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CHARGE DENSITY WAVE AND SUPERCONDUCTIVITY: LOCALIZED BIPOLARONS VERSUS EXTENDED COOPER PAIRS?

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The electron-phonon coupling in conducting materials may produce at low temperature either a charge density wave (CDW) or standard superconductivity (SC). We analyse the limits of validity of these two kinds of theories which up to now are not well-defined on the particular HOLSTEIN model (where the non-interacting electrons are described by a tight binding model and are coupled to a dispersionless optic mode by on-site electron-phonon coupling). In the adiabatic limit (no quantum lattice fluctuations), we prove rigorously at any dimension that when the electron-phonon coupling is large enough, the ground-state and the first excited states can be described as a superlattice of bipolarons pinned to the lattice which forms the CDW. Then, the low temperature thermodynamics of the electron-phonon system can be described by an Ising spin Hamiltonian (lattice gas model). When the quantum lattice fluctuations are taken into account, this Hamiltonian becomes a quantum model with spins 1/2. For smaller electron-phonon coupling, we argue that the CDW with gapless phasons (or FROHLICH modes usually considered in the litterature) are unstable against quantum lattice fluctuations. A superconducting phase may then appear. The conjectured phase diagram at 0K thus involves either superconducting phase or pinned bipolaronic structures (CDW).

1-Introduction

At low temperature, the electron-phonon coupling is known to be responsible of very different physical phenomena in systems with conducting electrons, such as standard Superconductivity (SC) and Charge Density Waves (CDW). The standard theory of superconductivity (BCS)[1] assumes first that the electron-phonon coupling which is treated as a perturbation, can be eliminated by a canonical transformation and replaced by an effective attractive electron-electron interaction. Next, it is shown that the collective pairing of electrons with opposite spins takes place and forms the BCS superconducting state through an optimized variational form excluding other types of instabilities. The quantum character of the phonons is essential in this theory because the electron-electron attraction originates by the exchange of one or few quanta of phonon energy only.

By contrast, the standard theories of CDW[2] neglect the quantum character of the phonons. Then, it is shown that in some conditions which are favored by the low

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dimensionality of the systems, the energy cost due to a periodic lattice distortion which raises the degeneracy of the electrons at the Fermi surface ("nesting") may become negative. The system then exhibits a CDW associated to a periodic lattice distortion (PLD).

These two theories are based on different assumptions which seems to be incompatible. Thus, in order to get a better understanding of the competition between CDW and SC, it is useful to first focus our attention on one of the simplest models (Holstein model). We analyse the stability of the CDW structures against quantum fluctuations of the lattice. The aim of this talk is to shed a new light on this problem through a new approach based on previous works on incommensurate and chaotic structures in PEIERLS models[4] which is currently under development.

2-Definition of the HOLSTEIN Hamiltonian

The Holstein Hamiltonian is the sum of three terms

$$H = H_k + H_{ep} + H_p \tag{1}$$

H_k is the Hamiltonian of a single band of non-interacting electrons (within a tight binding approximation)

$$H_k = -T \sum_{<i,j>,\,\sigma} c^+_{i,\sigma}\, c_{j,\sigma} \tag{2-a}$$

where T is the exchange constant between neighboring sites <i,j> on a d-dimensional square lattice and σ is the electron spin $\pm\frac{1}{2}$ noted $\uparrow$ or $\downarrow$. $c^+_{i,\sigma}$ and $c_{i,\sigma}$ are the creation and annihilation Fermions operators of an electron at site i with spin σ respectively. The band width is thus 4T d.

H_p is the Hamiltonian of quantum phonons corresponding to a dispersionless optical branch

$$H_p = \sum_i \hbar\omega_0 \left(a^+_i a_i + \frac{1}{2} \right) \tag{2-b}$$

where a^+_i and a_i are the creation and annihilation boson operators of phonons at site i respectively. The on-site electron-phonon interaction with constant g is represented by the Hamiltonian

$$H_{ep} = g \sum_i n_i \left(a^+_i + a_i \right) \tag{2-c}$$

the electronic density operator at site i is

$$n_i = c^+_{i,\uparrow} c_{i,\uparrow} + c^+_{i,\downarrow} c_{i,\downarrow} \tag{2-d}$$

In this model, two dimensionless independant parameters can be defined. They are:

$$\gamma = \frac{\hbar\omega_0}{T} \tag{3-a}$$

which measures the "quantum character" of the phonons and

$$(3\text{-b}) \qquad k = \frac{2g}{\sqrt{\hbar \omega_0 T}}$$

which is the reduced electron-phonon coupling constant in the classical phonon limit. By setting, the position operator

$$(4\text{-a}) \qquad u_i = \frac{\sqrt{\gamma}}{2} \, (a_i^+ + a_i)$$

and its conjugate operator (the commutator is $[u_i, p_i] = i$)

$$(4\text{-b}) \qquad p_i = \frac{i}{\sqrt{\gamma}} \, (a_i^+ - a_i)$$

the initial Hamiltonian (1) becomes in unit of energy 2T:

$$(5) \quad \hat{H} = \frac{H}{2T} = -\frac{1}{2} \sum_{<i,j>\,\sigma} c^+_{i,\sigma} c_{j,\sigma} + \frac{k}{2} \sum_i n_i u_i + \frac{1}{2} \sum_i (u_i^2 + \frac{\gamma^2}{4} p_i^2)$$

It is useful first, to analyse the limit cases γ small (adiabatic limit) and γ large (antiadiabatic limit) and next to try to understand the crossover between these two limits.

3-Adiabatic Limit

For most real systems, the phonon energies with typical energies $\hbar \omega_0$ are of the order of magnitude of several hundred degrees K. These energies are much smaller than the typical band energies T by factors 100 or more.

The adiabatic limit is obtained by setting γ=0 in (5). Standard theories of CDW use this adiabatic approximation. As we shall see in the next, this approximation is different of the Born-Oppenheimer approximation which takes partially the quantum lattice fluctuations into account. Thus, the adiabatic solution may be *qualitatively* modified by quantum lattice fluctuations even for very small values of γ.

For γ=0, $\{u_i\}$ can be considered as scalar variables in (5) instead of being an operator. Then, the creation operator of an electron in the eigenstates μ and spin σ is:

$$(6\text{-a}) \qquad c^+_{\mu,\sigma} = \sum_i \Psi^i_\mu \, c^+_{i,\sigma}$$

where $\{\Psi^i_\mu\}$ fulfills the eigen equation

(6-b) $$- \sum_{j:<i,j>} \Psi_\mu^j + k\, u_i \Psi_\mu^i = E_\mu(\{u_i\})\, \Psi_\mu^i$$

depends on the variables $\{u_i\}$. (The sum in (6-b) $\sum_{j:<i,j>}$ is done for the nearest neighbouring sites j of i). For the ground-state and the low-lying excitations of this model, there are two electrons with opposite spins which occupy the eigenstates with energy smaller than the Fermi energy E_F. Then the energy (5) is a function $\Phi(\{u_i\}))$ of the scalar variables $\{u_i\}$

(6-c) $$\Phi(\{u_i\})) = \frac{1}{2} \sum_i u_i^2 + \sum_{E_\mu<E_F} E_\mu(\{u_i\})$$

The ground-state of the system is obtained by minimizing this variational form. When this functional has many local minima with energy close to the energy of the ground state, these states which are metastable, corresponds to the low-lying excitations which determine the low-temperature behavior of the system. In one dimension, this model has been studied and it has been shown that the ground-state exhibits a Transition by Breaking of Analyticity (TBA).

4-The Transition by Breaking of Analyticity in One-dimensional Peierls Models

The ground-state of (6-c) exhibits a periodic lattice distortion (PLD) with wave-vector $2k_F$ (k_F is the Fermi wave-vector) associated with a CDW. When the electronic band filling (number of electron pairs per site) is an irrational number ζ, the CDW-PLD state is degenerate with respect to the variation of its phase α. Then, it has been observed that this model exhibits at zero K, a transition by breaking of analyticity (TBA) as a function of the electron-phonon coupling constant k. This TBA was found to be perfectly similar to those of the Frenkel-Kontorowa (FK) model (with the same critical behavior). But, this last model has the advantage that the existence of the TBA and of many of its characteristics can be analysed *rigorously* [4,9].

Let us briefly recall here, the main characteristic of the TBA in this Holstein model. When k is smaller than some critical value $k_c(\zeta)$, the coordinates of the incommensurate CDW-PLD ground-state depend analytically on the phase ("analytic state"), and in addition this state is phase *undefectible* (or charge undefectible). This means that there exists no local minima of (6-c) with the same band filling ζ, not belonging to the family of incommensurate ground-states with arbitrary phase α. For example, if one adds to the system a *localized spinless* pair of electrons, it cannot remain localized in a metastable state and extends over the whole system. In that regime, the CDW phonon excitations which correspond to phase fluctuation are gapless and form the well-known phason branch.

(However, let us remark that taking into account the pair breaking of the electrons, the CDW-PLD ground-state is spin-defectible, which means that that this state can accept a localized defect with a spin $\pm\frac{1}{2}$ as in the well-known SSH model for polyacethylene. But in that case, there is no chaotic states with spins when the system is charge undefectible).

When k is larger than the critical value $k_c(\zeta)$, the coordinates of the incommensurate CDW-PLD depend discontinuously on the phase ("non-analytic state"), and then this state is phase *defectible* (or charge defectible). The functional energy (6-c) has infinitely many metastable states (local minima) with the same band filling ζ, other than the incommensurate ground-states. Most of these states are chaotic with a finite entropy. In that situation, the ground-state accepts *localized charge defects* and these configurational excitations are *gapless* while the phonon phase excitations have a finite gap (unlike the "analytic" regime). In this regime, the CDW and its metastable states can be interpreted as an incommensurate order of *localized bipolarons* and as glass of bipolarons respectively. (We extend the concept of a bipolaron which is usually well defined only when it is single). Then, it corresponds to a pair of electrons with opposite spins localized in a potential well due to the lattice distortion created self-consistently by the electron-phonon coupling. In section 6, we shall show that we can still define explicitly the bipolaronic lattice distortion of a single bipolaron for a many bipolarons system) .

Charge undefectibility characterizes experimentally the conducting materials while charge defectibility characterizes the insulating materials. Thus the TBA is a metal-insulator transition at zero degree K. More details about this transition were given in refs.3,4 .

Standard theories of CDW assume implicitely that the CDW is always in the conducting "analytic regime" ($k<k_c(\zeta)$). Up to now, our arguments for proving the existence of a TBA in a CDW model which is in contradiction with these standard theories, were essentially numerical and thus could be controversed. We can now provide a theorem valid at any dimension for the adiabatic Holstein model, which gives a *rigorous* fundation to this conjecture.

5- Bipolaronic States in the Adiabatic Holstein Model

When the electron-phonon coupling is large enough in the adiabatic Holstein model *at any dimension* , we prove the following theorem:

Theorem (1989)[6]

Let us choose an arbitrary configuration of pseudo-spins $\{\sigma_n = 0 \text{ ou } 1\}$ *such that :* $\sum_n \sigma_n = r$ *be the number of electron pairs of a finite system with s sites with for example periodic boundary conditions (s is arbitrarily large and* $n \in \mathbb{Z}^d$ *is a site of the d dimensional square lattice). Then, for*

(7-a) $$k > 2\sqrt{5d}$$

there exists one and only one local minima $\{u_n\}$ *of the functional energy* (6-c) *of the adiabatic Holstein model, such that*

(7-b) $$u_n = -k\ \rho_n$$

where ρ_n *is the electronic density of the* CDW *at site n and such that*

(7-c) $$|\rho_n - \sigma_n| \leq \frac{1}{4}$$

These metastable states have the following properties:

1-The electronic eigenstates exhibits a finite gap in energy with a lower band occupied and an upper band empty.

2- The phonon excitations exhibits a finite gap in frequency

3- The amplitude of a local perturbation at site n obtained for example by fixing an atom i *at a position different of its equilibrium position , decreases faster than a exponential of the distance between site i and n.*

The detailed proof of this theorem is too long for being described here and will be given elsewhere[6]. Let us just give the basic strategy which we used for proving this theorem:

1-We define an non-linear operator O in a Banach space for which the fixed points are the extrema of the variational form (6-c)

2- For each pseudospin configuration, we associate a domain $\mathcal{D}(\{\sigma_n\})$ by condition (7-c). When k is large enough, we prove that this domain $\mathcal{D}(\{\sigma_n\})$ is invariant by this operator O and

3- that the spectral norm of the linearized operator ∇O is strictly smaller than 1 inside this domain..

Consequently, this operator contracts the metric distance in the domain so that the Banach fixed point theorem (1921) can be applied. This theorem asserts the existence of a unique fixed point inside each domain $\mathcal{D}(\{\sigma_n\})$ which thus proves the main part of the theorem.

Let us note that the same strategy applied to the FK model allows one to prove the existence of chaotic states but in that case, the proof is much easier and shorter. Extensions and variations of this theorem and of its methods of proof to other adiabatic models and related results seem to be possible and are currently under study (but there will be slight changes in the theorems, for example when the phonons are not dispersionless).

• Let us discuss some physical implications of this theorem. In this specific Holstein model, for any pseudo-spin configuration $\{\sigma_n\}$, there exists a metastable state of the variational energy form (6-c) such that the r maxima and the s-r minima of the associate electronic density $\{\rho_n\}$ corresponds to the sites where $\sigma_n=1$ and $\sigma_n=0$ respectively. All the occupied electronic states are below an energy gap while the unoccupied electronic states are above. In other words, one can choose arbitrarily the location of the peaks of the electronic density which physically means the location of the bipolarons. More precisely, we can say that site n is occupied by a localized bipolaron when the pseudospin σ_n is 1 and that this site n is empty when $\sigma_n=0$. (However, it is clear that the bipolarons are not strictly localized on single sites but extend more or less around the occupied sites.)

Athough the bipolarons can be considered as localized in the real space (because one can choose arbitrarily their location), the electronic eigenstates are not necessarily localized. (Nevertheless, the proof of this theorem works whatever the electronic states are). For example if the configuration of pseudo spins $\{\sigma_n\}$ is chosen periodic, the electronic states are extended Bloch eigenstates. If this configuration is random, they might be exponentially localized. In fact, in this model, the concept of localization of the electrons is physically meaningless when the (undistinguishable) electrons *are not independant particles*. The excitations of these metastable states are global excitations of the whole system of electrons coupled with the phonons which thus involve self-

consistently the electronic density and the potential created by the lattice deformation. In such a situation, *the concept of defectibility replaces and generalizes the concept of localization.* Then, it is clear that these bipolaronic states are insulating at zero degree.

In the adiabatic approximation, the phonon frequencies ω_ν which correspond to small motions of the atoms (which are assumed to have a unit mass) are determined by the eigen equation

$$\omega_\nu^2 \, u_n^\nu = \sum_m \frac{\partial^2 \Phi(\{u_i\})}{\partial u_n \partial u_m} u_m^\nu \tag{8}$$

We have proven for this theorem, that the set of eigenvalues ω_ν^2 of the quadratic form associated with the second order expansion of the adiabatic energy are strictly larger than a finite positive value ω_G^2 which implies the absence of a gapless phason branch. In addition, the local perturbation created for example by an impurity at some site n decays faster than an exponential far away from this impurity. In fact, numerical studies in 1-d have shown that the decay is exponential and the corresponding characteristic length is the coherence length ξ. (However ξ depends on k and on the pseudospin configuration $\{\sigma_n\}$).

Although we know that there exist many other metastable states involving atomic distortions with smaller amplitudes, for large enough k, the ground-state presumably belongs to the set of metastable states defined by this theorem, but for a special choice of the pseudo-spins $\{\sigma_n\}$. We have not proven this conjecture but we have numerical evidence and a strong physical intuition that it should be true.

As for the FK model[7], for large enough electron phonon coupling k, these chaotic states are those which are relevant for studying the thermodynamics of the system in 1,2 or 3 dimensions at low temperature. Since the configuration of the sytem are represented by a pseudo-spin configuration, the initial Hamiltonian can be expanded on this basis as a lattice gas model. This is the representation which has been also found by Alexandrov et al [8] within a completely different approach. In ref [4], we found the beginning of the expansion of this Hamiltonian as a function of $\frac{1}{k}$. In one-dimension, this Hamiltonian has indeed an incommensurate ground-state with the expected modulation wave-vector $2k_F$. But let us emphasize that for two or three dimensional models, the ground-state which corresponds to a particular arrangement of the bipolarons is unknown. They could interpret observed CDWs in 2-d systems.

As in the one dimensional model, when the electron-phonon coupling k becomes smaller, it can reasonably be expected that the bipolaron size also diverges in d-dimensional models at some critical value of k (which depends on the band filling) Then all the metastable states disappear after complex cascades of inverse bifurcations as for the FK model! . But unlike the 1-d adiabatic model which is always unstable against a CDW-PLD, the adiabatic ground-state should present for smaller k another transition to a state with no lattice distortion.

6- Effective Bipolaronic Shape in the Adiabatic 1-d Holstein Model

Since we have noted the similarity of the 1-d Holstein model and of the FK model, it is tempting to check if the "decomposition theorem" obtained for the FK model applies here. For the FK model, this theorem asserts that the "non-analytic" incommensurate ground-states can be decomposed as a *linear superposition of effective discommensurations*. The shape of these discommensurations is not those of a single one but is effective because it depends on the presence of the other discommensurations. The size of this discommensuration is the coherence length ξ and diverges when k approach $k_c(\zeta)$ from above. We found numerically the same result in the adiabatic Holstein model (and hope to prove it rigorously). More precisely, the PLD of the groundstate can be written as

(9-a) $$u_n = - \sum_i \sigma_i \, b_{n-i}$$

with a sequence of pseudospins $\{\sigma_i\}$ defined as

(9-b) $$\sigma_i = \chi (i\zeta+\alpha)$$

where function $\chi(x)$ has period 1, is equal to 1 for $0<x\leq\zeta$ and to zero for $\zeta<x\leq1$. α is the arbitrary phase. The sequence $\{b_i\}$ is shown for two examples, figure 1. This result suggests that the bipolaron are well-defined objects at least for the "non-analytic" incommensurate ground-states and its excited states. It size diverges at $k_c(\zeta)$ and it does not exist below $k_c(\zeta)$ when the ground-state is incommensurate and "analytic".

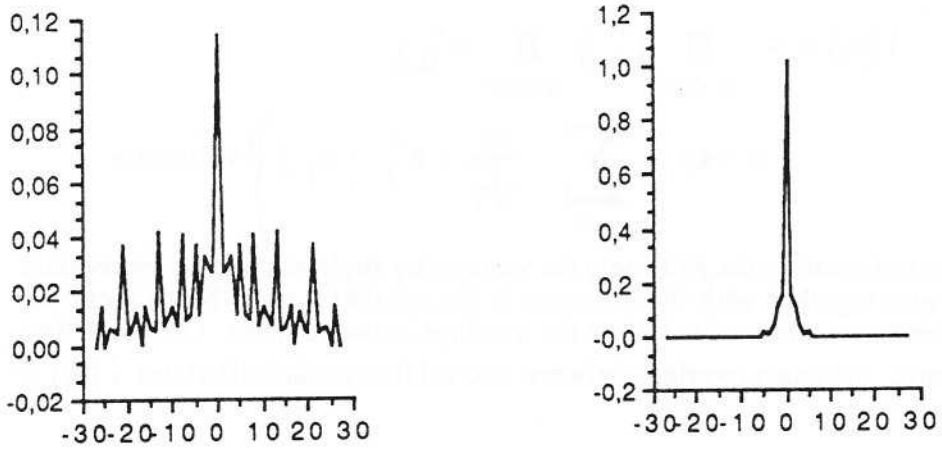

Figure 1: *Bipolaron shape* $\{b_i\}$ *versus* i *in the 1-d adiabatic Holstein model for* $\zeta = \frac{\sqrt{5}-1}{2}$ *for* $k=1.58\approx k_c(\zeta)$ *(left) and* $k=1.7$ *(right). This bipolaron is well localized far above the TBA and diverges when approaching* $k_c(\zeta)$ *from above. It is undefined for* $k<k_c(\zeta)$.

Let us now examine, the effect of quantum lattice fluctuations on the adiabatic solutions.

7- Quantum lattice fluctuations for small γ

The Born-Oppenheimer approximation consists of quantizing the atomic motion in the adiabatic potential with the Hamiltonian

$$(10\text{-a}) \qquad H = \frac{\gamma^2}{4} \sum_i \frac{1}{2} p_i^2 + \Phi(\{u_i\})$$

Within this approximation, the electrons are always in adiabatic equilibrium with the atomic configuration. Clearly, they cannot order by themselves into a superconducting order. The global wave function of the system has the form

$$(10\text{-b}) \qquad \Psi = \Psi(\{u_i\}) \prod_{\mu \text{ occ},\sigma} c^+_{\mu,\sigma}(\{u_i\}) \; |\text{vacuum}\rangle$$

where $c^+_{\mu,\sigma}(\{u_i\})$ is defined by (6-a) and (6-b). In fact, this approximation is in general very hard to handle explicitly because the adiabatic energy $\Phi(\{u_i\})$ is a highly non-linear functional as suggested by the above theorem. In practice, one can use quadratic expansions around the local minima of $\Phi(\{u_i\})$.

When the bipolaronic structure is "non-analytic" (large k), we can perform a more suitable approach. The adiabatic states are represented as coherent states

$$(11\text{-a}) \qquad |\{u_i\}\rangle = \prod_{\mu \text{ occ}} c^+_{\mu,\uparrow} \prod_{\mu \text{ occ}} c^+_{\mu,\downarrow} \times \exp\left(\sum_i \frac{u_i}{\sqrt{\gamma}} (a^+_i - a_i) \right) |\text{vacuum}\rangle$$

which consists of creating the PLD onto the vacuum by application of an appropriate distortion operator together with the electrons in the adiabatic state. Since there are many metastable states, one can calculate the overlaps between them. One find after some calculations, the exact overlaps between two arbitrary adiabatic states $|\{u_i\}\rangle$ and $|\{u'_i\}\rangle$ which are

$$(11\text{-b}) \qquad \langle\{u'_i\}|\{u_i\}\rangle = (\det \bar{\bar{A}})^2 \exp\left(-\frac{1}{2\gamma} \sum_{i=1}^{s} (u_i - u'_i)^2 \right) \text{ and}$$

$$(11\text{-c}) \qquad \langle\{u'_i\}| \hat{H} |\{u_i\}\rangle = \left(s\frac{\gamma}{4} + \frac{\Phi(\{u_i\}) + \Phi(\{u'_i\})}{2} - \frac{1}{4} \sum_{i=1}^{s} (u'_i - u_i)^2 \right) \langle\{u'_i\}|\{u_i\}\rangle$$

($\det \bar{\bar{A}}$ is the determinant of the $r \times r$ matrix $\bar{\bar{A}} = (A_{\mu',\mu})$ defined as

$$(11\text{-d}) \qquad A_{\mu',\mu} = \langle \psi'^{\mu'} | \psi^{\mu} \rangle = \sum_i \psi'^{\mu'*}_i \psi^{\mu}_i$$

where $|\psi^{\mu}\rangle$ and $|\psi'^{\mu'}\rangle$ are the occupied electronic states given by (6-b) for the adiabatic state $|\{u_i\}\rangle$ and $|\{u'_i\}\rangle$ respectively.

For $k >> k_c(\zeta)$ and small γ, it is easy to check that these adiabatic states $|\{u_i\}\rangle$ are orthogonal one with each other because of the exponential in (11-b). For example, for two adiabatic states which differs only by the location of a bipolaron, one easily find the estimation

$$(12) \qquad \langle\{u'_i\} | \{u_i\}\rangle \approx \exp(-\frac{k^2}{\gamma}).$$

For other adiabatic states which differs by the location of more and more pseudospins, the overlap becomes smaller and smaller. To fix the ideas, $k=2$ and $g=10^{-2}$ yields an overlap of e^{-400} which clearly is physically neglegible. Clearly, the adiabatic metastable configurations are practically unaffected by the existence of small quantum lattice fluctuations.

When k approach $k_c(\zeta)$ from above, we noted that the bipolaronic size diverges, which implies that the overlap between close adiabatic states goes to 1.More precisely, the overlap calculated in (12) becomes roughly proportional to $\exp(-\frac{E_{PN}}{\gamma})$ where E_{PN} is the Peierls-Nabarro energy barrier that the height of the barrier which has to be overcome for moving a bipolaron of one lattice site. This feature is a proof that the above quantized adiabatic states (11-a) becomes very bad quantum states at the TBA.

In the bipolaronic regime $k>k_c(\zeta)$, one can consider the quantum lattice fluctuations as a correction to the lattice gas Hamiltonian of bipolarons. The extra terms appears as transverse spin operators which allows the tunnelling of bipolarons from one occupied site to an empty site. Setting

$$(13\text{-a}) \qquad S^z_i = \sigma_i - \frac{1}{2}$$

the spin Hamiltonian expands with the general form

$$(13\text{-b}) \qquad H = \sum_i h\, S^z_i + \sum_{i,j} J_{i-j}\; S^z_i\, S^z_j + \sum_{i,j,k} K_{i-k,j-k}\, S^z_i\, S^z_j\, S^z_k + \ldots.$$

$$- \sum_{i,j} \Gamma_{i-j} (S^x_i\, S^x_j + S^y_i\, S^y_j) + \ldots..$$

Thus for small γ, one can explicitly calculate the first terms of this expansion (see ref.4). At the lowest order, one finds again the same Hamiltonian as obtained by ALEXANDROV et al 1986 for describing bipolaronic superconductivity in this specific model. In their representation, superconductivity is obtained if the average of the transverse component $\langle S_i^x \rangle$ is non zero. However close to the adiabatic limit and the quantum terms in (13-b) which fulfills[4]

$$(13\text{-c}) \qquad \Gamma_{i-j} \approx J_{i-j} \exp\left(-\frac{4 E_{PN}}{\gamma}\right) \approx J_{i-j} \exp\left(-\frac{k^2}{\gamma}\right) \qquad \text{for large } k,$$

are negligible unless k becomes close above to $k_c(\zeta)$ so that $E_{PN} \to 0$. Then the bipolaronic model collapses because due to the divergence of the bipolaron size, the series in (13-b) tends to diverge.

Among the arguments which we developped in ref.6 for proving the unstability of "analytic" CDW under quantum lattice fluctuations, we suggested that for $k<k_c(\zeta)$, when the phase fluctuations of the incommensurate "analytic" CDW extends over a large enough distance (which is about the size of a Cooper pair!), then it gains more energy by quantum tunnelling than it looses by the elastic deformation.

We are currently performing more sophisticated expansions with respect to the quantum parameter γ. At the present stage of our work, all attempts confirm that the adiabatic "analytic" CDW are far from a good quantum state of the Hamiltonian. Although, we have no rigorous proof, our present conjecture remains that the existence of a gapless phason branch for a CDW structure makes this structure unstable with respect to quantum lattice fluctuation whatever γ is small (but non zero)!

On the other hand, as we demonstrate above, bipolaronic states should remain stable providing that the quantum parameter be not too large $\gamma << k^2$.

8- The antiadiabatic limit $\hbar\omega_0 >> T$

This regime is physically unusual but is plausible when the electrons are not coupled to phonons but to other excitations of the solid with much higher energies (plasmons, excitons?). In that regime, the Lang-Firsov unitary transformation $\exp(iS_{LF})$ (e.g. see ref. 8) transforms the Hamiltonian (1) into

$$(14\text{-a}) \qquad \hat{H} = \exp(iS_{LF})\, H \exp(-iS_{LF})$$

with

$$(14\text{-b}) \qquad i S_{LF} = \frac{g}{\hbar\omega_0} \sum_i \left(\sum_\sigma c^+_{i,\sigma} c_{i,\sigma} \right) \left(a^+_i - a_i \right)$$

One obtains

(14-c) $$\hat{H} = T \left[- \sum_{<i,j>,\,\sigma} \hat{t}_{i,j}\, c^{+}_{i,\sigma}\, c_{j,\sigma} - \frac{1}{4}\, k^2 \sum_i \left(\sum_\sigma c^{+}_{i,\sigma}\, c_{i,\sigma} \right)^2 \right.$$

$$\left. + \gamma \sum_i \left(a^{+}_i\, a_i + \frac{1}{2} \right) \right]$$

where

(14-d) $$\hat{t}_{i,j} = \exp\left(\frac{1}{2}\, \frac{k}{\sqrt{\gamma}} \left(\left(a^{+}_i - a_i \right) - \left(a^{+}_j - a_j \right) \right) \right)$$

When

(15-a) $$k^2 << \gamma$$

or equivalently $g << \hbar\omega_0$ (this is the regime opposite to the regime of stability of the bipolaronic structures against quantum fluctuations)

(15-b) $$\hat{t}_{i,j} = 1$$

Then, one obtain an Hamiltonian where the electrons and the phonons *exactly decouple*. An electron-electron interaction is left which is on-site with the coefficient

(15-c) $$-\frac{Tk^2}{2} = -U$$

This situation allows one to try the standard BCS form for superconductivity

(16-a) $$\prod_q \left(u_q + v_q\, c^{+}_{q,\uparrow}\, c^{+}_{-q,\downarrow} \right) \; | \text{vacuum} >$$

where c^{+}_q is now the creation operator of an delocalized electron with wave vector q and spin $\uparrow$. The modulation u(q) and v(q) now is in the reciprocal space. This specificity allows the existence of quantum states (under magnetic field) of the whole system with a permanent current and thus of superconductivity.

(16-b) $$\prod_q \left(u_q + v_q\, c^{+}_{q+\delta,\uparrow}\, c^{+}_{-q+\delta,\downarrow} \right) \; | \text{vacuum} >$$

NOZIERES et SCHMITT-RINK[10] have studied this negative U Hubbard model.

9-Conclusion:

As we suggested above, the "analytic" incommensurate CDW should be unstable with respect to quantum lattice fluctuations. Thus, we conjecture that the phase diagram at zero degree K of the Holstein model should be shared into two main regions according to the role of the quantum lattice fluctuations (cf. fig. 2):

1- The bipolaronic region which starts from the adiabatic line $\gamma=0$ and from the critical point of the TBA when the band filling ζ is irrational. In that region, sufficiently far away from the critical line (according to (13-c) and (15-a), $\gamma \approx 4E_{PN} \approx k^2$ for large k), the quantum lattice fluctuations have a small effect . (For the commensurate band filling, this line may reach the origin but should converge to the incommensurate line in the limit of high order commensurability.)

2- The superconducting region in the remainding part of the phase diagram, where the quantum lattice fluctuations always prevent the formation of a structure in real space.

It can be reasonably expected for models in more than 1 dimension, that there exist many other critical lines (perhaps infinitely many) inside the bipolaronic region which determine regions with different bipolaronic arrangements. In addition, superconductivity should behave very differently depending on the region of the phase diagram where it is supposed to exist (e.g. close to the adiabatic line for irrational ζ opposed to the region with both large k and $\gamma >> k^2$).

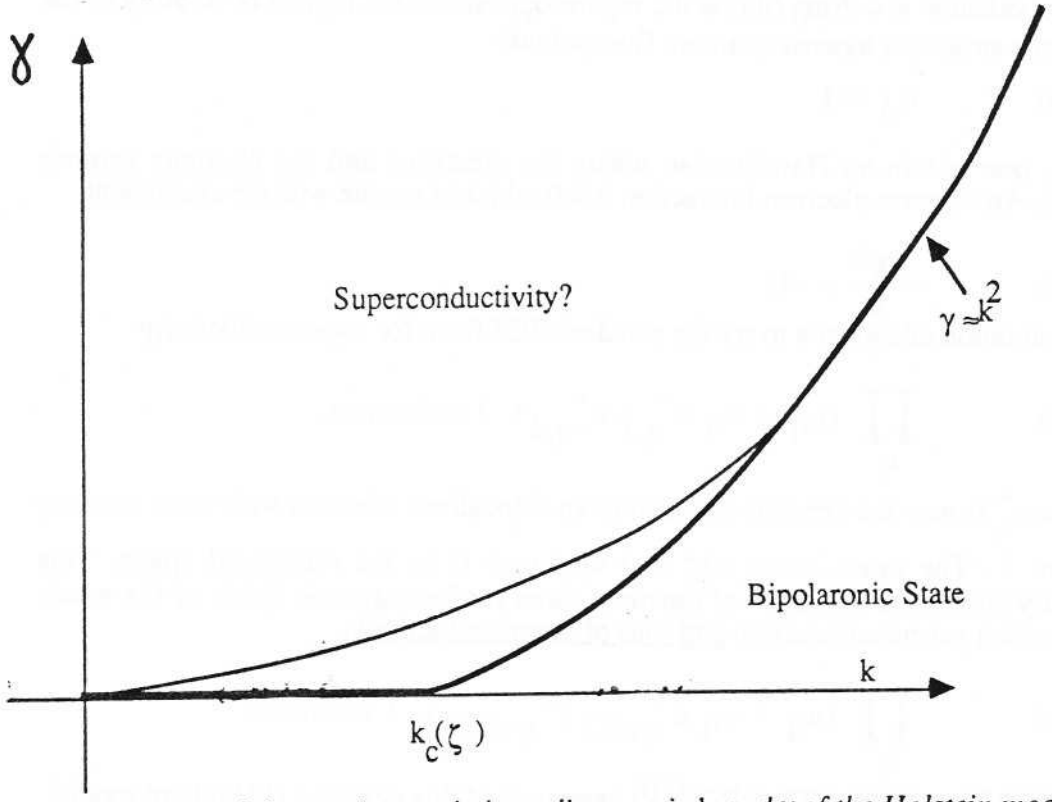

Figure 2 *Scheme of the conjectured phase diagram in* k *and* γ *of the Holstein model at zero degree K. A line separates the bipolaronic states from the superconducting region . For large* k, *this line follows approximately* $\gamma \approx k^2$ *but depends on the band filling for small* k. *For irrational band filling, it should reach (with an exponent?) the* k *axis at* $k_c(\zeta)$.

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