Nonadiabatic Pauli susceptibility in fullerene compounds

E. Cappelluti,[1] C. Grimaldi,[2] and L. Pietronero[1]
[1]*Dipartimento di Fisica, Universitá di Roma I "La Sapienza," Piazzale A. Moro, 2, 00185 Roma, Italy and Istituto Nazionale Fisica della Materia, Unitá di Roma 1, Italy*
[2]*Département de Microtechnique–IPM, École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*
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The Pauli paramagnetic susceptibility χ is unaffected by the electron-phonon interaction in the Migdal-Eliashberg context. The adiabatic assumption of Migdal's theorem, however, is of questionable validity when applied to very narrow-band systems as the fullerene compounds. In these materials therefore the nonadiabatic effects are in principle relevant and quantities such as χ could be seriously affected by the electron-phonon interaction. In this paper we investigate the Pauli spin susceptibility in the nonadiabatic regime by following a conserving approach based on Ward's identity. We find that a sizable renormalization of χ due to electron-phonon coupling appears when nonadiabatic effects are taken into account. The intrinsic dependence of χ on the electron-phonon interaction gives rise to a finite and negative isotope effect which could be experimentally detected in fullerides. In addition, we find an enhancement of the spin susceptibility with temperature increasing, in agreement with the temperature dependence of χ observed in fullerene compounds. The role of electronic correlation is also discussed.

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I. INTRODUCTION

The Pauli susceptibility χ of wide-band metals is usually considered as unrenormalized by electron-phonon effects. As pointed out some time ago by Fay and Appel,[1] χ is actually affected by the electron-phonon coupling, the effect having magnitude of order $\lambda\omega_{\rm ph}/E_F$, where λ is the electron-phonon coupling, $\omega_{\rm ph}$ is a typical phonon frequency, and E_F is the Fermi energy. For wide-band metals, $\omega_{\rm ph}\ll E_F$ and the electron-phonon renormalization of χ is therefore negligible. This result basically follows from the observation that the lowest-order electron-phonon correction to χ is a vertex diagram which, according to Migdal's theorem,[2] is of order $\lambda\omega_{\rm ph}/E_F$. Hence the absence of phonon effects in χ is just a manifestation of the electron-phonon adiabaticity of normal metals.

This situation may be somewhat different for weak ferromagnetic metals like $ZrZn_2$.[1,3] In this case, the electron-phonon corrections compete with the exchange term, leading to a possible phonon-dependent ferromagnetic transition temperature. Experiments have, however, reported only small isotope effects in $ZrZn_2$ with quite large error bars,[4] leaving the problem of phonon-corrected weak-ferromagnetism essentially unresolved. The recent discovery of giant isotope shifts in manganites proves, however, the existence of ferromagnetic materials with important electron-phonon effects in their magnetic properties.[5]

The possibility of having electron-phonon-dependent spin susceptibility is, however, not correlated exclusively to the vicinity of magnetic instabilities. In fact, as long as $\lambda\omega_{\rm ph}/E_F$ is not negligible, as in very-narrow-band systems, the electron-phonon vertex contributions are no longer unimportant so that χ is expected to acquire a phonon renormalization. The most promising candidates for the observation of this effect are the C_{60}-based materials. Fullerene compounds have in fact phonon modes extending up to 0.2 eV and Fermi energies of about 0.3 eV.[6] Therefore, $\omega_{\rm ph}/E_F$ is in principle large, suggesting that C_{60}-based metals could be labeled as nonadiabatic systems.

Recently, the finding of a superconducting transition at $T_c=52$ K in hole-doped C_{60} single crystals has raised a renewed interest in these materials.[7] Nonzero isotope effects and other properties strongly indicate that superconductivity in fullerides is driven by electron-phonon interactions.[6] However, the description of superconductivity of Rb_3C_{60} within the traditional Migdal-Eliashberg (ME) theory is found to be inconsistent with respect to the adiabatic hypothesis $\lambda\omega_{\rm ph}/E_F\ll 1$ which is at the basis of the ME theory itself.[8] Instead, by relaxing the adiabatic hypothesis, a generalized formulation which includes nonadiabatic channels in the electron-phonon interaction provides a more self-consistent description of superconductivity, and suggests that the key ingredient for the high values of T_c in C_{60}-based materials is a constructive nonadiabatic interference rather than strong electron-phonon couplings.[8,9]

In principle, the hypothesis that superconductivity in fullerene compounds is enhanced essentially by nonadiabatic electron-phonon effects can be sustained by the observation of independent signatures of nonadiabaticity. In this respect and according to what we have pointed out at the beginning, Pauli susceptibility is a quantity where such signatures could be found. In this paper, we provide extensive calculations of χ beyond the adiabatic limit by including nonadiabatic effects at different stages of a perturbation theory in $\lambda\omega_{\rm ph}/E_F$. We find that when $\omega_{\rm ph}/E_F$ is no longer negligible, (i) the paramagnetic spin susceptibility can be considerably reduced with respect to the adiabatic limit and (ii) it acquires a negative isotope effect and (iii) a possible anomalous temperature dependence at constant sample volume. These features are signatures of nonadiabatic electron-phonon interactions and predictions (ii) and (iii) are susceptible of experimental verification. Such results suggest therefore that an important role could be played by a nonadiabatic electron-phonon interac-

tion in determining magnetic properties and stimulate further theoretical and experimental work on this line.

II. PAULI SUSCEPTIBILITY BY WARD'S IDENTITY

As remarked in the Introduction, the effect of electron-phonon interactions on spin susceptibility has already attracted some interest in the past mainly in relation to weak antiferromagnets. However, different approaches led to different results,[1,3] reflecting the lack of a controlled theory.

In our paper we use the functional formalism based on the Baym-Kadanoff technique to derive a conserving derivation of Pauli susceptibility valid for both electron-phonon and electron-electron interactions. The Pauli susceptibility χ is calculated by knowledge of the spin vertex function which is related to the self-energy via a Ward's identity.

Following the Baym-Kadanoff technique, we introduce an external magnetic field H coupled with the electrons which induces a magnetization M. The interaction Hamiltonian describing the coupling of H with the electron spins is

$$H_h = -h\sum_{\mathbf{k},\sigma} \sigma c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}, \tag{1}$$

where $h=\mu_B H$ (μ_B is the Bohr magneton) and $c^{\dagger}_{\mathbf{k}\sigma}$ ($c_{\mathbf{k}\sigma}$) are creation (annihilation) fermionic operators for electrons with wave number $\mathbf{k}$ and spin index $\sigma=\pm 1$. The electron magnetization M due to Eq. (1) is given by $M=\mu_B \Sigma_\sigma \sigma n_\sigma$ where $n_\sigma=\Sigma_{\mathbf{k}}\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}\rangle$ and $\langle\cdots\rangle$ denotes the statistical average. We express now the magnetization M in terms of the finite-temperature single-electron propagator

$$G_\sigma(\mathbf{k},\tau) = -\langle T_\tau c_{\mathbf{k}\sigma}(\tau) c^{\dagger}_{\mathbf{k}\sigma}(0)\rangle, \tag{2}$$

where T_τ is the time ordering operator and τ is the imaginary time. Since $\langle c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}\rangle = G_\sigma(\mathbf{k},0^-)$, the magnetization can be expressed as

$$M = \mu_B \sum_{\mathbf{k},\sigma} \sigma G_\sigma(\mathbf{k},0^-) = \mu_B T \sum_n \sum_{\mathbf{k},\sigma} \sigma G_\sigma(\mathbf{k},n) e^{-i\omega_n 0^-}. \tag{3}$$

In the above expression, $G_\sigma(\mathbf{k},n)$ is the thermal electron propagator which satisfies the following Dyson equation:

$$G^{-1}_\sigma(\mathbf{k},n) = i\omega_n - \epsilon(\mathbf{k}) + \mu + h\sigma - \Sigma_\sigma(\mathbf{k},n), \tag{4}$$

where $\omega_n=(2n+1)\pi T$, $n=0,\pm 1,\pm 2,\ldots$, are Matsubara frequencies, $\epsilon(\mathbf{k})$ the electron dispersion, μ the chemical potential, and $\Sigma_\sigma(\mathbf{k},n)$ is the electronic self-energy due to the coupling to phonons and to the electron-electron interaction. The spin susceptibility χ is formally given by $M=\chi H$ where $\chi=[dM/dH]_0$ is the derivative of the magnetization at zero field. Hence, from Eq. (3), a general expression for χ is

$$\chi(T) = \mu_B^2 T \sum_n \sum_{\mathbf{k},\sigma} \sigma \left[\frac{dG_\sigma(\mathbf{k},n)}{dh}\right]_0$$

$$= -2\mu_B^2 T \sum_n \sum_{\mathbf{k}} G(\mathbf{k},n)^2 \Gamma(\mathbf{k},n), \tag{5}$$

where $G(\mathbf{k},n)$ is the electron propagator for zero magnetic field which satisfies the $H\to 0$ limit of Eq. (4):

$$G^{-1}(\mathbf{k},n) = i\omega_n - \epsilon(\mathbf{k}) + \mu - \Sigma(\mathbf{k},n). \tag{6}$$

In the second term of Eq. (5) we have introduced the spin-vertex function

$$\Gamma(\mathbf{k},n) = \frac{1}{2}\sum_\sigma \sigma \left[\frac{dG^{-1}_\sigma(\mathbf{k},n)}{dh}\right]_0. \tag{7}$$

In Eqs. (5) and (7), the notation $[\cdots]_0$ indicates that the quantity in brackets must be calculated for zero magnetic field. Plugging Eq. (4) into Eq. (7) the resulting vertex Γ satisfies the Ward's identity,

$$\Gamma(\mathbf{k},n) = 1 - \frac{1}{2}\sum_\sigma \sigma \left[\frac{d\Sigma_\sigma(\mathbf{k},n)}{dh}\right]_0, \tag{8}$$

and since $\Sigma_\sigma(\mathbf{k},n)$ is a functional of the electron propagator, a self-consistent relation between Eqs. (7) and (8) is obtained which permits us to calculate the spin susceptibility. At this point, the spin susceptibility χ can be calculated once the electron self-energy and its magnetic field dependence is known.

III. NONADIABATIC PAULI SUSCEPTIBILITY

The electron-phonon interaction is usually neglected in calculations of the Pauli susceptibility although it can strongly renormalize other physical quantities.[10] Indeed it can be shown that the electron-phonon self-energy depends on the external magnetic field h at least as

$$\lim_{h\to 0} \Sigma^{\mathrm{ep}}(\mathbf{k},n) \sim h O(\omega_{\mathrm{ph}}/E_{\mathrm{F}}), \tag{9}$$

where ω_{ph} characterizes the phonon frequency scale and E_{F} is the Fermi energy. In wide-band materials the adiabatic ratio $\omega_{\mathrm{ph}}/E_{\mathrm{F}} \ll 1$ and the electron-phonon contribution to χ can be consequently disregarded. From Eq. (9), we see that electron-phonon effects appear in χ only at a nonadiabatic level. Hence, any evidence of electron-phonon effects on the Pauli susceptibility could be a direct proof of a nonadiabatic electron-phonon coupling. This statement holds true as long as phonon-modulated spin-orbit effects,[11] not included here, can be neglected. This should be, however, the case for the fullerene compounds and other systems with light atoms.

In order to properly include electron-phonon interactions in χ we need then to explicitly specify the functional form of the nonadiabatic electron-phonon self-energy. Nonadiabatic effects enter in a twofold way in the electron-phonon self-energy: finite-bandwidth effects and vertex diagrams. These two kinds of effects, of course, are of the same order in ω_{ph}/E_F, and there is no justification for neglecting vertex corrections with respect to finite-bandwidth effects.

In this paper we consider two approximation schemes for the electron-phonon self-energy. The first one is essentially the mean-field theory which corresponds to the noncrossing approximation. It is diagrammatically equivalent to the Migdal-Eliashberg electron-phonon self-energy without,

FIG. 1. Electronic self-energy within the noncrossing approximation.

however, assuming an infinite electron bandwidth compared to the relevant phonon energies. At this level only finite-bandwidth nonadiabatic effects are considered. The second one includes first-order electron-phonon vertex corrections as well as finite-bandwidth effects in the framework on the nonadiabatic Fermi liquid picture.[9] Both approximation schemes reduce to the adiabatic Migdal-Eliashberg limit for $\omega_{\rm ph}/E_F \to 0$.

A. Noncrossing self-energy

The self-consistent noncrossing approximation neglects the vertex corrections in the electron-phonon self-energy and for $\omega_{\rm ph}/E_F \ll 1$ reduces to the ME theory of the electron-phonon-coupled system. The diagrammatic representation of the electron self-energy is shown in Fig. 1 where the wiggly line represents the phonon propagator and the dashed line the electron-electron Coulomb repulsion. The corresponding compact expression of the noncrossing self-energy reads

$$\Sigma_\sigma(k) = \sum_{k'} [V(k-k') + Ie^{-i\omega_m 0^-}] G_\sigma(k'), \tag{10}$$

where k and k' are fermionic four-vectors defined as $k \equiv (\mathbf{k}, i\omega_n)$ and $k' \equiv (\mathbf{k}', i\omega_m)$. Moreover, $\Sigma_k \equiv -T\Sigma_n \Sigma_{\mathbf{k}}$ and $V(k-k') \equiv |g(\mathbf{k}-\mathbf{k}')|^2 D(k-k')$, where $g(\mathbf{k}-\mathbf{k}')$ is the electron-phonon matrix element and $D(k-k')$ is the phonon propagator. I is exchange Coulomb interaction which gives rise to the Stoner enhancement factor.

By introducing Eq. (10) into the expression of the spin-vertex function (8) and using Eq. (7) we obtain

$$\Gamma(k) = 1 - \sum_{k'} [V(k-k') + Ie^{-i\omega_m 0^-}] \sum_\sigma \frac{\sigma}{2} \left[\frac{dG_\sigma(k')}{dh} \right]_0$$

$$= 1 + \sum_{k'} [V(k-k') + I] G(k')^2 \Gamma(k'). \tag{11}$$

The noncrossing approximation for the self-energy leads therefore to a self-consistent ladder equation for $\Gamma(\mathbf{k}, n)$ (Ref. 12) (Fig. 2). If we consider dispersionless phonons of frequency ω_0, the above ladder equations can be rewritten in the extended notation as

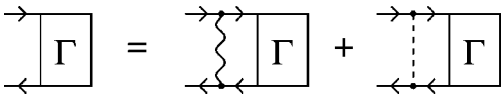

FIG. 2. Diagramatic self-consistent expression of the spin vertex in noncrossing approximation.

$$\Gamma(\mathbf{k}, n) = 1 - T \sum_{\mathbf{k}', m} [|g(\mathbf{k}-\mathbf{k}')|^2 D(n-m) + I] G(\mathbf{k}', m)^2 \Gamma(\mathbf{k}', m), \tag{12}$$

where

$$D(n-m) = \frac{\omega_0^2}{(i\omega_m - i\omega_m)^2 - \omega_0^2}. \tag{13}$$

The electron propagator appearing in Eq. (11) satisfies the Dyson equation (6) with the zero-field limit of the self-energy (10):

$$\Sigma(\mathbf{k}, n) = -T \sum_{\mathbf{k}', m} |g(\mathbf{k}-\mathbf{k}')|^2 D(n-m) G(\mathbf{k}', m) + IT \sum_{\mathbf{k}', m} G(\mathbf{k}', m) e^{-i\omega_m 0^-}. \tag{14}$$

In the simplest case of $\mathbf{k}$-independent Coulomb repulsion here considered the second term on the right side of Eq. (14) gives rise just to a constant term which can be absorbed into a redefinition of the chemical potential $\mu' \to \mu$. We can then neglect it since we shall consider only half-filled systems for which we set $\mu' = 0$.

In an isotropic system the angular dependence of the self-energy and of the vertex function in Eqs. (12) and (14) is negligible and it can be dropped. Therefore, following the same procedure reported in Ref. 12, we replace the electron-phonon interaction $|g(\mathbf{k}-\mathbf{k}')|^2$ by its average over the Fermi surface:

$$|g(\mathbf{k}-\mathbf{k}')|^2 \to \langle\langle |g(\mathbf{k}-\mathbf{k}')|^2 \rangle\rangle_{\rm FS} \equiv g^2, \tag{15}$$

where

$$\langle\langle |g(\mathbf{k}-\mathbf{k}')|^2 \rangle\rangle_{\rm FS} = \frac{\sum_{\mathbf{k},\mathbf{k}'} |g(\mathbf{k}-\mathbf{k}')|^2 \delta[\epsilon(\mathbf{k})] \delta[\epsilon(\mathbf{k}')]}{\sum_{\mathbf{k},\mathbf{k}'} \delta[\epsilon(\mathbf{k})] \delta[\epsilon(\mathbf{k}')]}. \tag{16}$$

In this way, the electron self-energy becomes independent of the momentum, $\Sigma(\mathbf{k}, n) = \Sigma(n)$, and the electron propagator becomes, at half-filling,

$$G(\mathbf{k}, n) = \frac{1}{iW_n - \epsilon(\mathbf{k})}, \tag{17}$$

where we have set $\Sigma(n) = i\omega_n - iW_n$. By using a constant density of states, N_0, the momentum summation in Eq. (14) is transformed as $\Sigma_{\mathbf{k}'} \to N_0 \int_{-E_F}^{+E_F} d\epsilon$ and, under integration over ϵ, the renormalized frequency W_n becomes

$$W_n = \omega_n - \lambda \pi T \sum_m D(n-m) \frac{2}{\pi} \arctan\left(\frac{E_F}{W_m} \right), \tag{18}$$

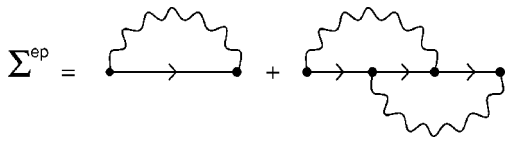

FIG. 3. Vertex-corrected electron-phonon self-energy.

where $\lambda = N_0 g^2$ is the electron-phonon coupling constant. By using Eqs. (15) and (17), also the spin-vertex function (12) becomes momentum independent and, by following the same steps as above, it reduces to

$$\Gamma(n) = 1 + T\sum_m [\lambda D(n-m) + I]\frac{2E_F}{W_m^2 + E_F^2}\Gamma(m). \quad (19)$$

Finally, since the self-energy and the spin-vertex function depend only on the frequency, the summation over **k** can be readily performed in Eq. (5) leading to

$$\chi(T) = \chi_P T\sum_n \frac{2E_F}{W_n^2 + E_F^2}\Gamma(n), \quad (20)$$

where $\chi_P = 2\mu_B^2 N_0$. The spin susceptibility is then obtained by the solution of Eqs. (18), (19), and (20).

B. Vertex-corrected self-energy

In the vertex correction approximation, the electron-phonon self-energy Σ^{ep} is modified with respect to the ME one by the inclusion of the first electron-phonon vertex diagram as shown in Fig. 3. By making use of the condensed notation introduced in Sec. III A, the vertex-corrected self-energy can therefore be written as follows:

$$\Sigma_\sigma^{ep}(k) = \sum_{k'} V(k-k')G_\sigma(k') \times\left[1 + \sum_q V(k-q)G_\sigma(q-k+k')G_\sigma(q)\right], \quad (21)$$

where $q \equiv (\mathbf{q}, i\omega_l)$. The derivative of $\Sigma_\sigma^{ep}(k)$ with respect to $h = \mu_B H$ calculated at zero magnetic field is

$$\left[\frac{d\Sigma_\sigma^{ep}(k)}{dh}\right]_0 = \sum_{k'} V(k-k')\left[\frac{dG_\sigma(k')}{dh}\right]_0 + \sum_{k',q} V(k-k') \times V(k-q)G(q-k+k')G(q)\left[\frac{dG_\sigma(k')}{dh}\right]_0 + \sum_{k',q} V(k-k')V(k-q)G(k')G(q) \times\left[\frac{dG_\sigma(q-k+k')}{dh}\right]_0 + \sum_{k',q} V(k-k')V(k-q) \times G(k')G(q-k+k')\left[\frac{dG_\sigma(q)}{dh}\right]_0 \quad (22)$$

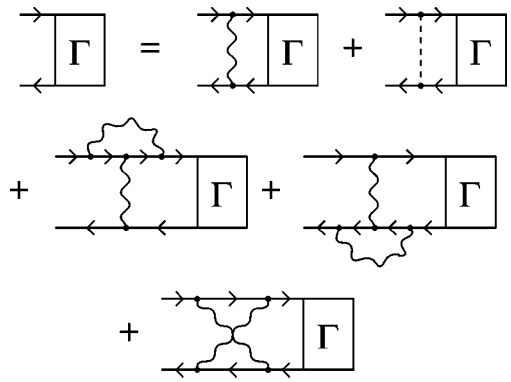

FIG. 4. Diagrammatic self-consistent expression of the spin vertex in the vertex-corrected theory.

and by rearranging the four-vector indices and using Eqs. (7) and (8) the spin-vertex equation reduces to

$$\Gamma(k) = 1 + \sum_{k'} [\tilde{V}(k,k') + I]G(k')^2\Gamma(k'), \quad (23)$$

where the electron-phonon kernel $\tilde{V}(k,k')$ is

$$\tilde{V}(k,k') = V(k-k')\left[1 + 2\sum_q V(k-q)G(q)G(q-k+k')\right] + \sum_q V(k-q)V(q-k')G(q)G(k+k'-q). \quad (24)$$

A graphical representation of Eqs. (23) and (24) in terms of Feynman diagrams is shown in Fig. 4.

In comparison with Fig. 2, the kernel of the spin susceptibility resulting from the vertex-corrected self-energy is modified by additional electron-phonon contributions represented by vertex and cross diagrams. This set of diagrams is therefore quite similar to those encountered in the theory of nonadiabatic superconductivity. They however differ in the orientation of the electron propagators (particle-hole rather than particle-particle contributions), leading to a different momentum dependence of the cross term. The evaluation of the kernel (24) can therefore be carried on by following a scheme similar to the one already employed in previous works.[9,13]

Let us first evaluate the zero-field self-energy entering the electron propagators in Eq. (24). As discussed before, the Coulomb self-energy provides just a constant term which can be absorbed into the definition of the chemical potential. The total self-energy reduces therefore to the electron-phonon one which from Eq. (21) can be written in the compact form

$$\Sigma(\mathbf{k},n) = -T\sum_m \sum_{\mathbf{k}'} |g(\mathbf{k}-\mathbf{k}')|^2 D(n-m) \times[1 + P(\mathbf{k},\mathbf{k}';n,m)]G(\mathbf{k}',m), \quad (25)$$

where we have introduced the vertex function given by

$$P(\mathbf{k},\mathbf{k}';n,m)=-T\sum_{l}\sum_{\mathbf{q}}|g(\mathbf{k}-\mathbf{q})|^2D(n-l)\times G(\mathbf{q}-\mathbf{k}+\mathbf{k}',l-n+m)G(\mathbf{q},l). \quad (26)$$

In analogy with the noncrossing approximation, we eliminate the angular dependence of the self-energy by replacing the whole electron-phonon matrix element, including the vertex correction, by its average over the Fermi surface:

$$\begin{aligned}&|g(\mathbf{k}-\mathbf{k}')|^2[1+P(\mathbf{k},\mathbf{k}';n,m)]\\&\quad\rightarrow\langle\langle|g(\mathbf{k}-\mathbf{k}')|^2[1+P(\mathbf{k},\mathbf{k}';n,m)]\rangle\rangle_{\mathrm{FS}}\\&\quad=g^2[1+\lambda P(Q_c;n,m)].\end{aligned} \quad (27)$$

An analytic expression of the Fermi-averaged vertex function $P(Q_c;n,m)$ can be obtained by using standard field theory techniques. The derivation follows closely the one outlined in Ref. 9, and here we present only the final result:

$$\begin{aligned}P(Q_c;n,m)=T\sum_{l}D(n-l)\Bigg(&B(n,m,l)\\&+\frac{A(n,m,l)-B(n,m,l)[W_l-W_{l-n+m}]^2}{(2E_FQ_c^2)^2}\\&\times\Bigg\{\sqrt{1+\left(\frac{4E_FQ_c^2}{W_l-W_{l-n+m}}\right)^2}-1\\&-\ln\left[\frac{1}{2}\sqrt{1+\left(\frac{4E_FQ_c^2}{W_l-W_{l-n+m}}\right)^2}\right]\Bigg\}\Bigg),\end{aligned} \quad (28)$$

where

$$\begin{aligned}A(n,m,l)=(W_l-W_{l-n+m})\Bigg[&\arctan\left(\frac{E_F}{W_l}\right)\\&-\arctan\left(\frac{E_F}{W_{l-n+m}}\right)\Bigg],\end{aligned} \quad (29)$$

$$\begin{aligned}B(n,m,l)=(W_l-W_{l-n+m})&\frac{E_FW_{l-n+m}}{[E_F^2+W_{l-n+m}^2]^2}\\&-\frac{E_F}{E_F^2+W_{l-n+m}^2}.\end{aligned} \quad (30)$$

The present expression of $P(Q_c;n,m)$ is an improvement of the previous calculation presented in the Appendix of Ref. 9 by explicitly including the self-consistent self-energy renormalization. The parameter Q_c is a dimensionless momentum describing the relevant momentum scale of the electron-phonon scattering process. In conventional metals, such as the low-temperature superconductors, the maximum exchanged phonon momentum q_c is about the Debye vector q_{D} and $Q_c\sim q_{\mathrm{D}}/2k_F\sim 1$. It is well known, however, that in systems with low density of charge carriers, as fullerene's compounds and cuprates, the electrons scatter only with charge fluctuations of long wavelength because of the strong electronic correlation. In fact, in strongly correlated systems, the electrons are surrounded by giant correlation holes which suppress charge density fluctuations with large momenta.[14–16] The relevant exchanged phonon scale is thus quite smaller than the Debye vector: $q_c<q_{\mathrm{D}}$ and $Q_c=q_c/2k_F<1$. As a consequence the effective electron-phonon interaction is suppressed for momenta larger than some cutoff q_c which depends on the filling factor and on the Coulomb repulsion. In Eq. (28) and in the following, we have modeled this situation by introducing a sharp momentum cutoff q_c. It can be used as a free parameter to distinguish between weak- and strong-correlation cases, where q_c is small for strong-correlated systems and of order 1 for weakly interacting electrons.

As usual, the self-energy effects can be expressed in a convenient form by means of the renormalized frequencies $W_n=\omega_n[1-\Sigma(n)/(i\omega_n)]$ obtained by integrating Eq. (25) over the energy. In the vertex-corrected theory they fulfill the self-consistent relation

$$W_n=\omega_n-\pi T_c\sum_{m}\lambda[1+\lambda P(Q_c;n,m)]\times D(n-m)\frac{2}{\pi}\arctan\left(\frac{E_F}{W_m}\right). \quad (31)$$

By using the momentum-independent self-energy (31), the spin-vertex function (23) can be rewritten as

$$\Gamma(\mathbf{k},n)=1-T\sum_{\mathbf{k}',m}[I+\tilde{V}(\mathbf{k},\mathbf{k}';n,m)]\frac{\Gamma(\mathbf{k}',m)}{[iW_m-\epsilon(\mathbf{k}')]^2}, \quad (32)$$

where the nonadiabatic electron-phonon spin kernel $\tilde{V}(\mathbf{k},\mathbf{k}';n,m)$ is given by

$$\begin{aligned}\tilde{V}(\mathbf{k},\mathbf{k}';n,m)=&|g(\mathbf{k}-\mathbf{k}')|^2D(n-m)[1+2P(\mathbf{k},\mathbf{k}';n,m)]\\&+C(\mathbf{k},\mathbf{k}';n,m).\end{aligned} \quad (33)$$

Here $P(\mathbf{k},\mathbf{k}';n,m)$ is again the first vertex correction given in Eq. (26) and $C(\mathbf{k},\mathbf{k}';n,m)$ is the cross correction whose explicit expression is given below:

$$C(\mathbf{k},\mathbf{k}';n,m)=T\sum_{l}\sum_{\mathbf{q}}|g(\mathbf{k}-\mathbf{q})|^2|g(\mathbf{q}-\mathbf{k}')|^2\times\frac{D(n-l)D(l-m)}{[iW_l-\epsilon(\mathbf{q})][iW_{n+m-l}-\epsilon(\mathbf{k}+\mathbf{k}'-\mathbf{q})]}. \quad (34)$$

Coherently with the approximations performed on the self-energy and with the Ward's relation (8), we evaluate $\Gamma(\mathbf{k},m)$ by replacing the kernel (33) by its momentum average over the Fermi surface:

$$\tilde{V}(\mathbf{k},\mathbf{k}';n,m)\rightarrow\langle\langle\tilde{V}(\mathbf{k},\mathbf{k}';n,m)\rangle\rangle_{\mathrm{FS}}=\tilde{V}(Q_c;n,m), \quad (35)$$

where

$$\tilde{V}(Q_c;n,m)=\lambda D(n-m)[1+2\lambda P(Q_c;n,m)] + \lambda^2 C(Q_c;n,m). \quad (36)$$

An explicit expression of $C(Q_c;n,m)$ can be also derived by using the same procedure as for $P(Q_c;n,m)$. We find

$$C(Q_c;n,m)=T\sum_l D(n-l)D(l-m)\Bigg\{2B(n,-m,l) + \frac{A(n,-m,l)-B(n,-m,l)(W_l-W_{l-n-m})^2}{2E_F^2Q_c^2|W_l-W_{l-n-m}|} \times \arctan\left(\frac{4E_FQ_c^2}{|W_l-W_{l-n-m}|}\right)\Bigg\}, \quad (37)$$

where the functions A and B are reported in Eqs. (29) and (30).

The final expression of the ladder vertex equation beyond the adiabatic approximation is readily obtained from Eqs. (32)–(36). The result of the integration over the electron energy gives

$$\Gamma(n)=1-\lambda T\sum_m [I+\tilde{V}(Q_c;n,m)]\frac{2E_F}{W_m^2+E_F^2}\Gamma(m). \quad (38)$$

Finally, the spin susceptibility in the vertex-corrected approximation is obtained by plugging Eq. (38) into Eq. (20).

IV. RESULTS

We are now in the position to calculate the Pauli susceptibility χ and to evaluate the effects on χ of the electron-phonon interaction, both in the noncrossing approximation and in the vertex corrected theory. Of course, when the adiabatic parameter ω_0/E_F or the electron-phonon coupling constant λ is turned to zero χ would reduce to the simple Stoner-enhanced susceptibility

$$\lim_{\lambda\to 0}\chi(T)=\lim_{\omega_0/E_F\to 0}\chi(T)=\frac{\chi_0(T)}{1-IN_0\dfrac{\chi_0(T)}{\chi_P}}, \quad (39)$$

where $\chi_0(T)$ is the free-electron Pauli susceptibility:

$$\chi_0(T)=-2\mu_B^2T\sum_n\sum_{\mathbf{k}}\frac{1}{[i\omega_n-\epsilon(\mathbf{k})]^2}=\chi_P[1-2f(E_F)], \quad (40)$$

where $f(E_F)$ is the Fermi distribution function at E_F.

In the following we will denote the zero electron-phonon limit in Eq. (39) as χ^{ee} and it will be used as a comparison element to evaluate the effects of the electron-phonon interaction.

In Fig. 5 we plot the total spin susceptibility (electron-electron + electron-phonon scattering) as function of the electron-phonon coupling and of the adiabatic parameter for zero temperature. Dashed lines are the results obtained within the noncrossing approximation while the solid lines are the data for the vertex-corrected theory. For this latter case we show the results for different values of the momentum cutoff Q_c ($Q_c=0.1$, 0.3, 0.5, 0.7, 1.0).

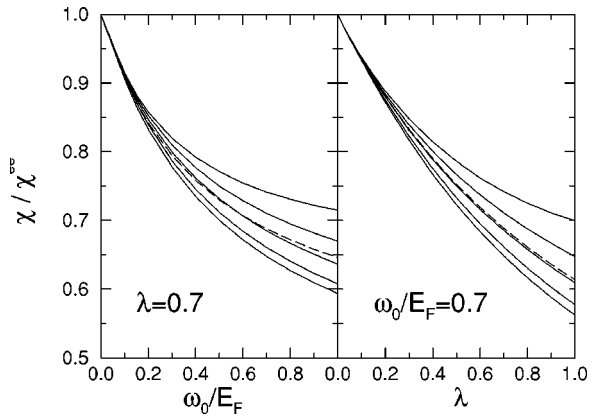

FIG. 5. Spin susceptibility as function of the adiabatic parameter ω_0/E_F and of the electron-phonon coupling λ for $N_0I=0.4$. Dashed lines represent the spin susceptibility for a noncrossing approximation, solid lines for the vertex-corrected theory (from the lower to the upper line: $Q_c=0.1$, $Q_c=0.3$, $Q_c=0.5$, $Q_c=0.7$, $Q_c=1.0$).

The first main result of Fig. 5 is that the inclusion of the electron-phonon coupling, *in the nonadiabatic regime* $\omega_0/E_F\geqslant 0$, yields a sensible reduction of χ with respect to the pure electronic spin susceptibility. As expected this effect vanishes as $\lambda\to 0$ (right panel) or $\omega_0/E_F\to 0$ (left panel). Note that both the noncrossing and vertex-corrected theories yield similar reduction. This is quite different from the situation encountered in the superconducting pairing channel,[8,9,13] where the effect of the vertex corrections is much stronger and highly dependent on Q_c.

The results of Fig. 5 suggest that some care is needed in estimating the value of the bare density of states from paramagnetic susceptibility measurements. Indeed, our analysis shows that χ is not simply related to the density of states by Eq. (39). Namely, disregarding the electron-phonon effects would lead to a substantial underestimation of the bare density of states from a spin susceptibility measurement as long as the electron-phonon interaction is in the nonadiabatic regime.

A more clear signature of the nonadiabatic effects is provided by the isotope dependence of the spin susceptibility. In Fig. 6 we report the numerical calculations of the isotope coefficient $\alpha_\chi=-d\log\chi/d\log M$, where M is the ion mass, as a function of the adiabatic ratio ω_0/E_F and of λ. Both the noncrossing (dashed lines) and the vertex-corrected (solid lines) theories predict negative values of α_χ. Compared to the noncrossing data, the vertex-corrected results show for small values of Q_c a stronger dependence on ω_0/E_F and λ, leading to α_χ of about -0.1. The observation of the isotope effect, which should be absent in metals fulfilling the ME framework, could be therefore stringent evidence of a nonadiabatic electron-phonon coupling. Note that a previous analysis of experimental data of some superconducting properties has demonstrated the failure of the ME theory for Rb_3C_{60}, pointing out the breakdown of Migdal's theorem in

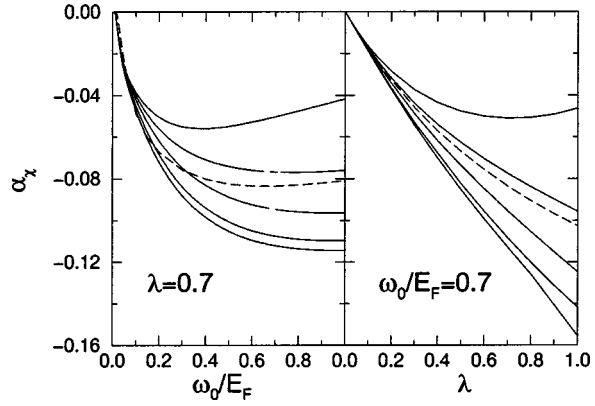

FIG. 6. Isotope effect on the spin susceptibility as a function of ω_0/E_F ($\lambda=0.7$, $N_0I=0.4$) and as function of λ ($\omega_0/E_F=0.7$, $N_0I=0.4$). Solid lines and dashed line as in previous figures.

fullerides. In this respect, a measurement of α_χ in C_{60} compounds would represent a direct and independent test of the relevance of the nonadiabatic electron-phonon interaction.

From a qualitative point of view, the zero-temperature behaviors of both the noncrossing and vertex-corrected theories give substantially similar results. An interesting differentiation among the two approximations arises in the temperature dependence of χ. In Fig. 7 we report the calculated temperature dependence of the Pauli susceptibility for different values of ω_0/E_F and λ. The noncrossing approximation (dashed lines) yields a monotone decreasing of χ as the temperature T increases. This is basically due to the phonon cutoff in the ladder equation for the susceptibility, Eq. (19). On the other hand, the vertex-corrected χ (solid lines) has a richer temperature dependence which becomes more important as $\lambda\omega_0/E_F$ increases. Starting from $T=0$, the basic feature is represented by an initial increase of χ with T followed by a decreasing for larger temperatures. Although the decreasing part is rather Q_c independent, the initial increase of

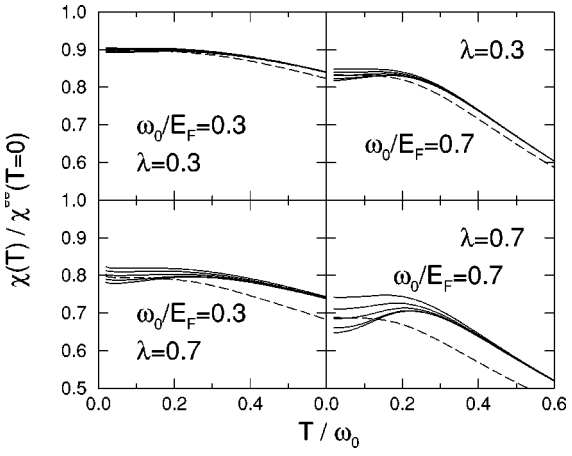

FIG. 7. Temperature dependence of the spin susceptibility for different values of ω_0/E_F and λ and $N_0I=0.4$. Solid lines and dashed line as in previous figures.

χ is steeper for lower values of the momentum cutoff. Note, for example, that for $\lambda=0.7$, $\omega_0/E_F=0.7$, and $Q_c=0.1$, at $T/\omega_0\simeq 0.2$ the susceptibility is enhanced by a 8%–9% of its value at $T=0$. Although this increase is rather small, it is nevertheless an interesting feature since it is not related to any increase of the lattice constant due to thermal expansion (the calculations reported here in fact are done for constant volumes). Experimentally, in fact, χ at $T\simeq 300$ K is found to be larger than its value at $T\sim 25$ K of $\sim 30\%$ and $\sim 40\%$ for K_3C_{60} and Rb_3C_{60}, respectively.[17,18] Moreover, recent data suggest a following decrease of χ in K_3C_{60} by increasing temperature.[19] The initial increase of χ with temperature is currently explained by a temperature enhancement of the density of states at the Fermi level due to the thermal expansion of the unit cell. Note, however, that a power law dependence of the density of state on the lattice constant is not sufficient to reproduce the observed increase of χ, while a stronger dependence like an exponential law needs a quite small value of the Stoner enhancement, $(1-N_0I)^{-1}\simeq 1.3$, to fit the experimental data of χ.[18] Such small Stoner enhancement is in contrast to recent Monte Carlo calculations which estimate $(1-N_0I)^{-1}\simeq 3$.[20] Although a detailed study of the effect of the thermal expansion on χ is beyond the scope of this paper, the results of Fig. 7 suggest that the electron-phonon contribution could be an additional source for the temperature dependence of χ. Of course, firmer evidence of the role of nonadiabaticity would be the measurement of the temperature dependence of χ for a constant sample volume, in the spirit therefore of the constant volume resistivity experiment of some years ago.[21]

An alternative explanation of the nonmonotone temperature dependence of χ has recently been attributed to possible effects of mobile ions K^+.[19] This argument would predict, however, a finite isotope effect on χ by alkali isotopic substitution and no carbon isotope effect whereas in nonadiabatic theory an opposite trend is expected.

V. CONCLUSIONS

In this paper the nonadiabatic theory of the Pauli spin susceptibility in narrow-band systems has been formulated in order to identify possible signatures of nonadiabatic electron-phonon coupling in fullerides. We have identified peculiar features that can be considered as hallmarks of a relevant electron-phonon coupling *in the nonadiabatic regime*. In particular, an effective reduction of the spin susceptibility by the nonadiabatic electron-phonon coupling has been found in contrast to the conventional ME framework where no electron-phonon renormalization is expected. In addition, we predict a finite negative isotope effect on χ which we suggest as a possible experimental test. The Pauli spin susceptibility of χ also acquires in nonadiabatic regime an anomalous temperature dependence in qualitative agreement with the experimental data.

The results of the present work are particularly interesting in the framework of the theory of nonadiabatic superconductivity, where the same nonadiabatic interferences, at the origin of the electron-phonon renormalization of χ, open new

attractive channels in the Cooper pairing and play therefore a positive role with respect to the superconducting onset. In this perspective the simultaneous effect of the nonadiabatic channels on the Cooper pairing and on the Pauli susceptibility in fullerenes is worth further investigation. Nonadiabatic effects could be, for instance, responsible for the anomalous T_c vs χ dependence in ammoniated alkali-doped fullerides.[22–24]

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