitude of V is close to the point where the charge-susceptibility diverges. Considering the experimental fact that the T_c is relatively high in the doped β-MNCl, and also that the pairing occurs in the spin singlet channel,[11] we believe that the single-band model does not suffice as an effective model for β-MNCl.

IV. TWO-BAND MODEL: CALCULATION RESULTS

A. Superconducting transition temperature

We now move on to the two-band model. In this model, we find that spin-singlet pairing strongly dominates over triplet pairing. In Fig. 4, we plot T_c of the singlet-pairing superconductivity as a function of the band filling. It can be seen that $T_c \sim 30$ K is obtained, which can be considered as

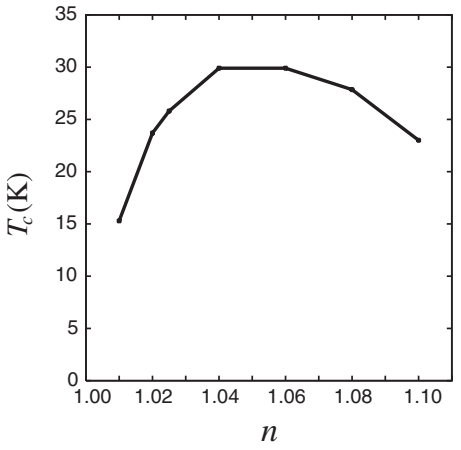

FIG. 4. T_c plotted as a function of the band filling for the two-band model on the HC lattice.

relatively high noting that $T_c \sim 100$ K (assuming $t \sim 0.4$ eV appropriate for the cuprates) is obtained by the same method for the Hubbard model on the square lattice.[22,25] The difference between the single- and two-band models is dramatic in that not only the T_c but even the leading pairing symmetry is different. This is in contrast to the case of the cuprates, where the single-band Hubbard and three-band d-p models is expected to give roughly the same conclusions.

The high T_c obtained in this model can be understood as a combination of several features of this model. Let us start with a canonical HC lattice, where $t'=0$ and $\Delta=0$. We show in Fig. 5(a) the FLEX result of the maximum value of the spin susceptibility as a function of temperature for the band filling of $n=1.08$. The spin susceptibility is nearly independent of T,[14,27] which is in sharp contrast with the Hubbard model on the square lattice. For example, for the square lattice with $n=0.7$, we have a strong enhancement of the spin susceptibility upon lowering the temperature. With further hole doping to $n=0.65$, the spin susceptibility is suppressed, but even there, the spin susceptibility moderately increases upon lowering the temperature. Also in Fig. 5(a), we show the eigenvalue λ of the linearized Eliashberg equation as functions of temperature for the HC and square lattices. T_c is the temperature where λ reaches unity. The DOS at E_F is nearly the same for the square lattice with $n=0.65$ and the HC lattice with $n=1.08$, and also the spin susceptibility has similar values at low temperature, but still, the HC lattice has higher T_c.[28]

So what is the origin of this high T_c? Figure 6(a) shows the contour plot of the Green's function squared, whose ridge corresponds to the Fermi surface, which consists of disconnected two-dimensional pockets centered around K and K′ points. The spin susceptibility in Fig. 6(b) is maximized at wave vectors that bridge the opposite sides of each pieces of the Fermi surface. As can be seen in Fig. 6(c) [and more clearly in Fig. 6(d)] the gap has a d-wave form, where the gap changes sign across the wave vector at which the spin susceptibility is maximized. In contrast to the case of the square lattice, one of the nodes of the d-wave gap does not intersect the Fermi surface because of its disconnectivity, i.e.,

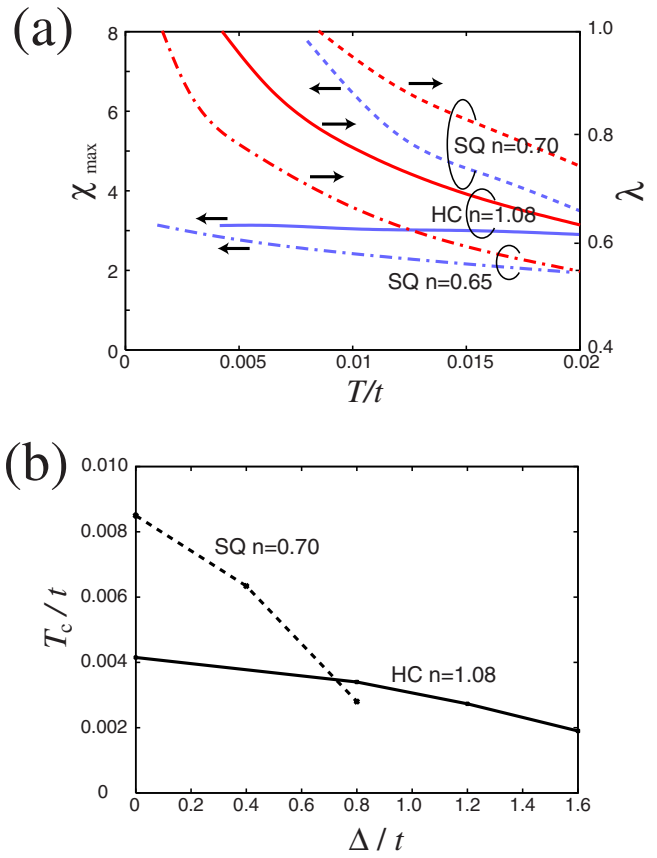

FIG. 5. (Color online) (a) The maximum value of the spin susceptibility (blue, left axis) and the eigenvalue of the Eliashberg equation (red, right) as functions of temperature for the square lattice with $n=0.7$ (dashed) or $n=0.65$ (dash dotted) and for the honeycomb lattice (solid) for $n=1.08$. $U=6t$ in all cases. HC and SQ stand for the honeycomb (with $t'=0$) and the square lattices, respectively. The temperature at which $\lambda=1$ is the T_c. (b) T_c as functions of the level offset Δ obtained by FLEX+Eliashberg equation for the square lattice with $U=6t$, $n=0.7$ (dashed) or for the HC lattice with $U=6t$ and $n=1.08$ (solid).

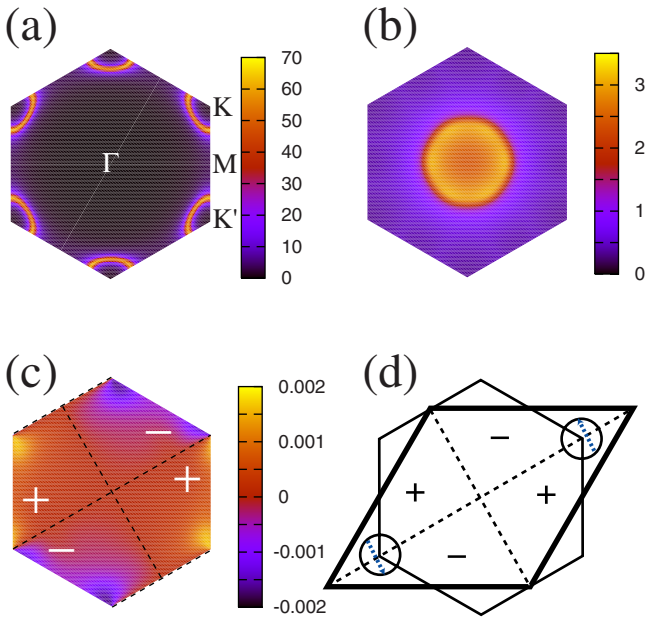

FIG. 6. (Color online) The contour plots of the FLEX result at the lowest Matsubara frequency for the honeycomb lattice ($t'=\Delta=0$) in the hexagonal Brillouin zone with $n=1.08$ and $T=0.01t$. (a) The Green's function of the upper band squared, (b) the spin susceptibility, (c) the superconducting gap of the upper band, and (d) the Fermi surface (the two circles) and the sign of the gap function are schematically shown in the extended zone scheme. The dashed arrows represent the wave vectors at which the spin fluctuations develop.

the gap is like "p wave" if we focus only on one of the Fermi surfaces. Since smaller number of gap nodes on the Fermi surface is favorable for superconductivity,[29] this can be a reason for high T_c despite the low DOS and the weak spin fluctuations.

We now introduce the M-N level offset Δ as in the model for β-MNCl. As shown in Fig. 5(b), we find that superconductivity is relatively robust against the introduction of Δ. This again is in sharp contrast with the square lattice, where finite Δ strongly suppresses superconductivity. The weak effect of Δ may be because the DOS of the HC lattice is small near the band center, so that the introduction of Δ, which opens up a gap at the center of the band, is less effective than in the case of the square lattice, where the DOS diverges at the band center for $\Delta=0$.

Let us now comment on the origin of the difference between the single- and two-band models, namely, low-T_c triplet pairing in the former model and the high-T_c singlet pairing in the latter. The difference mainly comes from the difference in the band filling, i.e., for the single-band model, the band filling is dilute, favoring ferromagnetic spin fluctuations, while in the two-band model, it is near half filling in favor of antiferromagnetic spin fluctuations. Of course, if the level offset Δ is large enough, the two-band case should tend to the single-band case, as have been shown in Ref. 26 but in the present case, the magnitude of Δ is small enough to retain the nature of the $\Delta=0$ situation.

B. Gap function

Apart from the T_c, another important issue is the form of the superconducting gap. By symmetry, there are two degenerate d-wave gaps, and the most probable form of the gap below T_c is the form $d+id'$, where the two d-wave gaps mix with a phase shift of $\pi/2$.[30–32] Since the two d-wave gaps have nodal lines at different positions, this kind of mixture leads to a gap that has a finite absolute value on the entire Fermi surface. The $d+id'$ form of the gap can be constructed from the d-wave gap obtained by solving the linearized Eliashberg equation. In the upper panels of Fig. 7, we show the contour plot of the $d+id'$ gap for the case of $n=1.06$ and 1.16 for the model for β-MNCl.

To clearly see the anisotropy on the Fermi surface, we plot the magnitude of the gap (normalized by the maximum value) in the lower panel of Fig. 7. It can be seen that the $d+id'$ gap is nearly isotropic for low-doping concentration, while it becomes more and more anisotropic for higher doping. The $d+id'$ pairing scenario is consistent with the experimental findings that the superconductivity occurs in the spin

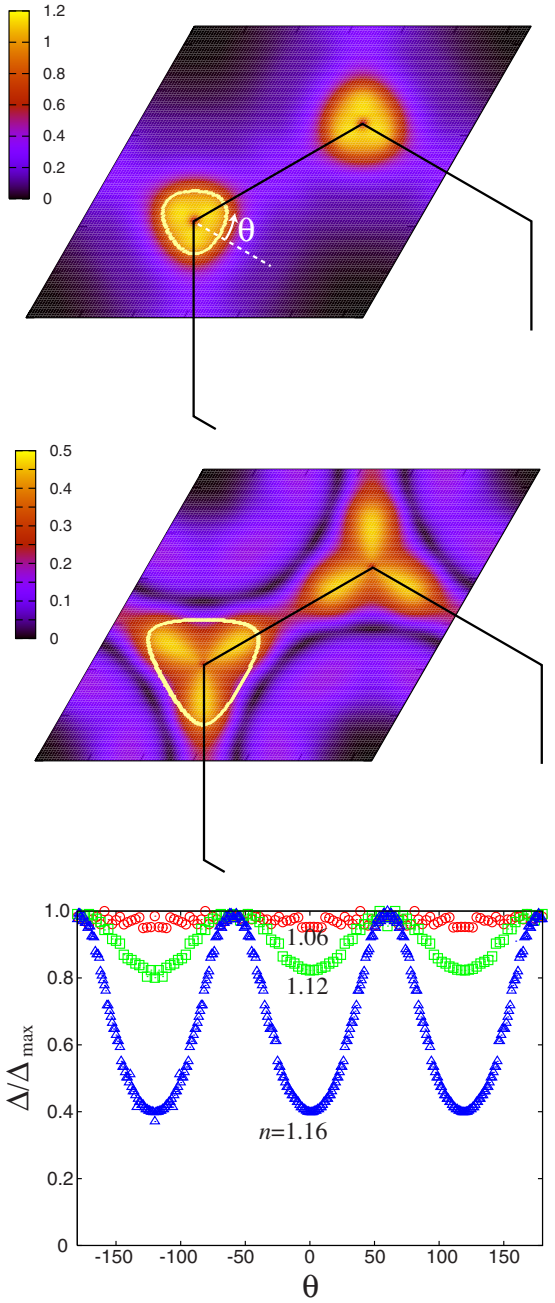

FIG. 7. (Color online) Upper two panels: the contour plots of the $d+id'$ gap for $n=1.06$ (top) and $n=1.16$ (middle). The model for β-MNCl is used. The yellow (light) solid line (closed around the Brillouin zone edge) represents the Fermi surface. Lower panel: the normalized $d+id'$ gap along the Fermi surface. θ is defined in the top panel.

singlet,[11] s-wave-like channel,[7,13,14] while the coherence peak in the spin-lattice-relaxation rate is absent.[16] It is also consistent with the experimental finding that the gap anisotropy increases upon doping.[14,15] Since the $d+id'$ state breaks time-reversal symmetry, it is interesting to experimentally investigate such a possibility.

C. Effect of dimensionality

Finally, we consider the effect of dimensionality by adopting a three-dimensional model where the two-band model on the HC lattice is connected by vertical (M-M and N-N) hopping integrals t_z (inset of Fig. 8). Here, we take up to $32\times32\times32$ k-point meshes and up to 8192 Matsubara frequencies. In Fig. 8, we plot T_c as a function of t_z for the band filling of $n=1.08$. As can be seen, the three dimensionality suppresses T_c, which is in agreement at least qualitatively with the experimental finding that the intercalation of THF molecules between the layers enhances T_c.[17]

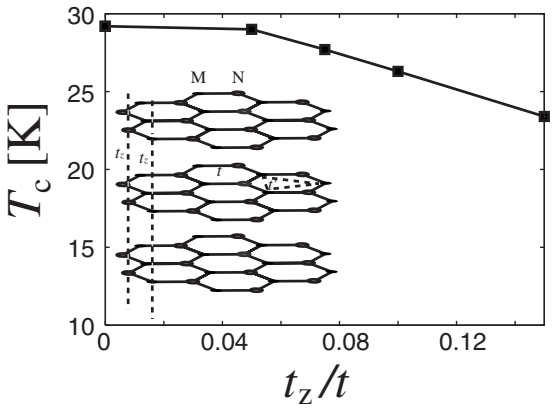

FIG. 8. T_c plotted as a function of t_z for the model shown in the inset. $t=1.2$ eV, $t'/t=0.35$, $\Delta/t=2.7$, $U/t=6$, and $n=1.08$.

In fact, the suppression of T_c with increasing three dimensionality is a general trend for spin-fluctuation-mediated pairing as has been studied in Refs. 33–35. Namely, since the pairing interaction is large around a certain wave vector in spin-fluctuation-mediated pairing, the fraction of the volume where the pairing interaction is large in the Brillouin zone becomes smaller as the three dimensionality increases.

We note here that although the introduction of t_z reduces the T_c, the reduction itself is not drastic (consistent with the experiments), and also the features of the spin susceptibility or the superconducting gap obtained for the purely two-dimensional model are not altered qualitatively. Therefore, the essence of the system is captured already within the two-band model on the two-dimensional honeycomb lattice.

V. CONCLUSION AND FUTURE PROBLEMS

In the present study, we have studied two kinds of models for the doped β-MNCl. We have found that the single-band model does not suffice as a minimal effective model of the material. On the other hand, from the study on the two-band model, we have proposed a possibility of spin-fluctuation-mediated $d+id'$-wave superconductivity in the doped β-MNCl. We have shown that the relatively high T_c is obtained as a characteristic feature of the spin-fluctuation-mediated superconductivity in a doped band insulator on the HC lattice. We have also found that the gap anisotropy on the Fermi surface strongly increases upon increasing the doping concentration, and the interlayer hopping suppresses superconductivity. These results are in qualitative agreement with the experimental findings.

As opposed to the present proposal, the possibility of charge-fluctuation-mediated pairing has been considered for this material from the early days.[18] As mentioned earlier, we

found it difficult to have charge-fluctuation-mediated pairing in the *single-band* model that considers the nearest-neighbor off-site repulsion, at least in the FLEX approximation. On the other hand, we have not investigated the effect of off-site repulsions in the two-band model, namely, the possibility of charge-fluctuation-mediated pairing in the two-band model is not ruled out. Nonetheless, even if the charge fluctuation plays a role in the occurrence of the superconductivity, we believe that the $d+id'$ is the most probable pairing state in this material considering the good agreement with various experiments.

Another remaining issue is the curious doping dependence of the superconductivity. For Li_xZrNCl, T_c shows an increase upon lowering the carrier concentration until a sudden disappearance of the T_c and a superconductor-insulator transition is observed.[36] Recently, this increase in T_c has been shown to be correlated with the increase in the uniform spin susceptibility, which can be considered as a support for the present spin-fluctuation scenario.[14] In fact, although the spin fluctuation is nearly *temperature* independent in our model (so that the development of the spin fluctuation upon lowering the temperature need not be observed experimentally), it is strongly *doping* dependent and is enhanced upon lowering the doping rate.[14] Nonetheless, the origin of the *sudden* drop of T_c at a certain doping rate cannot be explained within the present two-band model (see Fig. 4) and remains as an open problem. Also, the almost constant behavior of T_c in the heavily doped regime[17] is puzzling since for such a large doping, the Fermi level hits the second band and a large difference in the DOS should be detected (which is not the case experimentally). A rigid band picture may not be valid in the heavily doped regime, as suggested in some experiments[37] and band calculations.[38]

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