

## Electron-phonon interaction in correlated electronic systems: polarons and the formation of orbital ordering

Daniela Schneider, Karl-Heinz Höck, Klaus G. Ziegler

### Angaben zur Veröffentlichung / Publication details:

Schneider, Daniela, Karl-Heinz Höck, and Klaus G. Ziegler. 2004. "Electron-phonon interaction in correlated electronic systems: polarons and the formation of orbital ordering." *physica status solidi (c)* 1 (11): 2828–31. <https://doi.org/10.1002/pssc.200405359>.



# Electron-phonon interaction in correlated electronic systems: polarons and the formation of orbital ordering

D. Schneider\*, K.-H. Höck, and K. Ziegler

Institut für Physik, Universität Augsburg, Universitätsstrasse 1, 86135 Augsburg, Germany

The properties of a dilute electron gas, coupled to the lattice degrees of freedom, are studied and compared with the properties of an electron gas at half-filling, where spinless fermions with two orbitals per lattice site are considered. The simplest model which includes both the local electron-lattice interaction of the Jahn-Teller type and the electronic correlations is the  $E \otimes \beta$ -Jahn-Teller-Hubbard model. We analyze the formation and stability of Jahn-Teller polarons and bipolarons, respectively. Our approach is based on a hopping expansion in the strong-coupling regime. The results are compared with recently published findings for the Hubbard-Holstein model [1, 2]. The special case of the Jahn-Teller-Hubbard model at half-filling is mapped on a spin-1/2 Heisenberg model with phonon-dependent coupling constants. This has been derived within a projection formalism that provides a continued-fraction representation of the Green's function. We study the exact solution for two and three particles and compare it with the effective theory on the infinite lattice with one particle per site.

**1 The model** The Hamiltonian for fermions with spin  $\sigma$  and pseudospin  $\gamma = \theta, \epsilon$ , coupled to phonons, is given by  $H = H_t + H_0$ , where  $H_t$  is a nearest neighbor tunneling term for the fermions

$$H_t = -t \sum_{\langle \mathbf{j}, \mathbf{j}' \rangle} \sum_{\sigma=\uparrow, \downarrow} \sum_{\gamma=\theta, \epsilon} c_{\mathbf{j}\gamma\sigma}^\dagger c_{\mathbf{j}'\gamma\sigma} + h.c.$$

and  $H_0$  is local, containing a (Hubbard) interaction and a term for dispersionless phonons of energy  $\omega_0$  with  $H_0 = \sum_{\mathbf{j}} H_{0\mathbf{j}}$  and

$$H_{0\mathbf{j}} = \omega_0 b_{\mathbf{j}}^\dagger b_{\mathbf{j}} + g(b_{\mathbf{j}}^\dagger + b_{\mathbf{j}}) \sum_{\sigma} (n_{\mathbf{j}\theta\sigma} \pm n_{\mathbf{j}\epsilon\sigma}) + \sum_{\gamma} U n_{\mathbf{j}\gamma\uparrow} n_{\mathbf{j}\gamma\downarrow} + U_O (n_{\mathbf{j}\theta\uparrow} + n_{\mathbf{j}\theta\downarrow}) (n_{\mathbf{j}\epsilon\uparrow} + n_{\mathbf{j}\epsilon\downarrow}),$$

where the plus (minus) sign refers to Holstein ( $E \otimes \beta$  Jahn-Teller) electron-phonon coupling [3].  $H_{0\mathbf{j}}$  is diagonalized by a Lang-Firsov transformation [4] and has energies

$$E_{0\mathbf{j}} = \omega_0 n_{\mathbf{j}} - E_p \left[ \sum_{\sigma} (n_{\mathbf{j}\theta\sigma} \pm n_{\mathbf{j}\epsilon\sigma}) \right]^2 + \sum_{\gamma} U n_{\mathbf{j}\gamma\uparrow} n_{\mathbf{j}\gamma\downarrow} + U_O (n_{\mathbf{j}\theta\uparrow} + n_{\mathbf{j}\theta\downarrow}) (n_{\mathbf{j}\epsilon\uparrow} + n_{\mathbf{j}\epsilon\downarrow}) \quad (1)$$

if there are  $n_{\mathbf{j}} (\geq 0)$  phonons and  $n_{\mathbf{j}\gamma\sigma} (= 0, 1)$  electrons with (pseudo)spin  $\sigma$  ( $\gamma$ ) at site  $\mathbf{j}$ . Each electron has an energy gain  $E_p = g^2/\omega_0$ . The regime  $U < 2E_p$  has an attractive interaction, leading to an on-site bipolaron.

\* Corresponding author: e-mail: daniela.schneider@physik.uni-augsburg.de, Phone: +49 821 598 3221, Fax: +49 821 598 3262

**2 The bipolaron problem** To study the binding properties of polarons we consider two electrons on a lattice with two orbitals ( $\theta$  and  $\epsilon$ ) per lattice site. In a strong coupling approach we study the similarities and differences of the non-degenerate Holstein-Hubbard ( $n_\epsilon$  or  $n_\theta = 0$ ) (HH) and the  $E \otimes \beta$ -Jahn-Teller-Hubbard model (JTH). We restrict ourselves to the case of repulsive on-site interaction ( $U > 2E_p$ ). Therefore the ground state of  $H_0$  has single occupied sites. If both electrons are in the same orbital, the situation is the same as in the HH [1, 2] due to the orbital conserving hopping. We show in the following, however, that the situation for electrons in the  $\theta$  and the  $\epsilon$  orbitals is different. This is a consequence of the fact that we have a different distortion configuration associated with different orbitals.

For a hypercubic lattice a hopping expansion leads to an exponentially reduced hopping rate  $te^{-E_p/\omega_0}$  in first order. The results in second order for each possible hopping process can be written in general as

$$-\frac{t^2}{\omega_0} e^{-\frac{2E_p}{\omega_0}} \sum_{n=1}^{\infty} \frac{(-b)^n}{n! n} \quad \text{and} \quad -\frac{t^2}{\omega_0} e^{-\frac{2E_p}{\omega_0}} \sum_{n=0}^{\infty} \frac{(-b)^n}{n! (n+a)} = -\frac{t^2}{\omega_0} e^{-\frac{2E_p}{\omega_0}} \tilde{\gamma}(a, b). \quad (2)$$

The first series is the result of processes with empty and singly-occupied sites as excited states, while the second series takes doubly-occupied states into account.  $\tilde{\gamma}(a, b)$  is related to the incomplete Gamma function [5]. For electrons in the same orbital  $a$  is equal to  $(U - 2E_p)/\omega_0$  and for different orbital occupation  $a$  is given as  $(U_O + 2E_p)/\omega_0$ . Values of  $b$  are  $\pm E_p/\omega_0$  and  $\pm 2E_p/\omega_0$ . The positive sign corresponds only to processes that describe hopping in different orbitals. One process takes place in the  $\theta$ , the other in the  $\epsilon$  orbital. Hence they only appear in the JTH but not in the HH. The terms related to the processes explained above are related to processes that change the orbital occupancy at least at one site. This means, for instance, that the initial state has an electron in orbital  $\theta$  and the final state an electron in orbital  $\epsilon$  at one site. Only the electrons at nearest neighbor sites are available for these processes. If both hopping processes take place in the same orbital the minus sign appears. These are the only contributions in the HH. They also exist in the JTH if either both electrons are in the same orbital or if only one of the two electrons moves. The factor 2 arises if only two sites are involved in the process. While the series with  $b > 0$  reduces the exponential prefactor even further, the series with  $b < 0$  cancel it in part or, in the case of  $-2E_p/\omega_0$ , even completely. This means that hopping processes that do not change the lattice configuration, i. e. the orbital occupancy, are not suppressed exponentially in the strong coupling regime. Processes with exchange in the same orbital are included in this case but not those in different orbitals. The latter change the orbital occupancy and therefore the lattice configuration. In a strong coupling approach we neglect the exponentially reduced terms. For nearest-neighbor electrons in the same orbital we have the possibility of an exchange, which eventually leads to a nearest-neighbor singlet and triplet configuration as in the HH. The singlet, a nearest-neighbor bipolaron, is the ground state if  $U < 4E_p$ . The corresponding binding energy does not depend on the coordination number of the lattice. This is an artefact of our approximation, and it is  $\Delta = t^2/E_p - 4t^2/U$ . On the other hand, the exchange of electrons in different orbitals vanishes in the strong coupling regime. The lowest energy state of electrons in different orbitals is that of unbound polarons, and it is equal to that of unbound polarons in the same orbital. The nearest neighbor states are higher in energy with a finite energy shift with respect to the unbound states.

**3 Jahn-Teller effect for systems with one particle per site** For simplicity, the spin of the electron is ignored now. Formally, the orbital has a similar meaning as the spin in the sense that it appears as an additional degree of freedom. This gives us orbital-depending effects in tunneling, very similar to spin-dependent effects [6]. In this case the ground state for sufficiently small hopping rate  $t$  compared with  $E_p$ ,  $\omega_0$  and  $U_O$  is a singly occupied lattice without phonons.

In the following we shall consider the Green's function  $G(z) = (z - H)^{-1}$  and its projection to the subspace with one fermion per lattice site and no phonon  $P_0$ . The Green's function satisfies the operator identity [7]

$$P_0(z - H)^{-1}P_0 = (P_0(z - H)P_0 - P_0HP_1(z - H)^{-1}_1P_1HP_0)^{-1}_0 \equiv (z - P_0H_0P_0 - H_{eff})^{-1}_0, \quad (3)$$

where  $P_1$  projects to the complement:  $P_1 = 1 - P_0$ .  $(\dots)_j^{-1}$  is the inverse on the projected Hilbert space. Since the Hamiltonian  $H$  preserves the total number of fermions in the system, the matrix of  $H$  in fermion-number representation is given by diagonal blocks. Consequently, the projected matrix  $H_{eff} = P_0 H P_1 (z - H)^{-1} P_1 H P_0$  acts only on the Hilbert space with a total fermion number equal to the number of lattice sites. In other words, all virtual processes in  $H_{eff}$  are creation and annihilation processes of pairs of doubly occupied and empty sites. Equation (3) is a recursion relation on the projected Hilbert spaces and can be iterated. It provides a continued-fraction representation [7]. A truncation after the first iteration by replacing  $H \rightarrow H_0$  in  $(z - H)^{-1}$  leads to the approximation of  $H_{eff}$  by the XXZ Heisenberg model

$$H_{eff} \approx \frac{t^2}{2} \sum_{\langle j,j' \rangle} \left[ a_{\uparrow\uparrow} (S_j^z S_{j'}^z - 1) + a_{\uparrow\downarrow} (S_j^x S_{j'}^x + S_j^y S_{j'}^y) \right] \quad (4)$$

with coupling constants for a lattice with  $N$  sites

$$\begin{aligned} a_{\uparrow\uparrow}(z) &= -4 \frac{e^{-2E_p/\omega_0}}{\omega_0} \tilde{\gamma} \left( \frac{U_O - z - (N-2)E_p}{\omega_0}, -\frac{2E_p}{\omega_0} \right) \quad \text{and} \\ a_{\uparrow\downarrow}(z) &= -4 \frac{e^{-2E_p/\omega_0}}{\omega_0} \tilde{\gamma} \left( \frac{U_O - z - (N-2)E_p}{\omega_0}, \frac{2E_p}{\omega_0} \right). \end{aligned} \quad (5)$$

For weak and strong electron-phonon coupling we obtain the same behavior for  $a_{\uparrow\uparrow}$ :

$$a_{\uparrow\uparrow} \sim \frac{4}{z + (N-2)E_p - U_O} \quad (6)$$

but a different behavior for  $a_{\uparrow\downarrow}$ , where

$$a_{\uparrow\downarrow} \sim a_{\uparrow\uparrow} \quad (g/\omega_0 \ll 1), \quad a_{\uparrow\downarrow} \sim 0 \quad (g/\omega_0 \gg 1). \quad (7)$$

Thus the weak-coupling limit of  $H_{eff}$  gives an isotropic Heisenberg model, whereas the strong-coupling limit leads to an Ising model.

**3.1 Three-atomic molecules** As a simple example we study the effect of the geometry on properties in the case of a molecule with three atoms ( $N = 3$ ), either in a stretched (SM) or triangular (TM) configuration. The Hilbert space of the full Hamiltonian has infinite dimensions, whereas the Hilbert space of the Hamiltonian of the projected Green's function with one electron per site and no phonon has only dimension  $d = 8$ . The problem of diagonalizing the effective Hamiltonian is further reduced by the global spin-flip symmetry to  $d = 4$ . However, the eigenvalues of  $H_{eff}$  are complicated functions of  $z$  due to the virtual hopping processes which include the creation of an arbitrary number of phonons. The projected Green's function of Eq. (3) can be expressed in terms of eigenstates and eigenvalues of the effective Hamiltonian  $H_{eff}$  as  $\langle E_j | (z - H)^{-1} | E_j \rangle = 1/(z - E_j(z))$ . The energy functions  $E_j(z)$  for the stretched and for the triangular molecule are listed in Table 1. The geometric degeneracy of the TM gives a doubly degenerate ground state for  $E_0$ . Therefore, the energy  $E_2$  is absent in this case.

**3.2 Discussion of the pole structure** The energy levels of the molecules are poles of the projected Green's functions  $1/(z - E_j)$ , i.e. they are solutions of the equation  $z = E_j(z)$ . The parameter  $z$  appears in  $a_{\uparrow\uparrow}$  and  $a_{\uparrow\downarrow}$  (and therefore in  $E_j$ ) only in the combination  $z' = z + E_p$ . In general, we expect a complex solution  $z' = x + iy$  for  $z = E_j$ . However, it turns out from the properties of the incomplete Gamma function that the imaginary part is always  $y = 0$  and that the real part satisfies an equation of the form

$$x = \frac{t^2}{2} \sum_m \frac{c_m}{x - \alpha_m} \quad (c_m \geq 0, \quad \alpha_m = \omega_0 m - 2E_p + U_O). \quad (8)$$

Consequently, there are solutions  $x_m$  with  $\alpha_m < x_m < \alpha_m + \omega_0$ .

	TM	SM
$E_0$	$-3E_p + t^2(a_{\uparrow\uparrow} + a_{\uparrow\downarrow})/2$	$-3E_p - t^2\lambda_0/2$
$E_1$	$-3E_p + t^2(a_{\uparrow\uparrow} - a_{\uparrow\downarrow})$	$-3E_p + t^2a_{\uparrow\uparrow}/2$
$E_2$	—	$-3E_p - t^2\lambda_2/2$
$E_3$	$-3E_p$	$-3E_p$

**Table 1** Energies for the TM and the SM with  $\lambda_0 = -\frac{3}{2}a_{\uparrow\uparrow} - \frac{1}{2}\sqrt{a_{\uparrow\uparrow}^2 + 8a_{\uparrow\downarrow}^2}$  and  $\lambda_2 = -\frac{3}{2}a_{\uparrow\uparrow} + \frac{1}{2}\sqrt{a_{\uparrow\uparrow}^2 + 8a_{\uparrow\downarrow}^2}$ .

	TM/ WC	TM/ SC	SM/ WC	SM/ SC
$E_0$	3	2	3	2
$E_1$	0	2	1	1
$E_2$	—	—	0	1
$E_3$	0	0	0	0

**Table 2** Values of the parameter  $\lambda$  in Eq. (9).

**3.2.1 Pole structure in the asymptotics** In weak coupling (WC) as well as strong coupling (SC), the Green's functions have only two poles for each  $E_j$ . These poles are solutions of a quadratic equation. To study all elements of the Green's functions in one case, the parameter  $\lambda$  is introduced such that the Green's function reads

$$\langle E_j | (z - H)^{-1} | E_j \rangle = \frac{1}{z' - \frac{2t^2\lambda}{z' - 2E_p - U_O}}, \quad (z' = z + 3E_p) \quad (9)$$

with the values of the parameter  $\lambda$  given in Table 2. The poles in Eq. (9) are

$$z'_{1/2} = E_p + U_O/2 \pm \sqrt{(E_p + U_O/2)^2 + 2t^2\lambda}. \quad (10)$$

The ground state is related to  $\min\{z'_1, z'_2\}$ , i.e., it is  $z'_2$ . Moreover,  $\lambda$  must be maximal. Thus  $E_0$  is the ground state, except for TM/SC, where the ground state has an additional degeneracy due to  $E_0 = E_1$ .

**4 Conclusions** It is argued in [2] that in the strong coupling regime the main source of bipolaron formation is the non-exponential off-diagonal matrix element in second order related to the exchange of neighboring electrons. In the  $E \otimes \beta$  case we found exponential decay for this exchange in the situation of different orbital occupancy. Therefore the unbound state is preferred. For one particle per site we showed that  $H_{eff}$  of the projected Green's function yields an XXZ Heisenberg model. Furthermore it should be noted that the corresponding Holstein-Hubbard model leads to an isotropic Heisenberg term. This was also discussed for the bipolaron problem [1, 2]. As an application we have discussed the spectral properties for stretched and triangular molecular configurations of three atoms.

**Acknowledgements** This work was supported by the Deutsche Forschungsgemeinschaft through Sonderforschungsbereich 484.

## References

- [1] A. Macridin, G. A. Sawatzky, and M. Jarrell, Two-dimensional Hubbard-Holstein bipolaron, cond-mat/0401305.
- [2] J. Bonča, T. Katrišnik, and S. A. Trugman, Phys. Rev. Lett. **84**, 3153 (2000).
- [3] R. Englman, Jahn-Teller Effect in Molecules and Crystals, edited by J. B. Birks (Wiley-Interscience, London, New York, Sydney, Toronto, 1972).
- [4] I. G. Lang and Yu. A. Firsov, Sov. Phys. JETP **16**, 1301 (1963).
- [5] P. J. Davis, Handbook of Mathematical Functions, edited by M. Abramowitz and I.A. Stegun (Dover Publications, Inc., New York, 1965), chap. 6.
- [6] F. Meier et al., Molecular spintronics: Coherent spin transfer in coupled quantum dots, cond-mat/0401397.
- [7] K. Ziegler, Phys. Rev. A **68**, 053602 (2003).