From local to macroscopic coherence in systems with composite quasi-particles

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INTRODUCTION

Particles which locally strongly interact with their environment polarize this latter and finish up in self-trapped states. On a local level, the environment surrounding the particles can be described in terms of a coherent superposition (in form of Glauber states) of the elementary bosonic excitations of the bare environment. As a result, new composite entities form, given by the charge carriers and their surrounding clouds of bosonic excitations of the environment. It is at present a matter of dispute whether in the normal state such composite entities can exist as itinerant Bloch-like states or are purely diffusive. The retarded interaction between the charge carriers and the bosonic excitations of the environment favors the picture of diffusive motion caused by a dephasing of the dynamics of the two constituents.

The question we want to address here is whether this process of dephasing can be blocked, leading to a delocalization when such self-trapped composite entities condense into a macroscopic coherent quantum state. We investigate such a possibility in terms of a model of very dense into a superfluid state. Our interest here is to show how in such a macroscopic quantum state the internal structure of the local coherent quantum state of the bosonic excitations of the environment is modified.

We shall investigate this problem on the basis of a model of itinerant electrons, coupled to some purely local vibrational modes. For moderately strong and strong coupling, it results in self-trapped composite entities, given by on-site electron pairs, surrounded by local lattice deformations which are described by clouds of phonons in local coherent quantum states. In the intermediate coupling limit these localized entities can be considered as two-particle resonant states of the underlying system of itinerant electrons. They form out of pairs of such uncorrelated itinerant electrons and ultimately decay into them.

THE BOSON-FERMI MODEL

A model Hamiltonian which captures such physics is given by the boson-fermion model (BFM)

\[ H_{\text{BFM}} = (D - \mu) \sum_{i,\sigma} n_{i\sigma} - t \sum_{\langle ij \rangle,\sigma} c_{i\sigma}^\dagger c_{j\sigma} \]

\[ + (\Delta_B - 2\mu) \sum_i \left( \rho_i^+ \frac{1}{2} + \frac{1}{2} \right) + \nu \sum_i \left[ \rho_i^+ c_{i\downarrow}^\dagger c_{i\uparrow}^\dagger + \rho_i^- c_{i\uparrow}^\dagger c_{i\downarrow}^\dagger \right] \]

\[ - \hbar \omega_0 \alpha \sum_i \left( \rho_i^+ \frac{1}{2} \right) (a_i + a_i^+ \frac{1}{2}) + \hbar \omega_0 \sum_i \left( a_i^+ a_i^\dagger + \frac{1}{2} \right), \]

(1)

Here \( \rho_i^\pm \) denote the creation and annihilation operators of hard-core bosons (characterizing the self-trapped localized electron-pairs) on some effective sites \( i \), which should be understood as of being made up of small clusters of...
atoms rather than single atomic sites. Such hard-core bosons have spin-$\frac{1}{2}$ commutation relations $[\rho_i^+, \rho_i^-]_\pm = 2\rho_i^\pm$ and $[\rho_i^-, \rho_i^+]_\pm = \delta_{ij}$. $c_i^{(+)}$ denote the fermionic operators referring to annihilation (creation) of itinerant electrons with spin $\sigma$, $n_{i\sigma} = c_i^{\dagger \sigma} c_i\sigma$ being the number operator for such fermions. $a_i^{(+)}$ denote annihilation (creation) operators of the excitations of the environment, related to the local lattice displacements $X_i = (a_i + a_i^\dagger) / \sqrt{2M\omega_0/\hbar}$ where $M$ is some atomic mass characterizing the effective sites, and $\omega_0$ is the frequency of the local deformations of the lattice. The bare hopping integral for the itinerant electrons is given by $t$, the bare electron half bandwidth by $D = zt$ (which will serve as energy unit), $z$ denoting the coordination number. $\alpha$ denotes the coupling of the hard-core bosons to the deformations of the surrounding medium and $v$ the exchange coupling between the bosons and the pairs of itinerant fermions. The bare boson energy level is denoted by $\Delta_B$. The chemical potential $\mu$ is common to fermions and bosons such as to guarantee overall charge conservation.

The model defined by the Hamiltonian (1) was introduced shortly after the proposal of the concept of bipolaronic superconductivity [1] (appropriate to the extreme strong coupling anti-adiabatic regime) in an attempt to extend this concept to the regime of intermediate electron-lattice deformation coupling. This model is based on a conjecture, rather than on a derivation from an underlying basic electron-phonon Hamiltonian, such as for instance the Holstein model [2].

We shall in the following develop the ideas which had initially led us to this conjecture. Let us start from a simple Holstein model with purely local Einstein modes for the lattice vibrations. The corresponding Hamiltonian is given by

$$H = (D - \mu) \sum_{i\sigma} n_{i\sigma} - t \sum_{i\neq j, \sigma} (c_i^{\dagger \sigma} c_j\sigma + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow} - \bar{\alpha} \hbar \omega_0 \sum_{i,\sigma} n_{i\sigma}(a_i + a_i^\dagger) + \sum_i \hbar \omega_0 \left(a_i^\dagger a_i + \frac{1}{2}\right). \quad (2)$$

The meaning of the various operators and coupling constants is the same as in the Hamiltonian (1), except for the value of $\bar{\alpha}$ which is equal to $\frac{1}{4}\alpha$.

The electron-phonon interaction with coupling strength $\bar{\alpha}$ is taken to be local, in view of the interaction with primarily local phonon modes which favours small polaron formation. $U$ denotes some effective Coulomb repulsion - not to be confused with a Hubbard-type on-site interaction - on the effective sites, which, we stress again, ought to be considered as being composed of small molecular clusters such as a diatomic molecular unit or similar. Electrons described by this Hamiltonian can gain energy either by becoming itinerant and remaining essentially uncoupled to the lattice or, alternatively, getting self-trapped. In the first case the energy gain is described by the intrinsic band dispersion

$$\varepsilon_k = D(1 - \gamma_k) \quad \gamma_k = \frac{1}{z} \sum_\delta e^{ik\delta}, \quad (3)$$

$\delta$ denoting lattice vectors linking nearest-neighbor sites. For self-trapped polaronic states, the local part of the Hamiltonian (2) can be transformed by a shift in the phonon variables $a_i^{(+)} \to a_i^{(+)} + \bar{\alpha}(n_{i\uparrow} + n_{i\downarrow})$ upon which it acquires the form

$$H_{loc} = (D - \mu - \varepsilon_p) \sum_{i\sigma} n_{i\sigma} - (2\varepsilon_p - U) \sum_i n_{i\uparrow} n_{i\downarrow} + \sum_i \hbar \omega_0 \left(a_i^\dagger a_i + \frac{1}{2}\right), \quad (4)$$

with the polaron binding energy $\varepsilon_p = \bar{\alpha}^2 \hbar \omega_0$ denoting the gain in energy by the corresponding polaron level shift. The effective on-site interaction given by $(2\varepsilon_p - U)$ controls the possible formation of bipolaronic states

$$\rho_i^+ X_i^+ \equiv c_i^{\dagger \uparrow} c_i^{\dagger \downarrow} e^{-2i\delta(a_i - a_i^\dagger)} \quad (5)$$

when $2\varepsilon_p - U > 0$, with the single polaron states lying above such bipolaron states. For $2\varepsilon_p - U < 0$ we have the inverse situation with the bipolaronic level lying above that of the single polarons. What we are interested here is the first case.

Let us begin by considering the situation of sufficiently strong interaction such that the bipolaronic level lies below the bottom of the unrenormalized electron band, i.e., for $2\varepsilon_p - \frac{1}{4}U > D$, as illustrated in Fig. 1. In that case bipolarons can acquire itinerancy by decaying into virtual states of pairs of itinerant electrons and reassembling subsequently on some neighbouring site. Within the usual Lang-Firsov approximation, neglecting phonon creation as well as annihilation processes during the charge transfer from one site to the other, the corresponding effective bipolaron hopping integral is then given by

$$t^* \simeq t^2 e^{-2\alpha^2} / (2\varepsilon_p - \frac{1}{4}U - D).$$

Within this approximation one obtains a system of itinerant bipolarons on a lattice which, at low temperatures, can condense into a superfluid state, giving rise to what has been termed bipolaronic superconductivity. This theoretical proposition has ever since remained a matter of dispute, questioning if such a normal-state itinerant Bose liquid can be achieved in real materials. The main arguments against it is that the stability of local bipolarons requires a relatively high value of $\alpha^2 \geq (D + \frac{1}{4}U) / 2\hbar \omega_0$ for realistic values of $D$, $U$ and $\omega_0$ and hence leads to exceedingly small values of the bipolaron bandwidth. A further hampering factor for the realization of such bipolaronic itinerant band states is related to the retardation effects between the motion of the charge carriers and the lattice deformations which surround them. This gives rise to dephasing between the two, as shown by exact diagonalization approaches [3] and DMRG studies [4], indicating...
that incoherent processes in the optical conductivity are dominant. All these results suggest that coherent bipolaron motion is most unlikely to occur in real systems and it was for that reason that the Boson-Fermion model was initially proposed[3]. The aim was to capture the physics outside the regime of the extreme strong coupling antidiabetic limit and to describe the intermediate coupling case, where the bipolaron level lies inside the band of the itinerant electrons (see Fig. 2) and where the superconducting state is controlled by the phase correlations of these resonant bosonic bipolaron states.

Quite generally, this model can be considered as a paradigm for cross-over situations. For the specific case we are interested here, it describes a cross-over between a BCS-type superconductor of Cooper pairs in the weak coupling regime and the hypothetical, yet to be experimentally verified, *bipolaronic superconductivity* in the strong coupling regime. In a totally different physical situation, namely that of a strongly correlated electron system such as described by the Hubbard model, it has been recently shown that the intermediate coupling case can be mapped onto such a Boson-Fermion scenario. Such an effective Boson-Fermion Hamiltonian was also derived recently for the exchange interaction between spinon singlets of the RVB electron-pairs and holons[4]. The question to what extent such a BFM can emerge from a quite general class of interacting fermion systems, has been addressed from the point of view of a bosonization procedure of an intrinsically fermionic system. The presently much discussed molecular Bose-Einstein condensate, involving entangled atoms in squeezed states has also recently been analyzed[5] in the framework of such a BFM.

A derivation of the BFM for moderately strongly coupled electron-phonon systems, starting from the Holstein model, is so far not available. In order to provide the physically intuitive picture which initially had led us to propose this model, let us consider the case where the local bipolaron states are stable relative to single polarons. This eventuality can only be decided on the basis of direct experimental evidence for such local bipolaronic entities. The classical cases which initially had led to consider such a situation are systems like amorphous semiconductors[6], Ti$_4$O$_7$[7], Na$_x$V$_2$O$_5$[8], WO$_{3-x}$[9], and double valence fluctuation systems[10] like PbTe(Tl), just to name a few of them, and all of which represent such bipolarons as exclusively localized self-trapped states.

When these systems are doped, the electrons responsible for pairing are at the same time responsible for the - albeit diffusive - transport. It is hence tempting to hypothesize that such bipolaronic states can occur as resonant states inside a band of itinerant electrons. In more complex systems, such as ternary and quaternary compounds, different electrons come into play. Some of them are prone to form bipolarons and others will remain itinerant, but being coupled together via some hybridization term. In systems like that, such a Boson-Fermion scenario imposes itself more directly. It is the eventuality of bipolaronic resonant states rather than localized states, which was at the origin of our proposal of the Boson-Fermion scenario[3].

The level scheme for the local bipolaronic states inside the band of the bare itinerant electrons for this situation is depicted in Fig. 2. Filling up such a system gradually with electrons, it is evident from Fig. 2 that, as long as the Fermi level remains below the level of the local bipolaronic states, the charge carriers will be essentially bare itinerant electrons. But upon further increasing the concentration of charge carriers, the itinerant electrons will eventually overlap in energy with the intrinsically localized bipolaronic level.

The major supposition leading to this BFM is now to assume that this overlap can be simulated by some effective on-site hybridization describing a charge exchange between the bipolarons and pairs of itinerant electrons in the form of

$$v \sum_{i} \left[ \rho_i^+ X_i^+ c_{i\uparrow} c_{i\downarrow} + \rho_i^- X_i^- c_{i\uparrow}^+ c_{i\downarrow}^+ \right], \quad (6)$$

with the itinerant electrons and the localized bipolarons,
Eq. (5), treated as commuting fields. Such a supposition is supported by the fact that the spectrum of electrons, moderately strongly coupled to local modes, is known to be given by the bare spectrum for all k-vectors, except where their energy crosses the level of localized self-trapped electrons. In this regime of energies the electron spectrum is strongly modified, as shown by exact diagonalization studies [10, 11] as well as variational methods [12], with the spectrum becoming rather flat and approaching the local level of the self-trapped polaronic states. These are features which are characteristic of systems where itinerant charge carriers are hybridized with localized charge carriers, and this was the center of our initial assumption of an effective hybridization between localized bipolarons and pairs of itinerant electrons, as given by Eq. (6).

On a local level the dynamics of a polaronic system, involving two-particle resonant states, is controlled by the atomic limit of such a system, which we thus propose to describe by the following Hamiltonian

\[ H_{at} = (-4\varepsilon_p + U - 2\mu) \sum_i \left( \rho_i^+ \frac{1}{2} \right) + v \sum_i \left[ \rho_i^+ X_i^+ c_i^\dagger c_i^\gamma + \rho_i^- X_i^- c_i^\gamma^+ c_i^\dagger \right] + \sum_i \hbar \omega_0 \left( a_i^+ a_i + \frac{1}{2} \right) \]

with a bare bipolaron level given by \( \Delta_B = U \).

The equivalent formulation of this Hamiltonian [the second part of Eq. (7)] is obtained after employing an inverse Lang-Firsov-type transformation \( H_{at} \rightarrow U + H_{at} U \) with \( U = e^{\alpha(a_1^\gamma - a_1^\gamma^\dagger)(\varepsilon_1^p + \frac{1}{2})} \). Adding to this local Hamiltonian the term describing the kinetic energy of the itinerant electrons, leads immediately to the BFM Hamiltonian given by Eq. (1).

**COMPETITION BETWEEN LOCAL AND GLOBAL COHERENCE**

**THE PSEUDOGAP PHASE**

The basic physics contained in this BFM model originates from a competition of the local electron pairing, acting as a prerequisite for superconductivity, with the delocalizing effect of the itinerant electrons which favors non-local Cooper pairing as the temperature is lowered and the superconducting phase is approached. The change-over from local pairing to Cooper pairing is not only manifest in the electronic properties of such systems but is expected to be accompanied by corresponding lattice deformations, the study of which is the issue of the present paper.

We shall examine this feature for the two following extreme limits:

i) the normal-state phase, characterized by the existence of a pseudogap in the density of states (DOS) of the itinerant electrons, which will be treated here in an approximate fashion within the atomic limit of the BFM,

ii) the superconducting phase, which will be treated on a mean-field level, taking into account the phase locking of the bound electron pairs.

Throughout this study we choose as a representative example a set of parameters presenting a situation close to the fully symmetric case. This implies an effective bipolaron level (corresponding to the energy level for the bound pairs) given by \( \Delta_B - 4\varepsilon_p \simeq 0 \), resulting in a final state bipolarons level lying close to the center of the band of the itinerant electrons, after the boson-phonon coupling has been switched on. We moreover want to consider the case \( n_{tot} = 2n_F + n_B = 2 \), giving a concentration of electrons \( n_F \simeq 0.5 \) and a concentration of bound pairs \( n_B \simeq 0.5 \) and choose \( v = 0.25 \), \( \omega_0 = 0.1 \) and \( \alpha = 2 \).

Let us start from the purely local physics, described by the atomic limit (\( t = 0 \)) of the BFM corresponding to the Hamiltonian \( H_{at} \), Eq. (7). This limit has been studied by us previously [13] in connection with the pseudogap phenomenon and the incoherent background in the electron spectral function, which, in such a scenario, arise from phonon shake-off processes. For the choice of parameters adopted here, the physics controlling the change-over from the normal into the superconducting phase can essentially be represented by the one-, two- and three-particle eigenstates of \( H_{at} \)

\[ |1, l\rangle_{\sigma} = c_{\sigma}^+ |0\rangle |0\rangle_l > \]  \hspace{1cm} (8)  

\[ |2, l\rangle_{\sigma} = \sum_n \left( u_{\sigma}^{at} c_{\sigma}^+ c_{\sigma}^\dagger + u_n^{at} \rho^+ \right) |0\rangle |0\rangle |n\rangle > \] \hspace{1cm} (9)  

\[ |3, l\rangle_{\sigma} = \sum_n c_{\sigma}^+ \rho^+ |0\rangle |0\rangle \frac{(a^+ - \alpha)^l}{\sqrt{l!}} e^{-\frac{1}{2} \alpha^2} \frac{\alpha^n}{\sqrt{n!}} |n\rangle |10\rangle \] \hspace{1cm} (10)

with \( |i, j\rangle_{\sigma} \) denoting the \( i \)-particle \( j \)-th eigenstate. The opening of the pseudogap at a certain temperature \( T^* \) shows up in the rapid spectral weight increase, with de-
increasing temperature below \( T^\ast \), of the scattering amplitude involving transitions from the lowest energy two-particle bonding state \( |2,0\>^a_t \) to the lowest energy single-particle states \( |1,0\>^a_t \) and from the lowest energy three-particle states \( |3,0\>^a_t \) to \( |2,0\>^a_t \). These contributions to the DOS of the itinerant electrons are manifest by the wings (see Fig. 3) for energies below and above that of the DOS of the itinerant electrons are manifest by the wings of the DOS, thus resulting in the opening of a pseudogap. Taking into account the dispersion of the itinerant electrons in an approximate fashion, such as within a Coherent Potential Approximation (CPA) approach\[19\], the main features seen in Fig. 3 are maintained, simply leading to a smearing of the DOS.

The two-particle eigenstates of \( H_{at} \), Eq. (9), are obtained by expanding the local oscillator states in terms of the eigenstates \( |n\>_i \) of the undeformed harmonic oscillator at a given site. \( |0\> \) and \( |0\> \) denote respectively the vacuum states of the itinerant electrons and of the bound electron-pairs on such a site. The coefficients \( u_{in}^a \) and \( v_{in}^a \) are determined by exact diagonalization of this atomic limit problem within a restricted Hilbert space of those excited oscillator states. It is generally sufficient to take into account up to 50 such states in order to get converging results.

The qualitative changes in the lattice deformations expected to occur when going from the pseudogap phase into the superconducting phase are essentially described by the lowest energy one-, two- and three-particle states, which contain the information on the relative weight of the bound electron-pairs \( (v_{in}^a)^2 \) and of the induced pairing in the subsystem of the itinerant electrons \( (u_{in}^a)^2 \). The first ones are intrinsically coupled to the local lattice deformations while the second ones acquire such local lattice deformations due to the charge exchange mechanism acting between the two. As we shall see below, the efficiency of this transfer of polaronic features is noticeably different in the normal and in the superconducting phase. It shows up in the composition \( P(n) \) of the coherent phonon states describing the local phonon clouds surrounding the charge carriers on a given site \( i \), which in the normal state (atomic limit) is given by

\[
P_{at}(n) = \frac{1}{Z} \sum_{m,l} \exp(-\beta E_{ml}^{at}) \{m,l|a^+a|m,l\}^{at}, \tag{11}
\]

\( Z \) denoting the partition function and \( E_{ml}^{at} \) the eigenvalues of \( H_{at} \) associated with the eigenstates \( |m,l\>^{at} \). Upon decreasing the temperature, \( P_{at}(n) \) shows a redistribution of weight from high to small values of the phonon number \( n \) [see Fig. 4(a)]. In the temperature regime of interest here, i.e., from slightly above \( T^\ast \) all the way down to \( T=0 \), the contributions to \( P(n) \) come essentially from the lowest energy one-, two- and three-
particle states, Eqs. (8-10). As illustrated in Fig. 5, these various contributions have noticeably different features as far as the phonon distributions are concerned. The one-particle states are totally decoupled from the lattice and hence have a contribution only for $n = 0$, while the three-particle contribution are maximally coupled to the lattice showing a broad peak in this distribution which corresponds to that of a shifted oscillator ground state. The two-particle bonding state $|2, 0\rangle^{at}$ has features of a displaced oscillator which is particularly noticeable at low temperatures where it is the main contribution, together with a much reduced contribution from the two-particle antibonding state $|2, 1\rangle^{at}$. That latter contribution to $P(n)$, considered without thermal weighting factor, clearly shows strong contributions for low values of $n$, which is indicative of weak correlations between the electronic degrees of freedom and those of the lattice. The onset of the local coherence between bound pairs and of the induced pairing in the itinerant subsystem can be tracked in the pair distribution function (PDF) $g(X)$, given in the normal state (atomic limit) by

$$g_{at}(X) = \frac{1}{Z} \sum_{m,l} e^{-\beta E_{m,l}^{at}} \delta(X - X_i)[m,l]^{at}_i,$$

which describes the probability of finding a local deformation of size $X$ and which can be studied by EXAFS and pulsed neutron scattering experiments[20]. $g_{at}(X)$ is obtained by expressing the eigenstates of $H_{at}$ in terms of a real-space representation involving the harmonic oscillator wavefunctions. In Fig. 6(a) we illustrate the temperature evolution of the PDF. Its decomposition into the contributions arising from the one-, two- and three-particle states given in Eqs. (8-10) are illustrated in Fig. 7. The one- and three-particle states contribute to the PDF in form of peaks centered around $X = 0$ and around some finite value of $X$, respectively. This expresses the fact that the one-particle states (8) are completely decoupled from the lattice, while the three-particle states (10) contain a bound electron-pair and thus give a maximal shift in the PDF. The two-particle bonding and anti-bonding states, Eq. (9) with $l = 0, 1$, give contributions to the PDF with two peaks lying very close together, which is a result of the strong correlation between the bound electron pairs and the induced pairs in the itinerant electron system, characterizing the normal-state phase. At high temperatures, the PDF is thus given by a very broadened smeared out double-peak structure. But as the temperature is lowered below $T^*$, local coherence between the bound electron pairs and the pairs of the itinerant electrons sets in. As a result, the opening of the pseudogap in the local density of states is accompanied by a change-over in the PDF from this very smeared out double-peak structure to a rather well defined single-peak structure. The latter is essentially due to the lowest energy bonding two-particle eigenstate contributing to a PDF with a maximum at some finite value of $X$. This is a signature of the strong correlations, which gradually build up as we go below $T^*$, between the

FIG. 5: Decomposition, for different temperatures, of $P(n)$ in the normal state (full squares) into contributions coming from the one-, two- and three-particle states ($P_1(n)$, $P_2(n)$ and $P_3(n)$, respectively). $P_2(n)$ and $P_3(n)$ represent the contributions coming respectively from the lowest energy two-particle state (bonding) and from the first excited two-particle state (antibonding).

FIG. 6: Comparison of the PDF as a function of the displacement $X$, measured by the dimensionless parameter $\xi = X/\sqrt{M\omega_p/\hbar}$, in the atomic limit and in the superconducting phase for different $T$. 

$\xi$
time between bound pairs and pairs of itinerant electrons will
When the system undergoes a transition into a superfluid
each other via the hopping term of the itinerant electrons.
and let them interact with
tutions coming respectively from the two lowest bonding and
butions coming from the coefficients
and antibonding two-particle eigenstates of $H_{at}$.
bound pairs and the induced pairing in the itinerant electron subsystem leading to roughly equal weight coming from the coefficients $u_{\mu n}^{\ast}$ and $v_{\nu n}^{\ast}$.

THE SUPERCONDUCTING PHASE

Let us now put such single units, as described by $H_{at}$, together in an infinite lattice, and let them interact with each other via the hopping term of the itinerant electrons. When the system undergoes a transition into a superfluid state of the charge carriers, the on-site correlations between bound pairs and pairs of itinerant electrons will be weakened, since the $k$-space pairing of the itinerant electrons enters into competition with the strong on-site pairing which controls the physics in the normal state. As a result, upon entering the superconducting phase, a substantial modification of the coherent phonon states of the individual units will occur.

We shall, for a moment, make the assumption that the deformation of the medium surrounding the bosonic charge carriers is not influenced by boson-fermion charge fluctuations. In that case the coupling of the bound electron-pairs to the lattice deformations can be eliminated, along the lines usually adopted in studies of the polaron problem, leaving us with an effective Hamiltonian given by the first four terms of the Hamiltonian (1) and with a renormalized hybridization coupling $\nu e^{-a^2/2}$. Such a Hamiltonian has been studied in detail within different schemes, such as self consistent diagrammatic techniques [21], dynamical mean-field theory [22] and renormalization group procedures [23], and gives rise to the following physics. As the temperature is lowered below a certain characteristic temperature $T^*$, pairing of the itinerant electrons sets in, leading to an emptying out of the single electron spectrum near the chemical potential and thus to the opening of a pseudogap. Upon further decreasing the temperature these electron-pairs become itinerant [24], eventually condensing into a macroscopic superfluid ground state of the form

$$\prod_i [u_i e^{-i\phi_i/2} + v_i e^{+i\phi_i/2} \rho_i^+ ] |0\rangle_i$$  \hspace{1cm} (13)

($u_i$, $v_i$ being some model dependent parameters) and exhibiting soundwave-like collective excitations as a consequence of the rigidity of the local phases $\phi_i$.

In order to study how such a superfluid state affects the surrounding lattice deformations, we now investigate this problem on the basis of a mean-field treatment of the full Hamiltonian with the coupling of the phonons to the bound pairs explicitly taken into account. To this purpose, we introduce the following order parameters for the charge carriers [25]

$$x = \frac{1}{N} \sum_i \langle c_i^+ c_i \rangle, \quad \rho = \frac{1}{N} \sum_i \langle \rho_i^+ \rho_i^- \rangle$$  \hspace{1cm} (14)

which we assume to be homogeneous in space. The corresponding mean field Hamiltonian is given by:

$$H_{\text{MFA}} = H_F + H_B - \nu px + \frac{\hbar \omega_0}{2}$$

$$H_F = (D - \mu) \sum_{i,\sigma} c_{i\sigma}^+ c_{i\sigma} - t \sum_{\langle i \neq j \rangle, \sigma} c_{i\sigma}^+ c_{j\sigma} + \frac{\nu \rho}{2} \sum_i [c_{i\uparrow}^+ c_{i\downarrow} + c_{i\downarrow}^+ c_{i\uparrow}]$$

$$H_B = (\Delta_B - 2\mu) \sum_i \left( \rho_i^+ + \frac{1}{2} \right) + \nu x \sum_i [\rho_i^+ + \rho_i^-] - \hbar \omega_0 \alpha \sum_i \left( \rho_i^+ + \frac{1}{2} \right) (a_i^+ + a_i^-) + \hbar \omega_0 \sum_i a_i^+ a_i.$$  \hspace{1cm} (15)
On the level of this approximation, the effect of the charge fluctuations between bound pairs and pairs of itinerant electrons is diminished, being taken into account in a global, spatially independent, fashion. The itinerancy of the intrinsically localized bound electron pairs, induced by the exchange coupling \[21, 23\] is not contained in the present approximation and therefore we cannot account for collective soundwave-like excitations and the spatially correlated lattice deformations which result from them. Nevertheless, the present study does permit to investigate local excitations of the lattice, controlled by the local fluctuations of the order parameters to which they are coupled, and which can be related to a number of experiments.

Considering itinerant bosons on a deformable lattice with weak boson-phonon coupling \[23\], leads to a hybridization of the Bogoliubov modes (coming from the repulsive interaction between the bosons) and the Einstein-like phonon modes. At long wavelengths this results in a modified sound velocity of the Bogoliubov mode, with little spectral weight carried by the phonons, and a moderately renormalized Einstein mode, principally of phonon character. In such a context, what the present mean-field study permits us to do, is to investigate the effect of the superfluid state on such an Einstein-like mode. Due to the strong coupling between the bound electron pairs and the phonons, such an Einstein mode will undergo a substantial renormalization, as it will be shown below.

The fermionic part \(H_F\) of \(H_{MF}\), being dynamically decoupled from the bosonic part \(H_B\) and hence from the phonons, can be cast into the standard form \(H_F = \sum_k \xi_k(\rho) (\alpha_k^+ \alpha_k + \beta_k^+ \beta_k)\). The \(\alpha\)’s and \(\beta\)’s denote the operators of Bogoliubov quasi-particles having the dispersion \(\xi_k(\rho) = \pm \sqrt{(\epsilon_k - \mu)^2 + (\nu \rho)^2 / 4}\) which differs from the bare electron dispersion \(\epsilon_k\) by showing a gap of size \(\nu \rho\). The corresponding eigenstates of \(H_F\) are given by the standard BCS expression

\[
|\Psi^{BCS}\rangle = \prod_k \left[ u_k + v_k \epsilon_{k\uparrow}^+ \epsilon_{-k\downarrow}^+ \right] |0\rangle .
\]  

The bosonic part \(H_B\) of \(H_{MF}\), showing intrinsic coupling between the bosonic charge carriers and the phonons, is diagonalized in terms of a set of states of the form

\[
|l\rangle_i^{sc} = \sum_n \left[ u_{ln}^{sc}(i) \right | \psi_i^{sc}(n) \rangle |n >_i .
\]  

having eigenvalues \(E_i^{sc}(x)\). The eigenstates of the mean-field Hamiltonian, Eq. (15), are thus given by

\[
|\Psi^{BCS}\rangle \otimes \prod_i |l\rangle_i^{sc} .
\]  

As in the atomic limit, the diagonalization of \(H_B\) is carried out in a truncated Hilbert space spanned by the eigenstates \(|n >\) of the undeformed harmonic oscillator. We ultimately arrive at the following set of self-consistent equations determining the temperature dependence of the order parameters

\[
x = -\frac{\nu \rho}{4N} \sum_k \frac{1}{\xi_k(\rho)} \tan \frac{\beta \xi_k(\rho)}{2} ,
\]

\[
\rho = \frac{1}{N} \sum_n u_{ln}^{sc} v_{ln}^{sc} \exp \left[ -\beta E_i^{sc}(x) \right] ,
\]

\[
Z = \sum \exp \left[ -\beta E_i^{sc}(x) \right] \text{ denoting the partition function corresponding to } H_B .
\]

This set of equations must be solved with the constraint that the total density of bosons and fermions remains fixed, i.e. \(n_{tot} = n_F + 2n_B + \frac{1}{2} \rho^2\), where

\[
n_F = \frac{1}{N} \sum_{k\sigma} \langle c_{k\sigma}^+ c_{k\sigma} \rangle = \frac{1}{N} \sum_k \left( 1 - \frac{\xi_k}{\xi_k(\rho)} \tan \frac{\beta \xi_k(\rho)}{2} \right)
\]

\[
n_B = \frac{1}{2} + \frac{1}{N} \sum_i \langle \rho_i^2 \rangle
\]

\[
= \frac{1}{2} + \frac{1}{2Z} \sum_{ln} \left[ (u_{ln}^{sc})^2 - (v_{ln}^{sc})^2 \right] \exp \left[ -\beta E_i^{sc}(x) \right] .
\]  

Let us now examine the modification of the composition of the local coherent phonon states on the individual lattice sites (discussed above on the basis of the atomic limit for the normal state) when a macroscopic phase-coherent state of the charge carriers emerges and the bound electron-pairs condense into a superfluid state. The probability distribution of the phonons in the superconducting phase is then described by

\[
P_{sc}(n) = \frac{1}{Z} \sum_i \exp \left[ -\beta E_i^{sc}(x) \right] \left[ (u_{ln}^{sc})^2 + (v_{ln}^{sc})^2 \right] .
\]  

The evaluation of \(P_{sc}(n)\) in the superfluid phase follows closely the calculation for \(P_{at}(n)\) in the atomic limit. In fact, the secular equation determining the coefficients \(u_{ln}^{sc}\) and \(v_{ln}^{sc}\) is formally the same as in the atomic limit, with the states \(c_{ln}^{sc}|0 > \otimes |n >\) and \(|0 > \otimes \rho^+|0 > \otimes |n >\) spanning the Hilbert space being now replaced by \(|0 > \otimes |n >\) and \(|\rho^+ > \otimes |n >\), respectively. The only modifications which then appear in this set of equations are (i) a renormalized hybridization constant \(\tilde{\nu} \equiv \nu x\), instead of \(\nu\) in the atomic limit, and (ii) an energy level for the diagonal elements involving the states \(|0 > \otimes |n >\) being equal to \(\hbar \omega_n\), instead of \(2(D - \mu) + \hbar \omega_n\) for the diagonal elements involving the states \(c_{ln}^{sc} \otimes |0 > \otimes |n >\). This renormalization of \(\nu\) is important since it reduces the bare \(\nu\) by roughly an order of magnitude (for the set of parameters chosen throughout this paper), expressing the fact that the superfluidity leads to a considerable reduction of the on-site correlation between the configurations with bound electron-pairs present and with such pairs being absent. This has significant consequences on the composition of the local phonon clouds surrounding the individual sites and thus on their corresponding PDFs.
Just below the superconducting $T_c$ ($\simeq 0.0067$ in our present case), $P_{sc}(n)$ is determined by an incoherent superposition of essentially the two lowest eigenstates of the mean-field Hamiltonian, Eq. (15), one giving rise to a broad peak at some finite $n$ and the other to a sharp maximum at $n = 0$. At low temperatures, however, the ground state alone is sufficient to describe $P_{sc}(n)$. The contribution to $P_{sc}(n)$ leading to a peak at some finite value of $n$ (associated with the coefficients $v^{sc}_{0n}$) arises from the deformation induced by the presence of the bound electron pairs, while the contribution to $P_{sc}(n)$ leading to a maximum for $n = 0$ (associated with the coefficients $u^{sc}_{0n}$) arises from the absence of such an induced deformation for configurations where no bound pairs are present, with the oscillators being essentially undeformed. The coherent superposition of deformed and undeformed oscillator states in the ground state is a direct consequence of the macroscopic phase locking of the composite bosonic states in the superconducting phase, resulting from the variational ground state of $H_B$ of the form

$$\prod_i \left( e^{-i\phi_i/2} \bar{X}_i^- + e^{i\phi_i/2} \bar{X}_i^+ \right) |0\rangle_i \otimes |0 >_i . \quad (22)$$

The operators $\bar{X}_i^\pm$ are defined as

$$\bar{X}_i^- |0 >_i = \sum_n u^{sc}_{0n}(i) |n >_i \quad (23)$$
$$\bar{X}_i^+ |0 >_i = \sum_n v^{sc}_{0n}(i) |n >_i \quad (24)$$

and differ significantly from the usual polaron shift operators $X_i^\pm = e^{\mp \alpha (a_i^+ - a_i^-)}$, characterizing the deformations of a simply shifted oscillator state. Macroscopic phase locking and homogeneity imply that the coefficients $u^{sc}_{0n}(i)$ and $v^{sc}_{0n}(i)$ as well as phases $\phi_i$ are the same for all sites. Eq. (22) is a direct generalization of the superfluid ground state of a system of hard-core bosons on a lattice, which is of the form given in Eq. (13), and thus shows explicitly the locking together of the charge ($\rho_i^+$) and lattice ($\bar{X}_i^\pm$) degrees of freedom on a local level. We shall next address ourselves to the question of how such a macroscopic coherent quantum state of the phonons can be tracked experimentally and, in particular, what difference there is to be expected with respect to the normal state features of the PDF, examined in the previous section. The PDF in the superconducting phase is given by

$$g_{sc}(X) = \frac{1}{Z} \sum_i e^{-\beta E^{sc}_i(x)} \langle \delta(X - X_i) | l \rangle^{sc}_i . \quad (25)$$

In Fig. 8 we compare the PDF in the normal state and in the superconducting phase for different temperatures. The features examined above on the basis of the composition of $P_{sc}(n)$ are perfectly reflected in the PDF. In the normal state one observes a single peak at some finite $X$, indicative of well defined shifted oscillator states. In the superconducting phase, on the contrary, one observes a weakly temperature dependent double-peak structure, which, as we can see from Fig. 8, arises essentially from two contributions associated with the ground state and the first excited state of $H_B$. At temperatures just below $T_c$ this double-peak structure results from an incoherent superposition of these two states, corresponding to a situation where localized bipolarons are randomly distributed over the lattice. The statistical distribution of empty and doubly occupied sites will thus give rise to
that double-peak structure. In the limit of low temperature, on the contrary, the PDF arises exclusively from the ground state, with a peak centered near \( X = 0 \) (being determined by the coefficients \( u_{0}^{n} \)) and a peak centered around a finite value of \( X \) (being determined by the coefficients \( v_{0}^{n} \)). The appearance of a double-peak structure in the PDF has long since been recognized as a signature of delocalization of the charge carriers in polaronic systems\(^{[27]}\). The present study shows that, as the superconductive state sets in and is strengthened upon lowering the temperature, the macroscopic phase coherence of the system not only involves the charge carriers but also the deformations which surround them. This is only possible if at \( T = 0 \) these deformations are controlled by a single quantum state, i.e., the ground state \( \prod_{i} | 0 \rangle_{i}^{sc} \). The superconductivity-induced delocalization of the self-trapped entities which characterize the normal state, is caused by the locking together of the local lattice deformations in a coherent superposition of deformed and essentially undeformed oscillator states, resulting in the development of a double-peak structure in the PDF. At least in the scenario presented here, this result does not bear out any undressings\(^{[24]}\) of the charge carriers, stripping off partially the phonon clouds, as one enters the superconducting phase. Such an effect would show up in a PDF which, in the superconducting phase, would be given by essentially a single-peak structure centered around \( X = 0 \).

### DYNAMICAL LATTICE PROPERTIES

The discussion in the previous Section demonstrated the qualitative difference which exists between some basic lattice properties in the normal and the superconducting phase, and largely being controlled by the induced electron pairing in the subsystem of the intrinsically uncorrelated itinerant electrons. What results is a competition between the local pairing, characterizing the normal-state phase, and the \( k \)-space pairing in the superconducting phase, expected to be particularly relevant around the superconducting transition where the spatial phase fluctuations play a major role and residual local electron pairing\(^{[23]}\) persists in the superconducting phase. The qualitative difference in the structure of the PDF in the two phases leads us to expect squeezing effects of the local coherent phonon quantum states which are stronger in the pseudogap phase than in the superconducting phase. Associated with it, one should expect larger fluctuations of the local deformations of the environment and of the momenta of the ions representing it. These fluctuations are given by

\[
\Delta X_{i}^{2} = \langle X_{i}^{2} \rangle - \langle X_{i} \rangle^{2} , \quad \Delta P_{i}^{2} = \langle P_{i}^{2} \rangle \quad (26)
\]

with

\[
X_{i} = \sqrt{\frac{\hbar}{2 M \omega_{0}^{i}}} (a_{i}^{+} + a_{i}) \\
P_{i} = i \sqrt{\frac{\hbar M \omega_{0}^{i}}{2}} (a_{i}^{+} - a_{i}) . \quad (27)
\]

The expectation values in Eq. (27) have to be calculated with respect to the eigenvectors of \( H_{at} \) and \( H_{B} \) for, respectively, the normal and the superconducting phase. The results for these fluctuations in the two phases are illustrated in Fig. 11. In particular, the product \( \Delta X_{i}^{2} \Delta P_{i}^{2} \) clearly shows that the coherent phonon states in the normal state are considerably squeezed, i.e., this product is for \( T \to 0 \) very close to the lower limit of the Heisenberg uncertainty principle, equal to \( \hbar^{2}/4 \). In the superconducting phase, on the contrary, we observe practically no squeezing effect with decreasing temperature, the product \( \Delta X_{i}^{2} \Delta P_{i}^{2} \) being always bigger than what would be expected in the normal state in this temperature regime. A possible experimental verification of these features comes from neutron absorption measurements\(^{[15]}\) which measure the kinetic energy \( E_{kin} = \hbar^{2} \langle P_{i}^{2} \rangle / 2M \) of the local lattice vibrations. Indeed, the experimental results shown that, as the temperature is reduced, \( E_{kin} \) saturates in the superconducting phase and is distinctly above the values expected in the normal state at the same temperature. This fact is born out in our study, as can be seen by inspection of Fig. 11(b). Finally, an issue which ought to be of crucial interest in studying the lattice properties connected with the onset of the superconducting order or of the electron pairing at \( T^{*} \) in the normal state, is the question of the changes in the phonon spectral properties upon going from one phase to the other. Strong changes in the frequency of specific local modes, induced by strong electron-phonon coupling, have been well established by photoinduced infrared absorption measurements\(^{[30]}\) and been related to strong local displacements of the ions. In this context, the appearance of new additional modes occurring with changes from the normal into the superconducting phase have also been reported, although this is still a matter of debate\(^{[31, 32]}\). More established are the strong intensity changes of certain Raman active modes and the strong frequency renormalizations and lifetime broadening occurring at such transitions\(^{[33]}\). Such effects can best be studied by energy loss spectra in inelastic neutron scattering or Raman spectroscopy where phonons are emitted. The cross-section for it is given by

\[
A_{emi}(\omega) = \int dt \, e^{i \omega t} A_{emi}(t) \\
= \frac{1}{Z} \sum_{l,m} e^{-\beta \epsilon_{l}} | < l | a | m > |^{2} \delta(\omega + \epsilon_{m} - \Delta) \quad (28)
\]

with

\[
A_{emi}(t) = < a(0) a^{+}(t) > \theta(-t) . \quad (29)
\]
FIG. 10: Comparison of (a) the positional and (b) the momentum fluctuations of the lattice in the normal (N) and in the superconducting (SC) phase as a function of relative temperature $T/T_0$ ($T_0 = \{T_c, T^*\}$) and with $\lambda^2 = (1/2)M_0^2\omega_0^2\varepsilon_p$. (c) Degree of the squeezing of the coherent phonon states as a function of $T/T_0$.

Here $\varepsilon_l = \{E^{at}_l, E^{sc}_l(x)\}$ and $|l> = \{|l>^{at}, |l>^{sc}\}$ are the eigenvalues and eigenvectors of the Hamiltonians $H_{at}$ and $H_B$, respectively. We illustrate the results of such phonon emission spectra for the two phases in Figs. 11 and 12. Quite generally, one observes a quasi-elastic contribution at frequency zero, which is the standard signature of coherent phonon states. As the temperature is lowered below either $T^*$ or $T_c$ and the itinerant electrons get correlated by local pairing in the normal state or by Cooper pairing in the superconducting phase, the local phonon mode first splits into several modes and finally evolves into a well defined mode which is softer in the pseudogap phase and harder in the superconducting phase. These features of the phonon spectral properties are expected to hold qualitatively true. A quantitative link-up between the normal phase and the superconducting phase as the temperature is decreased, would require a full description of the local boson-fermion exchange correlations, incorporating the electron itinerancy in the normal state and allowing for local electron pairing in the superconducting phase. These are presently largely unresolved fundamental problems in such system where amplitude and phase order occur independently, as in the high-temperature superconductors with its pseudo-gap phase above $T_c$ where local electron pairing persists in an incoherent fashion.

FIG. 11: Evolution of the phonon spectrum in emission experiments as a function of temperature in the normal state.

FIG. 12: Evolution of the phonon spectrum in emission experiments as a function of temperature in the superconducting phase.

CONCLUSIONS

The present study is an attempt to examine the condensed state of a system of composite particles such as charge carriers surrounded by clouds of bosonic excitations of the neighboring environment to which they are coupled and to see to what extent a macroscopic coherent quantum state can be induced in the system of these
bosonic excitations. As a specific example we chose a system of electrons, moderately strongly coupled to local lattice deformations, which can be described in terms of resonant local-pair states (localized bipolarons) inside a Fermi sea of essentially uncorrelated itinerant electrons with which they interact via a charge exchange term. We find noticeable qualitative differences in the structure of the local coherent quantum states of the phonons in the superconducting phase and in the phase above it which corresponds to a normal state characterized by a pseudogap in the DOS of the single-particle spectrum. Several measurable quantities related to static and dynamic lattice properties were examined, such as the pair distribution function, the fluctuations in the lattice positions and the corresponding ion kinetic energies of the atoms making up such a lattice and, finally, the phonon spectral properties given by energy-loss spectroscopy (measurable by inelastic neutron scattering or Raman spectroscopy). The difference between local electron pairing in the normal state and k-space pairing in the superconducting phase has noticeable consequences on such measurable quantities, resulting in (i) a single-peak structure of the PDF in the normal state and a double-peak structure in the superconducting phase, (ii) distinctively larger positional fluctuations and kinetic energy of the atomic clusters (making up the lattice) in the superconducting phase than in the normal state and, (iii) associated with it, a more effective squeezing of the phonon coherent quantum states in that latter. For the superconducting phase, the present mean-field study is restricted to local phase fluctuations of the superconducting state given by a generalization of a phase-locked state of bosons on a lattice, Eqs. (17, 18, 22), in which these bosons correspond to composite particles in form of bipolarons [see Eq. (5)]. The examination of soundwave-like collective excitations of such a system is planned for some future work. There, the question arises if the local dynamical deformations do get spatially correlated upon entering the superconducting state, as possibly suggested by ion channeling experiments. A possible approach to solve this problem could be to integrate out the fermionic sector of the BFM Hamiltonian Eq. (1), resulting in a Gross-Pitaevskii Lagrangian for the effective itinerant bosonic charge carriers coupled to the phonons, followed by a treatment similar to that adopted for bosons on a defromable lattice.

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