PHYSICAL REVIEW B **74**, 012503 (2006)

Coexistence of spin polarization and pairing correlations in metallic grains

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(Received 13 June 2006; published 19 July 2006)

We investigate the competition between magnetic depairing interactions, due to spin-exchange mechanism and/or to spin-dependent asymmetric bandwidths, and pairing coupling in metallic grains. We present a detailed analysis of the quantum ground state in different regimes arising from the interplay between ferromagnetic and pairing correlations for different fillings. We find out that the occurrence of a ground state with coexisting spin-polarization and pairing correlations is enhanced when the asymmetric spin-dependent distribution of the single-particle energies is considered. The mechanisms leading to such a stable quantum state are finally clarified.

DOI: 10.1103/PhysRevB.74.012503 PACS number(s): 74.20.Mn, 75.10.Lp

The coexistence of ferromagnetic (FM) and superconducting (SC) ordering is a central problem in condensed-matter physics since these two cooperative phenomena are usually antagonists. The relevant investigation originated from the analysis of the detrimental effect of dilute magnetic impurities on superconductivity,[1] and the discovery of ternary FM superconductors stimulated further studies.[2] The coexistence phase in the latter case emerges via either a modulated spin structure, the so-called cryptoferromagnetism,[3] or a ferromagnetic spiral phase.[4] Recently, the coexistence of FM and SC phases has attracted renewed interest due to the unprecedented occurrence of the magnetic order with substantial FM component in high-temperature superconductors[5] and to the observation of superconductivity inside the FM region in heavy-fermion metals.[6] Although the pairing in the triplet channel does not impose strong limitations to the formation of a SC phase with FM correlations,[7,8] a singlet SC pairing requires special conditions to be fulfilled. In this context, it has been shown that the coexistence of an *s*-wave SC state with a Stoner ferromagnetism can be achieved,[9] although such a phase is not energetically stable.[10] The conditions for the coexistence become less severe if the itinerant ferromagnetism is due to a change in the bandwidth of electrons with opposite spins.[11] Thus, a stable FM-SC state is realized when the unpaired electrons, from which the spin polarization arises, have a feedback with kinetic gain on the carriers involved in the pairing.[12]

The aim of this paper is to consider the coexistence of FM and SC ordering in metallic grains, whose size is such that the average level spacing is comparable to the energy scale (i.e., the SC gap) for the onset of the macroscopic order. The starting point to address this issue is the analysis of an isolated grain where the interactions between electrons are reduced to the mesoscopic Stoner exchange and the pairing term.[13] Nevertheless, it is unlikely to have a ground state with a nonzero total spin, if one does not allow for mesoscopic fluctuations that include an ensemble of grains.[14] We attempt to explore a different mechanism that may enhance the probability for the occurrence of a ground state with a spin polarization and pairing correlations. To this end, we investigate the role of an asymmetric, and spin-dependent, distribution of the single-particle energies. As a general outcome, it turns out that a coexistence of FM and SC correlations is allowed for the cases of small or ultrasmall grains, depending on the asymmetric bandwidth ratio. In this mesoscopic regime, it is the occurrence of a nonuniform renormalization of the energy separation between different unpaired configurations that makes possible the formation of a FM-SC ground state. As for the reduced BCS model,[15] our problem can be exactly solved in the canonical ensemble. This possibility permits to assess different ground state (GS) configurations for the entire range of the pairing and magnetic interaction strengths and to clarify the mechanisms involved in transitions between different regimes.

The dynamics we refer to in this study is described by the following Hamiltonian

$$H=\sum_{j=1}^{\Omega}\sum_{\sigma=+,-} w_\sigma \epsilon_j c_{j\sigma}^\dagger c_{j\sigma} - g\sum_{j,j'} c_{j+}^\dagger c_{j-}^\dagger c_{j'-} c_{j'+} - J\hat{S}^2,$$

where $c_{j\sigma}^\dagger$ ($c_{j\sigma}$) is the creation (annihilation) operator for an electron on level j, and ϵ_j are the single-electron energies. The first term in the Hamiltonian H describes the spin-dependent kinetic energy, with w_σ indicating the factor controlling the bandwidth amplitude for different spin polarizations. The second and third terms of H describe the electron-electron interaction via pairing g and the FM exchange J, respectively. Here the pairing strength is $g=\lambda d$, with d being the mean level spacing and λ a dimensionless coupling constant. We have chosen the FM exchange to be the Casimir of the Lie algebra $SU(2)$ in the spin sector, with $\hat{S}_j^+=(\hat{S}_j^-)^\dagger=c_{j+}^\dagger c_{j-}$, and $\hat{S}_j^z=1/2(c_{j+}^\dagger c_{j+}-c_{j-}^\dagger c_{j-})$. Our application involves a uniformly spaced distribution of energy levels, $\epsilon_j=-\frac{1}{2}(\Omega+1-2j)d$, spreading symmetrically around $\epsilon=0$ and labeled by a discrete index $j=1,\ldots,\Omega$.

Let us briefly discuss the exact solution of the problem. Introducing another $SU(2)$ Lie algebra in the pairing sector T, with $\hat{T}_j^+=(\hat{T}_j^-)^\dagger=c_{j+}^\dagger c_{j-}^\dagger$, and $\hat{T}_j^z=1/2(c_{j+}^\dagger c_{j+}+c_{j-}^\dagger c_{j-}-1)$, one can rearrange the Hamiltonian H into two parts $H=H_T+H_S$, where

$$H_T = \sum_j (w_+ + w_-)\epsilon_j \hat{T}_j^z - \frac{1}{2} g \sum_{j,k} (\hat{T}_j^+ \hat{T}_k^- + \hat{T}_k^+ \hat{T}_j^-),$$

$$H_S = \sum_j (w_+ - w_-)\epsilon_j \hat{S}_j^z - J\hat{S}^2, \qquad (1)$$

up to an irrelevant constant. Since H_T and H_S commute with each other, the singly occupied levels do not participate in the pair scattering, thus staying "blocked" according to the Pauli principle. Similarly, the double (empty) states do not enter the spin dynamics. Another relevant feature in the structure of H is the correspondence between the spin and pairing sectors: the effective magnetic field $(w_+ - w_-)\epsilon_j$ in the spin channel corresponds to the kinetic energy $(w_+ + w_-)\epsilon_j$ of a pair, and the transverse part of the magnetic exchange J has its counterpart in the pairing amplitude g. This is crucial for the problem to be exactly solvable. Let us denote with $|n,m\rangle$ a generic eigenstate of H with $N=2(n+m)$ electrons. In this state, $2m$ electrons fill a set B of singly occupied (and thus blocked) levels, while the remaining n pairs are distributed among the set U of $N_U = \Omega - 2m$ unblocked levels. Then, following Richardson[16] (see also Ref. 17), one can show that a generic eigenstate of H can be expressed as $|n,m\rangle = \Pi_{\beta=1}^{m+S^z} |\psi_\beta\rangle \Pi_{\mu=1}^{n} |\psi_\mu\rangle$, where $|\psi_\beta\rangle = \Sigma_{j\in B} \frac{\hat{S}_j^+}{(w_+ - w_-)\epsilon_j - \bar{E}_\beta} |-\rangle$ and $|\psi_\mu\rangle = \Sigma_{j\in U} \frac{c_{j+}^\dagger c_{j-}^\dagger}{(w_+ + w_-)\epsilon_j - E_\mu} |0\rangle$. Here $|-\rangle = \Pi_{i\in B} c_{i-}^\dagger |0\rangle$, with $|0\rangle$ being the vacuum state, and S^z is the z projection of the total spin of the electrons in the blocked levels. Furthermore, the n parameters E_μ and the $(m+S^z)$ terms $\bar{E}_\beta$ are the solutions of the two sets of the Richardson equations

$$\frac{1}{g} + \sum_{\nu=1(\nu\neq\mu)}^{n} \frac{2}{E_\nu - E_\mu} = \sum_{j\in U} \frac{1}{(w_+ + w_-)\epsilon_j - E_\mu},$$

$$\frac{1}{J} + \sum_{\alpha=1(\alpha\neq\beta)}^{m+S^z} \frac{2}{\bar{E}_\alpha - \bar{E}_\beta} = \sum_{j\in B} \frac{1}{(w_+ - w_-)\epsilon_j - \bar{E}_\beta}.$$

Moreover, the total energy is obtained by summing up the contributions from both the spin and pairing sectors: $E(n,m) = \Sigma_{\mu=1}^{n} E_\mu + \Sigma_{\beta=1}^{m+S^z} \bar{E}_\beta - J(S^{z2} - S^z) + \Sigma_{j\in B} w_- \epsilon_j$. For the GS configurations, the blocked levels in B should be as close as possible to the effective Fermi energy of the uncorrelated N-electron Fermi sea. On the other hand, the two polarization mechanisms due to the asymmetric spin-dependent bandwidths (the w mechanism) and the ferromagnetic exchange coupling (the J mechanism) tend to align the spins of electrons in the blocked levels. Then, the effective dynamics is marked by a subtle balance between the spin terms and the pairing one in the total energy $E(n,m)$. This competition gives rise to different GS configurations, including the FM state, the SC state, and the coexisting FM-SC state. It is worth pointing out that, making use of the relation $d/\Delta = 2\sinh[1/\lambda]/\Omega$ (Δ being the BCS gap value), one can tune the value of the coupling λ in a way to pass from the ultrasmall grain regime ($d \gg \Delta$) to large grain size ($d \ll \Delta$) one. Hereafter, for convenience, the parameter $w = w_-/w_+$ is used for the ratio between the spin-dependent bandwidths, while the sum $w_- + w_+ = 2$ is kept fixed, and the scale unit is d.

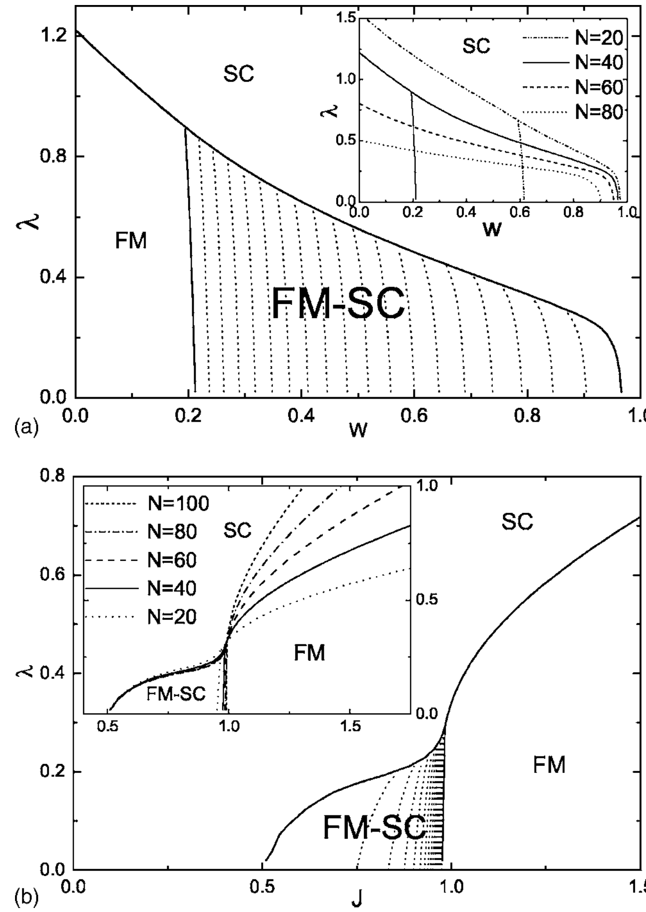

FIG. 1. (a) Representative diagram of the GS configurations in the $w-\lambda$ plane at zero exchange coupling for a density of $N=40$ electrons and 100 levels. (b) GS configurations with equal spin-bandwidths ($w=1$) and non zero magnetic exchange J. The dotted lines are the boundaries separating regimes with different numbers of depaired levels. The insets present the evolution of the GS by varying the electron densities.

Let us first study the effect of the asymmetric spin-dependent bandwidths on the spin polarization. The main features may be summarized as follows: (i) a coexisting FM-SC state always occurs when the system is away from half filling ($N=\Omega$); (ii) there are two different types of transitions between the FM and SC states driven by the relative strength of the pairing coupling with respect to the force for inducing a spin polarization; (iii) a fully polarized FM state occurs for a large spin-bandwidths asymmetry (i.e., small w) and low (high) electron densities (due to the electron-hole symmetry). At half filling, the FM-SC and FM states have always higher energy than the SC state, due to the loss of kinetic energy resulting from the presence of a nonzero polarization. Adding holes (electrons) to the half-filling configuration gives rise to different behaviors, depending on the values of w and λ. As plotted in Fig. 1(a), in the weak-coupling regime (small λ), the energy of the SC state gets higher if the ratio w reduces from 1 to 0, so the system arranges itself in a configuration where the paired levels are broken into the "blocked" levels, which occurs one by one.

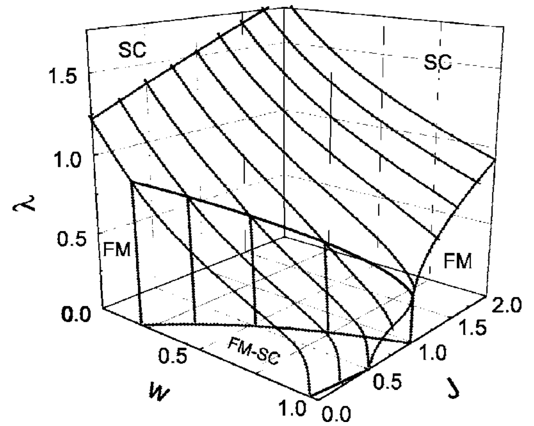

FIG. 2. Diagram of the GS configurations in the (λ, w, J) plane. We consider the case of $N=40$ electrons and 100 levels.

Hence, there is a discrete sequence of steps that link the SC state with zero magnetization to the fully polarized one, with a FM-SC configuration as an intermediate state. It should be emphasized that this happens only for pairing coupling that would correspond to the ultrasmall grain size. Indeed, for larger λ, the spin-bandwidth asymmetry induces a transition associated with a jump in the magnetization from zero to the intermediate or saturated value before the one-by-one depairing occurs. The strength of λ, at which the SC state is not anymore the lowest energy configuration, grows as w goes to zero, due to the increasing stiffness in pairing up the blocked levels. Concerning the evolution of the main boundaries as a function of the electron density, we notice that the FM-SC regime shrinks with respect to the w mechanism when the density changes from half-filling towards the completely empty (full) configuration, whereas the FM regime gets enhanced [see the inset in Fig. 1(a)]. Otherwise, with respect to the nonpolarized SC state, the FM-SC configuration gets more stabilized as a function of the pairing energy.

Now, we consider the effect of the ferromagnetic exchange coupling. The emerging picture reported in Fig. 1(b) is as follows: a coexisting FM-SC state is confined to a small portion of the phase diagram, i.e., only in proximity of the transition from an unpolarized state to a fully polarized one $(J \sim 1)$. For relatively strong pairing λ, the SC and FM states are energetically more favorable and their competition leads to a direct transition from the SC state to the FM one when J increases. The variation in the density does not significantly modify the FM-SC regime, but it does influence the boundary between the SC and FM states [see the inset in Fig. 1(b)]. Also in this case, there is a discrete sequence of steps from the SC state to the FM state, which goes via intermediate coexisting FM-SC configurations whose magnetization grows by breaking one pair at each step. The range of coupling where this change occurs is very narrow around the value $J \sim 1$. Moreover, the above studies show that it is intrinsic for the spin-exchange coupling J to hinder a GS configuration with coexisting polarized and paired electrons. Then, we simultaneously activate both the depairing mechanisms (with nonzero w and J). As plotted in Fig. 2, the effect of w in presence of the direct spin-exchange interaction J is to drive the system toward a fully polarized configuration. The J amplitude, below which a GS with coexisting FM-SC correlations is set in, renormalizes down to zero for small spin-bandwidth asymmetry (i.e., small w). Still, the FM configuration is favored by the concomitant action of J and w, as the values of J at the transition line that separates the FM-SC region from the FM one are reduced in presence of the spin-bandwidth asymmetry.

To further clarify the transitions between the different GS configurations, we compare the main contributions to the total energy as due to the various mechanisms involved. Our analysis starts from the limit of strong pairing coupling.[18] A simple argument shows that the lowest-order term for the condensation energy in a state with m broken pairs reads as $E_m^\lambda = (N/2-m)(\Omega - N/2 - m+1)g$. The amount of energy required to decouple m pairs with respect to the SC configuration $(m=0)$ is $\Delta E_m^\lambda = m(\Omega - m+1)g$. Taking this into account, one finds that, on average, the energy required to get one depaired state is higher than that required to break $m>1$ pairs, and this amount goes down with increasing m. This means that, for strong pairing interaction, when it becomes energetically favorable to depair one level, it is more advantageous to pass directly from the SC state to the FM state than any intermediate compromise. To understand the character of the transitions from one to another GS configuration in the intermediate coupling, one has to take the contribution from the magnetic gain into account. In this case, we have to solve the Richardson equation to obtain E_m^λ. We may distinguish three cases. First, we focus on the case with $w \neq 1$ and $J=0$. The energy gain for m pairs to be polarized due to the spin-bandwidth asymmetry $(w \neq 1)$ is $E_m^w = -m(G_w - m)d$, with $G_w = (\Omega - N)(1-w)/(1+w)$. Comparing the evolution of the energy cost to polarize m pairs in a complete paired background with the magnetic gain in the kinetic energy, it is possible to extract the value of λ_c and w_c for the transition between the SC and the FM state (or FM-SC). A transition occurs if $\Delta E_m^\lambda / m = -E_m^w / m$. In this circumstance, as plotted in Fig. 3(a), $\Delta E_m^\lambda / m$ varies, while $-E_m^w / m$ remains constant for fixed m, with a horizontal intercept that increases if m varies from $N/2$ to 1. When $w=0.1$ and λ is large, the first crossing between the two contributions is due to the $m=N/2$ line, indicating that the change in the GS configuration is from the SC state to the FM state. In the intermediate case $(w=0.5)$, the various E_m^λ are closer in energy for larger m. This is due to the weakening of the strength for pair-hopping processes between levels that are separated by the blocked sector. Thus, the scale that controls the pairing dynamics in presence of many depaired states gets renormalized down (almost unchanged) for large (small) m unpaired levels, respectively. Because of this mismatch, the first crossing between $\Delta E_m^\lambda / m$ and $-E_m^w / m$ occurs between the SC and a FM-SC state with a finite number of depaired electrons $(1 < m_0 < N/2)$, whose value depends on w.

Next we consider the role of the coupling J. The magnetic energy gain is $E_m^J = -Jm(m+1)$. As reported in Fig. 3(b), for large J, due to the almost linear behavior of E_m^λ and the constant behavior of the magnetic gain, the first crossing occurs for $m=N/2$, thus indicating a direct transition in the GS configuration from the SC state to the FM state. This still happens around $J \sim 1$. Nevertheless, below this value there is

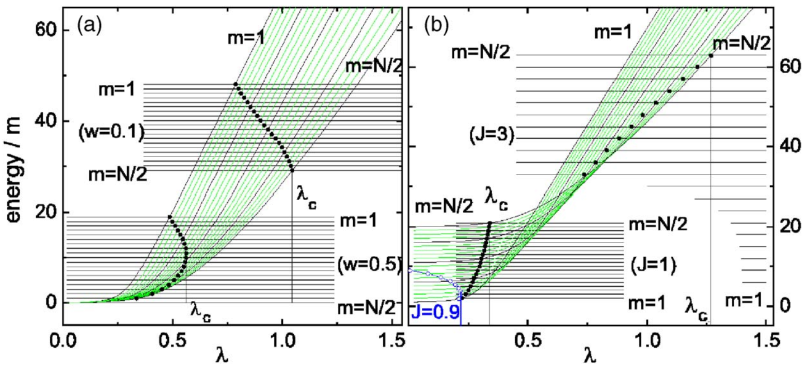

FIG. 3. (Color online) (a) Evolution of the average energy per depaired configuration vs λ for two values of w at zero exchange coupling. (b) As in (a) for three representative values of J at equal spin-bandwidths $(w=1)$. The competition between the amount of energy (green area) for creating unpaired polarized states within a paired background and that (black lines) due to the magnetic mechanism is also shown.

a change of curvature in the energy cost for depairing due to kinetic contribution of the polarized levels. Still, the nonuniform depairing cost, due to the blocking in the hopping process, has an intermediate first crossing with the uniform E_m^J. This leads to the occurrence of a transition between the SC and a FM-SC state with a finite number of unpaired electrons. When J is decreased, the magnetic part shrinks downward, there is no intersection, and the GS is SC.

Finally, we study the changeover that occurs inside the FM-SC regime by varying λ (w) and keeping fixed w (λ). There are separated energy scales in the spin and pairing sectors, related to the process of adding an extra unpaired state to a configuration with paired electrons and $2m_0$ polarized spins. In the pairing dynamics, the separation between levels with different m configurations gets very small due to the hopping of pairs that connects states beyond the blocked sector. By varying w, the large ratio between the scales mentioned before, allows to depair one level each step from the FM-SC state to the FM one. Otherwise, the lack of correspondence between the spin and pairing energy scales is responsible for allowing just one or zero depaired level as λ decreases.

In conclusion, we have investigated the GS configurations for a quantum problem where depairing processes contribute to the spin polarization and compete with the pairing correlations. The kinetic-induced polarization due to the asymmetric spin bandwidths is more favorable to get a FM-SC coexisting state, while the direct spin exchange is less adaptable to simultaneously accommodate the FM and SC states. The occurrence of a coexisting FM-SC state strongly depends on the size of the system as shown in the ground state diagram. It is intriguing to see that the enhanced quantum fluctuations, due to the reduced system size, weaken the competition between the magnetic and pairing correlations. In this context, when the pairing coupling and the magnetic energy compete with each other, the blocking-induced nonuniform renormalization of the energy separation, for different depaired configurations, is crucial for obtaining a GS configuration where polarized and paired electrons coexist.

[1] B. T. Matthias, H. Suhl, and E. Corenzwit, Phys. Rev. Lett. **1**, 449 (1958).

[2] See, for example, *Superconductivity in Ternary Compounds*, edited by M. B. Maple and F. Fisher (Springer-Verlag, Berlin, 1982).

[3] P. W. Anderson and H. Suhl, Phys. Rev. **116**, 898 (1959).

[4] E. I. Blount and C. M. Varma, Phys. Rev. Lett. **42**, 1079 (1979).

[5] J. Tallon, C. Bernhard, M. Bowden, P. Gilberd, T. Stoto, and D. Pringle, IEEE Trans. Appl. Supercond. **9**, 1696 (1999).

[6] S. S. Saxena, P. Agarwal, K. Ahilan, F. M. Grosche, R. K. W. Haselwimmer, M. J. Steiner, E. Pugh, I. R. Walker, S. R. Julian, P. Monthoux, G. G. Lonzarich, A. Huxley, I. Shelkin, D. Braithwaite, and J. Flouquet, Nature (London) **406**, 587 (2000).

[7] D. Fay and J. Appel, Phys. Rev. B **22**, 3173 (1980).

[8] V. P. Mineev, Phys. Rev. B **66**, 134504 (2002).

[9] N. I. Karchev, K. B. Blagoev, K. S. Bedell, and P. B. Littlewood, Phys. Rev. Lett. **86**, 846 (2001).

[10] R. Shen, Z. M. Zheng, S. Liu, and D. Y. Xing, Phys. Rev. B **67**, 024514 (2003).

[11] J. E. Hirsch, Phys. Rev. B **40**, 2354 (1989); **40**, 9061 (1989); **59**, 6256 (1999).

[12] M. Cuoco, P. Gentile, and C. Noce, Phys. Rev. Lett. **91**, 197003 (2003).

[13] I. L. Kurland, I. L. Aleiner, and B. L. Altshuler, Phys. Rev. B **62**, 14 886 (2000).

[14] G. Falci, R. Fazio, and A. Mastellone, Phys. Rev. B **67**, 132501 (2003); M. Schechter, *ibid.* **70**, 024521 (2004).

[15] J. von Delft and D. C. Ralph, Phys. Rep. **345**, 61 (2001); J. Dukelsky, S. Pittel, and G. Sierra, Rev. Mod. Phys. **76**, 643 (2004), and references therein.

[16] R. W. Richardson, Phys. Rev. **141**, 949 (1966); R. W. Richardson, J. Math. Phys. **18**, 1802 (1977).

[17] H.-Q. Zhou, J. Links, R. H. McKenzie, and M. D. Gould, Phys. Rev. B **65**, 060502(R) (2002).

[18] E. A. Yuzbashyan, A. A. Baytin, and B. L. Altshuler, Phys. Rev. B **68**, 214509 (2003).