

# Nuclear spin relaxation rate of disordered $p_x+ip_y$ -wave superconductors

Qiang Han<sup>1,2</sup> and Z. D. Wang<sup>1,2,\*</sup>

<sup>1</sup>Department of Physics, University of Hong Kong, Pokfulam Road, Hong Kong, China

<sup>2</sup>Department of Materials Science and Engineering, University of Science and Technology of China, Hefei 230026, China

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Based on an effective Hamiltonian with the binary alloy disorder model defined in the triangular lattice, the impurity scattering effects on the density of states and especially on the spin-lattice relaxation rate  $1/T_1$  of  $p_x+ip_y$ -wave superconductors are studied by solving numerically the Bogoliubov-de Gennes equations. In the clean limit, the coherence peak of  $1/T_1$  is observed as expected. More intriguingly, for strong scattering potential, the temperature dependence of  $1/T_1$  exhibits the two different power-law behaviors near  $T_c$  and at low temperatures, respectively, which is qualitatively consistent with the nuclear quadrupolar resonance measurement of the newly discovered superconductor  $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$  ( $x=0.35$ ). We argue that the disorder effect plays an important role in the thermodynamic properties of the  $p_x+ip_y$ -wave pairing state as indicated in this paper, as well as other superconducting states with unconventional pairing symmetries. Therefore further experimental exploration is expected to determine the actual pairing symmetry of this material.

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The superconductor  $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$  ( $x=0.35$ ), recently studied by Takada *et al.*,<sup>1</sup> has stimulated much theoretical and experimental interest in studying its spin and orbital symmetries of the Cooper pairs, which helps to explore and understand the underlying superconducting mechanism of this material. At present this issue is hotly debated, and there still exist controversies both theoretically and experimentally. Resonating valence bond<sup>2</sup> theories of the triangular lattice  $t$ - $J$  model<sup>3–5</sup> describe  $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$  as an electron-doped Mott insulator based on the fact that the  $\text{Co}^{4+}$  atoms has spin- $\frac{1}{2}$  as  $\text{Cu}^{2+}$  in high- $T_c$  cuprates. Such theories prefer the spin-singlet  $d+id'$ -wave pairing ( $d=d_{x^2-y^2}$  and  $d'=d_{xy}$ ). On the other hand, theories based on a combined symmetry analysis with fermiology<sup>6</sup> favor triplet over singlet pairing with the  $\mathbf{d}$ -vector perpendicular to the cobalt plane [most probably  $\mathbf{d}=\hat{\mathbf{z}}(p_x \pm ip_y)$ , i.e.,  $p_x \pm ip_y$ -wave pairing state]. As for the experimental aspect, a series of nuclear magnetic/quadrupolar resonance (NMR/NQR) measurements<sup>7–10</sup> also give divergent implication about the pairing symmetry in  $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ . Namely, for spin pairing Ref. 8 found the signature for the triplet pairing while Ref. 9 supported the singlet pairing, although both examined the temperature dependence of the Knight shift. For the orbital wave function of the Cooper pairs, a fully-gapped superconducting state<sup>7,8</sup> is inferred from the existence of the Hebel-Slichter coherence peak<sup>11</sup> of the spin-lattice relaxation rate ( $1/T_1$ ), while the opposite conclusion is drawn from the NQR experiment<sup>10</sup> that  $1/T_1$  decreases across  $T_c$  without the coherence peak and follows a power-law dependence deviating the exponential relation, which implies a nodal gap function.

Motivated by the novel power law behavior reported in Ref. 10 and the fact that various impurities or defects, such as intrinsic disorder associated with Na vacancies,<sup>12</sup> oxygen vacancies, substitution atoms in the  $\text{CoO}_2$  layer, etc., are present during the synthesis and handling of this compound, in this paper we investigate and elucidate the effect of disorder on the nuclear spin-relaxation rate of the  $p_x \pm ip_y$ -wave superconductor. Our results indicate that the seemingly in-

compatible experimental observations of the temperature dependence of  $1/T_1$  may be qualitatively understood within the picture of the *disordered* chiral  $p_x+ip_y$ -wave pairing state if the effect of impurities is properly considered.

Here we employ a mean-field Bogoliubov-de Gennes (BdG) Hamiltonian on a tight-binding triangular lattice with the nearest-neighbor (NN) hopping integral  $t$  and the NN bond pairing potential  $\Delta_{ij}$  resulting from the effective attractive interaction  $V$ . The model Hamiltonian,<sup>13,14</sup> which captures the essential physics of the present system with impurities, may be expressed as

$$\begin{aligned} H_{\text{eff}} = & - \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_{i,\sigma} (\epsilon_i - \mu) c_{i\sigma}^\dagger c_{i\sigma} \\ & + \sum_{\langle i,j \rangle} [\Delta_{ij} (c_{i\uparrow}^\dagger c_{j\downarrow}^\dagger + c_{i\downarrow}^\dagger c_{j\uparrow}^\dagger) + \text{h.c.}], \end{aligned} \quad (1)$$

where  $t_{ij}=t$  is the NN-hopping integral. In the remainder of this paper, we choose  $t<0$  according to the analysis on the band calculation<sup>15</sup> and the energies will be measured in unit of  $|t|$ .  $\mu$  is the chemical potential. Here, we adopt the binary alloy disorder model<sup>16</sup> where  $\epsilon_i$  is the  $\delta$ -functionlike *scalar* scattering potential and takes the value  $U_0$  on certain lattice sites according to the impurity concentration  $n_{\text{imp}}$  and zero elsewhere. The spin-triplet pairing potential  $\Delta_{ij}$  is defined as  $\Delta_{ij}=(V/2)(\langle c_{i\uparrow} c_{j\downarrow} \rangle + \langle c_{i\downarrow} c_{j\uparrow} \rangle)$ . Note that only the  $d_z$  component of the spin-triplet pairing is considered here in view of the experimental indication that the  $\mathbf{d}$  vector is parallel to  $z$ -axis according to the invariant behavior of the Knight shift for the in-plane magnetic field.<sup>8</sup> In the homogenous case with electron doping, the  $p_x+ip_y$ -wave pairing state is stable against the  $d+id'$ -wave in the triangle-lattice,<sup>14</sup> which is expressed as

$$\begin{aligned} \Delta_{p_x+ip_y}(\mathbf{k}) = & 2\Delta_p [\sin(k_x) + \sin(k_x/2)\cos(\sqrt{3}k_y/2) \\ & + i\sqrt{3}\cos(k_x/2)\sin(\sqrt{3}k_y/2)], \end{aligned} \quad (2)$$

where  $\Delta_p=(1/6N)\sum_{i,\delta}\Delta_{ii+\delta}e^{-i\theta(\delta)}$  with  $i+\delta$  the six NN sites

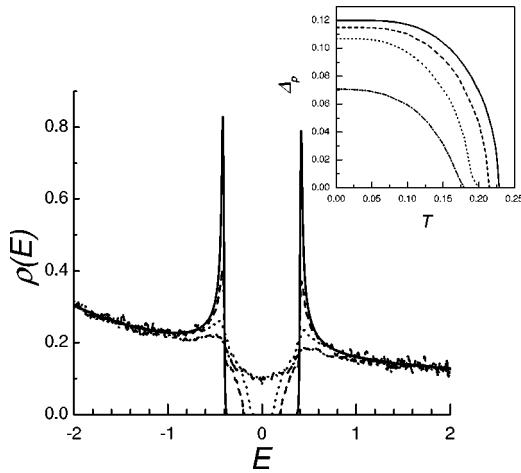


FIG. 1. Density of states as a function of energy of the disordered  $p_x+ip_y$ -wave pairing state for the clean limit (solid line),  $n_{\text{imp}}=2\%$ ,  $U_0=2$  (dashed line),  $n_{\text{imp}}=5\%$ ,  $U_0=2$  (dotted line),  $n_{\text{imp}}=5\%$ , and  $U_0=10$  (dash-dotted line). Also shown in the inset is the temperature dependence of the impurity-averaged order parameter  $\Delta_p(T)$ .

of  $i$ . By applying the self-consistent mean-field approximation and performing the Bogoliubov transformation, diagonalization of the Hamiltonian  $H_{\text{eff}}$  can be achieved by solving the following BdG equations:

$$\sum_j \begin{pmatrix} H_{ij} & \Delta_{ij} \\ -\Delta_{ij}^* & -H_{ij}^* \end{pmatrix} \begin{pmatrix} u_j^n \\ v_j^n \end{pmatrix} = E_n \begin{pmatrix} u_j^n \\ v_j^n \end{pmatrix}, \quad (3)$$

where  $u^n$ ,  $v^n$  are the Bogoliubov quasiparticle amplitudes with corresponding eigenvalue  $E_n$ .  $H_{ij}=-t_{ij}+\delta_{ij}(\epsilon_i-\mu)$ .  $\Delta_{ij}$  is calculated according to  $\Delta_{ij}=(V/4)\sum_n(u_i^n v_j^{n*}-u_j^n v_i^{n*})\tanh(E_n/2k_B T)$ . Throughout this work, we set  $V=2.3$  and  $\mu=1.0$  which gives rise to  $\Delta_p=0.12$  and the electron number per site is 1.39 in the absence of disorder.<sup>17</sup> Due to the vanishingly small anisotropic factor of the  $p_x+ip_y$ -wave pairing,<sup>18</sup> there is one  $s$ -wavelike full gap opened at approximately  $\Delta_{\text{Gap}}\approx 0.4$  (see Fig. 1 for the clean limit). We find that the energy-band width is 9.0 and the Fermi energy  $E_F\approx 3.8$  as measured from the bottom of the band, both being in one order of magnitude larger than the superconducting energy gap  $\Delta_{\text{Gap}}$ . Therefore, our numerical investigation does not violate the weak-coupling BCS theory as indicated by the characteristic quantity  $2\Delta_{\text{Gap}}/k_B T_c\approx 3.51$ .

Once the self-consistent quasiparticle spectrum is obtained, the nuclear spin relaxation rate is calculated according to,<sup>19</sup>

$$\begin{aligned} R(i,j) &= \text{Im } \chi_{-,+}(i,j, i\Omega_n \rightarrow \Omega + i0^+)/(\Omega/T)|_{\Omega \rightarrow 0} \\ &= \pi \int \int [\rho_{11}^{ij}(E)\rho_{22}^{ij}(-E') - \rho_{12}^{ij}(E)\rho_{21}^{ij}(-E')] \\ &\quad \times f(E)[1-f(E')]\delta(E-E')dEdE', \end{aligned} \quad (4)$$

$$R(i,i) = -\pi T \int_{-\infty}^{\infty} \rho_{11}^{ii}(E)\rho_{22}^{ii}(-E)f'(E)dE, \quad (5)$$

where  $\rho_{\alpha\beta}^{ij}(E)$  is expressed as

$$\begin{pmatrix} \rho_{11}^{ij}(E) & \rho_{12}^{ij}(E) \\ \rho_{21}^{ij}(E) & \rho_{22}^{ij}(E) \end{pmatrix} = \sum_n \begin{pmatrix} u_i^n u_j^{n*} & u_i^n v_j^{n*} \\ v_i^n u_j^{n*} & v_i^n v_j^{n*} \end{pmatrix} \delta(E-E_n).$$

For the unconventional pairing, the contributions from the off-diagonal elements  $\rho_{12}$  and  $\rho_{21}$  are absent in Eq. (5) due to the pairing symmetry, which forbids the on-site pairing amplitude.<sup>21</sup> The local density of states (DOS) is calculated according to

$$\rho(i,E) = 2\rho_{11}^{ii}(E) = 2\rho_{22}^{ii}(-E). \quad (6)$$

To numerically investigate the disorder effect on the electronic structure and accordingly the nuclear spin relaxation rate in the 2D system, for certain impurity content  $n_{\text{imp}}$ , typical DOS and  $1/T_1$  are obtained by averaging over 20 impurity configurations with the size of the supercell  $20\times 20$  and 100 wave vectors in the supercell Brillouin zone.<sup>16</sup> For each impurity configuration, self-consistent bond pairing potential  $\Delta_{ij}$  is obtained with the maximum relative error between two consecutive iteration steps is less than  $10^{-3}$ . In this work, we calculate the impurity-averaged  $1/T_1=T_1^{-1}(i)$  and  $\rho(E)=\rho(i,E)$ , where  $\langle \dots \rangle$  denotes averaging over space and impurity configurations.

In Fig. 1, we illustrate the dependence of the disorder-averaged DOS on the impurity content  $n_{\text{imp}}$  and the scattering strength  $U_0$ . The set of curves display clearly (i) the shrinking (and even vanishing) of the energy gap (ii) smearing and decreasing of the coherence peak as  $n_{\text{imp}}$  and  $U_0$  increase. As  $n_{\text{imp}}$  increases from 0 to 2% and 5% for the fixed  $U_0=2$ , the gap is filled from the gap edge, resulting in a reduced effective gap. And when  $U_0=10$  for  $n_{\text{imp}}=5\%$ , strictly speaking, the energy gap is closed with finite  $\rho(E)$  as  $E\rightarrow 0$  (although the DOS hump around  $\Delta_{\text{Gap}}$  can still be identified) and the residual DOS at the Fermi level is as large as 60% of the normal state value. The large residual DOS in the superconducting state is consistent with the experimental study of specific heat,<sup>20</sup> indicating the importance of inhomogeneity in this material. In the inset of Fig. 1, we give  $\Delta_p(T)$  in the clean limit,  $n_{\text{imp}}=2\%$ ,  $U_0=2$ ,  $n_{\text{imp}}=5\%$ ,  $U_0=2$  and  $n_{\text{imp}}=5\%$ ,  $U_0=10$ , showing that both the order parameter and the transition temperature are reduced and the decrease of  $\Delta_p$  as  $T\rightarrow T_c$  is also not as sharp in the disordered cases as in the dilute limit. These behaviors are consistent with the pair-breaking effect of nonmagnetic impurities in unconventional superconductors.

The evolution of the density of states originates from the impurities and depends on their content as well as strength. For the weak scalar scatterer with  $U_0=2$  (corresponds to  $c_s\simeq 0.5$  according to Ref. 22), there are two peaks of the local DOS around an isolated impurity near the gap edge with energies  $\omega_b/\Delta_{\text{Gap}}\simeq\pm 0.89$  highlighting the presence of an impurity bound state in the unconventional  $p_x+ip_y$ -wave superconductor<sup>21,22</sup> in remarkable contrast to the conventional  $s$ -wave superconductors, although both are fully gapped. And for strong potential, such as  $U_0=10$  (corre-

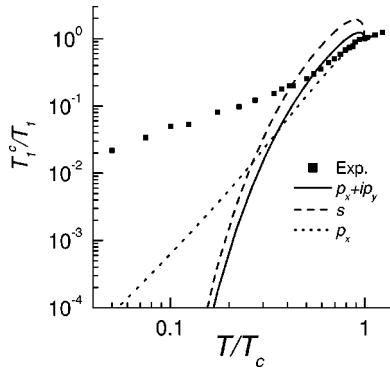


FIG. 2. Temperature dependence of  $1/T_1$  normalized by its value at  $T_c$  for the  $p_x+ip_y$ -wave (solid line),  $s$ -wave (dashed line), and  $p_x$ -wave (dotted line) superconductors. Solid squares represent the experimental data extracted from Fig. 2 of Ref. 10.

sponds to  $c_s \approx 2.5$ ), we have  $\omega_b/\Delta_{\text{Gap}} \approx \pm 0.37$ . For finite impurity density, to look into the closure of the energy gap induced by the impurities, we study the impurity-averaged self-energy of the quasiparticle, which is determined by the self-consistent equations:<sup>24,25</sup>

$$G(\mathbf{k}, i\omega_n) = [i\tilde{\omega}_n \sigma_0 - \xi(\mathbf{k}) \sigma_z - \Delta_{p_x} \sigma_x - \Delta_{p_y} \sigma_y]^{-1} \quad (7)$$

$$\Sigma(i\omega_n) = n_{\text{imp}} \frac{g(i\omega_n)}{U_0^{-2} - g^2(i\omega_n)}, \quad (8)$$

where  $i\tilde{\omega}_n = i\omega_n - \Sigma(i\omega_n)$  and  $\sigma_0$  is the unit matrix.  $\Delta_{p_x}$  and  $\Delta_{p_y}$  are real and imaginary parts of Eq. (2), respectively.  $\xi(\mathbf{k})$  is the normal state quasiparticle energy and  $g(i\omega_n) = \int d^2k / (2\pi)^2 G(\mathbf{k}, i\omega_n)$ . For the weak scatterers  $U_0^{-1} \gg g(i\omega_n)$ , the scattering rate  $\gamma$  [determined by  $\Sigma(\omega \rightarrow 0) = -i\gamma$ ] is  $\gamma = \sqrt{(\pi N_0 n_{\text{imp}} U_0^2)^2 - \Delta_{\text{Gap}}^2}$ , where  $N_0$  is the normal density of states per spin at the Fermi level. Therefore, the impurity parameter  $n_{\text{imp}} U_0^2$  must be larger than  $\Delta_{\text{Gap}}(T)/\pi N_0$  to entirely close the gap, i.e.,  $\gamma \neq 0$  is real. This effect of disorders for the  $p_x+ip_y$ -wave pairing is significantly different from that for the high- $T_c$  nodal  $d_{x^2-y^2}$ -wave pairing. In the  $d$ -wave pairing, infinitesimal  $n_{\text{imp}} U_0^2$  gives rise to finite density of states at the Fermi level. In the strong scattering limit,  $U_0^{-1} \ll g(i\omega_n)$ , we obtain  $\gamma = \sqrt{n_{\text{imp}} \Delta_{\text{Gap}} / \pi N_0}$ , which is the same as the result of  $d$ -wave superconductors with unitary impurities.<sup>26,27</sup> The above discussion qualitatively explains what we illustrate in Fig. 1.

The impurity effect on DOS is manifested by the variation of NMR relaxation according to Eq. (5). First, we address the temperature dependence of  $1/T_1$  in the absence of disorder (clean limit). When  $n_{\text{imp}}=0$ , the  $1/\sqrt{E^2 - \Delta_{\text{Gap}}^2}$  divergence of the DOS  $\rho(E)$  at the gap edge  $\Delta_{\text{Gap}}$  will lead to the Hebel-Slichter coherence peak of  $1/T_1$  just below  $T_c$  as shown in Fig. 2, although the jump of the peak is much lower than that of the  $s$ -wave pairing because the coherence factor changes from  $1 + \Delta_{\text{Gap}}^2/E^2$  in the  $s$ -wave case to 1 in the  $p_x+ip_y$ -wave case. Furthermore, when  $T \ll T_c$  and  $\Delta_{\text{Gap}} \gg T$ ,  $1/T_1$  of both the  $s$ -wave and  $p_x+ip_y$ -wave pairing states shows the behavior of  $e^{-\Delta_{\text{Gap}}/T}$  due to their fully gapped nature. Also shown in Fig. 2 is the behavior of  $1/T_1$  of the gapless  $p_x$ -wave pairing

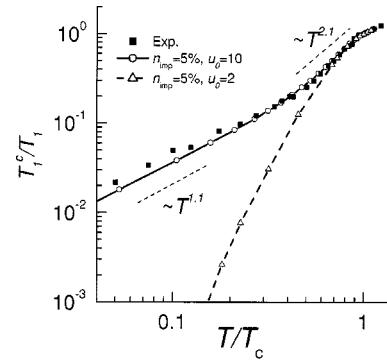


FIG. 3. Temperature dependence of  $T_1^c/T_1$  for the disordered  $p_x+ip_y$ -wave when  $n_{\text{imp}}=5\%$ ,  $U_0=10$  (solid line) and  $n_{\text{imp}}=5\%$ ,  $U_0=2$  (dashed line). The solid squares are the same as those of Fig. 2.

state for comparison. As for the nodal  $p_x$ -wave pairing [real part of Eq. (2)], the slower logarithmic divergent of  $\rho(E)$  and the halved coherent factor result in a much suppressed coherence peak, and in the low temperature region,  $\rho(E) \propto E$  gives rise to the  $T^3$  dependence of the nuclear spin relaxation rate as shown in the figure.

Figure 3 shows the nuclear spin-relaxation rate when the impurity scattering is present. Two typical results are compared with the experimental data.<sup>10</sup> As expected, the coherence peak of  $1/T_1$  disappears for both the weak and strong scattering cases, in accord with the finding that small concentration of impurities is able to smear the sharp divergence of the DOS near the gap edge. For strong disorders with  $U_0=10$ , our results of  $1/T_1$  as a function of temperature indicate a  $T^{1.1}$  dependence at the low temperature region and a  $T^{2.1}$  relation near  $T_c$ , being consistent with the results of an approximate constant  $\rho(E)$  near zero energy as shown in Fig. 1. More importantly, these results appear to be qualitatively consistent with the experimental observation of Ref. 10. The reason for the choice of such parameters is to take into account the strong scattering effect of the Co vacancies in the 2D triangular  $\text{CoO}_2$  lattice as an analogy to the cuprates, where voids in the  $\text{CuO}_2$  plane serve as important pairing breakers. On the other hand, the study of weak disorders with  $U_0=2$  exhibits that  $1/T_1$  first drops with a  $T^n$  ( $n=3$ ) law at the vicinity of  $T_c$  and then exponentially similar to its behavior in the clean limit, showing a downward curvature. The occurrence of the exponential behavior is due to the opening of the energy gap at certain temperature  $T^*$  for the weak scatterers. And  $T^*$  is governed by the solution of  $\Delta_{\text{Gap}}(T^*) = \pi N_0 n_{\text{imp}} U_0^2$ , which results in  $T^*/T_c \approx 0.8$  for  $U_0=2$  and  $n_{\text{imp}}=5\%$ . According to this, one will have a power-law dependence of  $1/T_1$  down to lower temperature region by simply increasing the impurity content  $n_{\text{imp}}$ . Notice that, our analytical discussion presented earlier gives complementary information to make our numerical results qualitatively not limited to the specific parameters we choose in the present paper.

In summary, we have elucidated the disorder effects on the electronic structure and nuclear spin-relaxation rate of the  $p_x+ip_y$ -wave pairing state, which is closely relevant to the new superconductor  $\text{Na}_{0.35}\text{CoO}_2 \cdot y\text{H}_2\text{O}$ . The experimentally

observed temperature dependence of  $1/T_1$  is explained qualitatively. It is also interesting to compare the present results with those for another layered superconductor,  $\text{Sr}_2\text{RuO}_4$  (Refs. 28 and 29) with a possible  $p_x+ip_y$ -wave pairing symmetry. We found that the gap in  $\text{Na}_{0.35}\text{CoO}_2\cdot y\text{H}_2\text{O}$  is highly isotropic<sup>18</sup> in the triangular lattice, while the gap in  $\text{Sr}_2\text{RuO}_4$  with the proposed  $p_x+ip_y$ -wave pairing is strongly anisotropic.<sup>28</sup> Therefore, we predict that future  $1/T_1$  measurements on high-quality samples of  $\text{Na}_{0.35}\text{CoO}_2\cdot y\text{H}_2\text{O}$  may observe both the coherence peak and the exponential dependence on temperature, in contrast to the  $T^{329}$  behavior of  $1/T_1$  in the significantly anisotropic  $\text{Sr}_2\text{RuO}_4$ , even if both are in the hypothetical  $p_x+ip_y$ -wave pairing states.

On the other hand, as far as the DOS-related physical quantities, such as  $1/T_1$ , are concerned, there should be no qualitative differences between the  $p_x+ip_y$ -wave pairing and the  $d+id'$ -wave pairing state in the sense that they are both fully gapped and have similar response to impurities.<sup>22,23</sup> Therefore, more definitive experimental measurements of the Knight shift are demanded to determine the symmetry of the spin part of the Cooper pair wave function. Moreover, the muon-spin-relaxation measurement,<sup>30</sup> which is sensitive to

the time-reversal-symmetry breaking effect in the  $p_x+ip_y$ - and  $d+id'$ -wave pairing states, and the phase-sensitive Josephson-tunneling related experiments<sup>31</sup> can give more decisive evidences to distinguish the gapped pairing states from the nodal ones, such as the  $p_x$  and  $d_{x^2-y^2}$  waves. The scanning tunneling microscopy experiments<sup>32</sup> are also able to shed light on the pairing symmetry by examining the energy and spatial pattern of the impurity<sup>22</sup> and vortex<sup>14</sup> states. Finally, we wish to stress that numerical calculations presented in the paper can be considered only as a qualitative illustration of physical properties of the considered superconducting state.

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\*Electronic address: zwang@hkucc.hku.hk

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- <sup>17</sup>In order to illustrate our numerical results with sufficient energy resolution, we have intentionally chosen a larger superconducting gap than the experimental data of this material.
- <sup>18</sup>We find that for  $\mathbf{k}$  on the Fermi surface, the gap anisotropy  $|\Delta_{p_x+ip_y}(\mathbf{k})|^{\max}/|\Delta_{p_x+ip_y}(\mathbf{k})|^{\min}$  varies from 1.08 (for 0.2 electron

dopping) to 1.02 (for 0.4 electron doping) for  $t < 0$ .

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- <sup>23</sup>After completing our work, we also noticed that Y. Bang, M. J. Graf, and A. V. Balatsky (cond-mat/0307510) studied the nuclear spin-lattice relaxation rate of a  $d+id$ -wave superconductor with impurities, where a self-consistent  $t$ -matrix method with an empirical temperature relations for both the gap function and the  $t$  matrix was used, and the strong fluctuation of the order parameter around unitary impurities was also neglected.
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