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Robustness of the single dressing fraction characterization of polaron structure in multi-mode partial dressing theory

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Abstract

In approaches to polaron theory which attempt to unify small and large polaron behavior, the notion of "partial dressing" plays a central role in determining the allocation of interaction energy between correlations of the small and large polaron types. This paper examines some of the consequences of generalizing from a single dressing fraction to a distribution of dressing fractions. Employing a distribution of dressing fractions allows distinct phonon modes to allocate interaction energy in distinct ratios, thereby supporting a more flexible description of polaron structure than that based on a single dressing fraction applied uniformly to all modes. The distribution of optimal dressing fractions can be characterized by a mean dressing fraction whose properties are in close agreement with the single dressing fraction used previously.

The determination of polaron structure depends centrally upon achieving an adequate description of the exciton-phonon correlations implicit in the optimal polaron state. For this purpose we distinguish between properly delocalized Schrödinger eigenstates and the localized functions which are primarily responsible for polaron structure. We obtain equations describing these localized functions and use these to derive structure information. We use the Fröhlich Hamiltonian [1]

$$H = \sum_m E a_m^+ a_m - \sum_m J a_m^+ (a_{m+1} + a_{m-1})$$
$$+ \sum_q \hbar \omega_q b_q^+ b_q + \sum_{qm} \hbar \omega_q (\chi_m^q b_q^+ + \chi_m^{q*} b_q) a_m^+ a_m. \quad (1)$$
$$\omega_q = \omega_B \sin\left(\tfrac{1}{2}|ql|\right),$$

$$\chi_n^q = \chi^q e^{-iqR_n} = \frac{-2i\chi \sin(ql)}{\sqrt{2NM\hbar\omega_q^3}} e^{-iqR_n} \qquad (2)$$

(Notation here and throughout this paper is that of Ref. [2].) To determine the optimal equations of motion we apply the time-dependent variational principle

$$\delta \int_{t_1}^{t_2} dt \langle \Psi(t) | i\hbar \frac{d}{dt} - H | \Psi(t) \rangle = 0, \qquad (3)$$

recently used by Skrinjar et al. [3] and Zhang et al. [4]. We define the trial state vector

$$|\tilde{D}(t)\rangle \equiv \sum_n \tilde{\alpha}_n(t) \tilde{a}_n^+ |0\rangle$$

$$\otimes \exp\left\{ \sum_q [\tilde{\beta}_q(t) \tilde{b}_q^+ - \tilde{\beta}_q^*(t) \tilde{b}_q] \right\} |0\rangle, \qquad (4)$$

employing "partially-dressed" operators [5-9]

$$\tilde{a}_m \equiv U a_m U^+ \equiv a_m \exp\left\{ \sum_q \delta_q (\chi_m^q b_q^+ - \chi_m^{q*} b_q) \right\}, \qquad (5)$$

$$\tilde{b}_q \equiv U b_q U^+ \equiv b_q + \sum_q \delta_q \chi_m^q a_m^+ a_m, \qquad (6)$$

$$U \equiv \exp\left[-\sum_{qm} \delta_q (\chi_m^q b_q^+ - \chi_m^{q*} b_q) a_m^+ a_m \right]. \qquad (7)$$

This state generalizes that used previously by Brown and Ivic [2] in using N dressing fractions δ_q rather than one. Applying Eq. (3) we obtain the formal evolution equations

$$i\hbar \dot{\tilde{\alpha}}_n(t) + i\hbar \sum_q \delta_q [\chi_n^q \dot{\tilde{\beta}}_q^*(t) - \chi_n^{q*} \dot{\tilde{\beta}}_q(t)] \tilde{\alpha}_n(t) = \frac{\partial \langle H \rangle}{\partial \tilde{\alpha}_n^*(t)}, \qquad (8)$$

$$i\hbar \dot{\tilde{\beta}}_q(t) - i\hbar \sum_n \delta_q \chi_n^q \frac{d}{dt} |\tilde{\alpha}_n(t)|^2 = \frac{\partial \langle H \rangle}{\partial \tilde{\beta}_q^*(t)}, \qquad (9)$$

in which the energy functional <H> is the expectation value of H in the $\tilde{D}$ state

$$\langle H \rangle = \sum_m [E - E_b^\sigma] |\tilde{\alpha}_m(t)|^2 - \sum_m \tilde{J} \tilde{\alpha}_m^*(t)$$

$$\times [\tilde{\alpha}_{m+1}(t) + \tilde{\alpha}_{m-1}(t)] + \sum_q \hbar\omega_q |\tilde{\beta}_q(t)|^2$$

$$+ \sum_{ql} (1 - \delta_q) \hbar\omega_q [\chi_l^q \tilde{\beta}_q(t)^*$$

$$+ \chi_l^{q*} \tilde{\beta}_q(t)] |\tilde{\alpha}_l(t)|^2, \tag{10}$$

where E_b^σ is the partially-reduced polaron binding energy

$$E_b^\sigma = \sum_q \delta_q (2 - \delta_q) |\chi^q|^2 \hbar\omega_q, \tag{11}$$

and $\tilde{J}$ is the renormalized transfer matrix element partially reduced from the bare value J by interactions with phonons

$$\tilde{J} = J e^{-\sigma}, \quad \sigma = 2 \sum_q \delta_q^2 |\chi^q|^2 \sin^2\left(\frac{1}{2} ql\right). \tag{12}$$

Under such circumstances that all the dressing fractions are equal ($\delta_q = \delta$) these quantities revert to the quantities appearing in [2]

$$E_b^\sigma \to \delta(2 - \delta) E_b, \quad \tilde{J} \to J e^{-\delta^2 S}, \tag{13}$$

where

$$E_b = \sum_q |\chi^q|^2 \hbar\omega_q, \quad S = 2 \sum_q |\chi^q|^2 \sin^2\left(\frac{1}{2} ql\right). \tag{14}$$

Integrating Eq. (9) and eliminating explicit phonon variables as in Ref. [2], we obtain an integro-differential equation for the dressed-exciton probability amplitudes in the form

$$i\hbar\dot{\tilde{\alpha}}_n(t) = \lfloor E - E_b^\sigma \rfloor \tilde{\alpha}_n(t) - \tilde{J}[\tilde{\alpha}_{n+1}(t) + \tilde{\alpha}_{n-1}(t)]$$

$$\sum_l K_{nl}^2(0) |\tilde{\alpha}_l(t)|^2 \tilde{\alpha}_n(t) + \sum_l K_{nl}^1(t) |\tilde{\alpha}_l(0)|^2 \tilde{\alpha}_n(t)$$

$$+ \int_0^t d\tau \sum_l K_{nl}^0(t - \tau) \frac{d}{d\tau} |\tilde{\alpha}_l(\tau)|^2 \tilde{\alpha}_n(t)$$

$$+ \tilde{f}_n(t) \tilde{\alpha}_n(t), \tag{15}$$

where

$$K_{mn}^{\mu}(t) = 2\sum_{q}(1 - \delta_q)^{\mu}\chi_m^q\chi_n^{q*}\hbar\omega_q\cos\omega_q t, \qquad (16)$$

$$\tilde{f}_n(t) = \sum_{q}\hbar\omega_q(\chi_n^q\tilde{\beta}_q^*(0)e^{i\omega_q t} + \chi_n^{q*}\tilde{\beta}_q(0)e^{-i\omega_q t}). \qquad (17)$$

This is the fundamental result of our time-dependent analysis. The sum rule

$$\delta_q(2 - \delta_q) + (1 - \delta_q)^2 - 1 \qquad (18)$$

holds mode-by-mode between the contributions to the polaron binding energy E_b^σ and the cubic non-linearity as represented by $K_{mn}^2(0)$. This implies that increases in δ_q transfer interaction energy borne by the mode q from the nonlinear terms *to* the linear terms, and vice versa. The fundamental difference between our present development and that of Ref. [2], etc. is that this redistribution of interaction energy may be nonuniform with respect to q. This nonuniformity has a significant impact on the structure of $K_{mn}^{\mu}(0)$ and hence on the form and magnitude of the nonlinear terms.

The time-dependent variation above was carried out for fixed, but arbitrary values of the dressing fractions δ_q, which can be used to "tune" the equations of motion. Determination of the optimal dressing fraction is a complex self-consistency problem. Because the q dependence of δ_q affects the form taken by the nonlinear terms in the evolution equations, it is not possible to usefully limit the class of possible solutions prior to determining δ_q; yet, δ_q, depends to some degree on the influence of the nonlinear terms. The case can be made, however, that the nonlinear terms are small and weakly structured, and therefore should not strongly shift the extrema of the energy in the large majority of cases [2]. This implies that the values of δ_q should be insensitive to the presence or absence of the nonlinear terms over large regions of parameter space. We proceed, therefore, by neglecting nonlinear terms in our determination δ_q. The impact of our approximations may be mitigated by limiting our discussion to the lowest lying states carrying the minimum kinetic energy; when the momentum contribution to the energy is significant, complexities can arise which call for further generalizations [11].

Caveats in hand, we obtain for the average energy

$$\langle H \rangle \approx E - E_b^\sigma - 2Je^{-\sigma} + \text{nonlinear terms}, \qquad (19)$$

We determine the optimal dressing fraction by minimizing <H> against variations in δ_q. We find the extrema of the energy to be given by

$$\delta_q = \frac{1}{1 + Be^{-\sigma}\sin|q/2|}, \qquad (20)$$

where $B = 4J/\hbar\omega_B$. Owing to the large number (N) of variational parameters (δ_q), qualifying extrema as maxima or minima by imposing conditions on second derivatives is impractical. Instead, we note that $\partial H/\partial\delta_q|_{\delta_q=0}$ is negative and $\partial H/\partial\delta_q|_{\delta_q=1}$ is positive for every δ_q, implying that the energy exhibits at least one proper minimum in the unit interval. In the computations which follow, this is sufficient information to allow us to classify the extrema we encounter. Fig. 1 illustrates the actual dependence of the energy on the adiabaticity B and exciton-phonon coupling strength S.

Eq. (19) can be solved by squaring and integrating over all phonon modes with the weight factors such that

$$\sigma = \sum_q \frac{2|\chi^q|^2 \sin^2(q/2)}{(1 + Be^{-\sigma}\sin|q/2|)^2}. \qquad (21)$$

In the limit $N \rightarrow \infty$, this yields

$$\sigma \equiv Sf(Be^{-\sigma}) \equiv \bar{\delta}^2 S, \qquad (22)$$

where

$$f(a) \equiv 3\int_0^{\pi/2} du\, \frac{\cos^2 u \sin u}{(1 + a\sin u)^2}, \qquad f(0) = 1$$

$$= \left\{ \frac{3\pi}{a^3} - \frac{6}{a^2} + \frac{6(a^2 - 2)}{a^3\sqrt{1 - a^2}} \arctan\sqrt{\frac{1 - a}{1 + a}} \right\}$$

$$\text{for } a < 1$$

$$= 3\pi - 9 \qquad \text{for } a = 1$$

$$= \left\{ \frac{3\pi}{a^3} - \frac{6}{a^2} + \frac{6(a^2 - 2)}{a^3\sqrt{a^2 - 1}} \operatorname{arctanh}\sqrt{\frac{a-1}{a+1}} \right\}$$

$$\text{for } a > 1. \quad (23)$$

The transcendental relation (22) can be solved for σ, or equivalently for $\bar{\delta}$, without specific knowledge of any of the individual δ_q's, which can be determined subsequently from Eq. (20). Since $\bar{\delta}$ is a weighted geometric mean of all δ_q's, it is reasonable to expect $\bar{\delta}$ to play the same role within this multiple-dressing fraction analysis as the single dressing fraction δ played in previous work [2]. Solving the relation (22) in terms of $\bar{\delta}$, we find that this is indeed so. The main result is a "phase diagram" which maps out the different characteristics of the polaron according to parameter regime (See Fig. 2).

The interpretation of this diagram is essentially the same as that given in Ref. [2] and in greater detail in Ref. [12]; noting the differing conventions for the adiabaticity parameter in the prior and present works, the critical point marking the onset of the self-trapping transition in our present analysis is within a few percent of our prior less-general treatment. Agreement in the balance of the phase diagram is similarly close. This demonstrates that the allocation of interaction energy between the small and large polaron correlation mechanisms depends primarily on the average dressing fraction and is only weakly sensitive to the detail of the organization of interaction energy according to phonon wave vector.

We can use these results to determine the complete set of dressing fractions through the relation (20). Illustrative results are shown in Fig. 3. We see that in general the degree to which a given mode participates in the dressing of the exciton depends on the self-consistent adiabaticity $Be^{-\sigma}$. To the degree that this measure of adiabaticity is small, all modes participate strongly in the dressing. The longest wavelength acoustic phonons remain strongly dressed for essentially all realizable parameters, though the mean dressing fraction can approach zero if the self-consistent adiabaticity is sufficiently strong. While the shifting of interaction energy among length scales implied by the variation of δ_q with wave vector affects the phase diagram only in an average sense, this variation should have a significant impact on the shape of the self-consistent wave function since an increase in δ_q (for example) diminishes the self-focussing effect contributed by nonlinear terms on that length scale. In particular, the pinning of δ_q near unity for small q implies an essential depletion of self-focussing on the longest length scales relative to more familiar nonlinear theories of the large polaron.

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