Nuclear spin-lattice relaxation rate in the D+iD superconducting state: implications for CoO superconductor

Yunkyu Bang*, M. J. Graf, and A.V. Balatsky
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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We calculated the nuclear spin-lattice relaxation rate $1/T_1$ for the D+iD superconducting state with impurities. We found that small amount of unitary impurities quickly produces the residual density of states inside the gap. As a result, the T-linear behavior in $1/T_1$ is observed at low temperatures. Our results show that the D+iD pairing symmetry of the superconducting state of Na$_{0.35}$CoO$_2$ · yH$_2$O is compatible with recent $^{59}$Co $1/T_1$ experiments of several groups.

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I. INTRODUCTION

The recent discovery of Na$_{0.35}$CoO$_2$ · yH$_2$O superconductor has renewed interest in the study of exotic superconductivity (SC). Motivated by the triangular lattice structure of spin-$\frac{1}{2}$ Co atoms of the reference CoO$_2$ layer, several authors have quickly proposed that it might be a realization of the long sought spin-liquid based superconductivity, originally proposed for high-T$_c$ superconductors by Anderson et al. Indeed several theoretical calculations indicate that the frustration of the antiferromagnetism (AFM) on the triangular lattice of Co favors the D+iD superconducting symmetry.

For a new superconducting material, the first step toward laying down the theory of the superconducting state is the experimental identification of the SC gap symmetry. Nuclear spin-lattice relation $1/T_1$ and Knight shift measurements have been the successful tools for this purpose and several measurements of $1/T_1$ and Knight shift of this compound have already been reported. At present, however, these reports appear inconsistent with each other. Regarding the coherence peak of Co$^{59}$ $1/T_1$, for example, Kobayashi et al. and Waki et al. reported the existence of a coherence peak; the maximum value of $1/T_1T$ is about 1.1 to 1.5 times of the normal state value while an ideal s-wave case has almost twice of the normal state value. However, Fujimoto et al. recently reported no coherence peak. Furthermore, they reported more precise Co $1/T_1$ below T$_c$ and found a substantial region of linear temperature dependence in $1/T_1$ at low temperatures and $T^3$ behavior near and below T$_c$. To fit their data a residual density of states (DOS) of 0.65N(0) was needed assuming an impure D-wave SC state. From this calculation, these authors concluded that the non-s-wave gap with the lines of nodes is most consistent with their $1/T_1$ data and the compatibility of the D+iD symmetry gap having a full gap remains as a question.

In this note, we calculated $1/T_1$ for D+iD gap with impurities using self-consistent T-matrix approximation and quantitatively compared our results with experiments reported up to date. Since the impurity effects on various symmetry gaps has been studied by many authors and the generic behaviors are well known, we summarize them. Within T-matrix approximation, the impurity scattering renormalizes the self-energies both of normal and anomalous components. In Nambu notation, they correspond to $\Sigma_0$, $\Sigma_2$, and $\Sigma_3$, respectively; $\Sigma_0$ and $\Sigma_2$ are the two separate components of the normal self-energy according to particle-hole transformation and $\Sigma_3$ is the anomalous self-energy component. Among them, the renormalization of $\Sigma_3$ usually vanishes for particle-hole symmetric band, which is a good approximation for most cases. For an isotropic s-wave gap, both $\Sigma_0$ and $\Sigma_2$ are renormalized and their effects largely cancel each other in the renormalized gap equation; as a result the impurity effect is null for low temperature SC properties such as the changes of the order parameter (OP) magnitude ($\Delta_0$) and $T_c$, and the residual DOS etc. For a sign changing OP such as the D-wave gap, the renormalization of $\Sigma_2$ vanishes, too, due to symmetry, and the impurity effects appear only in the renormalization of $\Sigma_0$ the normal self-energy. The case of D+iD gap is interesting in that it has a full gap all over the FS and should display similar thermodynamic behaviors as in an isotropic s-wave gap. However, regarding the impurity effects, it is expected to show a mixed behavior of a D-wave gap and a s-wave gap because the sign changing OP leads to a vanishing $\Sigma_2$ renormalization despite a full gap.

We find that for unitary impurities the residual DOS is created in the D+iD gap as easily as in the D-wave gap. This happens because the resonant scattering process of unitary impurity is more efficient at lower DOS. As a result, D+iD gap displays a similar T-linear behavior in $1/T_1$ at low temperatures with the same amount of unitary impurities as in the simple D-wave case. Therefore, the observation of the T-linear behavior in $1/T_1$ at low temperatures itself cannot rule out the D+iD symmetry gap on CoO$_2$ superconductor. Moreover, in contrast to the D-wave gap the D+iD gap with a smaller amount of impurities displays a reduced coherence peak reflecting full opening of the gap around the FS. Putting together our theoretical results, we conclude that the D+iD symmetry gap with unitary impurities provides the most consistent agreement with the currently available nuclear
spin-lattice relaxation data. We also studied the effects of Born limit impurity on both symmetry gaps. We find that the response to Born limit impurities is qualitatively different for D+iD and D-wave symmetry gaps, and both cases cannot fit the experimental data.

II. FORMALISM

The effect of the impurity scattering is included with T-matrix approximation\(^9\). As we explained above, for sign-changing OP and assuming particle-hole symmetric band, we just need to calculate \(\Sigma_0 = \Gamma T_0\), where \(\Gamma = n_i/\pi N_0\), \(N_0\) the normal DOS at the Fermi energy, \(n_i\) the impurity concentration. \(T_0\) is defined in Matsubara frequency as

\[
T_0(\omega_n) = \frac{g_0(\omega_n)}{[\hat{c}^2 - \tilde{g}_0^2(\omega_n)]},
\]

and

\[
g_0(\omega_n) = \frac{1}{\pi N_0} \sum_k \frac{i\omega_n}{\omega_n^2 + \epsilon_k^2 + \Delta_0^2(k)},
\]

where \(\omega_n = \omega_n + \Sigma_0\) and the scattering strength parameter \(c\) is related to the s-wave phase shift \(\delta\) as \(c = \cot(\delta)\). With this \(T_0\) the following gap equation is solved self-consistently.

\[
\Delta(k) = -N_0 \sum_{k'} V(k, k') \times T \sum_{\omega_n} \int_{-\omega_D}^{\omega_D} \frac{\Delta(k')}{\omega_n^2 + \epsilon_k^2 + \Delta_0^2(k')}. \tag{3}
\]

For D+iD gap equation, we assume the pairing potential \(V(k, k')\) to be a constant because thermodynamic properties of D+iD SC are determined by the absolute magnitude of the gap \(|D+iD|\) which is the same as the isotropic s-wave gap. The property of the D+iD gap in the above gap equation manifests itself only as the absence of \(\Sigma_2\) renormalization. For comparison, we also calculate the D-wave case with a proper pairing potential\(^11\), which also has no \(\Sigma_2\) renormalization due to the sign-changing OP. The only technical difference of the impurity effects on D+iD- and D-wave symmetry gaps is the normal self-energy correction due to impurities in the SC states with a full gap and lines of nodes in the gap, respectively.

With the gap function \(\Delta(k)\) and \(T_0(\omega)\) obtained from Eq.(1) \((T_0(\omega_n)\) is analytically continued from \(T_0(\omega_n)\) by Padé approximant method), we calculate the \(1/T_1\) nuclear spin-lattice relaxation rate\(^9,12\)

\[
\frac{1}{T_1} \sim \int_0^\infty \frac{\partial f(\omega)}{\partial \omega} ((Re \frac{\tilde{\omega}}{\sqrt{\omega^2 - \Delta_0^2(k)}} k)^2 + (Re \frac{\Delta(k)}{\sqrt{\omega^2 - \Delta_0^2(k)}} k)^2), \tag{4}
\]

where \(\tilde{\omega} = \omega + \Sigma_0(\omega)\) and \(\langle \cdots \rangle\) means the average over the FS. The first term in the bracket of Eq. (4) is \(N^2(\omega)\). The second term vanishes in our calculations because of the symmetry of the OP. To calculate \(1/T_1\) using Eq.(4), we need the full temperature dependence of the gap function \(\Delta(k)\). The gap equation Eq. (3) is basically the BCS gap equation, therefore it gives the BCS temperature dependence for \(\Delta(0)\) and \(\Delta_0/T_{sc} = 1.764\) and 2.14 for the D+iD- and D-wave gap solutions, respectively. We use the phenomenological formula, \(\Delta(k, T) = \Delta(k, T = 0) \Xi(T)\); \(\Xi(T) = \tanh(\beta \sqrt{T_{sc}/T - 1})\), with parameters \(\beta\) and \(\Delta_0/T_{sc}\). The temperature dependence of \(\Sigma_0(\omega, T) = \Gamma T_0(\omega, T)\) is similarly extrapolated: \(T_0(\omega, T) = T_0(\omega, 0) \Xi(T) + T_{normal}(1 - \Xi(T))\), where \(T_{normal} = \Gamma/(\epsilon^2 + 1)\) is the normal state \(T_0\). Then we only need to calculate \(\Delta(k)\) and \(T_0\) at zero temperature.

III. RESULTS AND DISCUSSIONS

In our numerical calculation, the BCS value \(\beta = 1.74\) and the ratio \(2\Delta_0/T_{sc} = 3.5\) for D+iD gap and \(2\Delta_0/T_{sc} = 5\) for D-wave gap are used, respectively. Note that the final results are insensitive to the precise value of \(\beta\). Fig.1(a) shows the normalized nuclear spin-lattice relaxation rate \(1/T_1\) for D+iD SC gap with varying impurity concentration of unitary impurities (\(c=0\)). The normalization is chosen so that the value of \(1/T_1\) at \(T_c\) is 100 in arbitrary units. The experimental data (black circles) are also normalized in the same fashion. The inset shows the corresponding DOS. The key result we find is that although D+iD gap develops a full gap as in an isotropic s-wave gap, the unitary impurity scattering produces resonance states inside the gap (at \(\omega = 0\) in our case of \(c=0\) and a particle-hole symmetric band). A finite amount of unitary impurities, therefore, quickly produces a residual DOS at \(\omega = 0\) inside the gap. As a result, the nuclear spin-lattice relaxation rate \(1/T_1\) develops a growing T-linear region at low temperatures with increasing impurity concentration. The high-temperature \(1/T_1\) below \(T_c\) initially follows the pure D+iD curve (black open squares) with a small amount of impurities, but with increasing impurity concentration it follows a nongeneric power law dependence\(^13\). With the impurity concentration of \(\Gamma/\Delta_0 = 0.32\) (\(\Delta_0\) is the absolute magnitude of the pure D+iD gap at zero temperature), the residual DOS at \(\omega = 0\) \(N(0)\) is about 0.63 \(N_0\) (\(N_0\) is the normal state DOS at Fermi level). Compared with the experimental data of \(^59\)Co \(1/T_1\) from T. Fujimoto et al. (Ref.[8]) it provides a reasonably good fit: the low temperature T-linear behavior and its magnitude, the high temperature power law (experimental fit was \(T^2\)) and the absence of the coherence peak. Therefore, the experimental observation of the substantial region of T-linear behavior\(^8\) in \(^59\)Co \(1/T_1\) of \(Na_{0.35}CoO_2\cdot yH_2O\) doesn’t rule out the possibility of D+iD gap in this compound.
Furthermore, our calculations for D+iD gap provide a resolution for the conflicting observation of the coherence peak by other groups\cite{5,6}. First, without impurities (black open squares) the height of the coherence peak is about 1.46 times of the value of $1/T_1$ at $T_c$, which is a little smaller than the value for isotropic s-wave case. This is because the second term in Eq.(4) vanishes due to the symmetry of the D+iD OP. With impurities the coherence peak is quickly suppressed and there is a small peak visible with a smaller amount of impurities (for example, see $\Gamma/\Delta_0 = 0.04$ (open purple squares); note the logarithmic scale). We notice that there is a systematic trend of the height of the coherence peak with $T_c$ of the samples used for $^{59}$Co $1/T_1$ measurement: no coherence peak with $T_c=3.9$ K\cite{8}; coherence peak with height 1.1 times of the value of $1/T_1$ at $T_c$ with $T_c=4.5$ K\cite{5}; coherence peak height 1.4 times of the value of $1/T_1$ at $T_c$ with $T_c=5.0$ K\cite{6}. Therefore, we can understand the seemingly inconsistent experimental reports on $^{59}$Co $1/T_1$ by different groups with D+iD gap and as a result of the different samples with different scattering rates\cite{14}. We also show the D-wave results with unitary impurities ($c=0$) in Fig.1(b). The overall shape of DOS for D-wave gap with and without impurities is qualitatively different from the case of D+iD gap. Nevertheless, the accumulation of the residual DOS $N(0)$ with impurities are very similar in magnitude, which is the main property determining the low temperature $1/T_1$ behavior. As a result, we can also fit the data of Fujimoto et al. with a D-wave gap with a similar amount of impurities (the necessary $\Gamma/\Delta_0$ value should be between 0.32 and 0.16, see Fig.1(b)); the residual DOS $N(0)$ with $\Gamma/\Delta_0=0.32$ is about 0.73 $N_0$. However, the D-wave results cannot explain the observation of the coherence peak seen by some experimental groups\cite{5,6}. For completeness, we show the same calculations in the Born limit ($c=1$) in Fig.2(a) and Fig.2(b). Both D+iD and D-wave gaps with Born limit impurities doesn’t produce a close fit to the experimental data unless $\Gamma/\Delta_0 \rightarrow 0.8$. Even with $\Gamma/\Delta_0 = 0.8$, the detailed line shape of the calculated $1/T_1$ doesn’t quite fit the experimental data for both gaps. Moreover, the reduction of $T_c$ and the gap magnitude $\Delta_0$ with $\Gamma/\Delta_0 = 0.8$ is about 50 % or more for both gap cases, which is not consistent with experiments.

IV. CONCLUSION

In summary, we have calculated the nuclear spin-lattice relaxation rate $1/T_1$ for both D+iD- and D-wave superconducting states with impurities. We found that unitary impurities produce the residual density of states inside the gap due to the resonant scattering for both cases. Interestingly, we found that the rate of accumulation of the residual density of states with unitary impurities is similar for both SC gaps. As a result, both D+iD and D-wave gaps produce the T-linear behavior in $1/T_1$ at low temperatures and can fit the experimental data of Fujimoto et al.\cite{8} with the unitary impurity concentration of $\Gamma/\Delta_0 \sim 0.3$. However, considering observations of the coherence peak in $^{59}$Co $1/T_1$ by other groups\cite{5,6} and a systematic trend of the height of the coherence peak with $T_c$ of the samples, we conclude that the D+iD symmetry gap is the most consistent with the currently available nuclear spin-lattice relaxation data of Na$_{0.35}$CoO$_2 \cdot y$H$_2$O.

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FIG. 1. (A) The normalized $1/T_1$ of D+iD gap with unitary impurities (c=0). Experimental data (black circles) are also normalized (T. Fujimoto et al. [Ref[8]]). Inset is the corresponding DOS. (B) The same plots as in (A) for a D-wave gap with unitary impurities (c=0).

Fig. 2. (A) The normalized $1/T_1$ of D+iD gap with Born limit impurities (c=1). Experimental data (black circles) are also normalized (T. Fujimoto et al. [Ref[8]]). Inset is the corresponding DOS. (B) The same plots as in (A) for a D-wave gap with Born limit impurities (c=1).

* Permanent address: Department of Physics, Chonnam National University, Kwangju 500-757, Korea.
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In fact, the temperature dependence of $1/T_1$ at high temperatures near $T_c$ is not a generic property for any symmetry gaps; it is determined by the temperature slope of $\Delta(T)$ near $T_c$, elastic and inelastic scattering rates, and the anisotropy of the gap, etc. Often quoted $T^3$ behavior of $1/T_1$ for a pure D-wave gap is a generic property at low temperatures reflecting the lines of nodes, but the same $T^3$ behavior observed at high temperatures is a result from combined effects mentioned above. Nevertheless, empirically once the coherence peak is suppressed the high temperature power law of $1/T_1 \sim T^\alpha$ can be fit with $\alpha$ between 2 and 3 for the dirty D+iD and D cases.

This conclusion needs further experimental verification since the measurements of Ref.[5] and Ref[6] have some ambiguity in extracting the $T_1$ relaxation time.