Pauli susceptibility of nonadiabatic Fermi liquids

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Abstract. – The nonadiabatic regime of the electron-phonon interaction leads to behaviors of some physical measurable quantities qualitatively different from those expected from the Migdal-Eliashberg theory. Here we identify in the Pauli paramagnetic susceptibility \(\chi\) one of such quantities and show that the nonadiabatic corrections reduce \(\chi\) with respect to its adiabatic limit. We show also that the nonadiabatic regime induces an isotope dependence of \(\chi\), which in principle could be measured.

When the Fermi energy \(E_F\) is anomalously small, as in high-\(T_c\) cuprates \(^1\) and in the fullerene compounds \(^2\), the Migdal-Eliashberg (ME) approach \(^3\) may result inadequate in describing the interplay between charge carriers and phonons. For example, the alkali-doped fullerenes \((A_3C_{60})\) have Fermi energies of order 0.25 eV \(^3\) and intramolecular phonon modes with frequencies \(\omega_0\) in the range between 20 meV and 0.2 eV \(^5\). In this case, the adiabatic parameter \(\omega_0/E_F\) lies somewhere between 0.1 and 0.9, depending on which phonon modes most couple to the electrons. The main consequence is that the electron-phonon vertex corrections may no longer be negligible, as assumed in the ME framework, and a generalization of the theory is required to include the nonadiabatic contributions \(^6\).

This generalization in terms of the electron-phonon coupling \(\lambda\) and the adiabatic parameter \(\omega_0/E_F\), the ME regime applies for \(\lambda \lesssim 1\) and \(\omega_0/E_F \ll 1\). Therefore, a generalization beyond the ME framework is required when \(\lambda \gtrsim 1\) and/or \(\omega_0/E_F\) is no longer negligible. However, when \(\lambda\) is larger than some critical value \(\lambda_c\) (which is of order one or larger), the system evolves toward a polaronic regime characterized by strong electron-lattice correlations. This holds true even in the adiabatic case in which the charge carriers acquire large effective masses. On the other hand, a region in the \(\lambda-\omega_0/E_F\) plane different from the one leading to polaronic states is defined by \(\lambda \lesssim 1\) and \(\omega_0/E_F\) finite. Within this region, where the charge carriers are weakly interacting nonadiabatically with phonons, the nature of quasiparticles is different from both the ME and the polaronic ones. In such a nonadiabatic regime we shall speak of nonadiabatic Fermi liquids (or nonadiabatic fermions), to stress the difference from the ME and
the polaronic limits. In practice, such a regime can be described by a perturbative approach where $\lambda \omega_0 / E_F$ plays the role of the small parameter of the theory. Various comparisons with exact results (for the one electron case) and quantum Monte Carlo calculations point toward the reliability of such a perturbative description.

At the zeroth order in $\lambda \omega_0 / E_F$, the nonadiabatic theory coincides with the ME limit while for finite values of $\lambda \omega_0 / E_F$ the nonadiabatic fermions display anomalous behaviors. In this situation, several properties are modified and a very important question regards the possibility to observe some fingerprints of such a nonadiabatic regime. Furthermore, in order to be considered as possible evidences, such fingerprints should be searched among those physical quantities for which some well established property in the ME regime results qualitatively modified in the nonadiabatic one. In order to clarify this statement, let us consider for example the electron-phonon renormalized charge carrier mass $m^*$. In the ME regime $m^* = (1 + \lambda)m$ [10], where $m$ is the bare mass and $\lambda$ is the electron-phonon coupling. Since $\lambda$ is independent of the ion-mass [11], no isotope effect is expected for $m^*$. However, when the nonadiabatic contributions are no longer negligible, $m^*$ acquires an ion-mass dependence which leads to a non-zero isotope coefficient $\alpha_{m^*}$ [12]. The effective mass $m^*$ represents therefore a clear example of a quantity for which a well established property in the ME regime ($\alpha_{m^*} = 0$) is drastically modified in the nonadiabatic one ($\alpha_{m^*} \neq 0$). So far, strong evidences for isotope-dependent $m^*$ have been reported for YBa$_2$Cu$_3$O$_{6+x}$ and La$_{2-x}$Sr$_x$CuO$_4$ [13] and theoretical calculations have shown that already the inclusion of the first nonadiabatic vertex correction to the ME limit provides values of $\alpha_{m^*}$ with sign and order of magnitude in agreement with those estimated by the experiments [12].

Another property typical in the ME regime which is instead strongly altered by the nonadiabatic contributions is the non-magnetic impurity dependence of the critical temperature $T_c$ of an homogeneous s-wave superconductor. For a conventional superconductor, weak disorder does not influence the critical temperature as stated by Anderson’s theorem [14]. On the contrary, since the electron-phonon vertex corrections are very sensitive to the amount of disorder, the critical temperature of a s-wave nonadiabatic superconductor can be strongly lowered by the impurities [14]. Such a peculiar behavior is also accompanied by an anomalous impurity dependence of the isotope coefficient of $T_c$. So far, reduction of $T_c$ driven by disorder for s-wave superconductors has been reported for K$_4$C$_{60}$ [15] and Nd$_{2-x}$Ce$_x$CuO$_4$ [16].

In this paper we consider another measurable quantity which could be considered as a test for the breakdown of Migdal’s theorem: the Pauli paramagnetic susceptibility $\chi$. Here, the characteristic feature in the ME regime ($\omega_0 / E_F \ll 1$) is that the electron-phonon interaction does not renormalize the Pauli susceptibility so that $\chi$ is independent of $\lambda$ and $\omega_0$ [10]. In the ME regime therefore $\chi \equiv \chi_P = \mu_B^2 N(0)$, where $\mu_B$ is the Bohr magneton and $N(0)$ is the electron density of states at the Fermi level. In principle, therefore, a measure of $\chi$ via for example electron paramagnetic resonance (EPR) is unaffected by the electron-phonon interaction and provides an estimate of the electronic density of states $N(0)$ which however is renormalized by many-electrons effects (Stoner enhancement) [11].

The interesting aspect of $\chi$ is that, as we show below, when $\omega_0 / E_F$ is no longer negligible $\chi$ acquires a phonon renormalization and becomes dependent on both $\lambda$ and $\omega_0$. This result can be of importance for two reasons. First, it leads to re-consider the estimates of the electron density of states obtained by EPR measurements, since these estimates have been based on the phonon-independent ME form of $\chi$. Second, and more importantly, the nonadiabatic renormalization of $\chi$ induces a non-zero isotope effect which, in principle, could be measured.

To evaluate the Pauli susceptibility we make use of the static limit of the Kubo formulæ [18].

\[^{(1)}\] In the present discussion we shall consider the many-electrons effects as being already contained in $N(0)$. 

\[ \chi(T) = \lim_{q \to 0} \mu_B^2 \int_0^\beta d\tau \langle T_\tau S_z(q, \tau) S_z(-q, 0) \rangle, \]  

where \( \beta \) is the inverse temperature \( T \) and

\[ S_z(q) = \sum_{k, \sigma = \pm 1} \sigma c_{k+q, \sigma}^\dagger c_{k, \sigma}, \]

where \( c_{k, \sigma}^\dagger \) (\( c_{k, \sigma} \)) is the creation (annihilation) operator for electron with momentum \( k \) and spin direction \( \sigma = \pm 1 \).

In what follows, we shall focus on the evaluation of Eq.(1) for a system of electrons interacting with phonons through the coupling \( g(q) \). In terms of electron and phonon Green’s functions, Eq.(1) reduces to the following general expression:

\[ \chi(T) = \lim_{q \to 0} \mu_B^2 T \sum_m \sum_k G(m, k + q) G(m, k) \Gamma(k + q, k; m), \]

where \( \omega_m = (2m + 1)\pi T \) and

\[ G(m, k) = \left[ i\omega_m - \epsilon(k) - \Sigma(m, k) \right]^{-1}, \]

is the Green’s function for an electron with dispersion \( \epsilon(k) \) and electron-phonon self-energy \( \Sigma(m, k) \). In Eq.(3), \( \Gamma(k + q, k; m) \) is the irreducible electron-phonon vertex function which is given by all diagrams which cannot be separated into two different parts by cutting a single electron or phonon propagator line. The reducible part of the vertex function gives in fact zero contribution when the summation over the spin index is performed in eqs.(1-2) [18].

In this paper we compute eq.(3) by employing a self-consistent calculation which amount to evaluate \( \Sigma(m, k) \) in the non-crossing approximation. For dispersionless phonons with frequency \( \omega_0 \), we consider therefore the electron-phonon self-energy as given by:

\[ \Sigma(n, k) = T \sum_{m, k'} g(k - k')^2 \frac{\omega_0^2}{(\omega_n - \omega_m)^2 - \omega_0^2} G(m, k'). \]

In the above equation we have implicitly assumed that the phonons are already renormalized and that \( \omega_0 \) is a dressed phonon frequency. In a conserving approach, the vertex function resulting from the non-crossing approximation for \( \Sigma(m, k) \) is given by all the ladder contributions. Therefore the vertex function satisfies the following ladder equation:

\[ \Gamma(k + q, k; n + m, n) = 1 + T \sum_{m, k'} g(k - k')^2 \frac{\omega_0^2}{(\omega_n - \omega_m')^2 + \omega_0^2} G(m', k') G(m' + m, k' + q) \]
\[ \times \Gamma(k' + q, k'; m' + m, m'). \]

Actually, from eq.(3), to evaluate \( \chi \) we only need to retain the static limit of eq.(6) which is given by setting first \( \omega_n = 0 \) and after \( q = 0 \). As already shown in Refs. [7], if we exchange the order of the two limits, the resulting dynamical limit of the vertex will be in general different from the static one. Therefore setting \( \omega_n = 0 \) and \( q = 0 \) in both hand sides of eq.(3) may give
a non well defined result because in that point the vertex in non-analytic. However, as we shall show below, the computing procedure we employ in handling the vertex function automatically provides the correct static limit by simply setting \( \omega_m = 0, q = 0 \) in eq.\( \text{(6)} \), regardless of the order of the two limits. Therefore, by setting \( \lim_{q \to 0} \Gamma(k + q, k; n, n) = \Gamma_s(k, n) \), the static limit of eq.\( \text{(6)} \) reduces to:

\[
\Gamma_s(k, n) = 1 + T \sum_{m'k} g(k - k')^2 \frac{\omega_0^2}{(\omega_n - \omega_{m'})^2 + \omega_0^2} G(m', k')^2 \Gamma_s(k', m').
\]

Without loss of generality, the solution of the set of equations \( \text{(3)}, \text{(4)} \) and \( \text{(7)} \) can be found by using a structureless electron-phonon interaction \( g(q) \equiv g^2 \). The resulting self-energy is then momentum independent and for a system with a half-filled electron band of constant DOS over the entire bandwidth \( 2E_F \), the self-energy can be written as \( \Sigma(n) = i\omega_n - iW_n \), where

\[
W_n = \omega_n + \lambda \pi T \sum_m \frac{\omega_0^2}{(\omega_n - \omega_m)^2 + \omega_0^2} \frac{2}{\pi} \arctan \left( \frac{E_F}{W_m} \right),
\]

is the renormalized electron frequency and \( \lambda = g^2N(0) \) is the electron-phonon coupling. Within the same approximation scheme, \( \Gamma_s(k, n) \) becomes momentum-independent and the resulting vertex function \( \Gamma_s(n) \) satisfies the following equation:

\[
\Gamma_s(n) = 1 - \lambda T \sum_{m'} \frac{\omega_0^2}{(\omega_n - \omega_{m'})^2 + \omega_0^2} \frac{2E_F}{W_{m'}^2 + E_F^2} \Gamma_s(m').
\]

We can verify that the above equation gives indeed the static limit of the vertex by neglecting the renormalization of the frequency, \( W_{m'} \to \omega_{m'} \), and by performing the zero temperature limit. In this way, to the first order in \( \lambda \) and at zero external frequency, Eq.\( \text{(6)} \) becomes:

\[
\Gamma_s(0) = 1 - \lambda \int \frac{d\omega}{2\pi} \frac{\omega_0^2}{\omega^2 + \omega_0^2} \frac{2E_F}{\omega^2 + E_F^2} = 1 - \lambda \frac{\omega_0}{\omega_0 + E_F}.
\]

\( \Gamma_s(0) \) coincides therefore with the static limit already calculated in the perturbation theory. [3]

We are now in the position to evaluate the Pauli susceptibility. Since both the self-energy and the vertex function are independent of the momentum, equation \( \text{(6)} \) can be analytically integrated over the energy and the final expression for \( \chi(T) \) reduces to:

\[
\chi(T) = \chi P T \sum_m \frac{2E_F}{E_F^2 + W_m^2} \Gamma_s(m),
\]

where \( \chi_p = \mu_B^2N(0), \) and \( W_m \) and \( \Gamma_s(m) \) are the solution of equations \( \text{(3)} \) and \( \text{(4)} \), respectively.

We solve the set of equations \( \text{(8)}, \text{(9)} \) and \( \text{(10)} \) for a temperature \( T/\omega_0 = 0.02 \) and different values of \( \lambda \) and \( \omega_0/E_F \). The frequency summations appearing both in the self-energy \( \text{(8)} \) and in the vertex function \( \text{(9)} \) is truncated at the frequency cut-off \( \omega_c = (2N + 1)\pi T \) with \( N = 400 \) corresponding to \( \omega_c \approx 50\omega_0 \). The solutions of \( \text{(8)} \) and \( \text{(9)} \) are then calculated by iteration and the results are plugged into eq.\( \text{(10)} \). The high-frequency part \( (\omega_m > \omega_c \gg \omega_0) \) of the summation in eq.\( \text{(10)} \) is calculated by setting \( W_m = \omega_m \) and \( \Gamma_s(m) = 1 \), since in this
high-frequency region the contribution from the electron-phonon coupling is negligible. The
procedure outlined above permits to estimate the zero temperature susceptibility \( \chi \) also for
the smallest value of \( \omega_0 / E_F \) we used in the calculations (\( \omega_0 / E_F = 0.01 \)).

In Fig. 1 we show the zero temperature calculated Pauli susceptibility as a function of the
adiabatic parameter \( \omega_0 / E_F \) and for different values of the electron-phonon coupling constant
\( \lambda \). When \( \omega_0 / E_F \) \( \rightarrow \) 0, \( \chi \) approaches its free-electron value \( \chi_P \), irrespectively of the value of
\( \lambda \) and we recover therefore the result of the ME theory. Instead, when \( \omega_0 / E_F \) is larger than
zero, \( \chi \) becomes dependent of \( \lambda \) and results to be always lowered with respect to \( \chi_P \). In Fig. 2
\( \chi / \chi_P \) is plotted as a function of the electron-phonon coupling \( \lambda \) for different values of \( \omega_0 / E_F \).
For small values of \( \omega_0 / E_F \), \( \chi / \chi_P \) decreases almost linearly with \( \lambda \). The main result of our
calculations is therefore that \( \chi(0) / \chi_P < 1 \) as soon as \( \omega_0 / E_F > 0 \). Preliminary calculations
including higher orders vertex corrections confirm this feature.

The reduction of the Pauli susceptibility induced by the electron-phonon interaction when
\( \omega_0 / E_F \) is finite requires to re-consider the estimates of the electron density of states based on
EPR measurements [19, 20]. In these estimates, in fact, the measured \( \chi \) is fitted with the ME
expression of the susceptibility

\[
\chi \propto N(0) \sim \frac{N_0(0)}{1 - I},
\]

where in the last equality we have explicitly separated \( N(0) \) into the free-electron form \( N_0(0) \)
and the Stoner enhancement \( 1/(1 - I) \) given by the many-electrons effects. Theoretical estima-
tions of \( 1/(1 - I) \) permit therefore to obtain \( N_0(0) \) from the experimental \( \chi \) [21]. However,
this procedure may systematically underestimate \( N_0(0) \) if \( \omega_0 / E_F \) is no longer negligible like in
the fullerene compounds. In fact, in view of the previous results, \( N_0(0) \) of eq. (12) should be
replaced by \( N_0^*(0) \simeq N_0(0) f(\lambda, \omega_0 / E_F) \), where the function \( f \) takes into account the phonon
renormalization effects and is less than the unity. From the calculations showed in Figs. 1
and 2, \( f \) can be as small as \( \sim 0.8 - 0.7 \), leading to an underestimation of the bare density of
states \( N_0(0) \) of \( \sim 20 - 30\% \).

Another remarkable feature of the nonadiabatic phonon renormalization is the lattice in-
duced isotope effect on \( \chi \). From Fig.1 in fact it is obvious that a change in frequency \( \omega_0 \)
induces a lowering of \( \chi \). Such a change of \( \omega_0 \) can be induced by isotope substitution leading
therefore to a non-zero value of the isotope coefficient:

\[
\alpha_\chi = - \frac{d \log \chi}{d \log M} = \frac{1}{2} \frac{d \log \chi}{d \log(\omega_0 / E_F)},
\]

where \( M \) is the ion mass and, in the last equality, we have used \( \omega_0 \propto (M)^{-1/2} \) (note that
in the nonadiabatic regime \( \chi \) depends also on \( \lambda \), however \( \lambda \) is independent of \( M \)). In Fig. 3
we show the numerical evaluation of eq. (13) as a function of \( \omega_0 / E_F \) and for different values of
\( \lambda \). As expected, the resulting isotope coefficient \( \alpha_\chi \) vanishes at the adiabatic limit. However,
for nonzero values of \( \omega_0 / E_F \), it becomes negative and for ordinary values of \( \lambda \) can be of
order \(-0.05\). This is a rather small value, nevertheless it provides a clear indication of
nonadiabaticity. It would be extremely interesting to investigate experimentally the presence
or the absence of an isotope effect on \( \chi \) in the fullerene compounds. The outcome of such
kind of experiment could provide us with an estimate of \( \omega_0 / E_F \) and therefore of the degree of
nonadiabaticity in such narrow band materials.

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Fig. 1. – $\omega_0/E_F$ dependence of the Pauli susceptibility $\chi$ for different values of the electron-phonon coupling constant $\lambda$.

Fig. 2. – Pauli susceptibility $\chi$ as a function of the electron-phonon coupling constant $\lambda$ for different values of the adiabatic parameter $\omega_0/E_F$. 
Fig. 3. – Isotope coefficient $\alpha_\chi$ of the Pauli susceptibility for different values of the electron-phonon coupling $\lambda$. 

$\omega_0/E_F$