ANALYTICAL PROPERTIES OF POLARON OBSERVABLES: ON THE PERTURBATION SERIES OF THE POLARON FUNCTIONAL INTEGRAL

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ABSTRACT

It was frequently claimed that polarons should undergo a formal phase transition from a mobile to a localized state, if the electron-phonon coupling parameter α exceeds some critical value. We disprove this assertion for the class of Fröhlich models. To do so, we analyze the perturbation series of the reduced propagator up to infinite order in α and establish its analytical behaviour for all values of α. Our results can be generalized to the case of magnetopolarons, polarons in an external potential and to polaronic excitons.

1. Introduction

Polarons are the eigenstates of a coupled electron-(LO)phonon system. They are quasiparticles in the sense of Landau's[1] definition and well established by many experiments. The theoretical basis for a description of polarons was provided by a famous paper of Fröhlich, Pelzer and Zienau[2]; these authors proposed a specific particle-field model, which now bears Fröhlich's name. Before long, this model proved to be of basic importance for various branches of solid-state physics and has attracted the attention of numerous physicists until today. The corresponding Hamiltonian reads as follows:

$$H \;:=\; H_P + H_{Ph} + H_I \,, \tag{1}$$

where

$$H_P \;=\; \frac{\mathbf{p}^2}{2m} \,, \tag{2}$$

$$H_{Ph} = \sum_{\mathbf{k}} \hbar\omega(k)\, a^{\dagger}(\mathbf{k})\, a(\mathbf{k})\,, \tag{3}$$

$$H_I = \sqrt{\frac{\alpha}{V}} \sum_{\mathbf{k}} \{\, g(k)\; a(\mathbf{k})\, e^{i\mathbf{k}\cdot\mathbf{r}} + h.c.\,\}\,. \tag{4}$$

Here, $\mathbf{p}$ and $\mathbf{r}$ indicate momentum and position of the electron; $\mathbf{k}$, $\omega(k)$ and $a(\mathbf{k})$ denote the wave vector, dispersion and the field operator of the phonon, V is the quantization volume. The space dimension D is arbitrary. Finally, $g(k)$ is the electron-phonon coupling function, a dimensionless coupling parameter $\sqrt{\alpha}$ being extracted. Without loss of generality, we may assume $g(k)$ to be real. The standard model, introduced by Fröhlich, Pelzer and Zienau[2] assumes additionally

$$D = 3, \qquad \omega(k) = \omega, \qquad g(k) = \sqrt{4\pi}\, \sqrt[4]{\frac{\hbar}{2m\omega}}\, \frac{\hbar\omega}{k}\,. \tag{5}$$

Our discussion, however, is not restricted to that case (see sections 3 and 4).

We insert a technical remark: At the moment, the volume V will be kept finite to guarantee a discrete wave-vector spectrum. The reason is that thereby we can avoid technical subtleties in connection with trace operations, which will be introduced below. We stress, however, that there exists no principle difficulty to use a mathematically well defined, continuous $\mathbf{k}$-vector version of H from the very beginning. Formally, all $\mathbf{k}$-summations in eqs. (3) and (4) have to be replaced by integrations and in addition $g(k)/\sqrt{V}$ by $g(k)/(2\pi)^{3/2}$. Publications of a more mathematical character usually prefer this starting point (see section 2). Later on (in section 3) we shall evaluate all $\mathbf{k}$-summations in the limit $V \to \infty$.

In this article we shall discuss the analytical properties of polaron observables as functions of α and other parameters. Functional-integration techniques will prove extremely useful, as can generally be experienced in connection with polaron physics. The extraordinary power of the method was already demonstrated in the early, pioneering paper of Feynman[3]. In combination with a variational principle, Feynman received results for the ground-state energy, which represent the state of the art until today. We choose the same starting point as Feynman and introduce the matrix element

$$U(\mathbf{r};\alpha,B) \;:=\; \mathrm{tr}_{Ph} < \mathbf{r}\,|\, e^{-BH}\,|\,\mathbf{0} > \tag{6}$$

of the reduced propagator. Here, $\mathrm{tr}_{Ph}\ldots$ indicates the trace operation concerning phonons, $\mathbf{r}$ and $\mathbf{0}$ are particle positions and B is a positive parameter; this may be interpreted as formal inverse temperature. Static and dynamical properties of the system are derivable from $U(\mathbf{r};\alpha,B)$ in a familiar way. One should notice that

$$U(\mathbf{r}-\mathbf{r}';\alpha,B) \;=\; \mathrm{tr}_{Ph} < \mathbf{r}\,|\,e^{-BH}\,|\,\mathbf{r}' > \tag{7}$$

holds because of translational symmetry; insofar eq. (6) covers the general case.

It proves useful to relate $U(\mathbf{r};\alpha,B)$ to the readily accessible quantity $U(\mathbf{r};0,B)$ of an uncoupled electron-phonon system. Then, functional integration may be used to derive (see, e.g. Schultz[4])

$$U(\mathbf{r};\alpha,B) \;=\; U(\mathbf{r};0,B)\times Z(\mathbf{r},\alpha,B)\,, \tag{8}$$

where

$$Z(\mathbf{r},\alpha,B) \;=\; \langle\,\exp(-S_I)\,\rangle\,. \tag{9}$$

The right-hand side is a specific expectation value; the general definition is

$$\langle\,A\,\rangle \;:=\; \frac{\int \delta^D R\,\exp(-S_0[\mathbf{R}])\,A[\mathbf{R}]}{\int \delta^D R\,\exp(-S_0[\mathbf{R}])}\,. \tag{10}$$

In (10), $\int \delta^D R\ldots$ is to indicate Wiener-Feynman integration over all real, closed D-dimensional paths $\mathbf{R}(\tau)$ with $\mathbf{R}(0)=\mathbf{R}(1)=\mathbf{0}$. Moreover,

$$S_0[\mathbf{R}] \;:=\; \int_0^1 d\tau\,\frac{\gamma}{2}\,\dot{\mathbf{R}}^2(\tau) \qquad,\quad \gamma := \frac{m}{\hbar^2 B}\,, \tag{11}$$

$$S_I[\mathbf{R}] \;:=\; -\frac{\alpha B^2}{V}\sum_{\mathbf{k}}|g(k)|^2\int_0^1\!\!\int_0^1 d\tau d\tau'\;G(\tau-\tau')\,e^{i\mathbf{k}\cdot[\mathbf{R}(\tau)-\mathbf{R}(\tau')+(\tau-\tau')\mathbf{r}]} \tag{12}$$

and

$$G(\tau) \;:=\; \frac{\cosh\left[B\hbar\omega(k)\,(1/2-|\,\tau|)\right]}{2\sinh\left(B\hbar\omega(k)/2\right)}\,. \tag{13}$$

The functional integral (9), containing the noninstantaneous self interaction $S_I[\mathbf{R}]$, is the central quantity of this article. To the best of present knowledge, it cannot be evaluated in closed form. However, one should notice that eq. (9) is ideally suited as a starting point for a perturbative treatment of the electron-phonon interaction: Inserting the power series for $\exp(-S_I)$, one arrives immediately at a power-series representation of $Z(\mathbf{r}, \alpha, B)$ and $U(\mathbf{r}; \alpha, B)$ as a function of α, if the integration can be done term by term and if the generated series exists. Provided this is true, one may use the familiar mathematical theorems for power series to derive the analytical properties of many polaron observables of interest. This will be done in section 3. We found it useful to premise a short compilation of functional-analytic results concerning the Hamiltonian (1); the main reason is that these results seem to be widely unknown, but are highly relevant in the context of analyticity problems. In section 4 we list some extensions of the results, which we shall present in section 3.

2. Some analyticity results of operator theory

As indicated above, we use a continuous **k**-vector version of the Hamiltonian H. Because of translation symmetry H commutes with the operator

$$\mathbf{P}_{tot} \;:=\; \mathbf{p} + \int d^Dk\; \hbar\mathbf{k}\; a^\dagger(\mathbf{k})\, a(\mathbf{k}) \;=\; \mathbf{p} + \mathbf{P}_{Ph} \tag{14}$$

of total momentum. Following Lee, Low and Pines[5], we can profitably use this fact to eliminate the electron coordinates from H; we define the unitary transformation

$$U \;:=\; \exp\Big(-\frac{i}{\hbar}\mathbf{r}\cdot\mathbf{P}_{Ph}\Big) \tag{15}$$

and calculate $\mathbf{P}'_{tot} := U^{-1}\mathbf{P}_{tot}U$ and $H' := U^{-1}HU$. The result is

$$\mathbf{P}'_{tot} \;=\; \mathbf{p}\,, \tag{16}$$

$$H' \;=\; H'_P \;+\; H_{Ph} \;+\; H'_I\,, \tag{17}$$

where

$$H'_P \;:=\; \frac{(\mathbf{p}-\mathbf{P}_{Ph})^2}{2m}\,, \tag{18}$$

$$H'_I \;:=\; \sqrt{\alpha}\int d^Dk\;\{g(k)\; a(\mathbf{k}) + h.c.\}\,. \tag{19}$$

Conservation of total momentum is now equivalent to $[H', \mathbf{p}] = \mathbf{0}$ and permits us to restrict H' to that subspace of the total Hilbert space, which is spanned by the eigenfunctions of $\mathbf{p}$ with a given eigenvalue $\hbar\mathbf{Q}$. This restriction leads to the Hamiltonian

$$H'(\mathbf{Q}) \;=\; \frac{(\hbar\mathbf{Q} - \mathbf{P}_{Ph})^2}{2m} + H_{Ph} + H'_I \;=:\; H'_0(\mathbf{Q}) + H'_I \qquad (20)$$

of Lee, Low and Pines. Clearly, $H'(\mathbf{Q})$ is defined on the Fock space of phonons alone, the electron coordinates being eliminated. Moreover, it is sufficient to discuss $H'(\mathbf{Q})$ instead of H. Let $E(\alpha, \mathbf{Q})$ be the ground-state energy of $H'(\mathbf{Q})$. The reader will notice that the existence of $E(\alpha, \mathbf{Q})$ is directly connected with a proper mathematical definition of $H'(\mathbf{Q})$, which in turn presupposes the specification of admissable functions $\omega(k)$ and $g(k)$. For the moment, we take the existence of $E(\alpha, \mathbf{Q})$ for granted. Even more: assume $E(\alpha, \mathbf{Q})$ to be a simple eigenvalue of $H'(\mathbf{Q})$, the corresponding eigenfunction being $\psi(\alpha, \mathbf{Q})$. One may then ask: What is the domain of analyticity of $E(\alpha, \mathbf{Q})$ and $\psi(\alpha, \mathbf{Q})$ as functions of $\alpha, \mathbf{Q}$? The complete answer is contained in the following statement: *Assume*

$$\omega(k) \geq \omega > 0\,, \qquad \omega(k_1) + \omega(k_2) \geq \omega(|\mathbf{k}_1 + \mathbf{k}_2|)\,, \qquad (21)$$

$$\int d^D k \, \frac{|g(k)|^2}{1 + (ak)^2} \;<\; \infty \qquad (22)$$

to be valid, where $a := \sqrt{\hbar/m\omega}$ *is the polaron radius. Then, the ground-state energy* $E(\alpha, \mathbf{Q})$ *of* $H'(\mathbf{Q})$ *exists and is an isolated, simple eigenvalue for* $0 \leq \alpha < \infty$, $\hbar^2 Q^2/2m < \hbar\omega$. $E(\alpha, \mathbf{Q})$ *and* $\psi(\alpha, \mathbf{Q})$ *are real analytic functions of* $\alpha, \mathbf{Q}$ *in the specified domain* (as for a proof, we refer to Fröhlich[6] and the detailed discussion of Spohn[7] and Gerlach and Löwen[8]).

Clearly, this statement provides a qualitative analytical analysis of the standard model (5) for optical polarons. More general, one will notice that many ground-state observables can be calculated as derivatives of $E(\alpha, \mathbf{Q})$ with respect to $\mathbf{Q}$ or as expectation values of type $\langle\, \psi(\alpha, \mathbf{Q}) \,|\, X \,|\, \psi(\alpha, \mathbf{Q}) \,\rangle$, X being an operator independent of α and $\mathbf{Q}$. As examples, we mention the polaron mass, the polaron radius and the mean phonon number associated with $\psi(\alpha, \mathbf{Q})$. The above statement assures us that these observables are smooth functions of $\alpha, \mathbf{Q}$, provided $\hbar^2 Q^2/2m < \hbar\omega$ is valid. Interestingly enough, the latter condition can totally be removed for $D = 1, 2$ and under slightly stronger conditions as (21), (22); for $D = 3$, the domain of Q can be extended, but Q remains finite (see Spohn[9]).

3. On the Pertubation Series of the Polaron Functional Integral

In this section we perform a detailed perturbative discussion of the matrix element $U(\mathbf{r}; \alpha, B)$, which we introduced in eq. (6) and represented by a functional integral in eqs. (8) and (9). Clearly, it is sufficient to study the properties of $Z(\mathbf{r}, \alpha, B)$. To do so, let us consider

$$Z_N(\mathbf{r}, \alpha, B) \;:=\; \langle \sum_{n=0}^{N} \frac{1}{n!}(-S_I)^n \rangle \;=\; \sum_{n=0}^{N} \frac{1}{n!} \langle (-S_I)^n \rangle \; . \tag{23}$$

Now, $-S_I$ is positive definite; this can easily be proven, if one inserts a Fourier series representation for $G(\tau - \tau')$ into eq. (12). Moreover, direct inspection of S_I shows that we may represent

$$\frac{1}{n!} \langle (-S_I)^n \rangle \;=:\; \alpha^n f_n(\mathbf{r}, B) \, , \tag{24}$$

where $f_n(\mathbf{r}, B)$ is some positive function to be discussed below. Combining (23) and (24), we find that $Z_N(\mathbf{r}, \alpha, B)$ is strictly positive and monotonically increasing as function of N ($\mathbf{r}, \alpha$ and B being fixed). If we can prove that $Z_N(\mathbf{r}, \alpha, B)$ is uniformly bounded from above by some function $C(\mathbf{r}, \alpha, B) < \infty$, we can apply the monotone convergence theorem to assure that $\lim_{N\to\infty} Z_N(\mathbf{r}, \alpha, B) =: Z_\infty(\mathbf{r}, \alpha, B)$ exists and

$$Z(\mathbf{r}, \alpha, B) \;=\; \sum_{n=0}^{\infty} \alpha^n f_n(\mathbf{r}, B) \, . \tag{25}$$

On the other hand, we may assume that $Z(\mathbf{r}, \alpha, B)$ exists. Then, $Z(\mathbf{r}, \alpha, B) \geq Z_N(\mathbf{r}, \alpha, B)$ is automatically true and (25) holds again. Insofar, $Z(\mathbf{r}, \alpha, B)$ exists if and only if this is true for $Z_\infty(\mathbf{r}, \alpha, B)$.

Consequently, we examine (25) in more detail. We anticipate that the functional-integral part in $f_n(\mathbf{r}, B)$ can be evaluated in closed form (see eq. (30) below). This leaves us with a finite-dimensional integral, which depends analytically on B, if $B > 0$ is fulfilled.

We now perform an analytical continuation: Despite its introduction as a function of positive α and B, the right-hand side of (25) may be discussed as an infinite series of complex α and B. This complex series converges absolutely for all α and $0 < \mathrm{Re}B < \infty$, if (and only if) the original series (25) converges for $0 \leq \alpha < \infty$, $0 < B < \infty$. To prove this, one has to recall that $f_n(\mathbf{r}, B)$ is positive, if B is positive; for complex B, one derives

$$|f_n(\mathbf{r},B)| \leq \left(\frac{|B|}{\mathrm{Re}B}\right)^{2n} f_n(\mathbf{0},\mathrm{Re}B) \tag{26}$$

by direct inspection.

Having in mind that $f_n(\mathbf{r},B)$ is an analytical function of B for $0 < \mathrm{Re}B < \infty$, we may state the following result: *(25) exists as a complex series for all α and $0 < \mathrm{Re}B < \infty$ and represents an analytical function of α and B in the quoted domain, if (25) exists as a real series for $0 \leq \alpha < \infty$, $0 < B < \infty$ and arbitrary $\mathbf{r}$.*

The remaining task is to assure the convergence of the real series (25). We mention two *sufficient conditions for the existence of (25) as a real series of α, B and $\mathbf{r}$ for $0 \leq \alpha < \infty$, $0 < B < \infty$ and arbitrary $\mathbf{r}$:*

$$\int d^Dk \, \frac{|g(k)|^2}{\omega(k)} < \infty \qquad \text{(short-range case)}\,, \tag{27}$$

or

$$\omega(k) \geq \omega > 0\,, \qquad |g(k)|^2\, k^{D-1} \leq const. \qquad \text{(long-range case)}\,. \tag{28}$$

To prove this assertions, we begin with the comparatively simple case of short-range coupling. To get an upper bound on an arbitrary term in the series (25), we may replace the exponential factor in $S_I[\mathbf{R}]$ by 1 (see eq. (12)). Using condition (27), a positive function $C(B) < \infty$ exists such that

$$f_n(\mathbf{r},B) < \frac{C(B)^n}{n!}\,. \tag{29}$$

Clearly, this inequality proves the convergence of (25).

The case of long-range coupling is much more involved. Inserting the formula (12) for S_I into $\langle (-S_I)^n \rangle$, the functional integral is of Gaussian type and can be evaluated. We find for general $g(k)$ and $n > 0$ ($f_0 = 1$):

$$\begin{aligned} f_n(\mathbf{r},B) = {} & \frac{1}{n!}\left(\frac{B^2}{(2\pi)^3}\right)^n \int d^Dk_1 \ldots d^Dk_n \; |g(k_1)|^2 \ldots |g(k_n)|^2 \\ & \times \int_0^1 d\tau_1 \ldots d\tau_{2n} \, \exp\left\{ -\frac{\hbar^2 B}{m} \sum_{j,l=1}^{n} A_{jl}(\tau_1,\ldots,\tau_{2n})\, \mathbf{k}_j \cdot \mathbf{k}_l \right\} \\ & \times \prod_{j=1}^{n} G_j(\tau_{2j-1}-\tau_{2j}) \exp\left\{ i\,(\tau_{2j-1}-\tau_{2j})\, \mathbf{k}_j \cdot \mathbf{r} \right\} \end{aligned} \tag{30}$$

Here, the matrix A_{jl} is defined as

$$A_{jl}(\tau_1,\ldots,\tau_{2n}) \;:=\; \frac{1}{4}\Big\{|\tau_{2j-1}-\tau_{2l}|+|\tau_{2l-1}-\tau_{2j}|-|\tau_{2j-1}-\tau_{2l-1}| \\ -|\tau_{2j}-\tau_{2l}|-2(\tau_{2j-1}-\tau_{2j})(\tau_{2l-1}-\tau_{2l})\Big\} \qquad (31)$$

and $G_j(\tau)$ is the kernel function (13), k being replaced by k_j. We remark that inequality (26) can directly be derived from eq. (30).

In a former publication[10] two of the present authors studied the diagonal element $U(\mathbf{0};\alpha,B)$ and calculated a uniform upper bound on $f_n(\mathbf{0},B)$. Because of $f_n(\mathbf{r},B) < f_n(\mathbf{0},B)$ the present problem to find an upper bound on $f_n(\mathbf{r},B)$ can be reduced to the former one. The final result is

$$f_n(\mathbf{r},B) \;\leq\; C_1(B)\cdot\frac{C_2(B)^n}{\sqrt{n!}}\,, \qquad (32)$$

where $C_1(B)$ and $C_2(B)$ are finite positive functions of B. Therefore, convergence of (25) is guaranteed again.

We add a few comments: Firstly, the results of this section complement nicely those of section 2. A direct comparison of the former conditions (eqs. (21), (22)) and the present ones (eqs. (27), (28)) demonstrates that the "finite temperature" version of the analyticity proof is surprisingly general. A main reason for this fact is the simple α- and B-dependence of the actions S_0 and S_I. Secondly, we can exclude any phase-transition like behaviour in the standard polaron systems; they are all covered by the stated conditions. Phenomena as self trapping and mass stripping do not occur within this frame. It is well known that a nonanalytical behaviour was reported in many publications. Most of these results were obtained by variational methods; a survey of the existing literature can be found in ref.[8]. Without underestimating the merits of the variational results as such, the nonanalyticities have to be classified as artifacts of the approximations made.

4. Extensions

In this section we indicate some generalizations of the results in section 3. The simplest one is concerned with an electronic coupling to several phonon branches, e.g. acoustical and optical ones. For a long time such a system was thought to be a candidate for phase-transition like behaviour. One proves by direct inspection of S_I, that the above analyticity statements can be correspondingly generalized, if the quoted conditions (27) or (28) are fulfilled for every branch.

It is also simple to remove the condition of isotropy for $\omega(\mathbf{k})$ and $g(\mathbf{k})$. The conditions (27) or (28) for $\omega(k)$ and $g(k)$ have to be replaced by generalized ones for $\omega(\mathbf{k})$ and $g(\mathbf{k})$. A specific example, which was extensively treated in the literature, is $g(\mathbf{k}) \propto |M\mathbf{k}|^{-1}$. Here M is a real, symmetric matrix with strictly positive eigenvalues.

Polarons in an external field have attracted a particular attention. We firstly turn to magnetopolarons. A homogeneous magnetic field $\mathbf{H}$ will lead to a matrix element $U(\mathbf{r}, \alpha, \mathbf{H}, B)$ instead of $U(\mathbf{r}, \alpha, B)$. Eq. (9) will change insofar, as S_I has to be supplemented by a purely imaginary term, which depends linearly on $\mathbf{H}$. Revisiting our estimation procedure, one finds that $|Z_N(\mathbf{r}, \alpha, \mathbf{H}, B)|$ is bounded by $Z_N(\mathbf{r}, \alpha, B)$. Therefore, the convergence of the power series for $Z(\mathbf{r}, \alpha, \mathbf{H}, B)$ is guaranteed, if the convergence of the power series for $Z(\mathbf{r}, \alpha, B)$ is established. We conclude that magnetopolaron observables as energy and mass show an analytical behaviour as functions of $\alpha, \mathbf{H}, B$ for $0 \leq \alpha < \infty$, $0 < |\mathbf{H}| < \infty$, $0 < B < \infty$, if the inequalities (27) or (28) are fulfilled.

A scalar potential $\lambda v(\mathbf{r})$ is the second example for an external field. We have extracted a dimensionless coupling constant λ, which we assume to be positive. Again one can generalize the analyticity proof, now for a matrix element $U(\mathbf{r}, \alpha, \lambda, B)$, if the additional inequality

$$|\tilde{v}(\mathbf{k})|\, k^2 \;\leq\; const. \tag{33}$$

is true, where $\tilde{v}(\mathbf{k})$ denotes the Fourier transform of $v(\mathbf{r})$.

It is interesting to compare these results with the ground-state results of operator theory. They are completely analogeous with the exception of the last example. If one discusses $E(\alpha, \lambda)$, the ground-state energy as function of α and λ, and assumes $v(\mathbf{r})$ to be of short-range attractive type (strictly speaking, $v(\mathbf{r})$ should be an element of the Rollnik class), a nonanalyticity may occur at a certain value of λ. This phenomenon is a so-called pinning transition (as for details, we quote ref.[10]). Pinning does not occur for a long-range potential of e.g. Coulomb type. In view of our above results we conclude that finite temperature destroys the pinning transition under all circumstances, provided, inequality (33) is guaranteed.

We close with a short comment on excitons. Introducing a electron-hole potential $\lambda v(\mathbf{r})$, we can repeat the analyticity results for the bound polaron.

5. Acknowledgements

We are grateful for financial support by "Deutsche Forschungsgemeinschaft" and "Volkswagenstiftung".

6. References

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