

Electronic structure of Sr_2RuO_4 by means of local-density approximation plus strong correlation effects

A. Pérez-Navarro, J. Costa-Quintana, and F. López-Aguilar

Grup d'Electromagnetisme, Edifici Cn, Universitat Autònoma de Barcelona, 08193 Bellaterra, Barcelona, Spain

(Received 4 November 1998; revised manuscript received 16 September 1999)

The electronic structure of Sr_2RuO_4 according to experimental spectroscopic data presents strong correlation in the electrons at energies close to E_F . We investigate this point by analyzing the electronic structure calculated from several complementary procedures. The first of these is performed by means of the local density approximation (LDA). In a second calculation we develop an LDA+ U calculation and finally, we include the self-energy effects obtained from a multiband Hubbard model. Our results for the first calculation are similar to those previously performed within the LDA, and these results are used for obtaining the self-energy effects included in the second calculation. The inclusion of these effects yields an electronic structure that seems to be in reasonable agreement with spectroscopic experimental data.

I. INTRODUCTION

Sr_2RuO_4 (SRO) is a layered compound whose crystal symmetry is equal to that of La_2CuO_4 (LCO).¹⁻⁴ In SRO the RuO_2 planes play a similar role to that of the CuO_2 ones in LCO. The dynamics of the charges within CuO_2 planes are responsible for the high T_c superconductivity since they are a common structural element of high-temperature superconductors.⁵ It seems reasonable to conjecture that the clues for the superconducting behavior of SRO should also be looked for in these sheets in a way analogous to that used in high- T_c systems. On the other hand, the normal state of these cuprates is very unusual, and it seems clear that it is fundamental to understand these anomalous normal state properties in order to have grounds for comprehending the possible mechanisms of superconductivity.

The electronic structure of SRO has some *a priori* differences with respect to that of LCO systems; this is interesting to analyze in order to obtain different routes for explaining the high- T_c phenomena.¹ The $3d^9$ configuration of the copper layer is substituted by four $4d$ electrons of Ru. The antibonding orbitals at E_F are derived in the ruthenates from d_{xy} , d_{xz} , and d_{yz} rather than from the $d_{x^2-y^2}$ orbital as in the cuprates. However, these different configurations for the corresponding metallic atoms do not hinder the fact that in both materials there are similar van Hove singularities near E_F .⁶ The strong correlation effects are present in both electronic structures,^{1,7-9} although these are more subtle in SRO than in LCO. Recent papers claim that the electronic correlations in SRO play a quantitative role, but from a qualitative point of view their effects are more modest.¹⁰ This is the reason for which the local density approximation (LDA) is able to explain some properties of the electronic structure^{10,11} although, as is well known, LDA fails in other properties in a similar way to that of the cuprates.

One important feature that still remains unexplained is the different Fermi surface obtained by means of the de Haas van Alphen and angle-resolved photoemission.¹² The different result seems to be due to the fact that the first gives the bulk properties and the second the surface properties.^{10,11} The LDA calculations are only able to fit the first. In addition,

it is important to say that, as in the high- T_c cases, the number of states located at E_F is one of key data for explaining their superconducting behavior and LDA is clearly unable to fit this number of states, since all calculations in this approximation yield a density of states (DOS) at the Fermi level, $N(E_F)$, more than three times larger than that of the experimental spectroscopic measurements.⁹

Obviously, the similitude of crystal symmetries and electronic structure (although with some differences), implies that the physical properties present analogies and differences. The resistivity above T_c is, in both LCO and SRO, clearly anisotropic. In the case of SRO, it is three orders of magnitude larger in planes than in chains.¹ This property is coherent with the two-dimensional character of this system. The normal state of SRO is a metallic Pauli paramagnetic material in opposition to LCO which is an antiferromagnetic insulator.⁸ The superconductivity in SRO occurs by itself without requiring doping as in LCO system.¹ The T_c in SRO is less than 1 K (0.93 K)¹ and, in contrast to the doped LCO system, lies between 30 and 40 K.

As has been said above, the DOS at the Fermi level obtained from the LDA calculation is three times larger than that obtained from spectroscopical measurements.⁹ This leads us to think about the requirement of including strong correlation effects within the band-structure calculation. However, we will see that these strong-correlation effects introduce important quantitative variations into the DOS which imply an improvement of our results that are closer to the experimental data than those arising from the LDA calculations. Achieving the electronic structures by considering strong correlation of the particles close to E_F is an initial point that should be treated in this system^{1,6-9,13} and that we introduce in the analysis of the electronic structure of SRO. However, although the spectroscopical data suggests the inclusion of U effects, on the other hand, the specific heat measurement^{1,14} leads us to think that other different effects, probably magnetic fluctuations, should be included in order to conciliate the thermodynamical data with the photoemission measurements. The analysis of the influence of spin fluctuations is an issue that lies beyond the aims of this paper.

We start our work by calculating the electronic structure of the ground state of the non-interacting system by means of a LDA calculation, which is shown in Sec. II. The reasons for beginning our analysis with this calculation is to obtain a noninteracting system (i.e., without self-energy effects), and to compare our LDA results with other previous papers^{2,3} and with spectroscopic data.

In Sec. III, a first correction to LDA is introduced, the so-called LDA+ U ,¹⁵ which is based on a first order perturbation theory considering the Hubbard Hamiltonian. This correction is able to yield the Hubbard-like splitting, but it is not yet able to show the dynamic effects that may substantially modify the pattern of the electronic structure.

In Sec. IV, we consider the LDA results in order to determine the analytical expression of the self-energy in the random phase approximation (RPA) within the multiband Hubbard model.¹⁶ This third calculation using the self-energy function describes the system of interacting particles by means of a system of quasiparticles whose states are not stationary but are quasistates with finite half life. The self energy obtained for determined band parameters, violates one of the conditions of the Luttinger theorem. According to this theorem, the zero of the imaginary part of the self-energy corresponds to the Fermi level, and the self-energy introduced can present more than one zero. Obviously, this failure of the self energy in this case, makes it invalid for determining the quasiparticle system. Fortunately, the deviations of the self energy with respect to that analytical functions that satisfy the Luttinger condition are relatively small. There is, then, a compromising situation between the correctness of self-energy expressions concerning the Luttinger theorem, and obtaining this with realistic band parameters that, as indicated above, should be obtained by fitting the LDA calculation. Following this, we have studied the dependence of the DOS on the variations of the band parameters included within the calculation of the self energy. This allows us to calibrate the validity of our results, as well as to estimate their stability, since good results obtained with a fitted self energy could be qualified as fortuitous and even dependant on the fitting parameters. However, our conclusion is that the results obtained with self energy represent an improvement of LDA calculation in any point of the region of the space parameters in which we have calculated the self energy, and we are reasonably sure that the LDA band parameters can be represented by a point in this region.

II. LOCAL DENSITY APPROXIMATION CALCULATION

The crystal structure of Sr_2RuO_4 is the same as that of K_2NiF_4 with the $I4/mmm$ group. The crystallographic parameters have been taken from Vogt and Buttrey¹⁷ at 0.35 K: $a = 3.86111 \text{ \AA}$, $c = 12.7219 \text{ \AA}$. At this temperature, the position of the atoms are: $z(\text{Sr}) = 4.49458 \text{ \AA}$, $x(\text{O}_1) = 1.93056 \text{ \AA}$ and $x(\text{O}_2) = 2.07109 \text{ \AA}$. The muffin-tin radii taken in our calculations are: $R_{\text{Sr}} = 2.908 a_0$, $R_{\text{Ru}} = 2.070 a_0$, $R_{\text{O}(1)} = R_{\text{O}(2)} = 1.474 a_0$; where a_0 is the Bohr radius. These data are similar to that used in previous calculations,^{2,3} although small variations can be seen since Oguchi takes the parameters at 100 K. The resulting band structure is shown in Fig. 1. The main reason for performing this first calculation is to compare both results and check that our starting

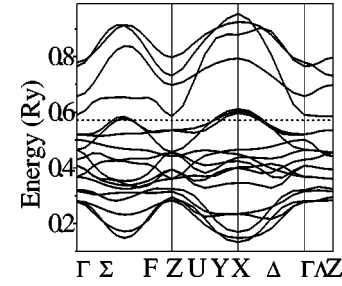


FIG. 1. Band structure of Sr_2RuO_4 . The dotted horizontal line denotes the Fermi level.

noninteracting system contains a similar band structure. This allows us to be sure that the differences found in the following calculations give an estimation of the strong correlation effects and are not due to a different method used in the solution of the LDA Hamiltonian.

Comparing Fig. 1 with that obtained by Oguchi² and Singh³ we see that our result is in reasonably good agreement with them. The first twelve bands mainly correspond to bonding d states of Ru and O p orbitals, and the nonbonding p states of O. Around Fermi level we find three antibonding states coming from d_{xz} , d_{xy} , and d_{yz} of Ru and $p \pi$ orbitals of oxygen. Above Fermi level, we find anti-bonding bands coming from orbitals d_{3z^2-1} and $d_{x^2-y^2}$ of Ru, d of Sr, and $p\sigma$ of O. This is an important difference with cuprates where the bands at Fermi level are of kind $d_{x^2-y^2}$. Below Fermi level, and away from it, some irrelevant differences appear that can be attributed to the different standard methods considered in both calculations (we used the symmetrized augmented plane wave method, Ref. 16). Another important coincidence between our results and previous theoretical works^{2,3} is that three bands cross Fermi level at line ΓX and two at long-line ΓZ , and the small dispersion in c -axis, as can be seen along the short-line ΓZ , which confirms the bidimensionality of the material. This bidimensionality can also be seen in the quasi-mirror symmetry of bands with regard to the middle point of long-line ΓZ .

In Fig. 2 we show the DOS corresponding to that band structure. In agreement with theoretical^{2,3} and experimental data,⁴ we find hybridization from O(1) $2p$ and Ru $4d$ states. We obtain that the peaks at E_F have d_{xz} , d_{xy} and d_{yz} character, hybridized mainly with orbitals p_y and p_z of O(1), and below Fermi level, the character of π orbitals of O(2) is also important. In Refs. 2 and 3 a pseudogap appears 1.3 eV below Fermi level. In our case this gap is more narrow and a new peak, marked in the figure as *II* appears. These results are still compatible with experimental DOS results,^{8,9} since the experimental broadening of the peaks cannot decide the existence of this new peak. The results from Schmidt *et al.* show 4 main features called peaks A, B, C, and D. In our case, the structure formed by peaks *Ia* and *Ib* can correspond to peak A, and that formed by peaks *II* and *III* to peak B. Peaks C and D can correspond to the structure of peaks that is found below peak *III*.

The main physical properties depend on the electronic structure exactly at Fermi level and at this location some important differences appear between the experimental data^{9,18} and the LDA calculations. Experimentally, the number of states at Fermi level is found to be $N(E_F) = 19$

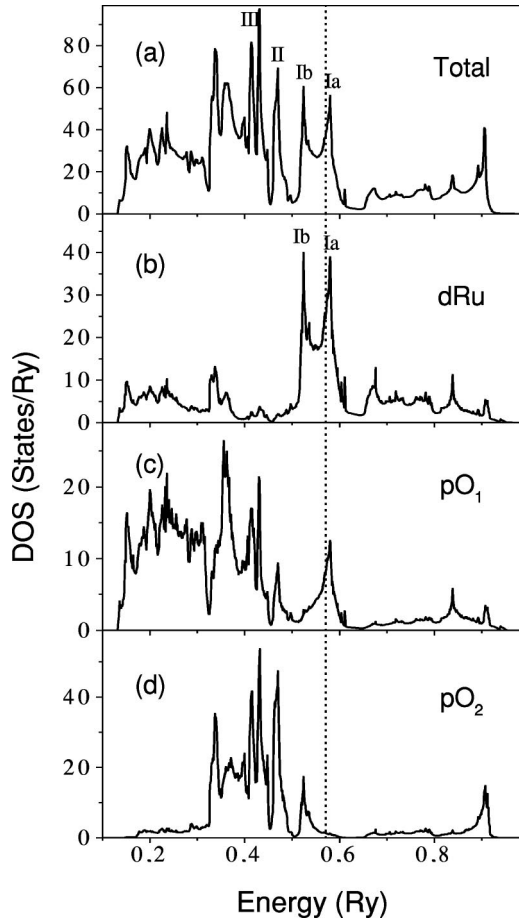


FIG. 2. LDA density of states of Sr_2RuO_4 (a) Total DOS; (b) partial $4d$ DOS of Ru; (c) partial p DOS of O_1 ; and (d) partial p DOS of O_2 .

states/Ry (1.4 states/eV),⁹ which is between three and four times less than that of the LDA calculations: Oguchi² give 59.4 states/Ry (4.4 states/eV) and Singh³ 56.2 states/Ry (4.1 states/eV), and in our calculation we obtain 36.8 states/Ry (2.7 states/eV), which is a better result, but is still twice the experimental result. The reason for the disagreement between previous theoretical results and ours could be the differences in the method used in every case, as well as in the different parameters introduced. Since there is a peak very close to the Fermi level, small variations in the position of this peak give strong differences in the number of states at Fermi level. In fact, in Fig. 2 it can be seen that the Fermi level do not fall exactly at the peak, but between two peaks. On the other hand, the origin of the disagreement between all the theoretical results and the experimental ones can arise from the above-mentioned intra-atomic correlations produced by the relative localization of the $4d$ electrons of Ru. These intra-atomic interactions produce a tendency to split in lower and upper Hubbard bands. But since the width of the $4d$ band is large enough, this Hubbard correlation cannot yield the total upper and lower splitting, but can produce a certain pseudogap and therefore a decreasing of DOS at E_F . The existence of this tendency to form the electronic correlation Pseudogap may be the reason for which the strong correlation band structure calculation improves the results of the LDA.

In LDA, the correlations are introduced via exchange and

correlation local potential. However, as is well known, this way to include these correlations between d electrons can be largely inappropriate (larger localization implies larger inability in the LDA interpolation formula for determining the electronic correlation). This is so, because LDA assume a fixed $4d^x$ configuration, x being the non integer number of $4d$ electrons. When there is localization and movement between lattice sites of these $4d$ electrons, as with the analyzed case, this x number can fluctuate in one electron. This fluctuation implies severe and sharp modification in the interaction energy within the site, equivalent to the familiar U energy, that in this material is around 2.5 eV. Therefore, the localization plus the dynamic transference of electrons lead to the necessity of introducing dynamic correlation effects.

In all experimental studies it is recognized that evidence for correlation effects have been found^{1,6,8,9,18,19} and today the importance of such effects in Sr_2RuO_4 is accepted.²⁰ However, it is also recognized that the influence of correlations will not be as important as in cuprates. In any case, the correlated electrons are those in the $4d$ orbitals, which present their main contribution at the Fermi level, as can be seen in Fig. 2. The inclusion of correlation effects will not therefore markedly affect the structure away from Fermi level, but the variations around it can be very important, as we will see in Sec. IV.

III. LOCAL DENSITY APPROXIMATION PLUS U CALCULATION

As a first approximation to include the correlation effects in d orbitals, we develop an LDA+ U calculation. We take the experimental value,²⁰ $U=2.4$ eV (0.18 Ry), for all d orbitals. The resulting DOS is shown in Fig. 3. The inclusion of these effects displaces the Fermi level and the structure around it from 0.57 to 0.68 Ry. Despite this displacement in the energies of the structure, the main features remain invariable with only some differences. Below the Fermi level, a gap appears, as a consequence of the standard Hubbard U splitting between upper and lower bands. The gap is situated 0.1 Ry below the Fermi level and its width is 0.08 Ry. Singh³ and Oguchi² find a gap 0.08 Ry below Fermi level and width of 0.08 Ry applying only an LDA calculation. The origin of both gaps is different, because the LDA+ U gap appears, as stated above, because of the splitting of the d bands where U was applied. But as we indicated in the previous section, there is no experimental evidence for its existence. Although correlation has been introduced only in the d orbitals, the gap can be seen not only in the partial d structure shown in Fig. 3(b), but also in the partial p of $\text{O}(1)$ and partial p of $\text{O}(2)$ because of the hybridization. The two peaks around the Fermi level found in the LDA calculation are displaced to the right, which reduces the number of states at Fermi level to 32.2 states/Ry (2.4 states/eV), which is an important improvement if we compare with that obtained by means of the LDA calculation. However, the bandstates calculated with LDA+ U are stationary and therefore, this method is not able to show the dynamic effects which, in this case, could be important. In the next section we refine the calculation with the inclusion of a self energy.

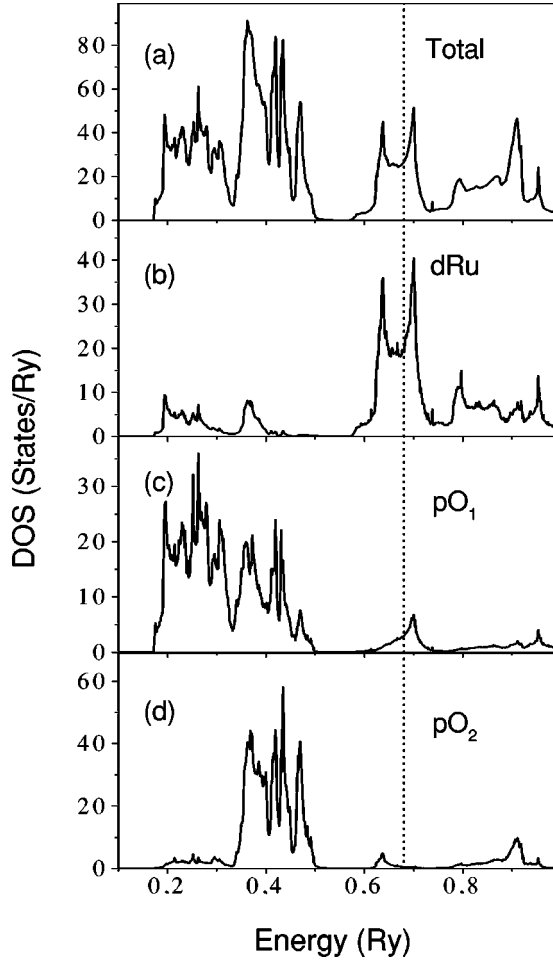


FIG. 3. (a) Total DOS of Sr_2RuO_4 obtained by an LDA+ U calculation with $U=0.18$ Ry; (b) partial $4d$ DOS of Ru; (c) partial p DOS of O_1 ; and (d) partial p DOS of O_2 .

IV. SELF-ENERGY EFFECTS

The dynamic effects in the interacting system are calculated from the self energy, which is added to band Hamiltonian by considering the LDA electronic structure as non-interacting ground state. The self energy will transform the particles into quasiparticles with a non-infinite half life. As we saw in Fig. 2(b) for the partial density of states of orbitals d , the main features of these correlated orbitals are the double peak around the Fermi level. Thus, if the self energy is introduced into the d orbitals, the states at Fermi level will become non-stationary quasistates with a half-life and therefore, the contribution to the DOS of the each d eigenvalues will be smaller. On the other hand, the spectral function of each strongly correlated d eigenvalue can present several peaks and therefore for each stationary state of the LDA groundstate can correspond a quasistate that contains several significant contributions to the DOS can be centered at different energies, whose differences are of the order of U . Therefore, each d orbital can produce several resonances (we call resonances each peak in the spectral function yielded by the self energy). As a consequence, we can find at E_F states whose characters correspond to different d -orbitals which could be absent in the LDA calculation where the spectral function for each stationary state is an only delta function. Therefore *a priori*, the number of states at Fermi level can

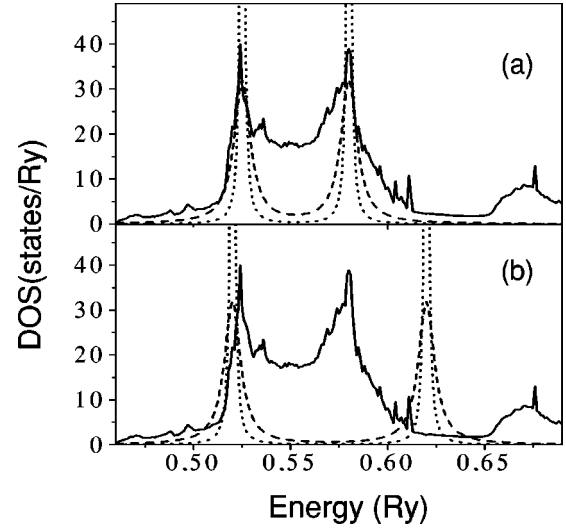


FIG. 4. Several double Lorentzians compared with the partial DOS of Ru of the LDA calculation. In all cases the weight of the Lorentzians is the same for both peaks, i.e., $n=0.5$. (a) Asymmetric double Lorentzian with every peak situated exactly at the same point as the peaks of the partial DOS: $\lambda=0.045$ Ry and $\xi=0.01$ Ry referenced to the Fermi level; (b) Symmetric double Lorentzian with $\lambda=\xi=0.05$ Ry. In both cases the dashed line corresponds to a width of $\Lambda=\Xi=0.005$ Ry and the dotted line to $\Lambda=\Xi=0.001$ Ry.

increase, decrease, or remain the same as in the LDA calculation.

The self-energy is calculated at $T=0$. We list below certain approximations that we have borne in mind:

(i) As is well known, the dependence on \mathbf{k} in strongly correlated systems is much smaller than the frequency dependence.^{16,21-23} Therefore, we only take into account the ω -dependence by considering average values of the self energy in the first Brillouin zone. The \mathbf{k} dependence has been avoided in some other works too, as for example in Ref. 10. (ii) In the partial density of states of d -orbitals in Fig. 2(b) there is a double peak around Fermi level that could be well fitted by a double Lorentzian. The reason for considering the fitting of the DOS in the proximity of E_F is to obtain analytical expressions for the self energy, which markedly reduce the complexity of the calculation without losing reliability. This allows us to obtain many calculations for different self energies without excessive computation time, and to perform an exhaustive analysis of the dependence of the results on the band parameters of the noninteracting ground state.

Figure 4 shows this partial DOS over-struck with some double Lorentzians. With these two approximations, we obtain the following self energy within the random phase approximation¹⁶

$$\Sigma(\omega) = -\frac{U^2 f}{2} \left[\frac{1}{\Omega_1} \left(\frac{n}{\omega + \Omega_1 + \lambda - i\Lambda} + \frac{1-n}{\omega - \Omega_1 - \xi - i\Xi} \right) + \frac{1}{\Omega_2} \left(\frac{n}{\omega + \Omega_2 + \lambda - i\Lambda} + \frac{1-n}{\omega - \Omega_2 - \xi - i\Xi} \right) \right], \quad (1)$$

where U is the Coulomb correlation, $f=2gn(1-n)$; $g=(\lambda+\xi)-i(\Lambda+\Xi)$, and λ , ξ and Λ , Ξ correspond respectively, to positions and widths of the Lorentzians that have

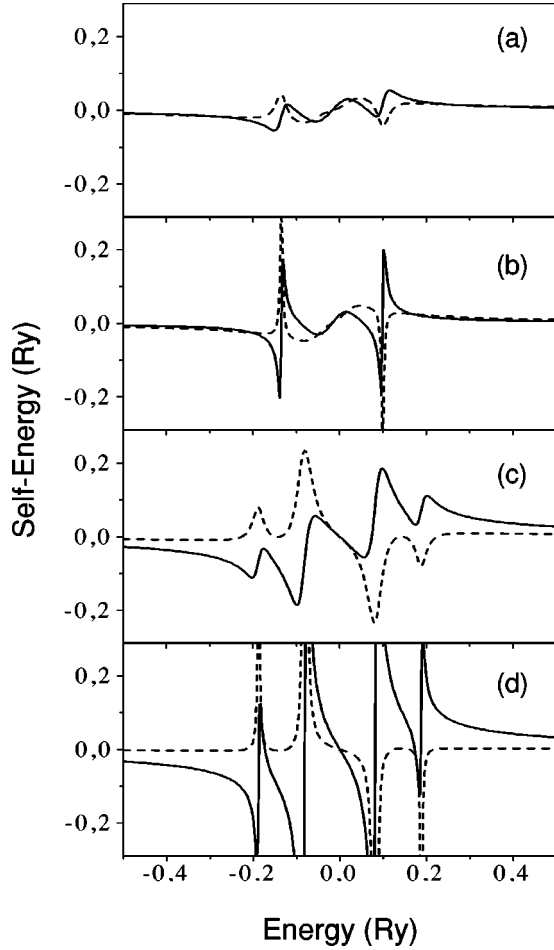


FIG. 5. Self-energies corresponding to the Lorentzians represented in Fig. 4. In all cases $U=0.18$ Ry and $n=0.5$. (a) $\lambda=0.045$ Ry, $\xi=0.01$ Ry and $\Lambda=\Xi=0.005$ Ry; (b) $\lambda=0.045$ Ry, $\xi=0.01$ Ry and $\Lambda=\Xi=0.001$ Ry; (c) $\lambda=\xi=0.05$ Ry and $\Lambda=\Xi=0.005$ Ry; (d) $\lambda=\xi=0.05$ Ry and $\Lambda=\Xi=0.001$ Ry. It can be seen as (a)–(b) violates the Luttinger condition while (c)–(d) do not.

been used as non-interacting DOS, as can be seen in Fig. 4; n is the occupation number that gives the weight of each Lorentzian; and $\Omega_1^2=g^2-Uf$ and $\Omega_2^2=g^2+Uf$ are the plasmon frequencies. We diagonalize the interacting Green functions as is explained in Ref. 24. We wish to emphasize that Eq. (1) arise from the summation of the infinite series of diagrams corresponding to the RPA. The resulting self energy may appear to correspond to a second order diagrams with a bare U interaction due to the U^2 dependence. But this is an erroneous estimation since, in Eq. (1) the “plasmon poles,” Ω_1 and Ω_2 also depend on the U energy.

The parameters to introduce in the self-energy are U , λ , Λ , ξ , Ξ , and n . We consider $U=0.18$ Ry, as we did in LDA+ U method. As a first step, we will take $n=0.5$, i.e., we will consider half-filling. The center of the peaks of the double Lorentzian, λ and ξ , will be taken as the centers of the two d peaks as shown in Fig. 4, (i.e., $\lambda=0.045$ Ry and $\xi=0.01$ Ry, with respect to the Fermi level), and $n=0.5$ because the two peaks are very similar. We can take different values of the width of the different peaks $\Lambda=\Xi=0.005$ Ry, and $\Lambda=\Xi=0.001$ Ry, as can be seen in Fig. 4. Figures 5(a) and 5(b) show the drawing of the self energies corresponding

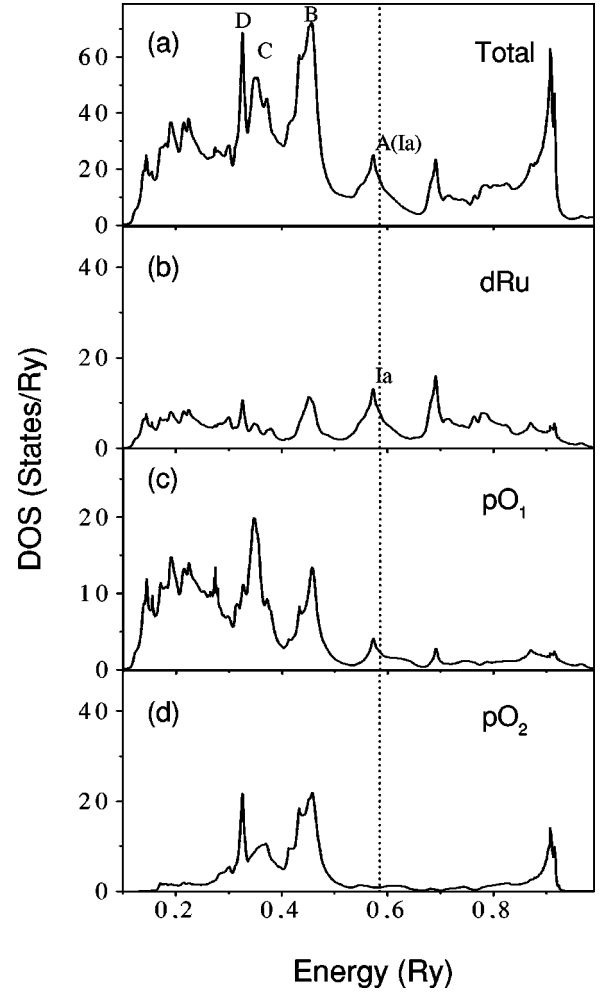


FIG. 6. The DOS of Sr_2RuO_4 taking into account the self-energy effects With $U=0.18$ Ry, $n=0.5$, $\lambda=\xi=0.05$ Ry and $\Lambda=\Xi=0.001$ Ry. (a) Total DOS; (b) partial $4d$ DOS of Ru; (c) partial p DOS of O_1 ; and (d) partial p DOS of O_2 .

to these parameters. The solid line (dashed line) represents the real (imaginary) part of the self-energy. We can see that in both cases the imaginary part is slightly negative in several intervals of the frequency and therefore, presents more than one zero and, according to the Luttinger theorem, the only zero of the imaginary part of the self energy should correspond to the Fermi level. Thus, these parameters give a self energy that slightly violates the Luttinger sum rule. Therefore, we should make small variations in the band parameters which define the self energy in order to satisfy the Luttinger condition. Analyzing the expression of the self energy, we can see that the reason for this violation is that the two Lorentzians are too near to each other, i.e., $\lambda+\xi$ is too small. We slightly separate the Lorentzians until the Luttinger condition is satisfied and find that the following parameters: $\lambda=0.05$ Ry and $\xi=0.05$ Ry. In Figs. 5(c) and 5(d), they have been represented the resulting self-energies with $\Lambda=\Xi=0.001$ Ry and $\Lambda=\Xi=0.005$ Ry respectively.

Figure 6 represents the resulting DOS for the self energy shown in Fig. 5(c). It presents two important new features: (i) In Fig. 6, the peak Ib of Fig. 2(a) centered at 0.05 Ry below the Fermi level (exactly where one of the peaks of the Lorentzians has been centered) has disappeared and now

there is a single peak below E_F ; and (ii) the peak *II* of Fig. 2(a) has disappeared too. Both below and above E_F there are pseudogaps originated by the strongly correlation effects. In general the structure with the inclusion of the self energy is smoother due to the non-stationary condition of the quasistates whose finite half life decreases their influence in the DOS. In addition, in Fig. 6 the splitting of every d state in several resonances has also influence in the DOS. However, as indicated above, the strong correlation effects produce quantitative changes in the DOS, but it maintains qualitative similitude with the electronic structure calculated with LDA. Looking at the partial density of states, we see that the origin of the peaks is the same as that of the LDA calculation. The p partial DOS is smoother too, although small changes can be observed which are due to the hybridization. As will be explained in the following paragraph, the improvements are more quantitative than qualitative, except around Fermi level where the influence of correlations is more important.

The results of DOS of Fig. 6 are in reasonable agreement with experimental data,^{4,8,9} above all with respect to the results obtained by Schmidt *et al.*⁹ We have identified the experimental peaks (*A*, *B*, *C*, and *D*) in our results of Fig. 6 in order to show this agreement by comparison. As was expected, as we move away from Fermi level, the agreement between theory and experiment decreases, but this zone is where our study is less reliable. What we have called peak *A* is experimentally found just below Fermi level. This is the peak responsible for the number of states at Fermi level, and in this case we find that it is $N(E_F) = 16.3$ states/Ry. It is an important improvement with respect to all the previous calculations. However, as has been explained in Sec. I, as the Fermi level is very close to a peak, a small variation in the position of this peak can imply that the number of states undergo a great variation. In order to see the effects of the self-energy, we can also look at the height of the peak: in the LDA calculation, shown in Fig. 2(a), the peak close to Fermi level, marked as *Ia* corresponded to $N_{Ia} = 56.9$ states/Ry. In the LDA+ U calculation $N_{Ia} = 51.6$ states/Ry and the height of the peak in the present case is $N_{Ia} = 25.0$ states/Ry. We can see then, that the effect of including correlation by means of the self energy reduces the number of states at Fermi level, since the correlated orbitals, which are the d orbitals, presented the more important contribution to the density of states around Fermi level.

To determine the influence of different parameters which define the self energy, we have carried out many other calculations and we present some of them in Figs. 7 and 9. Figure 7 shows the DOS obtained with the self-energy drawn in Fig. 5(d), i.e. taking a width of $\Lambda = \Xi = 0.005$ Ry, five times that introduced in Fig. 6. In this case the number of states at Fermi level increases to $N(E_F) = 30.9$ states/Ry, which still is an improvement with respect to the LDA calculation, and the height of the peak is $N_{Ia} = 35.5$ states/Ry. It is clearly worse than the previous result, although it is an important improvement with respect to LDA and LDA+ U calculations. Qualitatively Figs. 6 and 7 present the same characteristic, compatible with the experimental results, i.e., a peak very close to the Fermi level and below it. Looking at the experimental results⁹ we see that the height of the peak is in the order of two times the value of the number of states at Fermi level, and this restriction is not accomplished in this

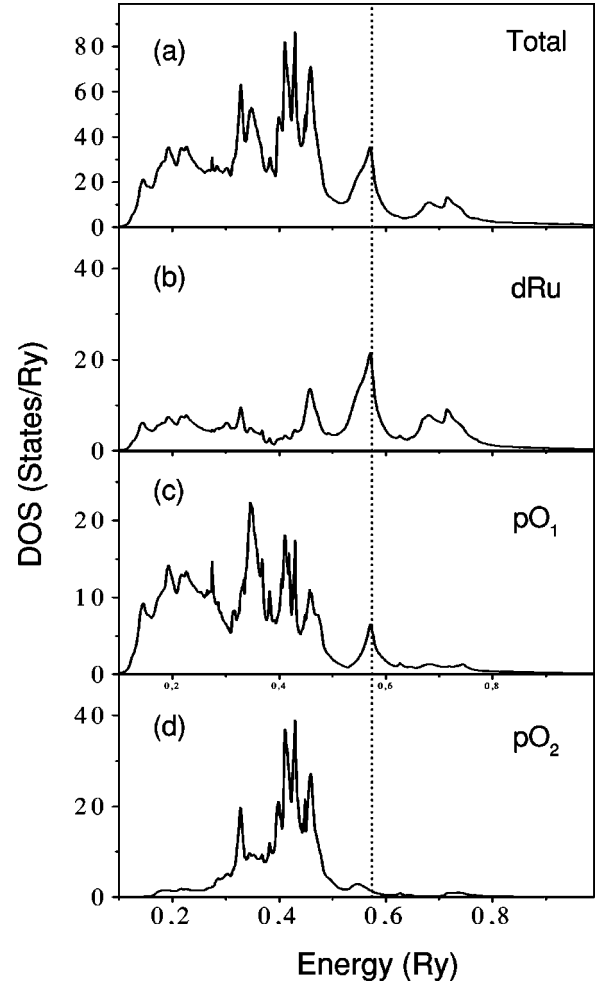


FIG. 7. The DOS of Sr_2RuO_4 taking into account the self-energy effects with $U=0.18$, $n=0,5$, $\lambda=\xi=0.05$ Ry and $\Lambda=\Xi=0.005$ Ry. (a) Total DOS; (b) partial $4d$ DOS of Ru; (c) partial p DOS of O_1 ; and (d) partial p DOS of O_2 .

case, given that the peak is too close to the Fermi level. Although the value of the number of states at Fermi level is very different, results from Figs. 6 and 7 are very similar, and in both cases the number of states at the Fermi level is reduced. The fit to the experimental results is better when the height of the Lorentzians is similar to the height of the peaks of the partial d DOS. Therefore, we conclude that despite the dependence of the self-energy with the width of the Lorentzians, the resulting DOS is not so dependent.

In Figs. 6 and 7, there is a feature that would be important to remark, which is the disappearance of peak *Ib*, precisely where one of the peaks of the Lorentzians is centered. As this peak has the origin on the d orbitals where the self energy have been applied, the effect of splitting of the d states into quasistates is more important as the peak *Ia* is of character d too, it is useful to see the effects of a self energy with one of the peaks of the Lorentzians centered at *Ia*. In order to find a self energy able to satisfy this condition and the Luttinger sum rule, we will vary the parameter n , as well as the width of the Lorentzians, in such a way that the resulting one would be as close as possible to the double Lorentzian introduced in Fig. 6. The resulting self-energy is shown in Fig. 8 with the following parameters: $\lambda = 0.045$ Ry and $\xi = 0.01$ Ry in order that the center of the Lorentzians would be at the

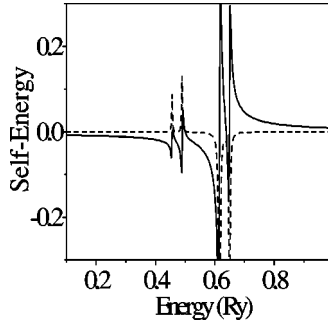


FIG. 8. Asymmetric self-energy with the peaks centered above the peaks of the partial d DOS shown in Fig. 2(b) and accomplishing the Luttinger condition. $U=0.18$ Ry, $n=0.1$, $\lambda=0.045$ Ry, $\xi=0.01$ Ry, $\Lambda=0.0001$ Ry and $\Xi=0.001$ Ry.

same place than the peaks Ia and Ib of Figs. 2(a) and 2(b); $\Lambda=0.0001$ Ry, $\Xi=0.001$ Ry and $n=0.1$. With these parameters the double Lorentzian is similar to that introduced in Fig. 6 and the self energy is Luttinger coherent. In Fig. 9, we show the corresponding DOS. In this case the features are

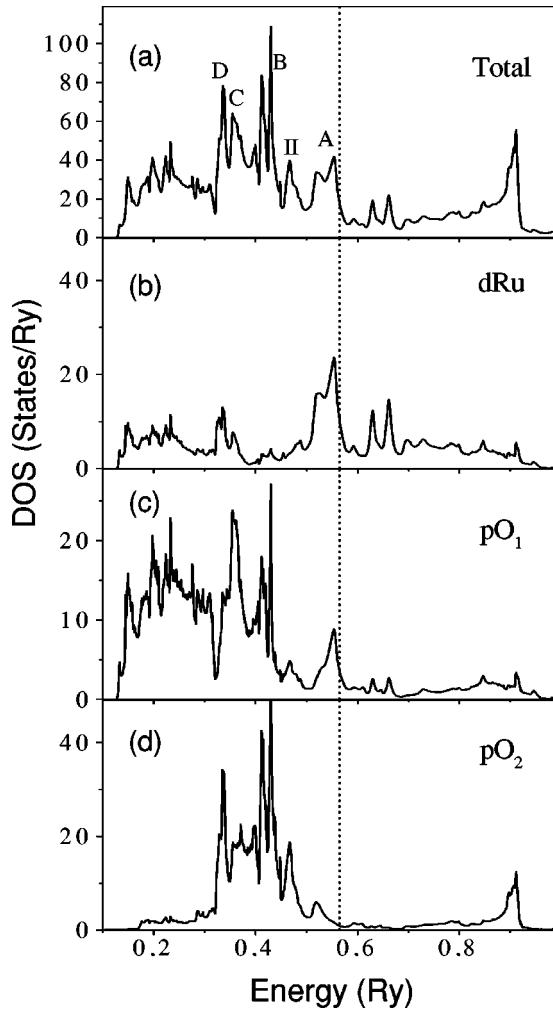


FIG. 9. The DOS of Sr_2RuO_4 taking into account the self-energy effects of Fig. 8, i.e., with $U=0.18$ Ry, $n=0.1$, $\lambda=0.045$ Ry, $\xi=0.01$ Ry, $\Lambda=0.0001$ Ry and $\Xi=0.001$ Ry. (a) Total DOS; (b) partial $4d$ DOS of Ru; (c) partial p DOS of O_1 ; and (d) partial p DOS of O_2 .

somewhat different from that of Figs. 6 and 7, and have more similar aspects to that of Fig. 2, i.e., the LDA calculation. In Figs. 6 and 7, as the peak Ib disappeared, the remaining peak was considered peak A. In the case of Fig. 9, an overly small double peak structure appears that can be considered as peak A, and is very close to the experimental results obtained by Schmidt *et al.*⁹ As in the spectroscopic data, it is below the Fermi level and with negative slope at that point. The number of states is $N(E_F)=18.1$ states/Ry, is closer to the experimental result [$N(E_F)=19$ states/Ry] than the previous calculations. In this case, The height of the peak is of $N(\text{Peak A})=41.7$ states/Ry, higher than in Fig. 6 but smaller than in the LDA.

In all the cases where the self energy have been included, the experimental structure around Fermi level have been quantitatively improved. Other self energies have been tested, symmetrical as that shown in Figs. 5(c)–5(d) and non-symmetrical as that shown in Fig. 8. In all cases have been found very similar results to that shown in Figs. 6 and 9 when the height of the Lorentzians is similar to the height of the peaks of the partial d density of states shown in Fig. 2. In Fig. 9, as well as in Figs. 6 and 7, the structure of peaks has suffered more quantitative than qualitative changes and the origin of the peaks is in all cases the same than commented in section II for the LDA calculation.

V. CONCLUSIONS

In this paper, we have seen that the LDA and LDA+ U calculation fails when trying to fit the spectroscopic data quantitatively, although in both cases we find reasonably good qualitative agreement with the experimental results. In this context, the inclusion of correlation effects find its justification. They have been included by means of a self-energy obtained by averaging to \mathbf{k} 's and introducing a double Lorentzian as a non-interacting DOS. The parameters of this Lorentzian have been obtained by fitting the double central peak of the partial d density of states arising from the LDA calculation. The self-energy has only been introduced into the d orbitals, which are the localized and, therefore, strongly correlated states. The main effect is the reduction of the number of states at the Fermi level, i.e., the height of the double peak of the d orbitals at the Fermi level is reduced. The results are more similar to the experimental ones according to whether the double Lorentzian is closer to the double peak. When the height of the peaks of the Lorentzians are similar to those of the LDA peaks, the partial d DOS are similar, and the number of states at the Fermi level remains around the experimental value. On the other hand, the structure of peaks next to the Fermi level is also qualitatively improved, in comparison with the experiment. The conclusion we obtain from this paper is that although the correlation effects are not particularly large, they need to be included in describing the features of the electronic structure, and have a certain quantitative importance in explaining the experimental results of Sr_2RuO_4 , and, above all, in explaining the results of the electronic structure near E_F .

ACKNOWLEDGMENTS

This work has been financed by the DGES (PB96-1143). One member of the team (A.P.N.) acknowledges the grant from CUR de la Generalitat de Catalunya.

- ¹Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. G. Bednorz, and F. Lichtenberg, *Nature (London)* **372**, 532 (1994).
- ²T. Oguchi, *Phys. Rev. B* **51**, 1385 (1995).
- ³D.J. Singh, *Phys. Rev. B* **52**, 1358 (1995).
- ⁴E.Z. Kurmaev, S. Stadler, D. L. Ederer, Y. Harada, S. Shin, M. M. Grush, T. A. Callcott, R. C. C. Perera, D. A. Zatsarinin, N. Ovednchikina, M. Kasai, Y. Tokura, T. Takahashi, K. Chandrasekaran, R. Vijayaraghavan, and U. V. Varadaraju, *Phys. Rev. B* **57**, 1558 (1998).
- ⁵E. Dagotto, *Rev. Mod. Phys.* **66**, 763 (1994).
- ⁶T. Yokoya, A. Chainani, T. Takahashi, H. Katayama-Yoshida, M. Kasai, and Y. Tokura, *Phys. Rev. Lett.* **76**, 3009 (1996).
- ⁷A.P. Mackenzie, S. R. Julian, A. J. Diver, G. J. McMullan, M. P. Ray, G. G. Lonzarich, Y. Maeno, S. Nishizaki, and T. Fujita, *Phys. Rev. Lett.* **76**, 3786 (1996).
- ⁸T. Yokoya, A. Chainani, T. Takahashi, H. Katayama-Yoshida, M. Kasai, Y. Tokura, N. Shanti, and D. D. Sarma, *Phys. Rev. B* **53**, 8151 (1996).
- ⁹M. Schmidt, T. R. Cummins, M. Brk, D. H. Lu, N. Ncker, S. Schuppler, and F. Lichtenberg, *Phys. Rev. B* **53**, R14 761 (1996).
- ¹⁰I.I. Mazin and David J. Singh, *Phys. Rev. Lett.* **74**, 733 (1997).
- ¹¹Canio Noce and Mario Cuoco, *Phys. Rev. B* **59**, 2659 (1999).
- ¹²A.P. Mackenzie, S. Ikeda, Y. Maeno, T. Fujita, S. R. Julian, and G. G. Lonzarich, *J. Phys. Soc. Jpn.* **67**, 385 (1998).
- ¹³D.F. Agterberg, T.M. Rice, and M. Sigrist, *Phys. Rev. Lett.* **78**, 3374 (1997).
- ¹⁴T. Katsufuji, M. Kasai, and Y. Tokura, *Phys. Rev. Lett.* **76**, 126 (1996).
- ¹⁵For a review, see V.I. Anisimov, F. Aryasetiawan, and A.I. Lichtenstein, *J. Phys.: Condens. Matter* **9**, 1997 (1997).
- ¹⁶J. Costa-Quintana, F. López-Aguilar, and L. Puig-Puig, *Phys. Rev. B* **48**, 1139 (1993); J. Costa-Quintana, M.M. Sánchez-López, and F. López-Aguilar, *ibid.* **54**, 10 265 (1996); J. Costa-Quintana, F. López-Aguilar, A. Pérez-Navarro, and L. Puig-Puig, *Physica B* **230-232**, 929 (1997).
- ¹⁷T. Vogt and D.J. Buttrey, *Phys. Rev. B* **54**, R9843 (1995).
- ¹⁸T. Yokoya, A. Chainani, T. Takahashi, H. Ding, J. C. Campuzano, H. Katayama-Yoshida, M. Kasai, and Y. Tokura, *Phys. Rev. B* **54**, 13 311 (1996).
- ¹⁹G.M. Luke, Y. Fudamoto, K. M. Kojima, M. I. Larkin, J. Merrin, B. Nachumi, Y. J. Uemura, Y. Maeno, Z. Q. Mao, Y. Mori, H. Nakamura, and M. Sigrist, *Nature (London)* **394**, 558 (1998).
- ²⁰Y. Maeno, *Physica C* **282-287**, 206 (1997).
- ²¹V. Zlatic, S.K. Ghatak, and K.H. Benermann, *Phys. Rev. Lett.* **55**, 1263 (1986).
- ²²C.M. Varma, *Phys. Rev. Lett.* **55**, 2723 (1985).
- ²³A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, *Rev. Mod. Phys.* **68**, 13 (1996).
- ²⁴F. López-Aguilar, J. Costa-Quintana, and M.M. Sánchez-López, *Phys. Rev. B* **56**, 1335 (1997).