Impurity effect on the metal-insulator transition in Kondo insulators

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(Received 22 July 1999; revised manuscript received 24 February 2000)

In this paper we study the effect of transition impurities on the metal-insulator transition in Kondo insulators. We use a two-subband model Hamiltonian in which a strongly correlated electron narrow subband is coupled to a wide conduction-electron subband. In the model, the disorder is treated by using the off-diagonal coherent potential approximation with a simple parametrization of the electron energy hopping. We obtain a criterion for the opening of the hybridization energy gap as a function of impurity concentration. In the particular case of $CeRh_{1-x}Pd_xSb$, the insulator-metal transition occurs at critical Pd concentration of 10%, in agreement with experimental data.

I. INTRODUCTION

Kondo insulators, extensively studied in the literature, present a wide range of interesting physical properties.[1–23] In these materials, the variation of external pressure, external magnetic field, temperature, and doping can destroy the energy gap and produce an insulator-metal transition.[15–21] The origin of the energy gap, which is usually associated with the mixing of conduction electrons with local spins, and many aspects of the metal-insulator transition in Kondo insulators were covered in several theoretical works based on the Anderson lattice Hamiltonian, the Kondo lattice Hamiltonian, and the picture of the two-subband model.[7,14,22] Despite the great achievements of those models, many other interesting characteristics of Kondo insulators still remain open for discussion. For instance, the existence of a gapless phase in anisotropic Kondo insulators and the effects of impurities on its physical properties are not yet fully understood. Recent theoretical works[22,23] based on the periodic Anderson model discussed the effects of impurities such as La, entering substitutionally in the Ce site of Ce-based Kondo insulators. In those works it was argued that there is a formation of an impurity band in the hybridization gap when the metal-insulator transition develops. In the case of Kondo insulators doped with a transition element, such as $CeRh_{1-x}Pd_xSb$, $CeNi_{1-x}Pd_xSn$, and $CeNi_{1-x}Cu_xSn$, the complexity of the electronic structure makes a theoretical description of the problem a difficult task, and sometimes it is necessary to develop alternative models to gain some insight into the physical process involved.

Motivated by these discussions, in this paper we examine the effect of transition impurities on the formation of the energy gap, and its influence on the metal-insulator phase transition in Kondo insulators. To this end we use a two-subband model, in which a strongly correlated electrons narrow subband is coupled to a wide conduction-electron subband via a constant hybridization term. In our model, impurities entering into the system directly affect the wide subband and indirectly affect the narrow one through the hybridization between them. The impurities introduce both diagonal and off-diagonal disorder in the Hamiltonian. The diagonal disorder modifies the local energy of the conduction electrons, which is reflected in the position of the center of the wide subband. The off-diagonal contribution modifies the energy hopping and plays a key role in the metal-insulator phase transition described by our model. Through a simple parametrization of the electron energy hopping we obtain a criterion for the opening of the energy gap as a function of the impurity concentration. The numerical results obtained for the particular case of $CeRh_{1-x}Pd_xSb$ show that a metal-insulator transition occurs at a critical Pd concentration of 10%, in good agreement with experimental data.[20] The critical exponent of this disorder-driven metal-insulator phase transition indicates that it belongs to the same class of universality as the density-driven transition predicted by scaling theory.[24,25]

II. FORMULATION

We begin with the following two-subband model Hamiltonian, in which a wide conduction-electron subband containing disorder is coupled to a strongly correlated narrow subband via an isotropic hybridization term. In the slave-boson mean-field approach[26,27] for strong Coulomb correlation, our model Hamiltonian reads

$$H=\sum_i \varepsilon_i^{c\gamma}|i^c\rangle\langle i^c|+\sum_{ij} T_{ij}^{cc}|i^c\rangle\langle j^c|+\sum_i \varepsilon_0^f|i^f\rangle\langle i^f| + \sum_{ij} T_{ij}^{ff}|i^f\rangle\langle j^f|+V^{cf}\sum_{ij}(|i^c\rangle\langle j^f|+(|i^f\rangle\langle j^c|). \quad (1)$$

A similar model Hamiltonian was already used in the literature to study the clean limit of Kondo insulators.[7,8] Here ε_0^f is the renormalized energy of the narrow f subband, which should be self-consistently determined to assure the constraint $n_f=1$. $\varepsilon_i^{c\gamma}$ is the energy of the wide conduction-electron subband, where $\gamma=H$ or I depending on the occupancy of the site by a host (H) atom or an impurity (I) atom. The terms T_{ij}^{cc} and T_{ij}^{ff} represent the energy hopping between sites in the same subband, and V^{cf} is the renormalized hybridization between subbands.

The disorder in this Hamiltonian affects the energy $\varepsilon_i^{c\gamma}$, the hopping term T_{ij}^{cc}, and the hybridization between subbands. In order to treat this disorder, we extend the off-diagonal coherent potential approximation[28] for this two-

subband model. In this picture, it is considered that the impurities renormalize the electron energy hopping in the wide subband and the hybridization term as[28]

$$\tilde{V}^{cf}=\xi_i^c V^{cf}, \quad T_{ij}^{cc}=\xi_i^c T_0 \xi_j^c, \tag{2}$$

where T_0 is the reference energy hopping and the factor ξ_i^c is a parameter of the model that should be chosen consistently with the extent of the conduction-electron wave functions of impurities and host atoms. The renormalized hopping T^{cc} takes into account the difference in the energy hopping when the electrons jump from one site to another, occupied by atoms with different conduction-electron wave functions. With this assumption and introducing the quantity $[\xi]=\Sigma_{i^c}|i^c\rangle\xi_i\langle i^c|$, we can eliminate the off-diagonal disorder in the wide subband and write the Green's function associated with the mean-field Hamiltonian in the form[28]

$$\begin{aligned} g^{-1}=z-H=[\xi]\Bigg[\sum_i \tilde{\varepsilon}_i^{c\gamma}|i^c\rangle\langle i^c|-\sum_{ij} T_0|i^c\rangle\langle j^c|\Bigg][\xi] \\ +\sum_i (z-\varepsilon^f)|i^f\rangle\Bigg\langle i^f\Bigg|-\sum_{ij} T_{ij}^{ff}\Bigg|i^f\Bigg\rangle\langle j^f| \\ -\tilde{V}^{cf}\sum_i (|i^c\rangle\langle j^f|+(|i^f\rangle\langle j^c|), \end{aligned} \tag{3}$$

where $z=\varepsilon+i0$. This Green's function contains only diagonal disorder described by the effective energy $\tilde{\varepsilon}_i^{c\gamma}$, defined by

$$\tilde{\varepsilon}_i^{c\gamma}=\frac{z-\varepsilon_i^{c\gamma}}{\xi_c^2}. \tag{4}$$

In order to deal with this diagonal disorder, we replace the energy $\tilde{\varepsilon}_i^{c\gamma}$ in Eq. (3) by an effective medium with self-energy Σ to restore the translational invariance of the conduction electrons. In this manner and considering the two-subband character of the model, the Green's function for the narrow and wide subbands in k space, written in terms of the effective medium, are given by:[7]

$$g_k^{ff}(z)=\sum_k \frac{[\Sigma(z)-\varepsilon_k^c]}{(z-\varepsilon^f-\varepsilon_k^f)[\Sigma(z)-\varepsilon_k^c]-(\tilde{V}^{cf})^2}, \tag{5}$$

$$g_k^{cc}(z)=\sum_k \frac{(z-\varepsilon^f-\varepsilon_k^f)}{(z-\varepsilon^f-\varepsilon_k^f)[\Sigma(z)-\varepsilon_k^c]-(\tilde{V}^{cf})^2}. \tag{6}$$

The existence of two dispersion relations makes the calculation of these Green's functions much more difficult. In order to simplify this calculation, we use the homothetic band approximation[29] to treat the two subbands of the model. In this approximation, the dispersion relation of the wide subband is adopted as a model ($\varepsilon_k^c=\varepsilon_k$), and the other one, corresponding to the narrow subband, is taken as $\varepsilon_k^f=\alpha\varepsilon_k$, where α is a parameter smaller than 1. After transforming the k summation into an integral over energy, the Green's functions $g^{ff}(z)$ and $g^{cc}(z)$ in real space are given by

$$\begin{aligned} g^{ff}(z)=\frac{1}{\alpha(E_+-E_-)}\{[\Sigma(z)-E_-(z)]F[E_-(z)] \\ -[\Sigma(z)-E_+(z)]F[E_+(z)]\}, \end{aligned} \tag{7}$$

$$\begin{aligned} g^{cc}(z)=\frac{1}{\alpha[E_+(z)-E_-(z)]}\{(z-\varepsilon^f-\alpha E_-)F[E_-(z)] \\ -[z-\varepsilon^f-\alpha E_+(z)]F[E_+(z)]\}. \end{aligned} \tag{8}$$

In these expressions, $F[E_\pm(z)]$ is the Hilbert transform given by

$$F[E_\pm(z)]=\int \frac{\rho_0(\varepsilon)}{E_\pm(z)-\varepsilon}d\varepsilon, \tag{9}$$

where $\rho_0(\varepsilon)$ is a standard model density of states for the wide subband. The quasiparticle energies $E_\pm(z)$ are given by

$$\begin{aligned} E_\pm(z)=\alpha\Sigma(z)+(z-\varepsilon^f) \\ \pm\sqrt{[\alpha\Sigma(z)-(z-\varepsilon^f)]^2+4\alpha(\tilde{V}^{cf})^2}. \end{aligned} \tag{10}$$

In order to calculate the Green's functions g^{ff} and g^{cc}, it still remains to determine the self-energy Σ. To this end we follow the usual procedure of the coherent potential approximation (CPA), replacing, in a particular site of the Green's function, the self-energy Σ by the effective energy $\tilde{\varepsilon}_i^{c\gamma}$. In doing so, we create a Slater-Koster problem in the effective medium of the conduction electrons, so that the average conduction-electron Green's function can be written in a Dyson-like form as

$$\langle G_{ij}^{cc}(z)\rangle=\langle g_{ij}^{cc}(z)\rangle+\langle g_{i0}^{cc}(z)\rangle[\Sigma(z)-\tilde{\varepsilon}^{c\gamma}(z)]\langle G_{0j}^{cc}(z)\rangle, \tag{11}$$

where G_{ij}^{cc} is a perturbed Green's function for the disordered problem in the effective medium, and g_{ij}^{cc} is the unperturbed Green's function. After some simple algebra we obtain the off-diagonal CPA equation

$$\begin{aligned} (1-x)\frac{[\Sigma(z)-\tilde{\varepsilon}^{cH}(z)]}{1-[\Sigma(z)-\tilde{\varepsilon}^{cH}(z)]g^{cc}(z)} \\ +x\frac{[\Sigma(z)-\tilde{\varepsilon}^{cI}(z)]}{1-[\Sigma(z)-\tilde{\varepsilon}^{cI}(z)]g^{cc}(z)}=0, \end{aligned} \tag{12}$$

where x is the impurity concentration, and $\tilde{\varepsilon}^{cH}$ and $\tilde{\varepsilon}^{cI}$ represent the two different energies in the wide subband. This equation together with Eq. (8) self-consistently determine the self-energy Σ. The density of states of the narrow subband is given by

$$\rho^f(z)=-\frac{1}{\pi}\mathrm{Im}\, g^{ff}(z), \tag{13}$$

and the total density of state for the wide subband is given by

$$\rho^c(z)=(1-x)\Bigg[-\frac{1}{\pi}\mathrm{Im}\, G^{cH}(z)\Bigg]+x\Bigg[-\frac{1}{\pi}\mathrm{Im}\, G^{cI}(z)\Bigg], \tag{14}$$

where $G^{c\gamma}$ is the perturbed Green's function given by[28]

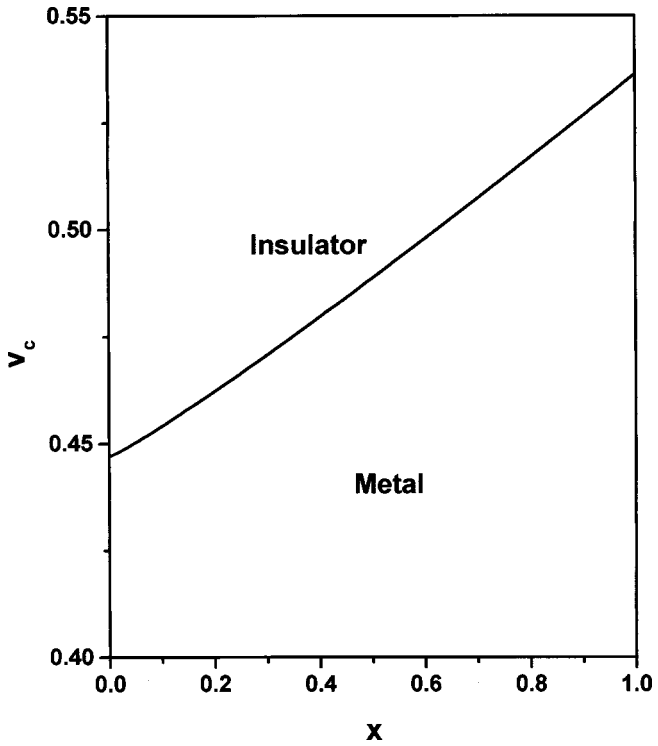

FIG. 1. Criterion for the opening of the energy gap as a function of the concentration.

$$G^{c\gamma}(z)=\frac{1}{\xi_c^2}\left[\frac{g^{cc}(z)}{1-[\Sigma(z)-\tilde{\varepsilon}^{c\gamma}(z)]g^{cc}(z)}\right]. \qquad (15)$$

Using the densities of states given by Eqs. (13) and (14), we are able to calculate the magnitude of the hybridization energy gap, if any, as a function of the impurity concentration. Alternatively, we can determine a criterion for the opening of the hybridization energy gap. In order to do so, we should remember that the energy gap between the quasiparticle energy subbands is calculated by taking the difference between the bottom of the higher-energy subband and the top of the lower-energy subband. Assuming that the center of the wide energy subband is not much affected by the presence of impurities the edges of the quasiparticle energy subbands in the case of half-filling of the wide subband are given by the condition $E_{\pm}=\pm W(x)/2$. Here $W(x)$ is the impurity-concentration-dependent width of the wide subband, which is self-consistently determined by the equations described before. With these assumptions and using Eq. (10), an energy gap develops when the renormalized hybridization satisfies the condition $\tilde{V}^{cf}\geqslant V_c=[W(x)/2]\sqrt{\alpha}$. This renormalized criterion for the opening of the energy gap, very similar to that obtained in Ref. 7, tell us that the modification of the bandwidth caused by impurities changes the critical value of the hybridization for the appearance of the insulating phase. In this way it is possible to obtain a metal-insulator transition by properly diluting impurities into the system.

III. RESULTS AND DISCUSSION

In this section we apply our model to the particular case of $CeRh_{1-x}Pd_xSb$. In order to do this we have to fix a set of model parameters. We use a parabolic model density of state for the wide subband at half-filling, and take the homothetic band parameter as $\alpha=0.2$. In the clean limit of the insulating phase, i.e., $x=0$, we take the parameter ξ_c as unity and the renormalized hybridization parameter as $\tilde{V}^{cf}=0.45$ in units of half bandwidth. When Pd is introduced into the compound, the energy hopping of electrons between sites involving Pd atoms becomes larger than the corresponding energy hopping between sites occupied only by Rh atoms. That is because the conduction-electron wave function of Pd is wider than the corresponding one of Rh. In our model we parametrize this effect by a proper adjustment of the parameter ξ_c. Here we take the parameter ξ_c to produce an increase of 5% in the energy hopping. In this way, as the concentration of Pd increases the density of states becomes wider, and the criterion for the opening of the energy gap becomes larger. So the region of metallic phase is enlarged as x goes to 1. In Fig. 1 we show a phase diagram for the set of model parameters described above.

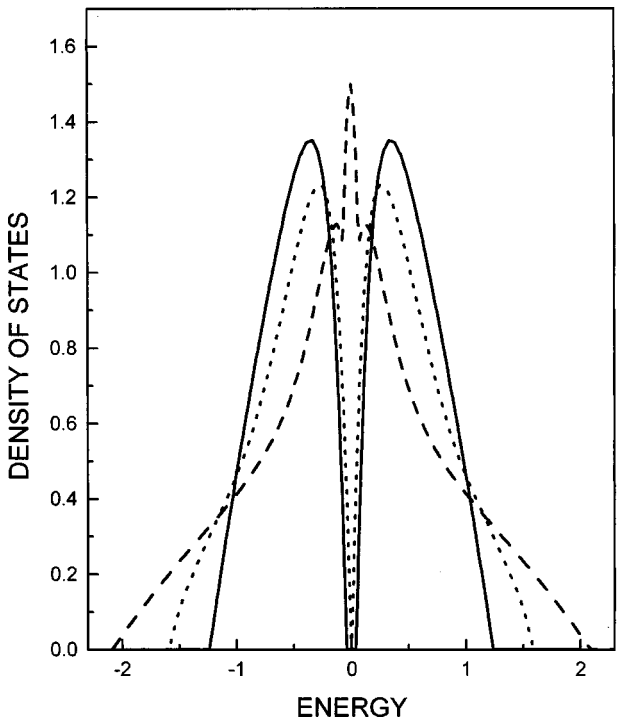

FIG. 2. Total density of states for the case of $CeRh_{1-x}Pd_xSb$ (energy is in units of the half bandwidth). The solid line represents the insulating phase for $x=0$, while the dashed line represents the metallic phase for $x=1$. The dotted line represents the critical concentration where the insulator-metal transition takes place. The Fermi level lies at zero energy.

Keeping the hitherto discussed model parameters fixed, we perform a self-consistent calculation of the densities of state as a function of the impurity concentration. For $x=0$ we obtain the total density of state with an energy gap shown by the solid line of Fig. 2. The Fermi level lies in the middle of the energy gap characterizing an insulating phase. At a critical Pd concentration of 10%, the renormalized hybridization ($\tilde{V}^{cf}$) becomes smaller than the criterion (V_c) for the opening of the energy gap, and the system undergoes an insulator-metal phase transition, as observed experimentally.[20] The dotted line in Fig. 2 shows the total density of states for the critical Pd concentration where the

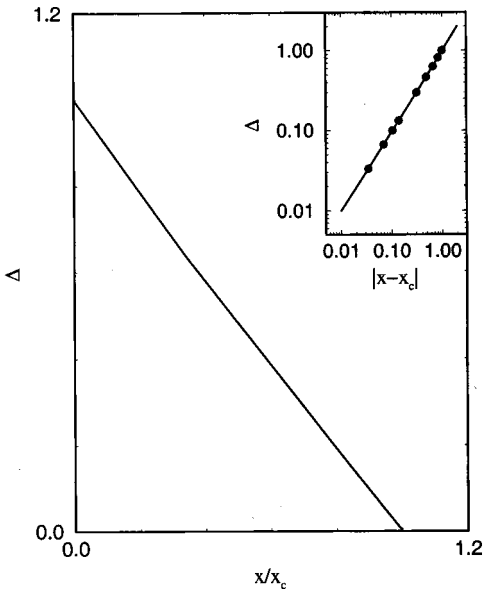

FIG. 3. Pd concentration dependence of the energy gap (Δ), in arbitrary units, for $CeRh_{1-x}Pd_xSb$. The inset shows a log-log plot for this curve where the circles are the calculated points, and the full line is a linear fit with a angular coefficient equal to 1.0

insulator-metal phase transition takes place. The dashed line represents the total density of states for the clean limit of a metallic phase ($x=1$). Using the calculated densities of states we obtain the magnitude of the energy gap. In Fig. 3 we plot the energy gap (Δ) as a function of Pd concentration. The insulator-metal phase transition occurs with a critical exponent $\mu=1$, as can be seen by the slope of the full line in the inset of this figure. This critical exponent indicates that this phase transition belongs to the same universality class as the density-driven transition predicted by scaling theory.[24,25] The case of other doped Kondo insulators such as $CeNi_{1-x}Pd_xSn$ and $CeNi_{1-x}Cu_xSn$ can be cast in the discussion of our model. However, we should mention that the case of $CeRh_{1-x}Ni_xSb$ is not explained by our model. According to our analysis, with the increase of Ni concentration, the bandwidth and the criterion for the opening of the energy gap decrease, so that the energy gap should be sustained. However, experimental data show that, above a critical Ni concentration of 10%, the gap is suppressed. This means that more details about the electronic structure of this compound should be considered in order to obtain a more realistic description of it. This interesting case will be investigated later.

The effect of impurities introduced in an anisotropic Kondo insulator is easily treated in this model, by including a k dependence in the hybridization between subbands. The present model can be also extended to deal with Ce-Ni-based Kondo insulators, in which impurities are introduced in both Ce and Ni sites. In these cases we expect, depending on the nature of the impurities, either cooperation or competition between the disorder effects in the formation of the energy gap. It is also worth mentioning that this two-subband model is straightforwardly extended to study effects of temperature and applied magnetic field on the metal-insulator transition of some doped Kondo insulators. In this case, starting in the insulating phase, there are two interesting situations to be discussed. In the first, impurities are introduced to favor the metallic phase. In this case, it is expected that the effects of impurities, temperature, and magnetic field will cooperatively work to close the hybridization energy gap. In the second situation, impurities are introduced to favor the insulating state. In this case, the effects of impurities tend to make the energy gap larger while the effects of temperature and magnetic field tend to annihilate the hybridization energy gap.[15,17] The combination of these two competing effects can produce some interesting aspects of the physical properties of Kondo insulators, such as magnetic susceptibility and specific heat. In addition, it is also interesting to study this model by using dynamical mean-field theory which becomes exact in the limit of infinite dimensions.[30–34] Calculations in all of these directions are now in progress, and will be published elsewhere.

In conclusion, in this paper we have discussed the effect of transition impurities on the metal-insulator transition in Kondo insulators by using a two-subband model. For the particular case of $CeRh_{1-x}Pd_xSb$ we found that a metal-insulator transition occurs at a critical Pd concentration of 10%, in good agreement with experimental data. It has also been shown that this phase transition belongs to the same universality class of density-driven transitions predicted by scaling theory.

ACKNOWLEDGMENTS

I would like to thank the Department of Physics and Astronomy at Rutgers University for their kind hospitality during my postdoctoral stay when this work was developed. This work was partially supported by a grant from the CNPq—Brazilian agency.

[1] G. Aeppli and Z. Fisk, Comments Condens. Matter Phys. **16**, 155 (1992), and references therein.

[2] Masatoshi Imada, Atsushi Fujimori, and Yoshimory Tokuro, Rev. Mod. Phys. **70**, 1039 (1998), and references therein.

[3] Masatoshi Imada, Fakher F. Assaad, Hirokazu Tsunetsugu, and Yukitoshi Motome, cond-mat/9808044 (unpublished).

[4] T. Sasso and Masatoshi Ito, Phys. Rev. B **53**, 6877 (1996).

[5] S. Doniach Castor Fu and S. A. Trugman, Physica B **199–200**, 450 (1994).

[6] Carlos Sanches-Castro, Kevin S. Bedell, and Bernard R. Cooper, Phys. Rev. B **47**, 6879 (1993).

[7] M. A. Continentino, G. M. Japiassu, and A. Troper, Phys. Rev. B **49**, 4432 (1994).

[8] M. A. Continentino, G. M. Japiassu, and A. Troper, J. Appl. Phys. **75**, 6734 (1994).

[9] M. Kyogaku, Y. Kitaoka, H. Nakamura, K. Asayama, T. Takabatake, F. Teshima, and H. Fujii, J. Phys. Soc. Jpn. **59**, 1728 (1990).

[10] T. Ohama, H. Yasuoka, and Y. Isikawa, J. Phys. Soc. Jpn. **64**, 4566 (1995).

[11] T. Takabatake, H. Tanaka, Y. Bando, H. Fujii, S. Nishigori, T. Suzuki, T. Fujita, and G. Kido, Phys. Rev. B **50**, 623 (1994).

[12] T. E. Mason, G. Aeppli, A. P. Ramirez, K. N. Clausen, C. Broholm, N. Stücheli, E. Bucher, and T. T. M. Palstra, Phys. Rev. Lett. **69**, 490 (1992).
[13] H. Ikeda and K. Myake, J. Phys. Soc. Jpn. **65**, 1769 (1996).
[14] J. Moreno and Piers Coleman, cond-mat/9807003 (unpublished).
[15] M. V. Tovar Costa, N. A. de Oliveira, A. Troper, G. M. Japiassu, and M. A. Continentino, Phys. Rev. B **57**, 6943 (1998).
[16] M. V. Tovar Costa, N. A. de Oliveira, A. Troper, G. M. Japiassu, and M. A. Continentino, Phys. Rev. B **60**, 1444 (1999).
[17] M. V. Tovar Costa, N. A. de Oliveira, A. Troper, G. M. Japiassu, and M. A. Continentino, J. Appl. Phys. **85**, 5332 (1999).
[18] K. Tsutsui, Y. Ohta, R. Eder, S. Maekawa, E. Dagotto, and J. Riera, Physica B **230–232**, 421 (1997).
[19] Latika Menon and S. K. Malik, Physica B **230–232**, 695 (1997).
[20] Latika Menon, F. E. Kayzel, A. de Visser, and S. K. Malik, Phys. Rev. B **58**, 85 (1998).
[21] A. Slebarski, A. Jezierski, S. Mahl, M. Neumann, and G. Borstel, Phys. Rev. B **56**, 7245 (1997).
[22] P. Scholtman, Phys. Rev. B **46**, 998 (1992).
[23] Ryousuke Shina, J. Phys. Soc. Jpn. **64**, 702 (1995).
[24] M. A. Continentino, Phys. Rep. **239**, 179 (1994).
[25] M. A. Continentino, Phys. Lett. A **197**, 417 (1995).
[26] P. Coleman, Phys. Rev. B **29**, 3035 (1984).
[27] Zlatko Tesanovic and Oriol T. Valls, Phys. Rev. B **34**, 1918 (1986).
[28] H. Shiba, Prog. Theor. Phys. **77**, 46 (1971); A. A. Gomes and C. A. da Silva (private communication).
[29] R. Kishore and S. K. Joshi, Phys. Rev. B **2**, 6943 (1970).
[30] T. Pruschke, M. Jarrell, and J. Freericks, Adv. Phys. **44**, 187 (1994).
[31] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1995).
[32] V. Dobrosavljevic and G. Kotliar, cond-mat/9706231 (unpublished).
[33] Ekkehard Lange and Gabriel Kotliar, cond-mat/9810207 (unpublished).
[34] R. Chiltra and G. Kotliar, cond-mat/9811144 (unpublished).