

PAPER

Theoretical investigation of excitonic magnetism in LaSrCoO_4

To cite this article: J Fernández Afonso *et al* 2018 *J. Phys.: Condens. Matter* **30** 135603

View the [article online](#) for updates and enhancements.



IOP | ebooks™

Bringing together innovative digital publishing with leading authors from the global scientific community.

Start exploring the collection—download the first chapter of every title for free.

Theoretical investigation of excitonic magnetism in LaSrCoO₄

J Fernández Afonso¹, A Sotnikov¹ and J Kuneš^{1,2} 

¹ Institute of Solid State Physics, TU Wien, Wiedner Hauptstr. 8, 1020 Vienna, Austria

² Institute of Physics, Czech Academy of Sciences, Na Slovance 2, 182 21 Praha 8, Czechia

E-mail: kunes@ifp.tuwien.ac.at

Received 24 November 2017, revised 15 January 2018

Accepted for publication 20 February 2018

Published 6 March 2018



CrossMark

Abstract

We use the LDA+U approach to search for possible ordered ground states of LaSrCoO₄. We find a staggered arrangement of magnetic multipoles to be stable over a broad range of Co 3d interaction parameters. This ordered state can be described as a spin-density-wave-type condensate of $d_{xy} \otimes d_{x^2-y^2}$ excitons carrying spin $S = 1$. Further, we construct an effective strong-coupling model, calculate the exciton dispersion and investigate closing of the exciton gap, which marks the exciton condensation instability. Comparing the layered LaSrCoO₄ with its pseudo cubic analog LaCoO₃, we find that for the same interaction parameters the excitonic gap is smaller (possibly vanishing) in the layered cobaltite.

Keywords: excitonic magnetism, Co³⁺, linear spin-wave approximation

(Some figures may appear in colour only in the online journal)

1. Introduction

Perovskite cobaltites from the La_{1-x}Sr_xCoO₃ family have attracted much attention due their peculiar magnetic and transport properties. In particular, the physics of the $x = 0$ member with Co³⁺ formal valence remains a subject of debate. Recently, we have proposed that mobility of intermediate spin (IS) excitations plays an important role in the physics of LaCoO₃ [1] and that the material is close to an excitonic condensation (EC) instability [2–4]. The EC scenario is also being discussed as the origin of the experimentally observed phase transition [5], in the materials from Pr_{0.5}Ca_{0.5}CoO₃ family [6–10].

The layered cobaltites La_{2-x}Sr_xCoO₄ are much less studied although they may exhibit similar physics. The parent compound La₂CoO₄, formal valence Co²⁺, is an antiferromagnetic insulator with a Néel temperature of 275 K [11]. The hole doping suppresses the Néel temperature and an incommensurate magnetic order appears for $x > 1/3$. The doping range $1/3 < x < 1/2$ is particularly interesting due to an hourglass-shaped spectrum of magnetic excitations [12] invoking similarities to curate superconductors. The stoichiometric compound LaSrCoO₄ with Co³⁺ formal valence is relatively less studied due difficulties in sample preparation [13]. The optical [14] and transport measurements [15, 16] for LaSrCoO₄ show a good insulator with a charge gap of about

1 eV. The inverse magnetic susceptibility has a concave shape with pseudo linear T -dependence below 150 K corresponding to $\mu_{\text{eff}} = 2.3\text{--}2.6 \mu_{\text{B}}$ and almost vanishing Weiss temperature ranging from 27 K [16, 17] to 30 K [15]. The authors of [17] reported magnetic anomaly at 7 K, which they interpreted as formation of spin glass. The specific heat data indicate only a modest entropy release of $0.06R$ at the supposed transition.

Theoretical studies of LaSrCoO₄ are even more limited. Wang *et al* [18] applied the unrestricted Hartree–Fock approach to a tight-binding model with Hubbard interaction in order to study various spin-state patterns and identified the high-spin–low-spin (HS–LS) order to be the most likely ground state.

In this work, we use the LDA+U density-functional method to investigate possible ordered ground states of LaSrCoO₄, in particular the excitonic condensate. Furthermore, we employ the strong-coupling expansion to derive a low-energy effective model in the Hilbert space spanned by the LS, IS, and HS states and compare it to an analogous model of LaCoO₃. Similarly to our results for LaCoO₃, we find a stable excitonic ground state for realistic on-site interaction parameters. Comparison to LaCoO₃ shows that for the same interaction parameters we obtain a smaller or vanishing excitonic gap in LaSrCoO₄, which implies that LaSrCoO₄ is closer to the excitonic instability or the excitonic condensate is actually realized.

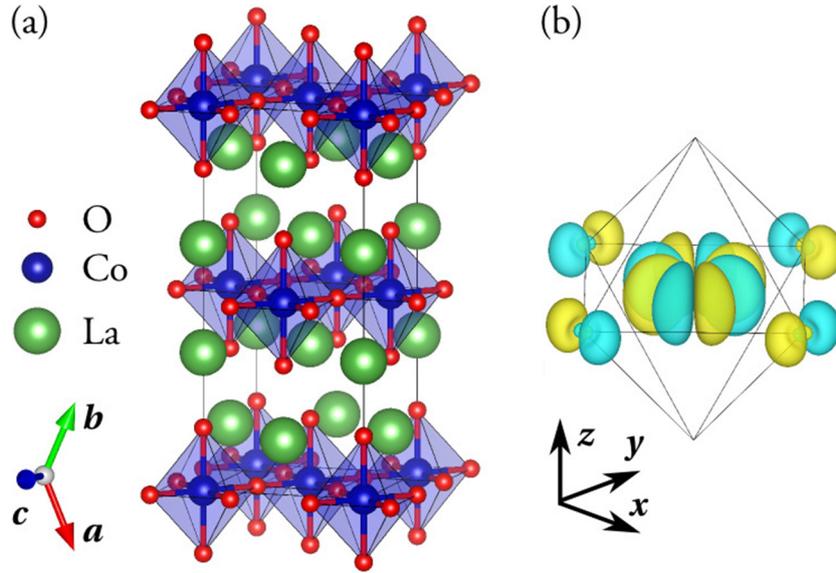


Figure 1. (a) Conventional unit cell of La_2CoO_4 . On the left side the coordinate basis (a, b, c) for the corresponding primitive unit cell with 2 f.u. is shown. (b) Isosurface of the excitonic spin-density distribution obtained around the cobalt atom. Blue and yellow colors represent the positive and negative spin projections, respectively. The spin density around oxygen atoms indicates that the exciton propagates only in the CoO_6 planes.

The paper is organized as follows: in section 2 we introduce the excitonic order parameter and explain the computational methods used in this work. In section 3.1 we present DFT calculations and investigate the stability of the excitonic condensate with and without spin-orbit coupling for different interaction parameters. Section 3.2 is devoted to the strong-coupling analysis of dispersion of bosonic excitations in the normal state, the softening of which marks the onset of condensation.

2. Computational method

2.1. LDA+U

Electronic structure calculations in this paper are performed in the framework of the density functional theory (DFT) by using local density approximation (LDA) [19, 20]. The effect of Coulomb interaction within the $3d$ shell of Co is described by means of LDA+U scheme with the so-called fully localized limit double-counting correction [21]. The spin polarization enters the orbital-dependent potential only while leaving the LDA exchange-correlation potential unpolarized. The calculations are carried out with the WIEN2k [22] package.

The $\text{La}_{2-x}\text{Sr}_x\text{CoO}_4$ structure consists of single layers of CoO_6 corner-sharing octahedra separated by a random distribution of La and Sr ions (see figure 1(a)). To simulate the LaSrCoO_4 compound, we employ the virtual crystal approximation (VCA) [23, 24] that sets the atomic number of La to 56.5. Anticipating the possibility of the staggered order [25], we choose a unit cell allowing for the $\sqrt{2} \times \sqrt{2}$ in-plane order. The unit cell vectors shown in figure 1(a) correspond to the structural parameters $a = 6.8019 \text{ \AA}$, $b = 6.8019 \text{ \AA}$, $c = 5.3796 \text{ \AA}$, $\alpha = \pi/2$, $\beta = \pi/2$, and $\gamma = 2.3284$. The Brillouin zone is sampled with the uniform $4 \times 4 \times 4$ k mesh.

The muffin-tin radii (in bohrs) are the following: 2.50 for La, 1.91 for Co, and 1.65 for O. The plane wave cutoff is set to $R_{\text{mt}}K_{\text{max}} = 6$.

In search for possible ordered solutions we start from several different initial states with broken symmetry: the staggered excitonic and spin-state (LS-HS) orders. The calculations are performed with and without spin-orbit coupling (SOC). The anticipated EC order with the $d_{x^2-y^2} \otimes d_{xy}$ orbital symmetry is described by a three-dimensional complex vector ϕ with components

$$\phi_\beta = \langle \hat{O}'_\beta \rangle + i \langle \hat{O}''_\beta \rangle = \phi'_\beta + i \phi''_\beta, \quad (1)$$

expressed in terms of the hermitian operators

$$\begin{aligned} \hat{O}'_\beta &= \frac{i}{4} \sum_{\sigma, \sigma'=\uparrow, \downarrow} (\tau_\beta)_{\sigma\sigma'} (\hat{c}_{2\sigma}^\dagger \hat{c}_{-2\sigma'}^\dagger - \hat{c}_{-2\sigma}^\dagger \hat{c}_{2\sigma'}^\dagger), \\ \hat{O}''_\beta &= \frac{1}{4} \sum_{\sigma, \sigma'=\uparrow, \downarrow} (\tau_\beta)_{\sigma\sigma'} (\hat{c}_{-2\sigma}^\dagger \hat{c}_{-2\sigma'}^\dagger - \hat{c}_{2\sigma}^\dagger \hat{c}_{2\sigma'}^\dagger), \end{aligned} \quad (2)$$

where $\hat{c}_{m\sigma}^\dagger$ ($\hat{c}_{m\sigma}$) are the creation (annihilation) operators for $3d$ electrons on the same Co atom (the site indices are not shown for the sake of simplicity). The orbital index m refers to the spherical harmonic $Y_{2,m}$ basis with the z quantization axis (see figure 1(b)). The Pauli matrices τ_β ($\beta = x, y, z$) capture the spin-triplet character of the EC order. A nonzero ϕ' gives rise to a spin-density distribution on the Co site with vanishing spin moment per atom that is shown in figure 1(b). For a nonzero ϕ'' , the spin density is identically zero, while the spin-rotational symmetry is broken due to presence of local spin currents.

2.2. Strong coupling expansion

The starting point of our strong-coupling analysis is the Hubbard Hamiltonian for the d -shells of Co

$$\hat{\mathcal{H}} = \sum_i \hat{\mathcal{H}}_{\text{at}}^{(i)} + \sum_{\mathbf{r}} \hat{\mathcal{H}}_{\text{t}}^{(\mathbf{r})}, \quad (3)$$

where

$$\begin{aligned} \hat{\mathcal{H}}_{\text{at}}^{(i)} = & \sum_{\alpha\beta} h_{\alpha\beta}^{ii} \hat{c}_{i\alpha}^\dagger \hat{c}_{i\beta} \\ & + \sum_{\alpha\beta\gamma\delta} U_{\alpha\beta\gamma\delta} \hat{c}_{i\alpha}^\dagger \hat{c}_{i\beta}^\dagger \hat{c}_{i\gamma} \hat{c}_{i\delta}, \end{aligned} \quad (4)$$

$$\hat{\mathcal{H}}_{\text{t}}^{(\mathbf{r})} \equiv \hat{\mathcal{H}}_{\text{t}}^{(ij)} = \sum_{\alpha\beta} h_{\alpha\beta}^{ij} \hat{c}_{i\alpha}^\dagger \hat{c}_{j\beta}, \quad i \neq j, \quad (5)$$

i and j are now the lattice-site indices (below, we account for local, $i = j$, and nearest-neighbor contributions only) that correspond to a particular bond index \mathbf{r} for $i \neq j$, and $\alpha, \beta, \gamma, \delta$ are the internal state (orbital and spin) indices. The local and nearest-neighbor hopping matrices $h_{\alpha\beta}^{ij}$ are provided by projection of the LDA band structure to Wannier orbitals. To this end, we use wien2wannier [26] and wannier90 [27] software. We use the Slater parameterization of the intra-atomic electron-electron interaction $U_{\alpha\beta\gamma\delta}$ of two tuneable parameters, average interaction \bar{U} and Hund's coupling \bar{J} , while fixing the ratio of Slater integrals $F_4/F_2 = 0.625$. While the parameterization of the local Coulomb interaction on Co is the same as the one employed in the LDA+U method, the values of the effective interaction parameters should be different since they refer to a different model.

We diagonalize the local Hamiltonian $\hat{\mathcal{H}}_{\text{at}}^{(i)}$ to obtain atomic eigenenergies $E_\gamma^{(q)}$ and eigenstates $|\Psi_\gamma^{(q)}\rangle$, where q is the number of electrons in the d -shell and γ is the state index. Next, we use the set of the lowest 25 states of the d^6 configuration containing LS, IS, and HS as an active space and treat the non-local terms $\hat{\mathcal{H}}_{\text{t}}^{(\mathbf{r})}$ as a perturbation. Performing the Schrieffer–Wolff transformation [28] to the second order, we arrive at the following bosonic Hamiltonian:

$$\begin{aligned} \hat{\mathcal{H}}_{\text{eff}} = & \sum_{ij, \alpha\beta} (\varepsilon_{1\alpha\beta}^{ij} \hat{d}_{i\alpha}^\dagger \hat{d}_{j\beta} \hat{s}_i \hat{s}_j^\dagger + \varepsilon_{2\alpha\beta}^{ij} \hat{d}_{i\alpha}^\dagger \hat{d}_{j\beta}^\dagger \hat{s}_i \hat{s}_j) \\ & + \text{H.c.} + \hat{\mathcal{H}}_{\text{int}}. \end{aligned} \quad (6)$$

Here, we consider the LS state as the bosonic vacuum, $|\emptyset\rangle_i = \hat{s}_i^\dagger |0\rangle$, and other states from the low-energy manifold of the d^6 configuration as different bosonic flavors α characterized by the corresponding creation (annihilation) operators $\hat{d}_{i\alpha}^\dagger$ ($\hat{d}_{i\alpha}$) on the lattice site i .

In equation (6) we distinguish three types of terms. The first one with the amplitude ε_1 corresponds to the *renormalized* on-site energies of bosons (for $i = j$),

$$\varepsilon_{1\alpha\beta}^{ii} = E_\alpha^{(6)} \delta_{\alpha\beta} + \sum_{\mathbf{r}, \nu, \nu=\pm 1} \mathcal{M}_{\alpha\beta, \nu\nu, \alpha\beta}^{(\mathbf{r})(6+\nu)} / \mathcal{E}_{\alpha\beta, \nu\nu}^{(6+\nu)}, \quad (7)$$

and their hopping amplitudes on the LS background (for $i \neq j$),

$$\varepsilon_{1\alpha\beta}^{ij} = \frac{1}{2} \sum_{\nu, \nu=\pm 1} \left(\frac{1}{\mathcal{E}_{\beta\nu, \nu\nu}^{(6+\nu)}} + \frac{1}{\mathcal{E}_{\alpha\nu, \nu\nu}^{(6+\nu)}} \right) \mathcal{M}_{\beta\nu, \nu\nu, \alpha\theta}^{(ij)(6+\nu)}. \quad (8)$$

The second term with the amplitude ε_2 corresponds to the non-local pair-creation processes,

$$\varepsilon_{2\alpha\beta}^{ij} = \frac{1}{2} \sum_{\nu, \nu=\pm 1} \left(\frac{1}{\mathcal{E}_{\theta\theta, \nu\nu}^{(6+\nu)}} + \frac{1}{\mathcal{E}_{\alpha\beta, \nu\nu}^{(6+\nu)}} \right) \mathcal{M}_{\theta\theta, \nu\nu, \alpha\beta}^{(ij)(6+\nu)}, \quad (9)$$

where

$$\begin{aligned} \mathcal{M}_{\gamma\delta, \nu\nu, \gamma'\delta'}^{(\mathbf{r})(6\pm 1)} = & \langle \Psi_{\gamma'}^{(6)} \Psi_{\delta'}^{(6)} | \hat{\mathcal{H}}_{\text{t}}^{(\mathbf{r})} | \Psi_\nu^{(6\pm 1)} \Psi_\nu^{(6\mp 1)} \rangle \\ & \times \langle \Psi_\nu^{(6\pm 1)} \Psi_\nu^{(6\mp 1)} | \hat{\mathcal{H}}_{\text{t}}^{(\mathbf{r})} | \Psi_\gamma^{(6)} \Psi_\delta^{(6)} \rangle, \end{aligned} \quad (10)$$

and $\mathcal{E}_{\gamma\delta, \nu\nu}^{(6\pm 1)} = E_\gamma^{(6)} + E_\delta^{(6)} - E_\nu^{(6\pm 1)} - E_\nu^{(6\mp 1)}$.

The last term $\hat{\mathcal{H}}_{\text{int}}$ in equation (6) characterizes all other processes with nonzero amplitudes that appear due to Schrieffer–Wolff transformation. Note that the main contributions to this term originate from (density-density and magnetic/orbital exchange) interactions between different bosonic flavors. Including the effect of $\hat{\mathcal{H}}_{\text{int}}$ on the exciton dispersion/spectra is a non-trivial task and goes beyond the scope of the current paper.

Here, we focus on the excitonic instability of the normal ground state that has dominantly the LS character. The small ε_2 allows us to neglect the density of d -bosons in the ground state as well as the in the lowest excited states. The elementary excitations are then approximately described by the bilinear part of the Hamiltonian (6), neglecting higher-order terms (with three and four operators \hat{d}_α) contributing to $\hat{\mathcal{H}}_{\text{int}}$. Using the same argument we drop the hard-core constraint on d bosons and proceed with the linearized spin-wave approach [29], which provides access to momentum dependencies of bosonic (IS and HS) excitations in the lattice. The excitation spectrum obtained in this way corresponds to one-boson excitations of the normal ground state, thus cannot account for effects originating from thermal occupation of IS and HS states.

3. Results

3.1. LDA+U calculations

LDA+U calculations without SOC lead to a non-zero ϕ' with staggered arrangement in the Co-O plane. We have verified that different spatial orientations of ϕ' are numerically equivalent. The ground state can be described as a polar excitonic condensate [30], or, more specifically, as a spin-density wave excitonic condensate in the classification of Halperin and Rice [4]. The corresponding spin-density distribution around Co site, which gives vanishing atomic moment, is shown in figure 1(b).

The stability of the EC long-range order as a function of U and J is summarized in figure 2. The overall shape of the phase diagram resembles the results for cubic LaCoO₃ [25]. We find ordered solutions in both the weak coupling (metallic) and the strong coupling regimes with a wedge of the normal phase at intermediate U and small J . The modification of spectral density due to the excitonic condensation is shown in figure 3.

Introducing SOC breaks the spin-rotational symmetry. The non-zero value of $\langle xy, \sigma | \hat{L}_z \hat{s}_z | x^2 - y^2, \sigma \rangle$ can be viewed as a source field for ϕ_z'' [25]. Upon inclusion of SOC, the EC state splits into two distinct stable solutions

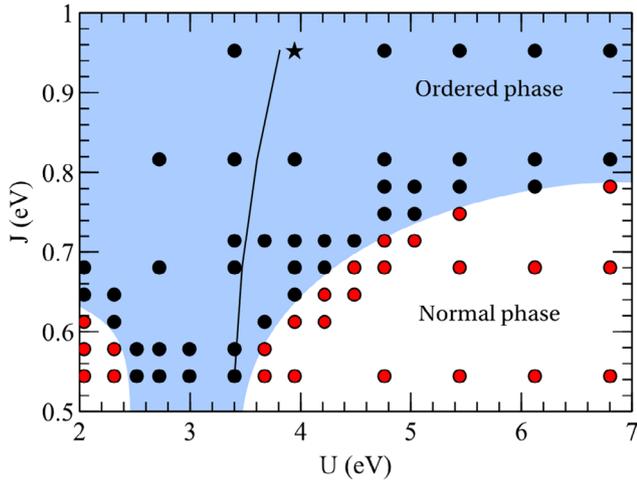


Figure 2. The U - J phase diagram indicating the local stability of the EC state obtained by the LDA+U approach. The symbols correspond to the actual calculations performed. The black line indicates the metal-insulator transition in constrained normal phase.

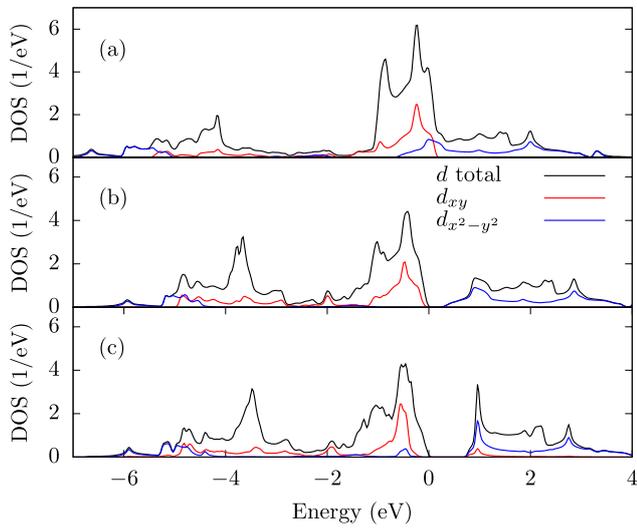


Figure 3. The spectral density of Co d states for the (a) LDA normal (N) solution, (b) LDA+U normal phase and (c) LDA+U EC phase obtained with $U = 3.95$ eV and $J = 0.95$ eV. Notice the hybridization between d_{xy} and $d_{x^2-y^2}$ states at low energies in the EC solution.

$$\begin{aligned}\phi_j^{\parallel} &= (-1)^j (0 \ 0 \ \lambda_{\parallel}^{\prime}) + i(0 \ 0 \ \lambda_{\parallel}^{\prime\prime}), \\ \phi_j^{\perp} &= (-1)^j (\lambda_{\perp}^{\prime} \ \lambda_{\perp}^{\prime\prime} \ 0) + i(0 \ 0 \ \lambda_{\perp}^{\prime\prime}),\end{aligned}\quad (11)$$

where $(-1)^j$ indicates the staggered in-plane arrangement. The ϕ^{\parallel} and ϕ^{\perp} states have different symmetries, reflected, for example, in the appearance of small but finite staggered moment $\mathbf{m}_j \propto \phi_j^{\prime} \times \phi_j^{\prime\prime}$ of $0.05 \mu_B$ for $\phi^{\perp} \neq 0$. No ordered moment arises in the ϕ^{\parallel} solutions. The total energies shown in table 1 for $U = 3.95$ eV and $J = 0.95$ eV (the star-shaped point in figure 2) favor the ϕ^{\parallel} order. In this parameter regime we have also searched for other possible two sublattices solutions such as antiferromagnetic or spin-state order (LS-HS) [18]. All our calculations converged to the ϕ^{\parallel} solutions. The calculations were performed without lattice relaxation.

Table 1. Comparison of the order parameters $|\phi^{\prime}|$ and $|\phi^{\prime\prime}|$ and the energy difference ΔE (compared to the normal state) of the ordered solutions at $U = 3.95$ eV and $J = 0.95$ eV.

Solution	Approach	$ \phi^{\prime} $	$ \phi^{\prime\prime} $	ΔE (meV f.u. $^{-1}$)
EC	LDA+U	0.289	0.000	-14.25
EC $^{\perp}$	LDA+U+SOC	0.274	0.098	-14.83
EC $^{\parallel}$	LDA+U+SOC	0.299	0.096	-17.58

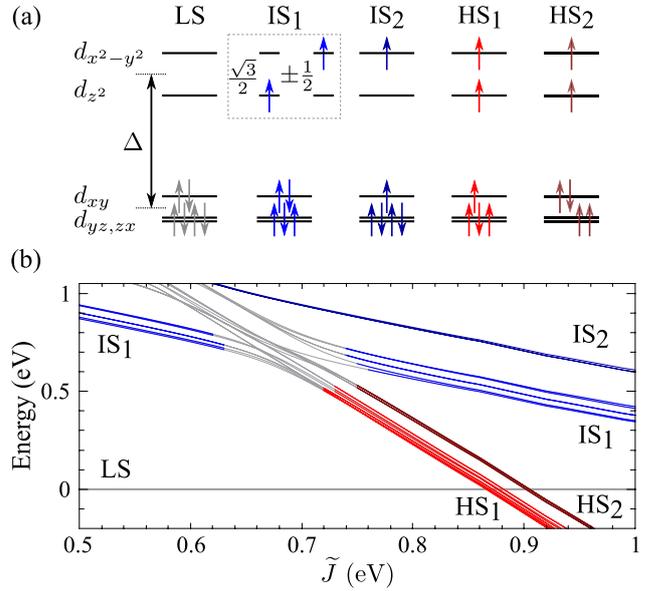


Figure 4. Spin configurations of the lowest atomic multiplets of La_2CoO_4 (a) and the corresponding atomic multiplet energies for SOC $\zeta = 56$ meV (b).

3.2. Strong coupling analysis

In figure 4 we show the set of eigenenergies $E_{\gamma}^{(6)}$ of the Hamiltonian (4) corresponding to the lowest atomic multiplets of LaSrCoO_4 as a function of the Hund's coupling \tilde{J} . The three intermediate-spin $d_{xy} \otimes d_{x^2-y^2}$ states (IS $_2$) have the highest atomic energies among 25 lowest states in the region of realistic values of Hund's coupling \tilde{J} due to cubic crystal field ($\Delta = 1.594$ eV) and additional tetragonal splittings of the t_{2g} and e_g states (0.103 eV and 0.384 eV, respectively). This seems to prevent the IS $_2$ states from condensation. However, as we show below, the renormalization of on-site energies by virtual hopping processes reverses the order of atomic multiplets, i.e. when placed on the lattice the IS $_2$ excitations have lower energies than the IS $_1$ ones.

Next, we compute the amplitudes (7)–(9) and diagonalize the bilinear part of the effective Hamiltonian (6). In figures 5(a)–(e) we show the dispersions of elementary (IS- and HS-like) excitations of (6) obtained for different values of \tilde{U} and \tilde{J} . While the IS excitations can move on the LS background, the HS excitations cannot, and the deviation from completely flat HS bands is due to HS-IS mixing through spin-orbit coupling. Note that the spin-orbital character of the excitations remains approximately fixed along the individual branches due to conservation of orbital and spin flavors in the nearest-neighbor kinetic exchange processes. Small mixing

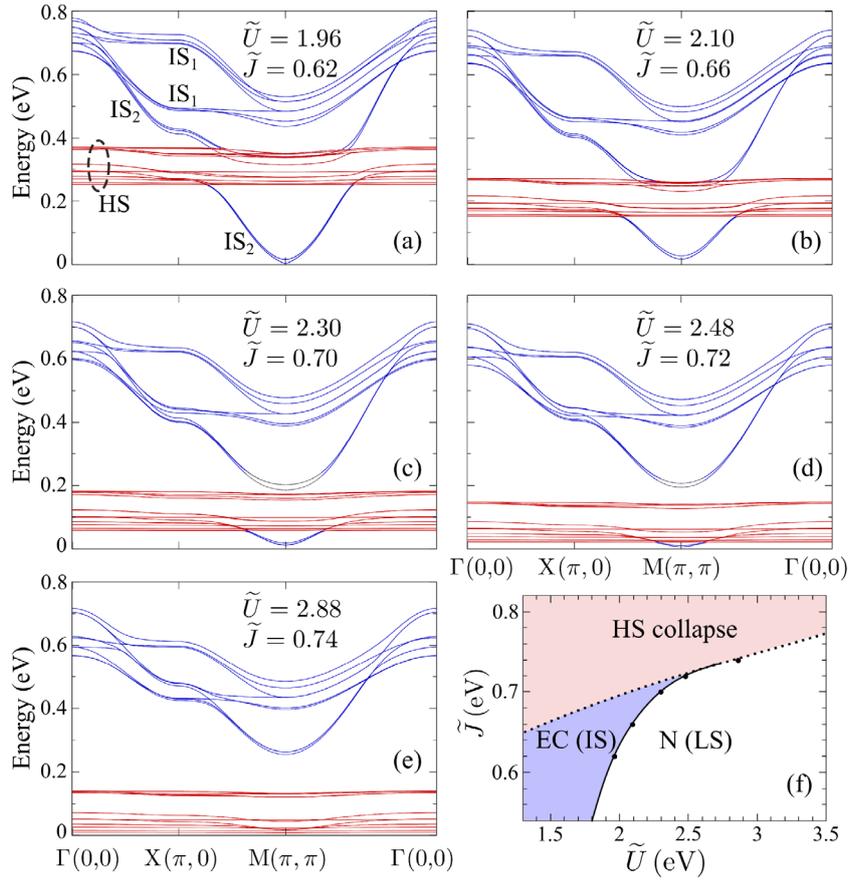


Figure 5. (a)–(e) Dispersions of the IS and HS excitations at different \tilde{U} and \tilde{J} (given in eV) and $\zeta = 56$ meV. (f) The corresponding phase diagram obtained following the disappearance of the exciton gap.

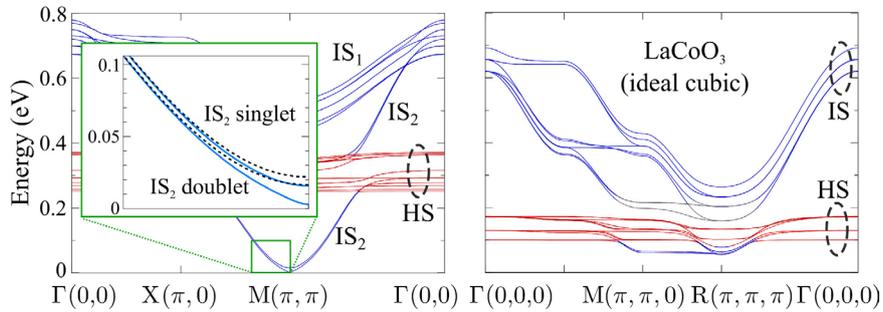


Figure 6. Dispersions of the IS and HS excitations for LaSrCoO₄ (left) and cubic LaCoO₃ (right) at $\tilde{U} = 1.96$ eV, $\tilde{J} = 0.62$ eV, and $\zeta = 56$ meV. Inset: IS₂ dispersion with (solid lines) and without (dashed lines) pair-creation contributions.

and thus momentum dependence arises due to spin-orbit coupling.

The interaction parameters were chosen so that the solution is at the verge of excitonic instability, i.e. normal solutions with a tiny gap, vanishing of which determines the phase boundary in figure 5(f). Variation of \tilde{J} enters predominantly through the changes of atomic multiplet energies in figure 4. Variation of \tilde{U} affects both the hopping amplitudes (bandwidths) and the renormalization of site energies (band shifts). The renormalizations of different

states scale differently with \tilde{U} depending of the number and amplitudes of the virtual hopping process for a given state on a LS background.

The influence of the pair-creation term is shown in the inset of figure 6. Due to bosonic nature of excitations, this term effectively acts as an ‘attraction’ between bands $\epsilon_\gamma(\mathbf{k})$ and their mirror images $-\epsilon_\gamma(\mathbf{k})$. Only when their separation is comparable or smaller than characteristic amplitudes determined by equation (9), the pair creation/annihilation processes become important. These amplitudes for the cases

under study are typically of the order of few meV, thus the effect becomes noticeable close to the minimum of the IS_2 energy dispersion.

To compare with $LaCoO_3$, we repeat the analysis for the hypothetical cubic structure of [25] and the same \tilde{U} and \tilde{J} values, see figure 6. We find the excitation gap is larger in the cubic structure due to narrower IS band. Moreover, the HS excitations in the layered structure are located at substantially higher energy than in its cubic counterpart. This is because the out-of-plane nearest-neighbor processes, which contribute to renormalization of the HS local energies in the cubic structure, are missing in the layered system. Similar processes play a minor role for the IS_2 excitations and thus the renormalizations in the cubic and layered structures are comparable.

More extensive numerical analysis confirms the general tendency that with a continuous decrease of \tilde{U} and \tilde{J} parameters, the EC instability first appears in the layered compound and only then in $LaCoO_3$. This agrees well with comparison of phase boundaries for critical U and J values in figure 2 with the values for $LaCoO_3$ published in [25] that are both obtained within the LDA+U approach.

4. Conclusions

We have performed LDA+U calculations for $LaSrCoO_4$ that amount to a static mean-field treatment of the excitonic order in this compound. We find excitonic condensate to be a stable solution with a total energy lower than the one of normal state over a large part of the studied U - J phase diagram. The generally used U and J values fall close to the boundary of the region of EC stability. The stable EC solutions are of the spin-density-wave type with a $d_{xy} \otimes d_{x^2-y^2}$ orbital symmetry and out-of-plane spin polarization. Comparison to $LaCoO_3$ by means of the linear spin-wave treatment of the effective strong-coupling model suggest that the layered cobaltite is closer to the excitonic instability or that the EC order is possibly realized. Investigations of behaviour under pressure and in high magnetic fields [31] are highly desirable.

Acknowledgments

The authors thank A Hariki, O Kaman and L H Tjeng for fruitful discussions. This work has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant agreement No. 646807-EXMAG). Access to computing and storage facilities provided by the Vienna Scientific Cluster (VSC) is greatly appreciated.

ORCID iDs

J Kuneš  <https://orcid.org/0000-0001-9682-7640>

References

- [1] Sotnikov A and Kuneš J 2016 *Sci. Rep.* **6** 30510
- [2] Mott N F 1961 *Phil. Mag.* **6** 287
- [3] Keldysh L V and Kopaev Y V 1965 *Sov. Phys. Solid State* **6** 2219
- [4] Halperin B and Rice T 1968 *Solid State Physics* vol 21, ed D T Frederick Seitz and H Ehrenreich (New York: Academic) pp 115–92
- [5] Tsubouchi S, Kyômen T, Itoh M and Oguni M 2004 *Phys. Rev. B* **69** 144406
- [6] Kuneš J and Augustinský P 2014 *Phys. Rev. B* **90** 235112
- [7] Nasu J, Watanabe T, Naka M and Ishihara S 2016 *Phys. Rev. B* **93** 205136
- [8] Tatsuno T, Mizoguchi E, Nasu J, Naka M and Ishihara S 2016 *J. Phys. Soc. Japan* **85** 083706
- [9] Yamaguchi T, Sugimoto K and Ohta Y 2017 *J. Phys. Soc. Japan* **86** 043701
- [10] Sotnikov A and Kuneš J 2017 *Phys. Rev. B* **96** 245102
- [11] Babkevich P, Prabhakaran D, Frost C D and Boothroyd A T 2010 *Phys. Rev. B* **82** 184425
- [12] Drees Y *et al* 2014 *Nat. Commun.* **5** 5731
- [13] Kaman O 2017 private communication
- [14] Moritomo Y, Arima T and Tokura Y 1995 *J. Phys. Soc. Japan* **64** 4117
- [15] Moritomo Y, Higashi K, Matsuda K and Nakamura A 1997 *Phys. Rev. B* **55** R14725
- [16] Chichev A V *et al* 2006 *Phys. Rev. B* **74** 134414
- [17] Guo H, Hu Z, Pi T-W, Tjeng L H and Komarek A C 2016 *Crystals* **6** 98
- [18] Wang J, Zhang W and Xing D Y 2000 *Phys. Rev. B* **62** 14140
- [19] Kohn W and Sham L J 1965 *Phys. Rev.* **140** A1133
- [20] Hohenberg P and Kohn W 1964 *Phys. Rev.* **136** B864
- [21] Liechtenstein A I, Anisimov V I and Zaanen J 1995 *Phys. Rev. B* **52** R5467
- [22] Blaha P, Schwarz K, Madsen G K H, Kvasnicka D and Luitz J 2001 *WIEN2K, An Augmented Plane Wave + Local Orbitals Program for Calculating Crystal Properties* (Austria: Karlheinz Schwarz, Techn. Universität Wien)
- [23] Nordheim L 1931 *Ann. Phys.* **401** 607
- [24] Muto T 1938 *Sci. Pap. Inst. Phys. Chem. Res.* **34** 377
- [25] Afonso J F and Kuneš J 2017 *Phys. Rev. B* **95** 115131
- [26] Kuneš J, Arita R, Wissgott P, Toschi A, Ikeda H and Held K 2010 *Comput. Phys. Commun.* **181** 1888
- [27] Mostofi A A, Yates J R, Pizzi G, Lee Y-S, Souza I, Vanderbilt D and Marzari N 2014 *Comput. Phys. Commun.* **185** 2309
- [28] Schrieffer J R and Wolff P A 1966 *Phys. Rev.* **149** 491
- [29] Sommer T, Vojta M and Becker K 2001 *Eur. Phys. J. B* **23** 329
- [30] Ho T-L 1998 *Phys. Rev. Lett.* **81** 742
- [31] Ikeda A, Nomura T, Matsuda Y H, Matsuo A, Kindo K and Sato K 2016 *Phys. Rev. B* **93** 220401