Effect of ferromagnetic spin fluctuations on the electronic states in Na_xCoO_2 based on two-dimensional triangular lattice 11-band d-p model

Yuki Yanagi [a] and Yoshiaki Ōno [a,b]

[a] *Department of Physics, Niigata University, Ikarashi, Nishi-ku, Niigata 950-2181, Japan*
[b] *Center for Transdisciplinary Research, Niigata University, Ikarashi, Nishi-ku, Niigata 950-2181, Japan*

Abstract

Electronic states of the CoO_2 plane in Na_xCoO_2 are investigated on the basis of the triangular lattice 11-band d-p model by using the second order perturbation (SOP) and the random phase approximation (RPA). Due to the effect of the in-plane ferromagnetic fluctuations enhanced for $0.6 \lesssim x \lesssim 0.75$, the quasiparticle dispersion near the Γ-point is strongly reduced to yield an almost flat dispersion just above the fermi level. With increasing x, the electronic specific heat coefficient γ increases towards the critical doping $x_c \sim 0.75$ above which the in-plane ferromagnetism takes place. We also find that the six small hole pockets near the K-point are stabilized due to the electron correlation effect within both the SOP and the RPA.

Key words: Na_xCoO_2; ferromagnetic spin fluctuations; RPA; electronic specific heat; renormalized band; mass enhancement

Recently, the layered cobalt oxides Na_xCoO_2 have been attracted much attention because of rich physical properties. The compound exhibits large thermopower S in the region $x \gtrsim 0.7$ [1,2] and an unexpected metal-insulator transition at $x = 0.5$ [3,4]. The superconductivity with $T_c \sim 5K$ was discovered in the H_2O intercalated compound $Na_xCoO_2 \cdot yH_2O$ for $x \sim 0.35$ and $y \sim 1.3$ [5]. Moreover, for $x \gtrsim 0.75$, Na_xCoO_2 shows a weak magnetic order below $T_m \sim$ 22K, where the ferromagnetic orderd CoO_2 planes couple antiferromagnetically with each other [6]. An anomalous metallic state is observed for $0.6 \lesssim x \lesssim 0.75$: the temperature dependence of the magnetic susceptibility is Currie-Weiss-like, and both of the electronic specific heat coefficient γ and the thermopower S increase with increasing x towards the magnetic ordered region [4].

Theoretical studies have been extensively performed for the electronic properties for both the normal and superconducting states in Na-poor region (i.e., $x \sim 0.35$). However, only a few works have been performed for the electronic states in Na-rich region (i.e. $x \sim 0.7$) where both of γ and S are largely enhanced. Koshibae *et al.* discussed the thermopower using Heikes formula [7] to include the spin-orbital degeneracy of the Co^{3+} and Co^{4+} together with the strong correlation effects. The band structure, however, was not considered there. Recently, Kuroki *et al.* proposed a peculiar shape of the band structure called "Pudding mold" band which yields a large thermopower based on the Boltzmann's equation [8]. The electron correlation effect, however, was not explicitly included there.

In this work, we study the electronic states of the CoO_2 plane in Na_xCoO_2 on the basis of the triangular lattice 11-band d-p model, where the tight-binding parameters are determined so as to fit the LDA band structure. The electron correlation effect is considered within the random phase approximation (RPA) to elucidate the effect of in-plane fer-

romagnetic fluctuations on the quasiparticle dispersion and the electronic specific heat. We also apply the second order perturbation (SOP) for comparison with RPA. The self-energy is calculated within the RPA and the SOP to obtain the quasiparticle dispersion, the mass enhancement factor and the damping rate.

We consider the two-dimensional triangular lattice d-p model which includes 11 orbitals: d_{xy}, d_{yz}, d_{zx}, $d_{x^2-y^2}$, $d_{3z^2-r^2}$ of Co and p_{1x}, p_{1y}, p_{1z} (p_{2x}, p_{2y}, p_{2z}) of O in the upper (lower) side of a Co plane. The Hamiltonian is given by

$$H = H_0 + H', \tag{1}$$

$$H_0 = \sum_{i,j,\ell,\ell',\sigma} (t_{ij}^{\ell\ell'} c_{i\ell\sigma}^\dagger c_{j\ell'\sigma} + h.c.), \tag{2}$$

$$H' = H_U + H_{U'} + H_J + H_{J'}, \tag{3}$$

where $c_{i\ell\sigma}^\dagger (c_{i\ell\sigma})$ is a creation (annihilation) operator for an electron with orbital ℓ (=d_{xy}, d_{yz}, d_{zx}, $d_{x^2-y^2}$, $d_{3z^2-r^2}$, $p_{1x}, p_{1y}, p_{1z}, p_{2x}, p_{2y}, p_{2z}$) and spin σ (=$\uparrow, \downarrow$) at site i. In eq. (3), $t_{ij}^{\ell\ell'}$, which can be written by the Slater-Koster parameters, is determined so as to fit the tight-binding energy bands to the LDA band structure [9]. H' is the on-site Coulomb interaction between d-electrons of Co, where H_U and $H_{U'}$ are the intra- and inter-orbital direct terms and H_J and $H_{J'}$ are the Hund's rule coupling and the pair-transfer term, respectively. These interactions are expressed by using Kanamori parameters, U, U', J and J'. We set the energy unit to be electron volt.

In the RPA, the spin and charge susceptibilities have 9×9-matrix forms as [10,11]

$$\hat{\chi}^s(q) = (\hat{1} - \hat{\chi}^{(0)}(q)\hat{S}^{(0)})^{-1}\hat{\chi}^{(0)}(q), \tag{4}$$

$$\hat{\chi}^c(q) = (\hat{1} + \hat{\chi}^{(0)}(q)\hat{C}^{(0)})^{-1}\hat{\chi}^{(0)}(q), \tag{5}$$

where $q = (\mathbf{q}, \omega_\ell = 2\ell\pi T)$ and the matrix elements $S^{(0)}_{\ell_1\ell_2\ell_3\ell_4}$ ($C^{(0)}_{\ell_1\ell_2\ell_3\ell_4}$) are U (U) for $\ell_1 = \ell_2 = \ell_3 = \ell_4$, U' ($-U' + 2J$) for $\ell_1 = \ell_3 \neq \ell_2 = \ell_4$, J ($2U' - J$) for $\ell_1 = \ell_4 \neq \ell_2 = \ell_3$, J' (J') for $\ell_1 = \ell_2 \neq \ell_3 = \ell_4$, and 0 for the others. Bare susceptibility $\hat{\chi}^{(0)}(q)$ is given by

$$\chi^{(0)}_{\ell_1\ell_2\ell_3\ell_4}(q) = -\frac{T}{N}\sum_k G^{(0)}_{\ell_2\ell_3}(k+q)G^{(0)}_{\ell_4\ell_1}(k), \tag{6}$$

where $k = (\mathbf{k}, \epsilon_n = (2n+1)\pi T)$, N is the total number of lattice points and $\hat{G}^{(0)}$ is the unperturbed Green's function. The self-energy is given by

$$\Sigma_{\ell\ell'}(k) = \frac{T}{N}\sum_q \sum_{\ell_1\ell_2} G^{(0)}_{\ell_1\ell_2}(k-q)V^{eff}_{\ell\ell_2\ell'\ell_1}(q), \tag{7}$$

$$\hat{V}^{eff}(q) = \frac{3}{2}\hat{V}^s(q) + \frac{1}{2}V^c(q), \tag{8}$$

$$\hat{V}^s(q) = \hat{S}^{(0)}\hat{\chi}^s(q)\hat{S}^{(0)} - \frac{1}{2}\hat{S}^{(0)}\hat{\chi}^{(0)}(q)\hat{S}^{(0)}, \tag{9}$$

$$\hat{V}^c(q) = \hat{C}^{(0)}\hat{\chi}^c(q)\hat{C}^{(0)} - \frac{1}{2}\hat{C}^{(0)}\hat{\chi}^{(0)}(q)\hat{C}^{(0)}, \tag{10}$$

In order to obtain dynamical quantities in the RPA, the analytic continuation from the imaginary Matsubara frequency to the real frequency is usually performed by using Padé approximation or the maximum entropy method. However, it is known that this procedure sometimes encounters numerical difficulties. To avoid the difficulties, we analytically carry out the summation over Matsubara frequencies and directly calculate the retarded self-energy in a real frequency formulation without numerical analytic continuation.

In the numerical calculations, we use 64×64 $\mathbf{k}$-meshes and 512 frequencies for $|\omega| < \omega_c = 3$. We set the interaction parameters: $U = U' + 2J$, $J = J'$, $U' = 1.3$, $J = 0.13$, which result in the in-plane ferromagnetism for $x \geq x_c \sim 0.75$ within the RPA, so as to reproduce the experimental result [6]. For simplicity, we exclusively consider the band nearest to the fermi level in the calculation of $\hat{G}^{(0)}$.

Fig. 1 shows the retarded self-energy $\Sigma^{\rm R}(\mathbf{k}, \omega)$ as a function of frequency ω for $x = 0.7$ at the Γ-point ($\mathbf{k} = (0, 0)$) and the M-point ($\mathbf{k} = (0, \frac{2\pi}{\sqrt{3}})$). We can see usual fermi liquid properties: ${\rm Re}\Sigma^{\rm R}(\mathbf{k}, \omega) \sim a\omega$, and ${\rm Im}\Sigma^{\rm R}(\mathbf{k}, \omega) \sim b\omega^2$, with $a, b < 0$ at low energy ($\omega \sim 0$). We find that the imaginary part of the retarded self-energy at the Γ point has the sharp peak around $\omega \sim 0.05$ due to the effect of the in-plane ferromagnetic fluctuations which becomes crucial near the critical doping $x \sim 0.75$ above which the in-plane ferromagnetism takes place.

The renormalized dispersion $\epsilon_{\mathbf{k}}$ is defined by the solution of the following secular equation:

$$\epsilon_{\mathbf{k}} - \epsilon^{(0)}_{\mathbf{k}} + \mu - {\rm Re}\Sigma^{\rm R}(\mathbf{k}, \epsilon_{\mathbf{k}}) = 0, \tag{11}$$

where $\epsilon^{(0)}_{\mathbf{k}}$ is the bare dispersion and μ is the chemical potential. Fig. 2(a) shows the renormalized dispersion $\epsilon_{\mathbf{k}}$ nearest to the fermi level together with the bare dispersion $\epsilon^{(0)}_{\mathbf{k}}$ for $x = 0.7$ along the $\Gamma - K - M - \Gamma$ line, where the K-point is $\mathbf{k} = (\frac{2\pi}{3}, \frac{2\pi}{\sqrt{3}})$. We find that the six small hole pockets near the K-point are stabilized due to the electron correlation effect within the RPA, which is consistent with the previous result obtained from the slave-boson approach for the similar d-p model [12]. We have also

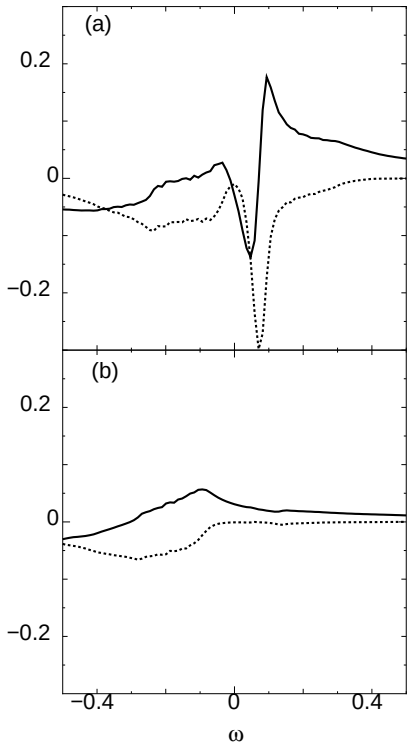

Fig. 1. The real part (solid line) and the imaginary part (dotted line) of the retarded self-energy $\Sigma^R(\mathbf{k},\omega)$ obtained from the RPA as functions of ω for $x = 0.7$ at the Γ-point (a) and at the M-point (b).

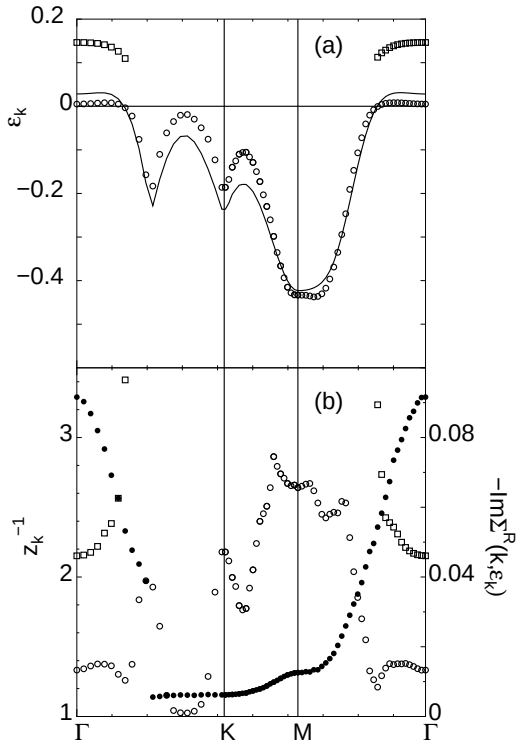

Fig. 2. The RPA results for the renormalized dispersion $\epsilon_{\mathbf{k}}$ (open circles and squares) together with the bare dispersion $\epsilon_{\mathbf{k}}^{(0)}$ (solid line) (a), and those for the mass enhancement factor $z_{\mathbf{k}}^{-1}$ (closed circles) together with the quasiparticle damping rate $-\mathrm{Im}\Sigma^{\mathrm{R}}(\mathbf{k},\epsilon_{\mathbf{k}})$ (open circles and squares) (b), along the $\Gamma - K - M - \Gamma$ line for $x = 0.7$.

confirmed that the similar tendency is obtained from the SOP as shown in Fig. 3(a).

Remarkably, the quasiparticle dispersion near the Γ-point is strongly reduced to yield an almost flat dispersion just above the fermi level as shown in Fig. 2(a). This is due to the effect of the in-plane ferromagnetic fluctuations as mentioned before. In addition, we find another branch of the dispersion which corresponds to the upper Hubbard band. In general, the self-energy corrections lead to two types of solution of the secular equation (11): the low energy solution defines quasiparticle excitations and the high-energy solution defines the excitations in the Hubbard bands [13].

In Fig. 2(b), the mass enhancement factor,

$$z_{\mathbf{k}}^{-1} = 1 - \left.\frac{\partial \mathrm{Re}\Sigma^{\mathrm{R}}(\mathbf{k},\omega)}{\partial\omega}\right|_{\omega=0}, \qquad (12)$$

together with the quasiparticle damping rate, $-\mathrm{Im}\Sigma^{\mathrm{R}}(\mathbf{k},\epsilon_{\mathbf{k}})$, is plotted along the $\Gamma - K - M - \Gamma$ line for $x = 0.7$. We can see that $z_{\mathbf{k}}^{-1}$ has a maximum value at the Γ-point. With increasing the doping x, $z_{\mathbf{k}}^{-1}$ near the Γ-point increases towards the critical doping $x_c \sim 0.75$ above which the in-plane ferromagnetism takes place. We note that, in the case with the SOP, $z_{\mathbf{k}}^{-1}$ has a maximum at the same Γ-point, but the value of $z_{\mathbf{k}}^{-1}$ is smaller than that from the RPA as shown in Fig. 3(b).

Finally, we calculate the electronic specific heat coefficient γ given by

$$\gamma = \frac{2\pi^2 k_B^2}{3}\frac{1}{N}\sum_{\mathbf{k}} z_{\mathbf{k}}^{-1}\left[-\frac{1}{\pi}\mathrm{Im}G^{\mathrm{R}}(\mathbf{k},0)\right], \qquad (13)$$

where $G^{\mathrm{R}}(\mathbf{k},\omega) = [\omega - \epsilon_{\mathbf{k}}^{(0)} + \mu - \Sigma^{\mathrm{R}}(\mathbf{k},\omega)]^{-1}$ is the renormalized retarded Green's function and k_B is the Boltzmann's constant. In Fig. 4, we plot the electronic specific heat coefficient obtained from the RPA γ_{RPA} and that from the SOP γ_{SOP} with that in the noninteracting system γ_0 together with the magnetic susceptibility $\chi^s(\mathbf{q} = 0, \omega = 0)$ obtained from the RPA as functions of the doping x. We see that γ_{RPA} is largely enhanced for $0.6 \lesssim x \lesssim 0.75$ where χ^s, *i.e.*, the in-plane ferromagnetic fluctua-

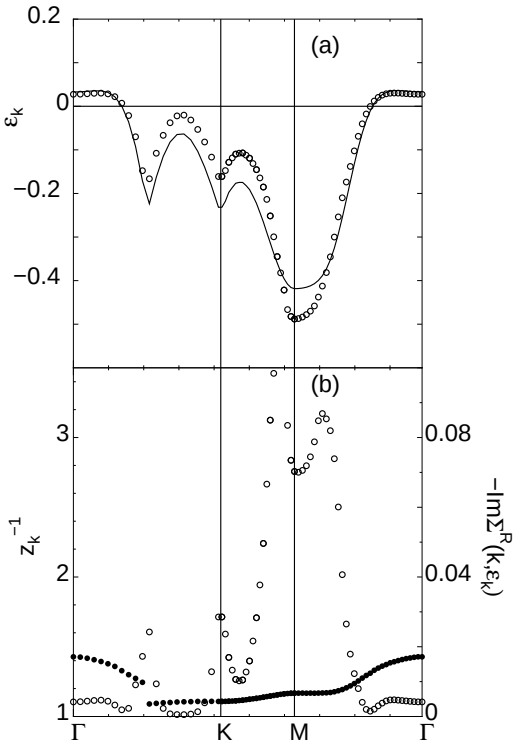

Fig. 3. The SOP results for the same quantities in Fig. 2.

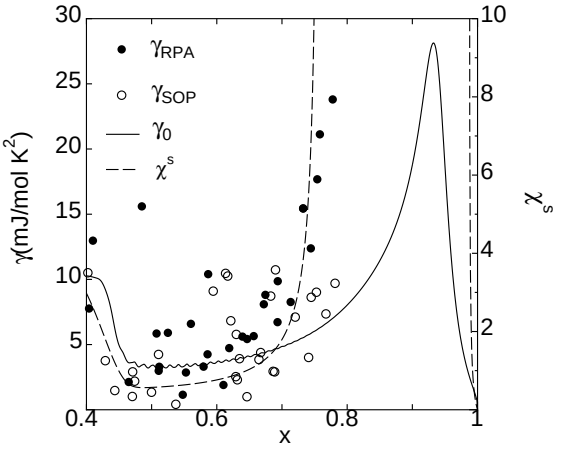

Fig. 4. The electronic specific heat coefficient γ and the magnetic susceptibility χ^s as functions of the doping x. Closed (open) circles are γ obtained from the RPA (SOP) and the solid line is that in the noninteracting system. χ^s is obtained from the RPA.

tion becomes large. γ_{RPA} increases with increasing x towards the ferromagnetic critical point $x_c \sim 0.75$ where χ^s diverges.

In summary, we have investigated the triangular lattice d-p model simulating the CoO_2 plane of Na_xCoO_2, and found that the in-plane ferromagnetic fluctuations play crucial roles for the electronic states for $0.6 \lesssim x \lesssim 0.75$, where γ increases with increasing x towards the critical doping $x_c \sim 0.75$ above which the in-plane ferromagnetism takes place. This behavior seams to be consistent with the experimental results for γ in the anomalous metallic region with $0.6 \lesssim x \lesssim 0.75$ [4]. In this region, the thermopower S is also enhanced and increases with increasing x as mentioned before. Therefore, we expect that the in-plane ferromagnetic fluctuations is important also for S. The explicit calculation for the transport properties including S is now under the way.

The authors thank H. Konatani and W. Koshibae for useful comments and discussions. This work was performed under the interuniversity cooperative Research program of the Institute for Materials Research, Tohoku University, and was supported by the Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology.

References

[1] I. Terasaki, Y.Sasago and K.Uchinokura, Phys. Rev. B **56**, R12685 (1997).

[2] M. Lee, L. Viciu, L. Lu, Y. Wang, M. L. Foo, S. Watauchi, R.A. Pascal Jr., R. J. Cava and N. P. Ong, Nature Materials **5**, 537 (2006).

[3] M. L. Foo, Y. Wang, S. Watauchi, H. W. Zandbergen, T. He, R. J. Cava and N. P. Ong, Phys. Rev. Lett. **92**, 247001 (2004).

[4] M. Yokoi, T. Moyoshi, Y. Kobayashi, M. Soda, Y. Yasui, M. Sato and K. Kakurai, J. Phys. Soc. Jpn **74**, 3046 (2005).

[5] K. Takada, H. Sakurai, E. Takayama-Muromachi, F. Izumi, R. A. Dilanian and T. Sasaki, Nature **422**, 53 (2003).

[6] T. Motohashi, R. Ueda, E. Naujalis, T. Tojo, I. Terasaki, T. Atake, M. Karppinen and H. Yamauchi, Phys. Rev. B **67**, 064406 (2003).

[7] W. Koshibae, K. Tsutsui and S. Maekawa, Phys. Rev. B **62**, 6869 (2000).

[8] K. Kuroki and R. Arita, J. Phys. Soc. Jpn. **76**, 083707 (2007).

[9] D. J. Singh, Phys. Rev. B **61**, 13397 (2000).

[10] K. Yada and H. Kontani, **74**, 2161 (2005).

[11] M. Mochizuki, Y. Yanase and M. Ogata, Phys. Rev. Lett. **94**, 147005 (2005).

[12] Y. Ōno, Physica C, in press.

[13] V. Zlatić, K. D. Schotte and G. Schliecker, Phys. Rev. B **52**, 3639 (1995).