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Local dynamic spin-fluctuations in YCo_2 compound

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Density-functional studies of the electronic structure, of YCo_2 compound are reported. The band structure near the Fermi energy shows rather heavy Co-d-derived bands. The resulting density of states is high, yielding a weak ferromagnetic instability via the Stoner mechanism. Many-body Hubbard-like correlation effects take place in the Co-d bands and they can be treated in the framework of Density Functional Theory (DFT) in combination with Dynamical Mean Filed Theory (DMFT) approximation. An interesting possibility is that the renormalized paramagnetic ground state may emerge, due to the combination of the weak competing ground states and quantum fluctuations.

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1. Introduction

In the normal metallic state the electrons form a Fermi liquid, a concept introduced by Landau [1]. In this state the excitation spectrum is very similar to that of a non-interacting Fermi gas. The basic excitations are weakly interacting fermionic quasiparticles which behave like normal electrons but have renormalized parameters like an effective mass. However, at low temperatures the Fermi liquid is potentially unstable against sufficiently strong interactions, and some type of brocken-state may form. This low temperature phase depends on the microscopic parameters of the material under consideration. Upon changing these microscopic parameters at zero temperature, e.g. by applying pressure and external field or by changing the chemical composition, the nature of the ground state may change and the system may undergo a quantum phase transition.

One of the most extensively studied materials exhibiting pressure tuned ferromagnetic quantum phase transition of itinerant electrons is probably the MnSi compound [2]. It belongs to the class of so called nearly or weakly ferroamgnetic materials. Common features of these metallic ferromagnets are: a low saturation moment at 0 K, low magnetic ordering temperature, high magnetic susceptibility at 0 K and negative magnetoresistance. The essential feature is that the physics is dominated down to quite low temperature (at the critical point down to 0 K) by quantum fluctuations at the critical point and its consequences are manifested in the magnetic, transport and thermodynamic properties. Actually the doped $Y(Co_{1-x}Al_x)_2$ is qualified in the category of phenomenon called the itinerant electron metamagnetism (IEM) which shows magnetic quantum phase transition. The IEM is attributed to sharp structures in the electronic density of states near Fermi energy. Being close to a ferromagnetic instability makes YCo_2 a good candidate for actually reaching the ferromagnetic quantum phase transition.

It is known that DFT-LSDA total energy calculations for the YCo_2 compound give a magnetic ground state for the experimental lattice parameter. On the other hand, for the equilibrium lattice parameter a non-magnetic ground state is obtained. In this paper we show that local quantum fluctuations described by DMFT stabilizes a renormalized paramagnetic ground state for the experimentally observed lattice parameter.

2. Theoretical model. Density functional, total energy calculations

Density functional theory [3] is in principle an exact ground state theory. It should, therefore, correctly describe the spin density of magnetic systems. However, common approximations to the exact density functional theory, such as the local spin density approximation (LSDA) and generalized gradient approximations (GGA) [4], neglect Hubbard correlations beyond the mean field level, with the well known result that the magnetic tendency of strongly, Hubbard, correlated systems is often underestimated. Narrow bands d-electron systems such as transition metals are their alloys are sometimes not accurately described by the LSDA approach.

One of the extensions of LSDA to include mean field Hubabrd type corrections is the LSDA+U method [5]. This method describes successfully many interesting effects, such as orbital and charge ordering in transition metal oxides [5, 6]. In a recent paper [7] we showed that the LSDA+U enhance the Stoner factor while reducing the density of states. It is arguable that the most important correlation effects in metals, fluctuation induced mass-renormalization and suppression of the Stoner factor are missing in the LSDA+U. For these materials the static mean-field type approximation is too crude and a more sophisticated approach is needed.

It is well known that that the most interesting correlation effects in quasiparticle spectra such as the mass enhancement, damping and life times, are connected with

the energy dependence of the self-energy $\Sigma(E)$, so one need to generalize the LSDA+U approach to include dynamical effects. We proposed recently a fully self-consistent LSDA+DMFT [8] approach which combines material dependent information through the electronic structure with the many-body DMFT solution of the Hubbard model.

The implementation of the LSDA+DMFT approach is based on an energy dependent wave function approach, the so-called Exact Muffin-tin orbitals (EMTO) method [9,10], a screened KKR-type of method within the LDA. The correlation effects are treated in the framework of Dynamical Mean Field Theory (DMFT) [11,12], with a spin-polarized T-matrix Fluctuation Exchange (T-FLEX) type of DMFT solver [13,14]. The SPT-FLEX approximation is a multiband spin-polarized generalization of the fluctuation exchange approximation (FLEX) of Bickers and Scalapino [15], but with a different treatment of particle-hole (PH) and particle-particle (PP) channels. Particle-particle (PP) channel is described by a T-matrix approach [16] giving a renormalization of the effective interaction. This effective interaction is used explicitly in the particle-hole channel. Justifications, further developments and details of this scheme can be found in Ref. [13,14].

In general, if the one particle Green's function G(k, z) can be found, the energy for a system of interacting electrons can be calculated using an expression derived by Galitskii-Migdal [17]:

$$E_{MB} = \frac{1}{2\pi i}\sum_{k,\sigma}\int[\varepsilon_k + \frac{1}{2}\Sigma(k,z)G(k,z)]dz \quad (1)$$

where $\Sigma(E)$ is the DMFT self-energy. The first term is the renormalized kinetic energy, and this contribution is included already in the LSDA part, meanwhile the second term is the extra contribution to be included. Thus, the second term of the above equation is calculated on the Matsubara axis, inside the many body solvers:

$$E_{DMFT} = \frac{T}{2}\sum_{nk,\sigma} Tr\Sigma^{\sigma}(i\omega_n)G^{\sigma}(k,i\omega_n) \quad (2)$$

3. Results

YCo_2 belongs to the $MgCu_2$ type structure. The corresponding Pearson symbol is cF24 and the space group is known as Fd-3m No. 227. This structure corresponds to the tetrahedral closed packed structures known as Laves Phase. The smaller Co atom forms a network of face- and corner sharing tetrahedra, which corresponds to the 16(d) positions (5/8, 5/8, 5/8). The larger Y atoms are accommodated by the large holes in this network, occupying the 8(a) positions (0, 0, 0). The sub lattice of the Y atoms has a diamond structure.

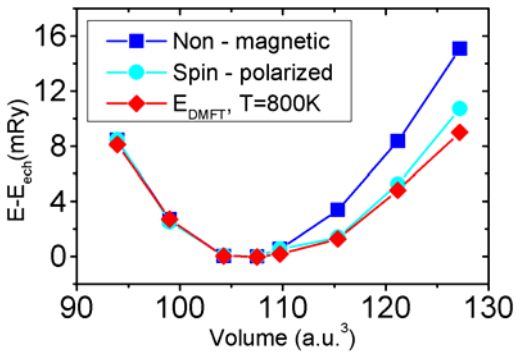

Fig. 1. Total Energy calculations in the LSDA – spin - polarized approach, the LDA – non - magnetic and LSDA+DMFT.

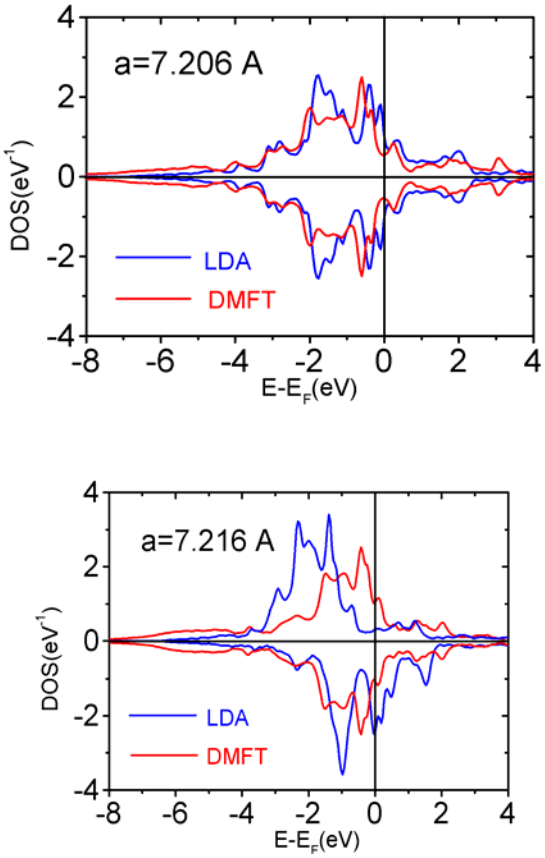

Fig. 2. Densities of states of YCo_2 for two lattice parameters. L(S)DA results–blue and LSDA+DMFT–red line.

To calculate the charge density we integrate along a contour on the complex energy plane which extends from the bottom of the band up to the Fermi level [10], using 30 energy points. For the Brillouin zone integration we sum up a k-space grid of 89 points in the irreducible part of the Brillouin zone. A cutoff of l_{max}=8 for the multi-pole expansion of the charge density and a cutoff of l_{max}=3 for the wave functions was used. The Perdew-Wang parameterization of the LDA to the exchange correlation potential was used. The LSDA+DMFT calcualtions were

performed for a T = 800 K and for the value of U=2eV for the average Coulomb interaction on the Co atoms and the corresponding exchange interactions J=0.9 eV. Unfortunately, there are no reliable schemes to calculate U in metals, and such intermediate values are accepted in the literature [13,14]. Fig. 1 showes the results for the total energy, for a large range of lattice parameters or atomic volumes. In the LSDA calculaltions (light blue) the kink around the value of 110 au^3 of Co atomic volume the magneitc state becomes stable, which contradicts the experimental results. On contrary the LSDA+DMFT calculations, produces a non-magnetic ground states, even for values of lattice parameters where the LSDA calcualtions fail to predict the proper ground state.

Density of states (DOS) are presented in Fig. 2. We choose to show the results for two different lattice parameters, namely a = 7.206 A and a = 7.216 A. In the framework of L(S)DA approach for the former lattice parameter a stable non-magnetic ground state is evidenced, meanwhile in the later case a magnetic ground states is more favourable energetically. In contrast in the LSDA+DMFT calculations at T=800 K a non-magnetic ground state is found for the entire range of studier lattice parameters. The LDA+DMFT total density of states, at the Fermi level, has a smaller value in comparison with the similar values found in L(S)DA calculations. For the larger lattice parameter a=7.216 Å the large value of DOS at the Fermi level is a signature of the magnetic instability. Many-body correlations, local but temporal fluctuations described by DMFT lead to the non-magnetic ground state.

In Fig. 3 we present the SPT-FLEX self-energy in the framework of LSDA+DMFT approach. The self-energy corresponds to the calculation in which the a = 7.206 A lattice paramater was used. The self-energy around the Fermi level shows a Fermi liquid behaviour.

$$\Im m\Sigma(E) \propto \left(E - E_F\right)^2$$
$$\frac{\partial \Re e\Sigma(E)}{\partial E} < 0 \qquad (3)$$

The energy dependence of self-energy for the t_{2g} and e_g orbitals is very similar, and practically in paramagnetic states the contributions in both spin channels are identical.

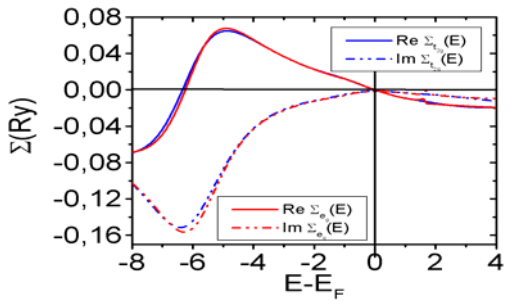

Fig. 3. Orbital resolved self-energy (the real part - continuous line and the imaginary part – dashed line).

4. Discussion

The temperature dependence of the macroscopic properties of the paramagnetic transition metals and their alloys presents a long-standing problem. The first approach was developed by Stoner [18], on the basis of Stoner theory who attributed the varying behaviour of magnetic susceptibility as function of temperature to the different structure of the density of state near the Fermi level. Detailed evaluation of this theory using accurately calculated densities of states for various metals are well known. A more evoluated version of the theory using a density functional theory at finite temperatures has been applied by Liu to Pd and Pt [19]. In general these calculations produce the correct trend but a detailed temperature dependence of susceptibility is not reproduced very well. It was shown by Beal-Monod [20] that in paramagnetic metals with strong exchange enhancement the low temperature T^2 term in χ is enhanced over its Stoner value due to the effect of long-wavelength spin fluctuations (paramagnons). This work constitutes the starting point for the development of the renormalized spin-fluctuation theory of Moriya and Kawabata [21]. Misawa [22] pointed out that there is a T^2 ln T term in the susceptibility of a Fermi liquid which leads to a maxima in the susceptibility and fits rather well the experiments on Pd and Rh. Later on, it was shown that the T^2 ln T term which might have been expected to be the dominant one cancels out due to the paramagnons. Anyway, is rather impossible task to calculate such a contribution from a first principle theory. Existing spin-fluctuation theories making use of realistic density of state curves were developed by Hubbard [23] and Hasegawa [24]. Recently, Burzo [25] and his co-workers applied successfully such an approach in the evaluation of the low temperature paramagnetic susceptibility to a large number of transition metal alloys. Shimizu [26] made calculations of susceptibilities based on the spin fluctuation theory in which the temperature dependence has two parts: a non-interacting electron system, Stoner type contribution and a part coming from the effect of the spin fluctuations. The spin fluctuation contribution can be resolved exactly at high temperatures and in such a way the square of the amplitude of local spin fluctuation can be determined. Burzo et. al. [27] analyzed several mechanism such as quenching, saturation or non-saturation of spin fluctuations.

5. Conclusions

According to our knowledge the present work is the first to try to combine the realistic electronic structure calculations with a fully self consistent procedure to solve the corresponding multiorbital Hubbard Hamiltonian. The many-body solver used in the preset calculations is a modified version of the Fluctuating exchange approximation (FLEX) very briefly presented above, which takes into account fluctuations in the particle-particle and particle-hole channel for the multiorbital case. Physically, this approximation describes dynamical spin

fluctuations, by calculating explicitly the longitudinal and transversal spin susceptibilities. These susceptibilities are used to calculate the fluctuating exchange potentials which are complex and energy dependent quantities leading to the self-energy $\Sigma(E)$. We showed that from a physical pint of view the existence of such fluctuating exchange potentials in YCo_2 determines a non-magnetic ground state for the complete lattice parameter range. Further theoretical calculations will be necessary to investigate, from such a microscopic theory, the temperature induced local moments in the paramagnetic state. Clearly more work on the susceptibility of paramagnetic metals and their alloys is required and further calcualtions along the presented lines will be of interest.

References

[1] A. L. Fetter, J. Walecka, Quantum Theory of Many-Particle Systems Mcgraw-Hill College 1971).

[2] C. Pfeiderer, G. J. MacMullan, S. R. Julian, G. G. Lonzarich Phys. rev. B **55**, 8830 (1997).

[3] P. Hohenberg, W. Kohn, Phys. Rev. **136**, B864 (1964); W. Kohn, L. J. Sham, Phys. Rev. **140**, A1133 (1965).

[4] R. O. Jones, O. Gunnarsson, Rev. Mod. Phys. **61**, 689 (1989).

[5] V. I. Anisimov, F. Aryasetiawan, A. I. Lichtenstein, J. Phys.: Condens. Mat. **9**, 767 (1997).

[6] M. Imada, A. Fujimori, Y. Tokura. Rev Mod Phys, **70**, 1039 (1998).

[7] A. G. Petukhov, I. I. Mazin, L. Chioncel, A. I. Lichtenstein, Phys. Rev. B **67**, 153106 (2003).

[8] L. Chioncel, L. Vitos, I. A. Abrikosov, J. Kollar, M. I. Katsnelson, A. I. Lichtenstein, Phys. Rev. B. 67 (2003) 235106; L. Chioncel, M. I. Katsnelson, R. A. de Groot and A. I. Lichtenstein, Phys. Rev. B 68, (2003).

[9] O. K. Andersen, O. Jepsen, G. Krier, in: V. Kumar, O. K. Andersen, A. Mookerjee (Eds.), Lectures on Methods of Electronic Strcuture Calcualtions, World Scientific, Singapore, pp. 63-124 (1994); O. K. Andersen, T. Saha-Dasgupta and S. Ezhov, {\it cond-mat/0203083} (2002).

[10] L. Vitos, H. L. Skriver, B. Johansson, J. Kollar, Computational Materials Science **18**, 24 (2000).

[11] W. Metzner, D. Vollhardt, Phys. Rev. Lett. **62**, 324 (1989).

[12] A. Georges, G. Kotliar, W. Krauth, M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996).

[13] M. I. Katsnelson, A. I. Lichtenstein, J. Phys. Condens. Matter **11**, 1037 (1999).

[14] A. I. Lichtenstein, M. I. Katsnelson, Phys. Rev. B **57**, 6884 (1998).

[15] N. E. Bickers, D. J. Scalapino, Ann. Phys. (NY) **193**, 206 (1989).

[16] M. I. Katsnelson, A. I. Lichtenstein, Eur. Phys. J. Phys. B. **30**, 9 (2002).

[17] V. M. Gaitskii, A. B. Migdal, Zh. Eksp. Teor. Fiz. **34**, 139 (1958) [Sov. Phys. JETP 7, (1958), 96].

[18] E. C. Stoner Proc. R. Soc. A **154**, 656 (1936).

[19] K. L. Liu, A. H. Mac Donalds, J. M. Daams, S. H. Vosko, D. D. Koelling, J. Magn. Magn. Matter. **12**, 43 (1979).

[20] M. T. Beal Monod, S. K. Ma, D. R. Fredkin, Phys Rev. Lett. **20**, 929 (1968).

[21] T. Moriya, A. Kawabata J. Phys. Soc. Japan **34**, 639 (1973); J. Phys. Soc. Japan **35**, 669 (1973).

[22] S. Misawa, K. Kanematsu, J. Phys F: Met. Phys. **6**, 2119 (1976)

[23] J. Hubbard, Phys Rev B. **19**, 2626 (1979); Phys Rev B. **20**, 4584 (1979); Phys Rev B. **23**, 5974 (1981).

[24] H. Hasegawa J. Phys. Soc. Japan **46**, 1504 (1979); J. Phys. Soc. Japan 49 (1980) 178; J. Phys. Soc. Japan **49**, 963 (1980).

[25] E. Burzo E. L. Chioncel, J. Optoelectron. Adv. Mater. **6**, 917 (2004); R. Tetean, R. E. Burzo, L. Chioncel, L et.al. Phys. Stat. Solidi A – Appl. Research **196**, 301 (2003); E. Burzo, S.G. Chiuzbaian, M. Neumann, et al. J. Appl. Phys. 92 (2002) 7362; R. Tetean, E. Burzo, L. Chioncel L, et al. J. Magn. Magn. Matter. 242 (2002) 836; R. Tetean, E. Burzo, Z. Sarkozi, et al. Materials Science Forum **373**, 661 (2001).

[26] M. Shmizu Rep. Prog. Phys. **44**, 329 (1981).

[27] E. Burzo, R. Tetean, Z. Sarkozi, L. Chioncel, M. Neumann M, J. Alloys and Compound. **323**, 490 (2001); J. Magn. Magn. Matter. **197**, 901 (1999); I. Lupsa, P. Lucaci, E. Burzo, J. Alloys and Compound **299**, 17 (2000).

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