Spin resonance peak in Na$_x$CoO$_2 \cdot y$H$_2$O superconductors: A probe of the pairing symmetry

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Motivated by the recent discovery of superconductivity in layered Na$_x$CoO$_2 \cdot y$H$_2$O compound, we investigate theoretically the spin dynamics in the superconducting state with three possible $p_s + ip_y$, $d + id'$- and $f$-wave pairing symmetries in two-dimensional triangular lattices. We find that a spin resonance peak, which is elaborated to have a close relevance to the relative phase of the gap function and the geometry of the Fermi surface, appears in both in-plane and out-of-plane components of the spin susceptibility $\chi$ for the spin-singlet $d + id'$-wave pairing, while only in the out-of-plane (in-plane) component of $\chi$ for the spin-triplet $p_s + ip_y$-wave ($f$-wave) pairing. Because the pairing symmetry in this compound is still hotly debated at present, this distinct feature may be used to probe or determine it unambiguously with future neutron scattering experiments.

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Recently, the superconductivity with $T_c \sim 5$K is found in the CoO$_2$-layered material, Na$_x$CoO$_2 \cdot y$H$_2$O [1]. This compound consists of two-dimensional(2D) CoO$_2$ layers where Co atoms form a 2D triangular lattice and is separated by a thick insulating layer of Na$^+$ ions and H$_2$O molecules. Although its $T_c$ is relatively low, it has still attracted much attention because it shares some similarities with high-$T_c$ cuprates, in particular it may be another unconventional superconductor evolving from the doped Mott insulator. Therefore, the investigation of this compound is expected to give insight on the mechanism of unconventional superconductivity. So far, several quite different theoretical proposals [2–7] for its pairing symmetry, such as the spin-singlet $d + id'$-wave and the spin-triplet $p_s + ip_y$-(or even $f$-)wave, have been put forward. On the other hand, experimental results on the pairing symmetry reported by different groups have also been controversial, even with the same experimental method [8–11].

In unconventional $d$- and $p$-wave superconductors, the gap function $\Delta(k)$ changes the sign (phase) around the Fermi surface and thus would lead to zeros (nodes) in the superconducting (SC) energy gap $|\Delta(k)|$. Therefore, one can in principle determine the gap function and consequently the pairing symmetry, by measuring the distribution of the phase and/or node positions. In practice, it is the node positions rather than the phase that can be inferred in usual thermodynamic, transport, and NMR experiments. Therefore, the probe of the node position has been mostly used in the clarification of the pairing symmetry in unconventional superconductors [12]. However, the much debated pairing symmetries so far proposed for the SC state of Na$_x$CoO$_2 \cdot y$H$_2$O are the broken-time-reversal-symmetry $d + id'$- and $p_s + ip_y$- waves. In this case, the energy gap $|\Delta(k)| = \sqrt{\Delta^d_\alpha(k) + \Delta^p_\alpha(k)}$ is nodeless. So, a probe which is directly related to the phase is of special importance for the determination of the pairing symmetry in this compound as well as in other unconventional superconductors. This is evident by noting the crucial impact of the tricrystal phase-sensitive experiment [13] on the determination of the dominating $d_{x^2-y^2}$ pairing symmetry in high-$T_c$ superconductors. In this Letter, by noting that a spin resonance peak appears in different components of the dynamical spin susceptibility $\chi$ for all three possible pairing symmetries in the 2D triangular lattice with the nearest-neighbor (n.n.) pairing interaction, we show that the identification of the spin resonance peak in the SC state, which can be carried out by neutron scattering experiments, may also provide an unambiguous clue to probe/determine the pairing symmetry in this compound. We elaborate that the occurrence of the spin resonance peak in a specific component of the dynamical spin susceptibility exclusively corresponds to a definite change of the phase of $\Delta(k)$.

To handle both the spin-singlet and spin-triplet superconductivity in the same model as well as to capture the essential physics of electron correlation, we employ a phenomenological $t-U-V$ model [14,15] on a 2D triangular lattice, in which an effective n.n. pairing interaction ($V$) is responsible for superconductivity and an on-site Hubbard $U$ for the electron correlation. After choosing the mean-field parameter $\Delta^p_{\alpha} = V(c_i^\dagger c_j + c_j^\dagger c_i) > 0$ as an example we write the effective Hamiltonian as,

$$H_{\text{eff}} = -\sum_{<ij>,\sigma} [t_{ij} c_i^\dagger c_{j\sigma} + \text{h.c.}] + U \sum_i n_{i\uparrow} n_{i\downarrow} + \sum_{<ij>} [\Delta^p_{\alpha} c_i^\dagger c_{j\alpha} + \pm c_i^\dagger c_{j\alpha} + \text{h.c.}]$$

where the upper sign is for the spin-triplet pairing state and the lower sign for the spin-singlet pairing state. Note that the actual superconducting pairing mechanism is still unclear at present and thus is not specified in this phenomenological model.
In the triangular lattice, the dispersion of quasiparticles is
\[ \epsilon_k = -2t[\cos k_x + 2 \cos \frac{k_x}{2} \cos \frac{\sqrt{3} k_y}{2}] - \mu \] (2)

For 2D triangular lattices with the n.n. superconducting pairing interaction, only \( d_{x^2-y^2} \pm i d_{xy} \)-wave, \( p_x \pm i p_y \)-wave, and \( f \)-wave pairing states may exist [16]:

i) \( \Delta^{d+id'}_k = \Delta_0 \left[ \cos(k_x) - \cos(k_x/2)\cos(\sqrt{3} k_y/2) \right] + i \sqrt{3} \sin(k_x/2) \sin(\sqrt{3} k_y/2) \),

ii) \( \Delta^{p_x+ip_y}_k = \Delta_0 \left[ \sin(k_x) + \sin(k_x/2)\cos(\sqrt{3} k_y/2) + i \sqrt{3} \cos(k_x/2) \sin(\sqrt{3} k_y/2) \right] \), and

iii) \( \Delta^f_k = \Delta_0 \left\{ \sin(k_x) - 2 \sin(k_x/2)\cos(\sqrt{3} k_y/2) \right\} \).

Given the attractive interaction \( V \) and with \( t < 0 \) [2,4,15], the mean-field calculation of Eq.(1) shows that nearly degenerate singlet \( d + id' \)- and triplet \( f \)-wave solutions are favored at doping \( n = 0.4 \), while the triplet \( p_x + ip_y \)-wave is stable at \( n = 1.35 \), where \( n \) is the average electron number per site [15,17]. In order to compare the results for different pairing symmetries with roughly the same SC gap, we have chosen \( V = 0.75t \) for \( n = 0.4 \) and \( V = 1.7t \) for \( n = 1.35 \), which gives \( \Delta_0 = 0.015t \) for \( f \)-wave, \( \Delta_0 = 0.014t \) for \( d + id' \)-wave \((n = 0.4) \) and \( \Delta_0 = 0.15t \) for \( p + ip \)-wave \((n = 1.35) \). The effective onsite Hubbard interaction is assumed to be \( U = 3t \) [18].

The bare spin susceptibility is given by,
\[ \chi^0_{ij}(q, \omega) = \frac{1}{4N} \sum_k \frac{C^{-}_{ij}(k, q)(F^+_{k,q} - 1)}{\omega - \Omega^{-}_{k,q} + i\Gamma} - \frac{C^{-}_{ij}(k, q)(F^-_{k,q} - 1)}{\omega + \Omega^-_{k,q} + i\Gamma} + \frac{2C^{-}_{ij}(k, q)(F^0_{k,q})}{\omega + \Omega^-_{k,q} + i\Gamma} \] (3)

where the coherence factors are,
\[ C^{-}_{ij}(k, q) = \left[ \frac{1 \pm \epsilon_{kE_{k+q}} + \text{Re}(\Delta_k \Delta^*_k)_{q}}{E_{k+q}E_k} \right] \] (4)
for the spin-singlet pairing and if \( ij = zz \) (the out-of-plane component of \( \chi \)) for the spin-triplet pairing, and
\[ C^\pm_{ij}(k, q) = \left[ \frac{1 \pm \epsilon_{kE_{k+q}} - \text{Re}(\Delta_k \Delta^*_k)_{q}}{E_{k+q}E_k} \right] \] (5)
if \( ij = \pm \) (the in-plane component of \( \chi \)) for the spin-triplet pairing. \( F^\pm_{k,q} = f(E_{k+q}) \pm f(E_k) \) and \( \Omega^\pm_{k,q} = E_k \pm E_{k+q} \), with \( E_k = \sqrt{\epsilon^2_k + |\Delta_k|^2} \) and \( f(E_k) \) the Fermi distribution function. Near \( T = 0 \), only the first term in Eq.(3) with the coherence factor \( C^- \), involving the creation of quasiparticle pairs, contributes to the spin susceptibility. An essential difference in the coherence factors between the spin-singlet pairing (or the component \( \chi_{zz} \) for spin-triplet pairing) and the component \( \chi_{+-} \) for spin-triplet pairing is a sign difference in front of \( \text{Re}(\Delta_k \Delta^*_k)_{q} \). This sign difference is the key point for our later discussions.

The many-body correction to the spin susceptibility is included by the random phase approximation [18]. In this way, the renormalized spin susceptibility is given by,
\[ \chi^0_{ij}(q, \omega) = \chi^0_{ij}(q, \omega)[1 - U\chi_{ij}(q, \omega)]^{-1}. \] (6)

The momentum dependences of the dynamical spin susceptibility for \( \omega = 0.02t \) in the SC state \((T = 0.0001t) \) are presented in Fig.1. For the \( p_x + ip_y \)-wave pairing, a peak near \( Q_1 = 0.94 \times (2\pi/3, 2\pi/\sqrt{3}) \) can be seen. While, for the \( f \)-wave and \( d + id' \)-wave pairings, a peak near \( Q_2 = (0, \sqrt{3}\pi/2) \) appears. An obvious feature, seen from the figure, is that these peaks depend only on the doping density and is irrespective of the symmetry of the pairing spin state and the components of \( \text{Im} \chi_\omega \). This is due to the fact that these peaks arise from the nesting of Fermi surface which is determined only by the doping. The nesting wavevectors \( Q_1 \) and \( Q_2 \) which correspond to these two peaks are denoted as dashed lines with arrows in Fig.2. We note that a much sharper peak occurs around \( q = (0,0) \) for the in-plane component \( \text{Im} \chi_{xx} \) in the case of the spin-triplet pairing. This peak already presents in the normal state (not shown here) and reflects an enhanced ferromagnetic fluctuations which may arise from the substantial density of state at the Fermi level. However, it is suppressed highly for spin-singlet pairing and for the out-of-plane component in the spin-triplet pairing as shown in Fig.1.

In Fig.3(a), we present the frequency dependence of \( \text{Im} \chi \) at \( Q_1 \) for the spin-triplet \( p_x + ip_y \)-wave pairing. It is seen that a spin resonance peak occurs near \( \omega = 0.05t \) for the out-of-plane component \( \text{Im} \chi_{zz} \), but is absent for the in-plane component \( \text{Im} \chi_{+-} \). However, in sharp contrast, the spin resonance peak appears in the in-plane component of \( \text{Im} \chi_{+-} \) rather than in the out-of-plane component for the spin-triplet \( f \)-wave pairing(Fig.3(b)). Moreover, the spin resonance peak can be found in both components for \( d + id' \)-wave pairing, via the relation \( \text{Im} \chi_{+-} = 2\text{Im} \chi_{zz} \) which holds for a spin-singlet state. These distinctly different features for all three possible pairing states are significantly important because they may be used as an unambiguous clue to probe/determine experimentally the pairing symmetry in this compound.

To understand our observation clearly, we plot the bare spin susceptibility \( \text{Im} \chi_0 \) in Fig.4. It is clear that a peak is evident at the spin gap edge followed by a step-like decrease just below the gap edge in the channels in which there is a spin resonance peak. Using the Kramers-Kroenig relation, we will obtain a logarithmic singularity in its real part \( \text{Re} \chi_0 \). Thus, the RPA correction will further magnify this effect and lead to a sharp peak near the gap edge. This indicates that a peak just above the spin gap edge is the source of the spin resonance.

From the BCS theory, the density of states(DOS) is divergent just above the SC gap edge, which is expected to affect some physical quantities. However, the effect is
limited by the coherence factor. The most evident effect of the coherence factor [Eqs.(4) and (5)] is for energies $E_k$ and $E_{k+q}$ near the quasiparticle gap edge $\Delta_0$, in which it is either $\sim 0$ or $\sim 1$, depending on the relative sign of $\Delta_k$ and $\Delta_{k+q}$. Specifically, $C^-$ is negligible unless $\Delta_k$ and $\Delta_{k+q}$ are of opposite signs for the spin-singlet pairing [19] and for the out-of-plane component in the spin-triplet pairing, or of the same sign for the in-plane component in the spin-triplet pairing. With these general considerations, let us now address why a peak appears in one component of the spin susceptibility, but is absent in the other. In Fig.2, we plot the phase(+ sign denotes the phase 0, - sign the phase $\pi$) and node position(dotted lines) for various terms of the three possible pairing symmetries. The Fermi surface for $n = 1.35$, where the $p_x + i p_y$-wave is favored, is a circle around $(0,0)$ point. For either the $p_x$ or $p_y$ term, the two half circles separated by the line nodes will have the opposite(different) sign(phase) of the gap function $\Delta_k$. Therefore, for the wave vector $Q_1 = 0.94 \times (2\pi/3, 2\pi/\sqrt{3})$, $\Delta_k$ and $\Delta_{k+q}$ have the opposite sign. According to Eqs.(4) and (5), the coherence factor $C^-$ is appreciable for $ij = zz$ and vanishes for $ij = + -$. As a result, the DOS peak shows up in the out-of-plane component $\text{Im}\chi_{zz}$ and does not in the in-plane component $\text{Im}\chi_{+-}$, as shown in Fig.4(a). Also for the spin-triplet pairing but with the $f$-wave, $\Delta_k$ and $\Delta_{k+q_2}$ connected by $Q_2 = (0, \sqrt{3}\pi/2)$ are of the same sign(phase)[Fig.2]. Therefore, the DOS peak exists in $\text{Im}\chi_{+-}$, instead of in $\text{Im}\chi_{zz}$[Fig.4(b)]. The most definite demonstration of this argument can be found in the case of the $d + id'$ wave, where an appreciable coherence factor $C^-$ [Eq.(4)] requires that $\Delta_k$ and $\Delta_{k+q}$ have the opposite sign. However, from Fig.2 one can see that, though the $\Delta_k$'s connected by wave vector $Q_1$ satisfy the requirement for the $d'$ term, those for the $d$ term do not. To see their effect, we have calculated the results for $d$ and $d'$ terms, separately. As shown in the inset of Fig.3(b), we find no peak for the $d$-wave term, but a sharp peak for the $d'$-wave term. Remembering that the term $\text{Re}[\Delta_k^{d+id'}\Delta_{k+q}^{d+id''}] = \Delta_k^d\Delta_{k+q}^d + \Delta_k^{d'}\Delta_{k+q}^{d'}$, one will expect that it is the effect of $d'$ term dominates for the $d + id'$ wave. The only difference between the cases of $d$- and $d'$-wave pairings is their sign (phase) of the gap function. So, it shows definitely that the spin resonance peak depends uniquely on the relative phase of the gap functions connected by the transition wave vector, when the Fermi surface is given. Therefore, the spin resonance peak appears in specific components of the dynamical spin susceptibility with distinctly different ways for the proposed possible pairing states. We suggest that neutron scattering experiments can probe the above mentioned spin resonance peak, as done for high-$T_c$ cuprates [20], and thus to identify the pairing symmetry of Na$_x$CoO$_2 \cdot y$H$_2$O superconductors.

Before concluding the paper, let us use the above idea to address the anisotropic suppression of the spin response at $q = (0,0)$ shown in Fig.1. At $q \sim (0,0)$, the two gap functions connected by $q$ will surely have the same phase. So, the coherence factor $C^-$ will be negligible for the spin-singlet pairing and the out-of-plane component of the spin-triplet pairing, but it is not for the in-plane component. Thus, comparing with that in the normal state and in the latter case, the spin response in the former case is strongly suppressed.

In conclusion, we have found that the spin resonance peak exists in distinctly different ways for three possible pairing symmetries proposed for newly discovered CoO$_2$ layer materials Na$_x$CoO$_2 \cdot y$H$_2$O, and suggested to use it as an unambiguous clue to probe/determine the pairing symmetry in future inelastic neutron scattering experiments. Moreover, we have elaborated that the spin resonance peak has a close relevance to the relative phase of the gap function and the geometry of the Fermi surface.

After the completion of this work, we noticed a preprint [21] by Li and Jiang, in which a spin resonance peak was predicted only for the spin-singlet $d + id'$-wave pairing in the framework of the $t - J$ model on triangular lattices and by using the slave-boson approach.

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For a spin-triplet pairing state, a three-component complex vector $\mathbf{d}(\mathbf{k}) = [d_x(\mathbf{k}), d_y(\mathbf{k}), d_z(\mathbf{k})]$ is adopted to represent its spin dependence. In this way, the gap function can be written as, $\Delta_{\alpha\beta}(\mathbf{k}) = \mathbf{d}(\mathbf{k}) \cdot \sigma_i \sigma_2^\alpha \Delta_{\beta}$, with $\sigma$ are the Pauli matrices. Following the argument in Ref. [6], we assume that the pairing state is unitary and the $\mathbf{d}$ vector is perpendicular to the cobalt plane, i.e., $\mathbf{d}||\mathbf{z}$. In this case, $\Delta(\mathbf{k}) = d_z(\mathbf{k})$. For a spin-singlet pairing, a single complex function $\Delta(\mathbf{k})$ is sufficient for the gap function.

In the case of electron doping, the average electron number $n = 1.35(n = 0.4)$ corresponds roughly to the experimental value in Na$_x$CoO$_2 \cdot y$H$_2$O with $x = 0.35(x = 0.6)$ for $t < 0(t > 0)$.

We also tried other values in the range of $U = t \sim 3.5t$, where the random phase approximation (RPA) is still valid. The conclusions obtained here do not change qualitatively. We note that the weak coupling approach (RPA) can still give accurate results for the spin susceptibility even for the intermediate coupling if an effective (reduced) $U$ is used, as shown by N. Bulut, D. J. Scalapino and S. R. White, Phys. Rev. B 47, 2742 (1993).

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Im\chi(q,\omega)

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Fig. 4, Li and Wang