Models of spin structures in $Sr_2RuO_4$.

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The mean field study of bands and the Fermi surfaces (FS) are carried out for the 3-band model of $RuO_4$ plane of ruthenate for various spin structures. In particular, the lowest spiral state with incommensurate vector $Q = 2\pi(\frac{1}{4}, \frac{1}{4})$, corresponding to nesting of bands, displays a set of FS’s with electron pockets for $\gamma$ band and additional shadow sheets along $\Gamma - M$ lines for $\alpha(\beta)$ bands. The latter may provide a new interpretation of the dispersionless peaks (so called SS features) observed in photoemission. For spiral state the polarisation asymmetry of the photoemission intensity is revealed. The calculated spin susceptibility for this state describes the low frequency magnetic peak in $\chi''(q,\omega)$ at $q = Q$ observed in neutron scattering.

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A single-layer quasi-2D ruthenate attracts attention as a superconductor ($T_c \sim 1K$) with a possible triplet type of pairing [1]. One of the arguments in favour of such type of pairing is the Knight shift behaviour [2]. It is suggested also that the pairing is governed by the ferromagnet (FM) fluctuations which are assumed in an analogy with the ferromagnetism of the parent cubic $SrRuO_3$. A knowledge of the normal state electronic and magnetic properties is a base for study the superconducting pairing. The band structure and magnetic fluctuations have been studied in [3][4]. The substituted compounds $Ca_{2-x}Sr_xRuO_4$ exhibit a complex phase diagram governed by the lattice distortions. It includes the phases of the paramagnet (PM) or FM metal or antiferromagnet (AFM) dielectric. Contrary to such behaviour the lattice of the $Sr_2RuO_4$ remains undistorted. The results on inelastic neutron scattering (INS) show magnetic peak at incommensurate momentum $Q \sim (0.3, 0.3, 1)$ (in units $(\frac{2\pi}{a}, \frac{2\pi}{a}, \frac{2\pi}{c})$). The static structure with the same period was observed in substituted $Sr_2Ru_{1-x}Ti_xO_4$ [5]. In [6] the peak position is connected with a structure of valence bands, namely with a nesting of $\alpha$ and $\beta$ bands of $d_{xz}$ and $d_{yz}$ nature, respectively. The nesting means an existence of the planar FS sections which coincide with each other under a translation $k \rightarrow k + Q$ on a nesting vector $Q$.

There are many ways to describe the correlated systems with nesting. One of them uses as a starting point the zero magnetic susceptibility $\chi_0(q,\omega)$ of Lindhard [7][11]. Nesting at some $q = Q$ leads to maximum in $\chi''_0(q,\omega)$ at $\omega \rightarrow 0$ and $q \rightarrow Q$. The subsequent renormalisation of type $\chi(q,\omega) = \chi_0/[1 - J(q)\chi_0]$ or $\chi(q,\omega) = \chi_0/[1 - U\chi_0]$ in the t-J or Hubbard model can explain the large observed density of the low frequency excitations only if the denominators in renormalised $\chi$ is small at $q = Q$. This indicates on the possible instability of the system with respect to formation of the spin structure with given $q = Q$. Then the renormalisation of $\chi$ on base of zero spectrum may be inadequate.

Another way of treating the nesting in correlated system is to accept a specific spin structure, which removes the induced instability, and then to carry out the mean field (MF) consideration taking into account the assumed spin order in zero approximation. In applications to cuprates it implies the using of the AFM or spiral states as a zero approximation [12][14]. Usually the homogeneous MF solutions are considered which suppose the infinite range of the spin order. In reality the doped compounds have a finite range of the spin correlations. Therefore the homogeneous MF solutions must be thought rather as local or temporary structures in system. A set of the possible spin structures for ruthenates is discussed.

Main features of bands and FS’s for $Sr_2RuO_4$ obtained from the magnetic quantum oscillations [15] and from the photoemission (ARPES) [16][17] are reproduced by a three-band strong coupling model proposed in [14][16]. However, the ARPES data reveal a set of shadow FS’s which may indicate on a periodic spin structure in sys-
tem. In particular, the sharp dispersionless peak, called in [14] the SS features, has been observed and connected with a FS along lines $\Gamma(0,0) - M(\pi,0)$. Interpretation of them is controversial yet. In [19] the SS features are attributed to the surface states arising due to a reconstruction of the surface layers. Such interpretation is argued by the fact that the temperature cycling removes SS features and other shadow FS’s, i.e. destroys the surface superlattices.

The aim of present work is to study how the various spin structures manifest in the form of the FS, in properties of shadow sheets of FS and in magnetic susceptibility of 3-band model of $Sr_2RuO_4$. The calculations are carried out in mean-field (MF) approximation. The PM, FM, AFM structures and spiral states with incommensurate vector $Q = 2\pi(\frac{1}{3}, \frac{1}{3})$ are considered. Such a spirality vector is chosen since it removes the nesting instability of both $\alpha$ and $\beta$ bands simultaneously. The possible bulk origin of the dispersionless SS features along $\Gamma$ is discussed. They are explained by Umklapp processes in periodic spiral structure. Calculations of spin susceptibility $\chi(q, \omega)$ is done on base of spiral ground state as a zero order state in RPA treatment. Thus, contrary to the standard renormalisation of $\chi$, the part of interaction is taken into account in a non-perturbative manner.

The valence bands in $Sr_2RuO_4$ is determined by electrons of $RuO_4$ plane. In approximate ionic model of $RuO_4 = Ru_d^4+(d^4)(O^{2-})_4$ the four electrons occupy three lower $d$- orbits $d_{xy}$, $d_{xz}$, $d_{yz}$ of $t_{2g}$ symmetry. These orbits together with $p_x$ orbits of oxygen generate three valence bands with the total occupancy $n = 4$ of electrons per unit site of $RuO_4$ plane. The strong coupling Hamiltonian for corresponding $\alpha$, $\beta$ and $\gamma$ bands of the $xz$, $yz$ and $xy$ nature are [13]

$$H = T + H_U; \quad T = \sum_{\nu, \sigma} \sum_k \epsilon_{\nu k} c_{\nu k}^{\dagger} c_{\nu k} + T_{12}$$

$$H_U = \sum_{n, \nu} \left\{ U_n n_{\nu n} + \sum_{\nu' \neq \nu} \frac{1}{4} [n_{\nu n} n_{\nu' n} - JS_{\nu n} S_{\nu' n}] \right\}$$

Here $U_2 = 2U - 5J$ and $n_{\nu n} = n_{\nu g n} + n_{\nu c n}$, $S_{\nu n}$ are the site operators of the occupancy and the spin corresponding to the $\alpha, \beta, \gamma$ bands ($\nu = 1, 2, 3$). Without interband hopping the zero band energies are

$$\epsilon_{\nu k}^{0} = -t_0^{\nu} - 2t_2^{\nu} \cos k_x - 2t_3^{\nu} \cos k_y + 4t_{xy}^{\nu} \cos k_x \cos k_y$$

For the model parameters $t_0, t_x, t_y, t_{xy}, U, J$ in (1,2) we use the values from [13]. In undistorted lattice the interband coupling can be only of the form $T_{12} = \sum_{k, \sigma} 4t_{\alpha\beta}^{\nu} \sin k_x \sin k_y (c_{\alpha k}^{\dagger} c_{\beta 2k}^{\dagger} + h.c.)$. A small value of $t_{\alpha\beta}^{\nu} \sim t_{xy}^{(2)} \sim 0.01eV$ is expected.

We search the MF solutions of model (1) with definite order of the local spins $< S_n >$. In absence of the spin-orbit interaction the orientation of $< S_n >$ is arbitrary. For simplicity we consider that the spin quantisation axis $z$ coincides with the crystal $c$ axis. With such agreement we define the following set of the average spins in different spin structures

$$< S_n^{\nu} > = d_{\nu} [e_x \cos Q n + e_y \sin Q n]$$

For the considered SP, FM and AFM structures the corresponding vectors are

$$Q = 2\pi(\frac{1}{3}, \frac{1}{3}), \quad Q = (0,0), \quad Q = (\pi, \pi)$$

For our choice of the quantisation axis the MF equations have similar form for all the states and they differ only in $Q$. Repeat that we choose the vector $Q$ of spiral state in such way that it removes of the nesting instability for both quasi-1D $\alpha$ and $\beta$ bands simultaneously.

The MF state $\Phi_Q$ is determined by occupancy of the eigenstates $b_{\lambda k}^{\dagger}$ (with spectrum $E_{\lambda k}$) of the linearised Hamiltonian $H_{lin}$

$$[H_{lin}, b_{\lambda k}^{\dagger}] = E_{\lambda k} b_{\lambda k}^{\dagger}$$

In the renamed basis set $a_{\nu k}^{\dagger}, i = 1, 2$, the corresponding eigen-operators are expressed as

$$b_{\lambda k}^{\dagger} = a_{\nu k}^{\dagger} S_{\nu \lambda}(k); \quad a_{\nu k}^{\dagger} = \{ c_{\gamma k}^{\dagger} c_{\lambda k}^{\dagger} \} i$$

Here $\lambda = 1, \ldots, 6$; the index $i = 1, 2$ corresponds to the spin projections and the $\nu = 1, 2, 3$ numerates the $\alpha, \beta, \gamma$ bands.

The energy $\bar{H}(\bar{T}, r_{\nu}, d_{\nu})$ averaged over the MF state with the spin order (3) is a function of one-electron means of a density $r_{\nu}$, of a local spin $d_{\nu}$ and of the mean kinetic energy $\bar{T}$ per site. In the basis set (6) the operators corresponding to the means $r_{\nu}$, $d_{\nu}$ are
\[ \hat{r}_\nu = \frac{1}{\hbar} \sum_{i=1,2} \sum_k a_{i\nu,k} a_{i\nu,k} \]
\[ \hat{d}_\nu = \frac{1}{\hbar} \sum_{i=1,2} \sum_k a_{i\nu,k} a_{3-i,\nu,k} \] (7)

Then the linearised Hamiltonian in (5) is equal to
\[ \hat{H}_{lin} = \hat{T} + \frac{\partial \hat{H}}{\partial \nu} \hat{r}_\nu + \frac{\partial \hat{H}}{\partial \mu} \hat{d}_\nu + \text{const} \] (8)

It determines the levels and eigen-operators in MF solution under a search.

A study of the main and shadow FS for each band \( \nu \) includes the calculation of the one-electron spectral functions \( A_\nu(k, \omega) \) at \( \omega = 0 \). In MF approximation one obtains
\[ A(k, \omega) = \sum_\sigma A_\sigma(k, \omega) \] (9)

\[ A_1(k, \omega) = \sum_{\nu, \lambda} |S_{i\nu,\lambda}(k, \omega)|^2 \delta (E_{\lambda k} - \mu - \omega) \]
\[ A_2(k, \omega) = \sum_{\nu, \lambda} |S_{2\nu,\lambda}(k - Q, \omega)|^2 \delta (E_{\lambda, k - Q} - \mu - \omega) \] (10)

Here \( E_{\lambda k} \), \( S_{i\nu,\lambda} \) are defined by (5) and the \( \delta \)-function is replaced by a broadened Lorentz-like function \( \tilde{\delta}(\epsilon) \) with width \( \eta \) or by a function \( \tilde{\delta}(\epsilon) = (4\eta)^{-1} \cosh^{-2}(\epsilon/2\eta) \).

Table below gives the values of energy (per site), the ratios of densities \( 2r_\alpha/r_\gamma \) and the values of local spins on the \( \alpha, \beta, \gamma \) orbits for the spiral, FM, AFM and PM states of model with parameters taken from [8] and \( t_{\alpha\beta} = 0 \).

| \( \tilde{H} \), [eV] | SP | FM | AFM | PM |
|---------------------|----|----|-----|-----|
| 2.708               | 2.7469 | 2.7552 | 2.7570 |
| 2r_\alpha/r_\gamma | 1.935 | 2.15 | 2.137 | 2.04 |
| \( d_\alpha = d_\beta \) | 0.234 | 0.254 | 0.078 | 0 |
| \( d_\gamma \) | 0.160 | 0.181 | 0.093 | 0 |

The states are listed in order of increasing of their energies. The lowest state is a spiral one with vector \( Q \), which removes the nesting instability for both \( \alpha \) and \( \beta \) bands. For all structures a value of the total local spin is of order of \( < S > = \sum_\nu d_\nu \sim 0.6 \). It is consistent with that obtained earlier for FM and AFM states. Note that all results refer to undistorted lattice.

In spite of small difference in the energies the states have very different structures of FS. For PM state one obtains the standard quasi-1D sheets of FS at \( k_{x(y)} = \pm 2\pi/3 \) for \( \alpha, \beta \) bands and the electron-like FS for \( \gamma \) band. The FS structure is in agreement with results of LDA band calculations [20,21] and with analysis of magnetic quantum oscillations. Similar picture of FS has just been obtained [14] from photoemission (ARPES) data for the samples undergone the temperature cycling in range \( T = 10 - 200K \), which removes the shadow FS from the observed map of FS. In [14] it has been suggested that the cycling destroys the surface superstructures responsible for the shadow FS.

A set of FS’s for the FM and AFM states is presented in Fig.2 where the images of spectral function \( A(k_x, k_y, \omega = 0) \) at \( \omega = 0 \) are shown. Corresponding broaden parameter \( \eta \) in \( \tilde{\delta} \)-function in (9) is \( \eta = 0.02 \text{ eV} \). For FM state one has a double set of FS since the bands for the up and down spins are different. In AFM state the FS of the \( \gamma \) band are the boundaries of the electronic pockets which are similar to the hole pockets of cuprates. Contrary to cuprates, here the shadow edges of the pockets are located on inner sides of the pockets closer to the centre of zone \( \Gamma(0,0) \). Due to magnetic doubling of elementary unit there exist also the shadow images of the main FS’s reflected with respect to the lines \( k_x \pm k_y = \pm \pi \). Note that such reflected shadow FS’s are really observed in ARPES map of \( \text{Sr}_2\text{RuO}_4 \). But the same shadow FS may originate from several other possible structures - from the surface (instead a bulk) AFM order or from a lattice reconstruction of the surface layers, as suggested in [19].

Fig. 2 presents the FS’s for the spiral state with \( Q = 2\pi(1/3,1/3) \). Several specific features must be emphasised.

1) The images of spectral functions (9) corresponding to the up and down spin polarisations differ drastically from each other. For instance, the image of \( A_1(k_x, k_y, \omega = 0) \) displays only a half of all FS’s seen in a map of the total spectral function (see. Fig.2). For opposite polarisation one has an inverted structure of FS’s since \( A_1(k_x, k_y, \omega) = A_1(-k_x, -k_y, \omega) \). Such polarisation asymmetry takes place only for the spin polarisations along the axis \( z \) which is normal to the plane of the local spin rotation in spiral state. This asymmetry in the photoemission intensities might be observable by methods of the polarisation photoemission only if an orientation of the spiral structure relative to the lattice remains fixed by a spin-orbit interaction.
2) The shadow FS's appear approximately along the lines \( k_x = 0 \) or \( k_y = 0 \). They can be explained by the Umklapp processes in periodic spiral structure with \( Q = 2\pi(1/3, 1/3) \) since they coincide with the main \( \alpha, \beta \) FS's after a shift \( q \rightarrow q \pm Q \). The position of these shadow FS almost coincides with position of the dispersionless peaks observed in photoemission along lines \( \Gamma(0,0) - M(\pi,0) \) \cite{11} and named the SS features. In \cite{11} the origin of these features has been connected with electronic surface states. It was supposed that these states arise due to a lattice reconstruction of the surface layers \( RuO_4 \) with doubling of elementary unit. Similar lattice distortion with staggered rotation of octahedra was observed in FM phase of the substituted compounds. One of arguments in favour of surface origin of SS patterns was a disappearance of them and the other shadow FS's after the temperature cycling of samples. Our calculations of spiral states allows to suggest a bulk origin of SS features due to spiral spin structure. Then disappearance of the SS features after the temperature cycling might mean a destroying the spiral spin (instead of surface) order in system.

It has been found also the MF solution with the periodic spin and charge structure containing 18 lattice sites in the elementary unit. Its MF energy is slightly higher than that of spiral state, but lower than energies of FM, AFM, PM states of model. In this solution the FS's of \( \alpha, \beta \) bands disappear due to formation of a gap along all former FS's of this bands and the FS of \( \gamma \) band has very complicated form. Both peculiarities are inconsistent with the observed FS's for all three bands.

It was interesting also to calculate the spin susceptibility \( \chi(q, \omega) \) of the three-band system in spiral state. For this aim we apply the RPA approach taken in a form different from the standard RPA. Our method uses the MF state with the spiral spin structure as a zero approximation. Thus the nesting instability is removed non-perturbatively already in zero order approximation. The consideration is inspired by similar treatment of the one-band Hubbard model \cite{12,14}. The details of calculations will be given elsewhere. Here in Fig. 3 we show only the calculated \( Im\chi(q, \omega) \) at small \( \omega = 0.02 \, eV \) for \( q \) varying along the contour \( \Gamma(\alpha,\alpha) - M(\pi,0) - Y_1(\pi,\pi) - \Gamma - Y_2(-\pi,\pi) \) (see insert in Fig.3). Besides the expected absorption peak at \( q = Q \), there is almost symmetric peak near the \( q = 2\pi(-1/3, 1/3) \). Both peak positions are in accordance with the positions of the observed magnetic peaks in INS on \( Sr_2RuO_4 \) at \( q \sim 2\pi(1/3, 1/3, 1) \). Note that an integral intensity \( \tilde{\chi}(\omega) = \int \chi''(q\omega)d^2q/4\pi^2 \) occurs to be of order \( \sim 1 \, eV^{-1} \) which is comparable with the similar quantity \( \sim 2 \, eV^{-1} \) in cuprates \cite{22}.

In conclusion, the main and shadow sheets of FS are obtained for the PM, FM, AFM and spiral states of three-band model of \( RuO_4 \) plane of \( Sr_2RuO_4 \). The map of FS's is highly specific to the spin structure. The lowest MF state appears to be the spiral state with the spiral- vector \( Q = 2\pi(\frac{1}{3}, \frac{1}{3}) \) removing the nesting instability for both quasi 1D \( (\alpha \) and \( \beta \) bands simultaneously. For spiral state the main FS's display in photoemission with intensity depending on the spin polarisation. Such polarisation asymmetry of photoemission spectra may be a test for the spiral structure in \( Sr_2RuO_4 \). In spiral state the shadow FS's along \( \Gamma - M \) line are revealed. Coincidence of their position with the position of the dispersionless SS peaks in ARPES map of \( Sr_2RuO_4 \) allows to suggest the bulk origin of the observed SS features as caused by Umklapp processes in spiral state. The calculation of the spin susceptibility \( \chi(q, \omega) \) for spiral state confirms the existence of the magnetic peak in the low frequency spin excitations at \( q \sim Q \) in accordance with the INS observations. If the spiral structure does exist in ground state of \( Sr_2RuO_4 \) the new questions can be put forward on the symmetry of superconducting pairing, on phase of quantum magnetic oscillations and so on.

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\[1\] Y.Maeno, T.M.Rice, M.Sigrist, Physics today, January 2001, 42-47.
\[2\] T.M.Rice, M.Sigrist J.Phys.:Cond.Mat. 7,L643 (1995)
\[3\] K.Ishida, H.Mukuda, Y.Kitaoka et al. Nature, 396, 658
Captions to Figures

Fig. 1. The Fermi surfaces for the FM (left) and AFM (right) spin structures of three-band model obtained as image of spectral function (9) at \( \omega = 0 \) and the broadening parameter \( \eta = 0.02\text{eV} \). The model parameters are those from [6] and \( t_{\alpha\beta} = 0.01\text{eV} \).

Fig. 2. The Fermi surfaces of system in spiral state with vector \( Q = 2\pi(\frac{2}{3}, \frac{1}{3}) \): 1) the image of spectral function \( A_{\uparrow}(k_x, k_y, \omega = 0) \) for the spin-up polarisation (left), 2) the same for the total spectral function (9) (right). The parameters are those from [6] and \( t_{\alpha\beta} = 0 \). Some nonlinear transformation of \( A(k, \omega) \) is applied to strength the shadow bands.

Fig. 3. The imaginary part of spin susceptibility \( \chi''(q, \omega) \) averaged over polarisations (full curve) and its components \( \chi''_{xx}(q, \omega) \) (dashed) and \( \chi''_{zz}(q, \omega) \) (dotted) at \( q \) varying along the contour \( \Gamma - Y_1 - M - G - Y_2 \) (see insert). The frequency and the broadening parameter are \( \omega = \eta = 0.02\text{eV} \).
\( \omega = 0.02, \gamma = 0.01 \)

\( Q = 2\pi(1/3,1/3) \)