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The Ordering of Polarons in the Holstein–t-J Model: An Application to $La_{2-x}Sr_xNiO_{4+y}$.

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Abstract. – The recently observed ordering of polarons in $La_{2-x}Sr_xNiO_{4+y}$ is investigated in terms of a simple one-band model of strongly correlated electrons at quarter filling. The lattice distorsions are described by an in-plane breathing mode which is treated in the adiabatic limit. We use exact diagonalization techniques on small lattices to obtain the ground-state configuration and the excitation spectrum of the Holstein–t-J model. For large enough electron-phonon coupling a state resembling the observed ordering of polarons appears as the ground state of the Holstein–t-J model. We discuss the dependence of the charge and magnetic structure factors on doping away from quarter filling. Within the limits of a finite-lattice calculation our results agree qualitatively with the experiment.

Antiferromagnetic insulating materials have received renewed attention in the search for the underlying mechanisms of high-T_c superconductors. Of particular interest has been the dependence of magnetic, charge and lattice fluctuations on doping [1-3]. As a typical and important example, the isostructural compounds La_2CuO_4 and La_2NiO_4 show a remarkable difference upon the substitution of La by Sr. Both materials become metallic upon doping, but in the nickelate a nearly total substitution of Sr for La is necessary. Also in $La_{2-x}Sr_xNiO_{4+y}$ no superconductivity is found for any x. A resolution of this problem might be given by extended local-density calculations [4], which show that the nickelates are much more susceptible to a breathing-polaron instability than the cuprates. In a recent electron diffraction experiment [3] on $La_{2-x}Sr_xNiO_{4+y}$ this idea is substantiated. As a function of Sr doping a superstructure spot at the (π, π) point is observed, which is interpreted as a sign of two-dimensional ordering of localized breathing polarons. The difference between the polaronic behaviour of the cuprates and the nickelates is also seen in the infrared optical response [2]. In the nickelates electronic transport and magnetic anomalies have recently been observed at $x = 1/2$ and $x = 1/3$ [5]. A theoretical model for these phenomena must contain both the coupling of electronic and lattice degrees of freedom and the correlated hopping of holes in an antiferromagnetic background. In this letter we show that an extension of the t-J model, the Holstein–t-J model [6-8] at quarter filling, exhibits a polaronic superlattice as observed in $La_{2-x}Sr_xNiO_{4+y}$.

The Hamiltonian of the Holstein–t-J model in the adiabatic limit is given by

$$H = -t \sum_{\langle ij \rangle, \sigma} (\tilde{c}^{\dagger}_{i,\sigma} \tilde{c}_{j,\sigma} + \text{H.c.}) + J \sum_{\langle ij \rangle} \left(\boldsymbol{S}_i \boldsymbol{S}_j - \frac{n_i n_j}{4} \right) - \alpha \sum_i \Delta_i n_i + \frac{K}{2} \sum_i ((\Delta_i^x)^2 + (\Delta_i^y)^2) .$$

The first two terms represent the standard t-J model. The third and fourth term take into account the coupling of a single dispersionsless optical-phonon branch to the electronic on-site energy and the elastic energy of a harmonic lattice, respectively. In the context of the nickel/copper oxides, the local Holstein coordinates $\{\Delta_i\}$ correspond to bond-parallel oxygen lattice displacements describing an in-plane oxygen breathing mode. To represent the breathing mode we introduced the collective coordinate $\Delta_i = \Delta_i^x - \Delta_{i-(1,0)}^x + \Delta_i^y - \Delta_{i-(0,1)}^y$ [7,8]. The effect of the electron-phonon coupling is that holes tend to move to sites where the oxygen atoms are displaced towards the nickel/copper site. Rescaling $\alpha\Delta_i^{x,y} \to \Delta_i^{x,y}$ we introduce the dimensionless coupling constant $\lambda = \alpha^2/K$. From now on all energies and interaction constants will be measured in units of the hopping amplitude t.

To investigate the ground-state properties of the Holstein-t-J model one needs to find those lattice distortions $\{\Delta_i^{x,y}\}$ that minimize the energy. The Hellmann-Feynman theorem leads to a set of $2N$ self-consistency equations. We solve these equations iteratively [7,8]. Each iteration step involves the diagonalization of the Hamiltonian, which can be represented as a large sparse matrix. For this purpose we use a standard Lanczos technique which only needs matrix vector multiplications as the basic ingredient. Since the possible spatial variation of the $\{\Delta_i^{x,y}\}$ breaks translational invariance, we cannot use momentum as a good quantum number to reduce the size of the Hamiltonian matrix. For the largest system sizes considered, $N = 16$, and for six holes, we are left with a matrix of dimension 2018016 in the $S^z_{\text{total}} = 0$ subspace. The state vectors are stored through a variant of a multispin coding technique [9].

In order to look for the formation of a polaronic superlattice in the Holstein-t-J model, we shall first present results exactly at quarter filling to show the dependence of the ground state on the parameters of the model and discuss the finite-size effects. Due to limited computer resources it is not possible to map out a complete phase diagram for the 4×4 site lattice. Instead the parameter dependence of the Holstein-t-J model at quarter filling is demonstrated for the $\sqrt{10} \times \sqrt{10}$ site square lattice. Figure 1 shows the phase diagram of the Holstein-t-J model for the $\sqrt{10} \times \sqrt{10}$ site lattice as a function of the electron-phonon coupling λ and the antiferromagnetic exchange interaction J. Apart from two types of phase-separated ground states (labelled PS1 and PS2) for unphysically large J, the phase diagram consists mainly of two phases separated by a line $\lambda_c(J)$. For large values of λ we observe a structure (labelled AB phase in fig. 1) which resembles a superlattice of self-trapped

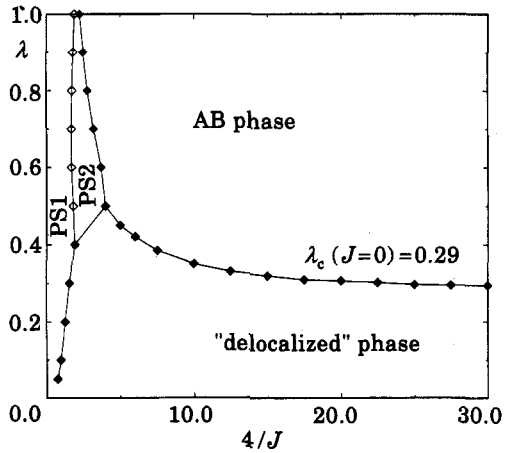

Fig. 1. – The phase diagram of the ten-site lattice at quarter filling. AB labels the phase with a polaronic superlattice, «delocalized» labels the phase that resembles the symmetries of the electronic system and PS1 and PS2 label two phase-separated states.

polarons. This phase corresponds to the polaronic superlattice observed in $La_{2-x}Sr_xNiO_{4+y}$. In terms of the on-site electron densities it appears as a charge density wave (CDW) with wave vector (π, π). A dopant-induced (π, π) CDW state around quarter filling was also predicted for the 2D Holstein-Hubbard model [10]. For small values of λ we find a ground state with finite «averaged» distortion $\Delta = (1/N) \sum_{i=1}^{N} |\Delta_i|$, which in terms of the local densities reflects the symmetry of the undistorted state at $\lambda = 0$, *i.e.* the ground state of the purely eletronic *t*-*J* Hamiltonian. Here a finite Δ is due to a non-trivial wave vector of the electronic part of the ground-state wave function. Similar behaviour was already seen close to the half-filled case [6-8]. A finite Δ in this «delocalized» phase depends strongly on the symmetries of the electronic wave function which in turn depend strongly on the system size. In contrast to the $\sqrt{10} \times \sqrt{10}$ site lattice the ground state of the $\sqrt{8} \times \sqrt{8}$ and the 4×4 site lattice at quarter filling is homogeneous and the phase for $\lambda < \lambda_c$ is undistorted. However, finite-size effects are small in the self-trapped AB phase at larger λ and not too large J, because the kinetic part of the energy is suppressed.

Having established the existence of a polaronic superlattice in the Holstein–*t*-*J* model for

TABLE I. – *On-site electron densities for the* 16*-site lattices for* $\lambda = 1.0$, $J = 0.4$ *and different hole doping. The sites of the* 4×4 *lattice are counted in such a way that the top row contains sites 1–4 from left to right, the row further down sites 5–8 and so on. For reasons of clarity only the first three digits are shown. Values within boxes indicate the sites which deviate from the ideal* AB *structure at quarter filling* ($N_h = 8$), *i.e. where hole sites are filled by electrons for* $N_h < 8$ *and where electron sites become empty for* $N_h > 8$. *The cases* $N_h = 5$ *and* $N_h = 11$ *describe different phases altogether.*

Site	On-site electron densities $\langle n_i \rangle$						
N_h	5	6	7	8	9	10	11
1	0.929	0.080	0.051	0.063	0.063	0.066	0.436
2	0.101	0.938	0.938	0.936	0.912	0.855	0.265
3	0.945	0.090	0.080	0.063	0.063	0.076	0.163
4	0.101	0.932	0.938	0.936	0.938	0.921	0.265
5	0.101	0.938	0.938	0.936	0.912	0.855	0.126
6	0.945	0.988	0.080	0.063	0.074	0.098	0.082
7	0.966	0.940	0.931	0.936	0.912	0.049	0.881
8	0.945	0.090	0.080	0.063	0.063	0.076	0.820
9	0.945	0.090	0.080	0.063	0.074	0.076	0.820
10	0.966	0.940	0.931	0.936	0.356	0.049	0.881
11	0.122	0.988	0.986	0.063	0.074	0.098	0.064
12	0.966	0.938	0.931	0.936	0.912	0.855	0.881
13	0.101	0.932	0.938	0.936	0.912	0.921	0.265
14	0.945	0.090	0.080	0.063	0.074	0.076	0.163
15	0.966	0.938	0.931	0.936	0.912	0.855	0.196
16	0.945	0.080	0.080	0.063	0.063	0.066	0.163

strong enough electron-phonon coupling, we shall now investigate this phase for the 4×4 site lattice at $J = 0.4$ and $\lambda = 1$ at quarter filling. The local electron densities $\langle n_i \rangle$ are tabulated in table I. At this filling one obtains only two different values for the local electron densities: $\langle n_i \rangle = 0.93661$ at the «electron sites» and $\langle n_i \rangle = 0.06338$ at the «hole sites». Note the small electron density at the hole sites, which only goes to zero as λ goes to infinity. Within the adiabatic approximation we study the basic excitations for this polaronic superlattice. For this purpose we need to obtain the spectrum of the Hamiltonian for a *fixed* set of $\{\Delta_i^{x,y}\}$. Since it is impossible to find all the eigenvalues of a mega-dimensional matrix exactly, we use a method that gives a polynomial approximation to the *many-body* density of states $N(E)$, which involves the stochastic evaluation of polynomial moments [11]. The resolution along the energy axis improves with the number of moments. In fig. 2*a*) we show the many-body density of states of the AB phase for the possible values of S^z_{total}. The overall structure of $N(E)$ consists of a set of «bands» above the ground state (labelled GS in fig. 2*a*)) separated by gaps. Our interpretation is that the first band above the ground state contains states, where one electron is moved from an electron site to hole sites, that the second band contains states, where two electrons are moved from electron sites to hole sites, and so on until all electrons are moved from electron sites to hole sites (of course the lattice distortion for highly excited states is extremely unstable, and the adiabatic approximation breaks down). When four electrons are moved to hole sites, the bandwidth and the density of states are largest, because the maximum number of electrons can interact via the antiferromagnetic exchange J, and the number of positions for the electrons to move to is maximum. When all electrons are on the «wrong» sites, we get only one state again. We checked this interpretation by selectively calculating states at the edges of the various bands. This $N(E)$ should be contrasted to the case of the purely electronic model, *i.e.* $\lambda = 0$, depicted in fig. 2*b*), where $N(E)$ has no gaps. It is interesting that for the AB phase the ground state is nearly degenerate with respect to S_z, *e.g.* the ground states for all of the different S_z subsectors are nearly degenerate and are at the value depicted by GS in fig. 2*a*). As λ goes to infinity this degeneracy becomes exact. In the purely electronic model the ground states for the different S_z sectors are well separated.

To study the stability of the polaronic superlattice *away* from quarter filling we calculated the ground state of the 4×4 site lattice at $J = 0.4$ and $\lambda = 1.0$ for all possible numbers of holes N_h. The on-site electron densities are summarized in table I for $N_h = 5$ to $N_h = 11$. At quarter filling

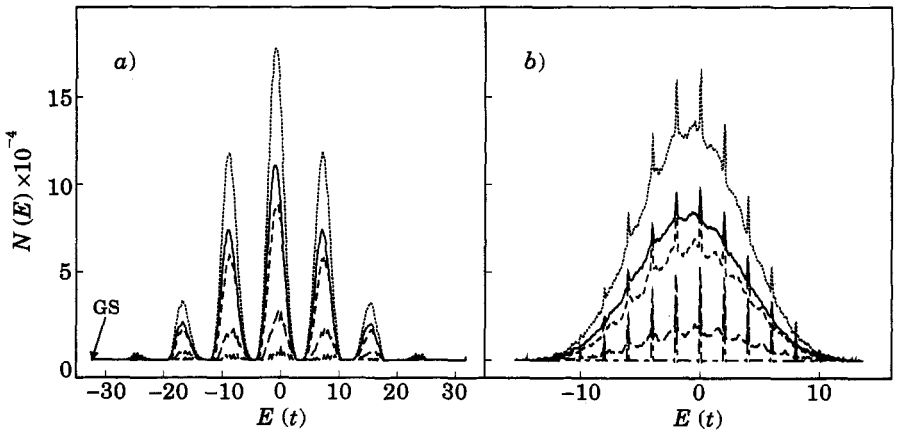

Fig. 2. – The many-body density of states $N(E)$ for the 16-site lattice at $J = 0.4$ and $\lambda = 1.0$ (*a*)) and $\lambda = 0.0$ (*b*)). —— $S_z = 0$, ····· $S_z = 1$, ---- $S_z = 2$, --- $S_z = 3$, ----- $S_z = 4$.

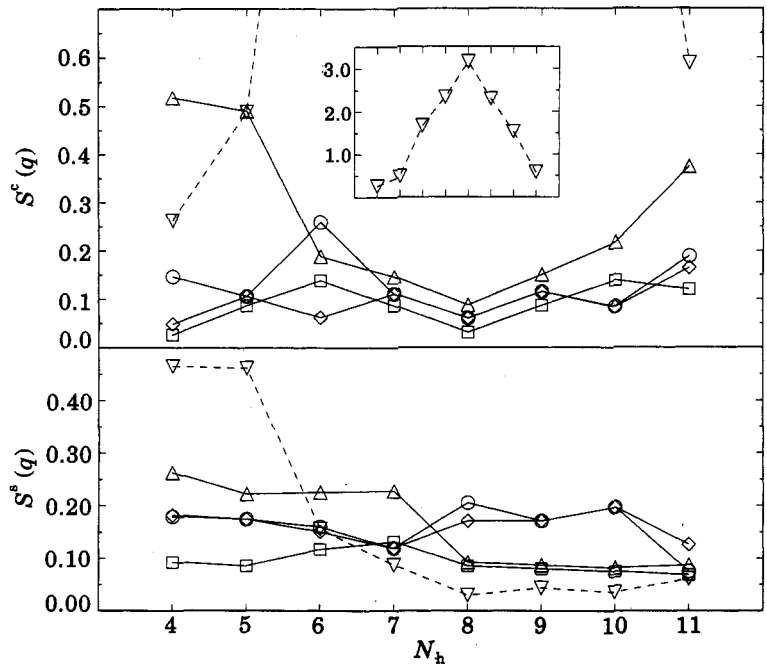

Fig. 3. – The charge and magnetic structure factors $S^c(q)$ and $S^s(q)$ as defined in the text *vs.* the number of holes N_h for the 16-site lattice at $\lambda = 1.0$ and $J = 0.4$. ▽---▽ $q = (\pi, \pi)$, △——△ $q = (\pi, \pi/2)$, ◇——◇ $q = (\pi, 0)$, □——□ $q = (\pi/2, 0)$, ○——○ $q = (\pi/2, \pi/2)$.

plus one hole ($N_h = 9$) one observes that one electron gets removed mainly from site 10 (for this configuration) and that the other sites remain to a large extent in the CDW state. Adding one further hole ($N_h = 10$) has a similar effect. Now sites 7 and 10 become additional hole sites. For $N_h > 10$ the polaronic superlattice breaks down. Adding electrons to the quarter-filled system causes the opposite effect. For $N_h = 7$ site 11 is occupied by the additional electron, for $N_h = 6$ sites 11 and 6. The CDW-like polaronic superlattice disappears for $N_h < 6$.

To discuss the density and spin correlations in more detail we present in fig. 3 the charge and magnetic structure factors,

$$S^c(\boldsymbol{q}) = \frac{1}{N}\sum_{i,j}\langle n_i n_j\rangle \exp\left[i\boldsymbol{q}(\boldsymbol{R}_i - \boldsymbol{R}_j)\right], \quad \text{and} \quad S^s(\boldsymbol{q}) = \frac{1}{N}\sum_{i,j}\langle S_i^z S_j^z\rangle \exp\left[i\boldsymbol{q}(\boldsymbol{R}_i - \boldsymbol{R}_j)\right],$$

as a function of the number of holes N_h at the allowed, non-trivial wave vectors of the 4×4 lattice. The pronounced peak in $S^c(\pi, \pi)$ at quarter filling reflects the commensurate ordering of localized polarons in the Holstein-t-J model. The (π, π) component of S^c is the dominant contribution in the relatively large regime from six to ten holes. For smaller hole densities a crossover to a density modulation with wave vector $(\pi, \pi/2)$ takes place around $N_h = 5$. In the experiment [3] on $La_{2-x}Sr_xNiO_{4+y}$ the wave vector of the charge modulation deviates strongly from its commensurate value (π, π) for $x < 0.5$, whereas it remains at (π, π) for larger doping. Here we seem to have a difference between the phase of the Holstein-t-J model on the (4×4)-lattice and the measured phases [3] in $La_{2-x}Sr_xNiO_{4+y}$. In a finite-lattice calculation the only possible phases correspond to the few wave vectors belonging to the reciprocal lattice. Also the region of stability of commensurate phases like the (π, π) phase gets artificially enlarged on finite lattices due to the small number of

commensurate phases available. But, if we take the wave vector where $S^c(q)$ is largest (*i.e.* $(\pi, \pi/2)$ for $N_h < 6$) as an indicator for the wave vector of a possible polaronic superlattice, we again find a phase that is similar to the experimentally observed one for $0.1 < x < 0.4$ [3]. For larger hole concentrations ($N_h > 11$) the structure in $S^c(q)$ disappears and we can no longer specify a definite wave vector. Since for these concentrations we have only a very small number of electrons, we do not believe that this effect mimics the disappearance of the superstructure spot in $La_{2-x}Sr_xNiO_{4+y}$ for $x \to 1$. The maximum in the magnetic structure factor $S^s(q)$ at the value (π, π) increases with the electron concentration indicating increasing antiferromagnetic correlations. This is consistent with the result close to half-filling [7,8]. Since the mobility of the electrons (holes) is dramatically reduced by the frozen-in lattice distortions, the ground state of the system might well be described by a Heisenberg model with immobile vacancies for $x < 0.5$. These vacancies are not at random positions, but are located in such a way that the total energy is minimized.

Calculating the ground state of the Holstein–t-J model on finite lattices, we have found convincing evidence that at quarter filling and large enough electron-phonon coupling the Holstein–t-J model exhibits a commensurate charge density wave with wave vector (π, π). This phase, which is related to a static lattice distortion, was experimentally observed [3]. To find such a CDW-like phase in the Holstein–t-J model, the electron-phonon coupling constant must exceed the (reasonable) value $\lambda \approx 0.35$ at $J = 0.4$. Since no such superstructure was observed in $La_{2-x}Sr_xCuO_{4+y}$, we must use a smaller electron-phonon coupling constant in $La_{2-x}Sr_xCuO_{4+y}$ than in $La_{2-x}Sr_xNiO_{4+y}$, in good agreement with local-density calculations [4]. Within the adiabatic approximation we have also calculated the many-body density of states and classified the basic excitations. According to the Frank-Condon principle they may be measured in absorption experiments. This might provide a method to quantify the value of the electron-phonon coupling constant in the framework of the Holstein–t-J model. The CDW state in the Holstein–t-J model at quarter filling was found to be nearly ferrimagnetic. The anisotropy in the doping dependence around the quarter-filled state agrees with the experimental findings. The incommensurate charge ordering away from quarter filling cannot be compared quantitatively with the experimental data due to the smallness of the lattices we can calculate.

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