Proof of the Nonexistence of Formal Phase Transitions in Polaron Systems, Exposed to a Homogeneous Magnetic Field

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Abstract

In the recent literature it was frequently claimed, that a polaron system, described by the parameters α (electron-phonon coupling), *B* (magnetic field) and *T* (temperature) should undergo a phase transition for certain values of (α, B, T) . As usual, such a transition would lead to a discontinuity in the free energy $F = F(\alpha, B, T)$ or its derivatives. We prove that no such transition exists. $F = F(\alpha, B, T)$ is a real analytical function in α and *B* for all *T* (including T = 0). The shortcoming of all previous "proofs" is, that they all use variational procedures, which may cause artificial discontinuities. In fact, some authors have mentioned themselves, that their results are variational, so that the discontinuities which they encounter could be artifacts of their approximation. Our results can readily be transferred to the case of lower spatial dimensions. Further extensions to related systems like quantum well structures are possible.

1. Introduction to the phase transition problem

The physics of polarons and polaron-like systems in a homogeneous magnetic field has gained considerable interest during the last years, both theoretically and experimentally. In the theoretical description of continuum (large) polarons, the Fröhlich Hamiltonian [1] was proven to be fundamental. In the effective mass approximation, it reads as follows

$$H = (\mathbf{p} + |\mathbf{e}|\mathbf{A}(\mathbf{r}))^2 / (2m) + \int d^d k \hbar \omega(\mathbf{k}) a^+(\mathbf{k}) a(\mathbf{k})$$
$$+ \int d^d k \alpha^{1/2} (\mathbf{g}(\mathbf{k}) a(\mathbf{k}) \exp(i\mathbf{k}\mathbf{r}) + h.c.)$$
(1)

Here, e, m, r, p are the charge, mass, the position and momentum operator of the electron respectively, whereas k, $\omega(k)$, g(k), $a^+(k)$, a(k) are the wave vector, frequency, coupling, creation and annihilation operators of the phonons. α is the dimensionless electron-phonon coupling parameter, ddenotes the spatial dimension; all vectors are d-dimensional. The vector potential A describes a homogeneous magnetic field B along the z-axis, B = (0, 0, B). Throughout this paper we work in the Landau gauge

$$A(\mathbf{r}) = \begin{cases} (0, Bx, 0) & \text{for } d = 3\\ (0, Bx) & \text{for } d = 2 \end{cases}, \quad B > 0, \tag{2}$$

x being the first component of the vector $\mathbf{r} = (x, y, z)$. The resulting elementary excitation is called magneto-polaron.

In the literature mainly Fröhlich-type models, describing the coupling of a conduction electron to one branch of optical phonons in polar semiconductors, are considered, i.e.,

$$\omega(\mathbf{k}) = \omega_0 > 0 \tag{3}$$

and

$$g(k) \sim |k|^{-(d-1)/2},$$
 (4)

but also the coupling to acoustical phonons may be important, see e.g., the recent work of Kato, Tokuda [2]. Therefore, we keep $\omega(k)$ and g(k) general, as far as possible.

For a small magnetic field, the magneto-polaron behaves qualitatively like a free polaron, whose physical properties are changed by B in a perturbational sense. However, if B gets very large, one expects a significantly anisotropic behaviour: Perpendicular to the field, the phonon cloud tends to be stripped off, the corresponding mass approaching a bare electron mass; parallel to the field, no such stripping occurs. This picture is strongly supported by calculations of Devreese and Peeters [3, 4].

Comparing the low-field and high-field case, the keyquestion to be answered is whether or not a discontinuous stripping transition at a critical value of *B* between these two extreme situations takes place. Such a formal phase transition is understood as a discontinuity in the ground state energy $E(\alpha, B)$ resp. the free energy $F(\alpha, B, T)$ of the magnetopolaron or their derivatives. We prove that no such transitions exists. It is clear that this question can only be decided within a careful mathematical analysis without using any a priori approximation.

Up to now this question was merely attacked by variational calculations, where in fact a nonanalytical behaviour of the physical quantities was found. However, our proof shows that the true physical quantities are smooth and that the discontinuities, found in several variational calculations, are artifacts of the approximation made. Concerning these calculations, we mention Peeters and Devreese, who calculate the ground state energy, free energy [3, 5], the polaron mass [3, 6], the polaron radius [4] as well as the number of virtual phonons in the ground state [4] and the magnetoabsorption spectrum [7] within the anisotropic Feynman approximation for a three-dimensional Fröhlich magnetopolaron. A similar non-analytical behaviour is found for a two-dimensional polaron (see the work of Wu Xiaoguang, Peeters and Devreese [8]). Peeters and Devreese indicate themselves, that this nonanalytical behaviour might be an artifact of their approximation.

Further variational calculations, which exhibit nonanalyticities, are due to Lepine, Matz [9] and Lepine [10].

Of course, the approximations indicate large changes in the polaron quantities, which may cause interesting experimental effects (see Devreese [11]) but these changes are continuous. From a fundamental point of view, however, the properties of Fröhlich-polarons cannot be classified within a phase transition concept.

4) Concerning the organization of the present paper, we

shall concentrate firstly on the zero-temperature (groundstate) case and study spectral properties of the momentumdecomposed Hamiltonian in the second chapter. We sketch the main steps of the functional analytical proof, which in mathematical detail will be published elsewhere [12]. The final result is, that the ground state is real analytic in all parameters.

In the third chapter we turn to the finite temperature case. This requires a technically different method, namely the path integral technique. We summarize results, which are partially published [13] and present a new functional integral expression for the momentum-decomposed partition function. Finally we consider a quantum-well structure in a magnetic field.

2. Spectral properties of a magneto-polaron (T = 0)

In this chapter, we assume an optical dispersion relation,

$$\min \omega(\mathbf{k}) = \omega_0 > 0, \quad \omega(-\mathbf{k}) = \omega(\mathbf{k}) \tag{5}$$

Moreover, the coupling should satisfy

$$\int d^{d}k |g(k)|^{2}/(1 + k^{2}) < \infty, \quad g(-k) = g(k)$$
(6)

We firstly treat the case d = 3.

We note that the second and third component of the total momentum P, defined as

$$\boldsymbol{P} = \boldsymbol{p} + \boldsymbol{P}_{\rm ph} \tag{7}$$

where

$$\boldsymbol{P}_{ph} = \int d^d \boldsymbol{k} \, \hbar \boldsymbol{k} a^+(\boldsymbol{k}) a(\boldsymbol{k}) \tag{8}$$

is the phonon momentum, commute with H and are conserved. To exploit this fact explicitly, we use the unitary Lee-Low-Pines-transformation $U = \exp(-iP_{ph}r)$ to get

$$U^{-1}HU = (\boldsymbol{G} - \boldsymbol{P}_{ph})^2 / (2m) + \int d^3k \ \hbar\omega(\boldsymbol{k})a^+(\boldsymbol{k})a(\boldsymbol{k}) + \alpha^{1/2} \int d^3k (g(\boldsymbol{k})a(\boldsymbol{k}) + \text{h.c.}), \qquad (9)$$

where

$$G = (p_1, |e|Bx + p_2, p_3).$$
(10)

Since eq. (9) does not depend on y and z, we may replace p_2 and p_3 , now playing the role of the total momentum, by c-numbers Q_2 and Q_3 . By this substitution we obtain the momentum-decomposed Hamiltonian H(Q), $Q = (0, Q_2, Q_3)$. It is useful to study the spectral properties of H(Q)instead of those of H, as the Hamiltonian H(Q) can be expected to have a discrete ground state which is energetically separated by a Q-dependent gap from the rest of the spectrum. This is desired to make the technical proof easier. On the other hand, we have introduced a new parameter Q and the spectrum of H(Q) has additionally to be studied as a function of Q.

Let E(Q) denote the ground state energy of H(Q) (we drop the α - and *B*-dependence for a while). Using functional analytical methods of Fröhlich [14] and equations (5), (6) from above, we can prove the *existence* of a ground-state, arriving at the following statement:

Let $\mathbf{k} = (0, k_2, k_3)$ and suppose that

$$\min \left[E(\boldsymbol{Q} - \boldsymbol{k}) + \hbar \omega(\boldsymbol{k}) \right] - E(\boldsymbol{Q}) \equiv \Delta(\boldsymbol{Q}) > 0 \quad (11)$$

The the energy interval $[E(Q), E(Q) + \Delta(Q)]$ contains merely discrete eigenvalues, i.e., the ground state is separated by a gap of magnitude of at least $\Delta(Q)$ from the continuous spectrum of H(Q).

For sake of brevity we only sketch the main ideas of the proof of this statement. For a complete mathematical proof the interested reader is referred to [12] and [15]. We firstly introduced a UV cutoff in the coupling g(k) and put the phonon momentum space on a lattice. Consequently, we arrive at a system with finite degrees of freedom, which has a discrete spectrum. Then we have to remove the lattice cutoff and the UV cutoff consecutively. Removing the lattice cutoff, the excitations with one real phonon present form a continuous spectrum, which is separated by a finite gap from the ground state. Taking the conservation of the second and third component of the total momentum into account, the magnitude of this gap is at least $\Delta(Q)$, where $\Delta(Q)$ is given by eq. (11). Hence, all that remains to do is to remove the UV-cutoff. It is known from an early paper of Gross [16] that this can be achieved without any divergencies by a canonical dressing transformation (see also Fröhlich [14]). As the UVcutoff is removed, the UV-cutoff Hamiltonian converges to $H(\mathbf{Q})$ in norm resolvent sense. Consequently, the corresponding energy interval $[E(Q), E(Q) + \Delta(Q)]$ remains discrete and the proof of our statement above is finished.

As a next step the *uniqueness* of the ground state of H(Q) is proven, using a generalization of the Perron-Frobenius theorem, i.e. by showing that the resolvent of H(Q) has a strictly positive numerical kernel in a fixed representation. As for details, we again refer to [12]. Then, to verify (11), we need more information about E(Q) as a function of Q. In [12], the following properties are proved:

$$E(\mathbf{Q}) = E((0, 0, |\mathbf{Q}_3|)) \tag{12}$$

$$E(\boldsymbol{O}) \leq E(\boldsymbol{Q}) \leq E(\boldsymbol{O}) + Q_3^2/(2m)$$
(13)

Equation (12) tells us that the ground state of H is highly degenerate, because is doesn't depend on the second component of the total momentum. For $\alpha = 0$ this fact is well-known, for $\alpha > 0$ this was already pointed out by Devreese [17].

Using eqs. (13) and (5), one deduces immediately that eq. (11) is fulfilled if

$$Q_3^2 < 2m\hbar\omega_0 \tag{14}$$

This means that there exists a discrete unique ground-state of H(Q), if eq. (14) is satisfied.

In Ref. [15] another bound for
$$E(\mathbf{Q})$$
 is derived, namely
 $E(\mathbf{Q}) \leq \min_{\mathbf{k}} \left[E(\mathbf{Q} - \mathbf{k}) + \hbar\omega(\mathbf{k}) \right] \leq E(\mathbf{O}) + \hbar\omega(\mathbf{Q})$ (15)

By a trial-function argument and an application of the minimax principle it can be proved (see again Ref. [15]), that the continuum edge of H(Q) begins exactly at the energy $E(Q) + \Delta(Q)$. $\Delta(Q)$ is exactly the gap energy which separates the ground state from the continuous spectrum. Physically, the continuum edge of H(Q) consists of scattering states, which are approximately a product of a one-phonon state of energy $\omega(k)$ and the ground state of H(Q - k), such that the total momentum is Q.

Now the fundamental spectral properties of H(Q) are well understood and we can turn to analytical properties by applying analytica perturbation theory due to Kato [18]. this theory is a rigorous formulation of the fact that a nonanalyticity of the ground state in a parameter A can only occur, if either two discrete energy bands cross each other as a function of A (forming a degenerate point) or if a discrete ground state disappears into the continuous spectrum as a function of A. If, on the other hand, we know that the ground state of H(Q) is nondegenerate and discrete for all $\alpha \ge 0$, B > 0, $Q_3^2 < 2m\hbar\omega_0$, analytical perturbation theory tells us that then the ground state is analytic in all parameters α , B, Q_3 for $\alpha \ge 0$, B > 0, $Q_3^2 < 2m\hbar\omega_0$. The perturbation in A, however, must not be too singular; in fact, it has to be form bounded with respect to the rest of the Hamiltonian.

For the α - and Q_3 -dependence this relative boundedness is readily shown. The *B*-dependence is more tricky. In [12] a scaling transformation is used to guarantee this relative boundedness. Consequently, the final result is that the ground state and the associated ground state energy of H(Q)are jointly real analytic functions in α , *B*, Q_3 for $\alpha \ge 0$, $Q_3^2 < 2m\hbar\omega_0$, B > 0.

We mention several consequences: Firstly, the ground state energy $E(\alpha, B)$ of H is analytic in α and B, for it is obtained by taking $Q_3 = 0$ in E(Q). Secondly, ground state expectation values are analytical in α , B, Q_3 in the quoted domain, if the operators of interest are independent of α and B.

We mention some interesting cases, which were discussed by Peeters and Devreese [4]: The mean number N of virtual phonons in the ground state, defined by

$$N = \left\langle \int d^3k a^+(\boldsymbol{k}) a(\boldsymbol{k}) \right\rangle, \qquad (16)$$

the self induced polaron potential, which is obtained by calculating

$$V(\boldsymbol{q}) = \left\langle -\alpha^{1/2} \int \mathrm{d}^3 k[g(\boldsymbol{k})a(\boldsymbol{k}) \exp\left(\mathrm{i}\boldsymbol{k}\boldsymbol{q}\right) + \mathrm{h.c.}] \right\rangle, \quad (17)$$

the anisotropic polaron radius, whose perpendicular component is given by

$$R^{\perp} = \langle \langle x^2 \rangle \rangle^{1/2} \tag{18}$$

Note that in eq. (18) we have taken x^2 and not $(x^2 + y^2)/2$ as Peeters and Devreese do, since we are working in the Landau gauge and not in the symmetrical gauge. Furthermore we have used slightly different definitions in order to get *a priori* well defined quantities; we have to take the ground state expectation value $\langle ... \rangle$ in eq. (16)–(18) with respect to H(O)i.e., we fixed the conserved components of the total momentum to be zero. Of course, other definitions are possible.

Another quantity, which is of interest, is the magnetic polaron mass. Peeters and Devreese [3] have defined parallel and perpendicular magnetic polaron masses in the aniso-tropic Feynman approximation. One way to define a parallel magnetic polaron mass m'' is:

$$1/m'' \equiv \partial^2 E(\mathbf{Q})/\partial \mathbf{Q}_3^2|_{\mathbf{Q} \equiv \mathbf{Q}}/\hbar^2$$
(19)

Another possibility to define a cyclotron mass m^* (depending on α and B) at weak or intermediate magnetic fields is:

$$E_{1}(O) - E(O) = \hbar |e|B/(2m^{*})$$
(20)

where $E_1(O)$ is the energy of the first excited state i.e., the second Landau level. It follows immediately that both masses m'' and m^* are analytical in α and B.

In view of this results, no dramatic behaviour is expected in the oscillator strength and resonance frequencies in the magneto-absorption spectrum of a magneto-polaron.

Finally, we mention that the two-dimensional case d = 2 can be treated quite analogously. It is even easier, since the Q_3 -dependence does not occur. Moreover, several branches of optical phonons can easily be included into the analyticity proof.

The treatment of acoustical phonons is much more difficult, since it is connected with an infrared problem. Then the ground state of H(Q) lies at the bottom of the continuous spectrum. It is no longer separated by a gap and the usual analytical perturbation theory breaks down. However, we believe, this is a technical problem and no indication that the true ground state energy may be nonalytical in the usual acoustical polaron models.

3. Finite temperature results (T > 0)

To take a finite temperature into account, we consider the formal free energy $F(\alpha, B, T)$ instead of the ground state energy. We prefer the attribute "formal", as we are not dealing with thermodynamics in a strict sense; we are treating a one-particle problem.

F is derivable from a formal partition function Z by

$$F(\alpha, B, T) = -1/\beta \cdot \ln (Z(\alpha, B, T)), \qquad (21)$$

where

$$Z(\alpha, B, T) = \operatorname{Tr} \exp(-\beta H(\alpha, B))/\operatorname{Tr} \exp(-\beta H(0, B))$$
(22)

with $\beta = 1/(k_B T)$, k_B being the Boltzmann constant. Note that we have written $H(\alpha, B) \equiv H$. Z is normalized to the $\alpha = 0$ case in order to get a well defined expression.

The properties of $F(\alpha, B, T)$ and $Z(\alpha, B, T)$ were extensively discussed by us in [13]. It turns out, that Z is real analytic in all parameters $\alpha \ge 0$, B > 0, $0 < T < \infty$, if the expression (22) is well-defined – obviously a very weak assumption. (In particular even acoustical dispersions were included in contrast to the T = 0 case). The basic method to prove this is to represent Z as a functional integral and then to expand Z in a power series of α . The mathematical theorem which quarantees such a development is known as dominated convergence theorem. Equation (22) is well defined if and only if the power series converges. This was explicitly shown in [13]. The coefficients are analytical functions in B and β for B > 0 and $0 < \beta < \infty$ and consequently Z and F are real analytic in all parameters α , B, T for $\alpha \ge 0$, B > 0, $0 < T < \infty$.

We add an additional remark, concerning thermodynamic (temperature dependent) expectation-values. The ground state expectation value has now to be replaced by the thermodynamic average

$$Tr[A \exp(-\beta H)]/Tr \exp(-\beta H), \qquad (23)$$

A being the variable of interest. As utilized by Peeters and Devreese [4], such interesting physical quantities as the temperature dependent mean-value of virtual phonons, the polaron radius and the self-induced potential are representable as derivatives of Z with respect to certain auxiliary parameters. Since Z is also analytic in these auxiliary parameters and in α , B, T, the thermodynamic averages are analytical in α , B, T, too.

928 B. Gerlach and H. Löwen

To show the analyticity of the parallel temperature dependent mass, one has to consider the partition function of the momentum decomposed Hamiltonian H(Q). This was not studied in [13]. Therefore, we give here the explicit expression and show, that some qualitative properties known from chapter 2 are also derivable, using this different technique.

It is convenient to assume firstly a discrete phonon momentum space for this approach and optical phonon dispersions. We define

$$Z(\boldsymbol{Q}, \alpha, \boldsymbol{B}, \boldsymbol{T}) = \operatorname{Tr} \exp\left(-\beta H(\boldsymbol{Q})\right)/\operatorname{Tr} \exp\left(-\beta H(\boldsymbol{Q})\right|_{\alpha \equiv 0}\right)$$
(24)

Together with H. Schliffke we derived a functional integral expression for Z for the B = 0 case [19]. For B > 0, this can be done analogously leading to (in natural units as in [19], ω being an arbitrary frequency)

$$Z(\boldsymbol{Q}, \alpha, \boldsymbol{B}, T) = \left\langle \exp\left(-\alpha S_{I}[\boldsymbol{R}, z] + iQ_{3}z\right) \times \prod_{\boldsymbol{k}} \left[1 - \exp\left(-\beta\hbar\omega(\boldsymbol{k}) + ik_{3}z\right)\right]^{-1}\right\rangle$$
(25)

where the average is understood as

$$\langle ... \rangle = \int dz \int_{\boldsymbol{O} \to (0,0,z)} \delta^3 \boldsymbol{R} ... \exp(-S_0[\boldsymbol{R}] - S_m[\boldsymbol{R}])/N$$
(26)

N is the obvious normalization factor such that $\langle 1 \rangle = 1$. $\int_{O \to (0,0,z)} \delta^3 R$ indicates Wiener integration over all paths R(t), $0 \leq t \leq \beta \hbar \omega$ with starting point R(0) = O and ending point $R(\beta \hbar \omega) = (0, 0, z)$. The actions are explicitly given by

$$S_0[\mathbf{R}] = \int_0^{\beta h \omega} dt \, \frac{1}{2} \dot{R}^2(t), \qquad (27)$$

$$S_{\rm m}[\boldsymbol{R}] = -i \int_0^{\beta\hbar\omega} \mathrm{d}t \ |\boldsymbol{e}|\boldsymbol{B} \cdot \dot{\boldsymbol{R}}_2(t) \boldsymbol{R}_1(t)/(m\omega), \qquad (28)$$

$$S_1[\boldsymbol{R}, z] = -\frac{1}{2} \int d^3 k |g(\boldsymbol{k})|^2 \int_0^{\beta \hbar \omega} dt \int_0^{\beta \hbar \omega} ds \exp(-|t - s| + i\boldsymbol{k}(\boldsymbol{R}(t) - \boldsymbol{R}(s))) \times \{\theta(t - s)\}$$

+
$$[\exp (\beta \hbar \omega(\mathbf{k}) - ik_3 z) - 1]^{-1} \}.$$
 (29)

Using this expression, qualitative properties can be studied in the same way as in [18]. For example, the Q_2 -degeneracy of $H(\mathbf{Q})$ is readily seen, since Z does not depend on Q_2 .

The definition of a temperature dependent parallel polaron mass $m''(\alpha, B, T)$ as a natural generalization of eq. (19) leads to the following expression

$$m''(\alpha, B, T) = \beta \hbar \omega m \langle \exp(-\alpha S_{1}[R, z]) \rangle / \\ \times \langle z^{2} \exp(-\alpha S_{1}[R, z]) \rangle$$
(30)

which is clearly real analytic in α , *B* and *T* for $\alpha \ge 0$, B > 0, $0 < T < \infty$.

Moreover, expanding the product in eq. (25), one may discuss the position of the continuum edge as in [19]. This yields the same result as in Chapter 2.

We also notice, that the formula (25)-(29) may be the starting point for a Monte-Carlo calculation of the free energy and the parallel polaron mass. For the zero field case this was already done in [19].

4. Quantum-well structures in a magnetic field

Our qualitative methods, described in Chapter 2 and 3, are relatively general and apply also to other related Hamiltonians.

In this chapter we mention the interesting example of a quantum well.

Quantum-well structures in a magnetic field have gained enormous interest during the last years, both experimentally and theoretically. In many cases, the magnetic field is positioned perpendicular to the layers. In a first approximation, we describe the quantum well structure by adding an external static attractive potential $\lambda V(z)$ to the three-dimensional Hamiltonian H, given by (1), λ being the extracted potential strength, $\lambda > 0$. For the sake of definiteness let $V(z) \leq 0$, $V(z) \neq 0$ and

$$dz V^2(z) < \infty \tag{31}$$

Note that the electron interacts only with the bulk phonons in this description. Now, only the second component of the total momentum is conserved and we restrict ourselves to the subspace $Q_2 = 0$. Of course the Q_2 -degeneracy is again present. Let us compile some spectral properties of this extended Hamiltonian; detailed proofs may be found in Ref. [15]. By $E(\alpha, B, \lambda)$ we denote the ground state energy of this Hamiltonian. Then there exists a discrete nondegenerate ground state with energy

$$E(\alpha, B, \lambda) \leqslant E(\alpha, B, 0) + E_0(\lambda), \tag{32}$$

where $E_0(\lambda) < 0$ is the negative binding energy of the oneparticle Hamiltonian $p_3^2/(2m) + \lambda U(z)$. The ground state and its energy are real analytical in all parameters α , B, $\lambda(\alpha \ge 0; B, \lambda > 0)$. Hence, even in this case no discontinuous stripping transition or another kind of phase transition exists.

The continuum edge of the quantum well Hamiltonian begins exactly at the energy

$$E_{\rm c} = \min[E(\alpha, B, \lambda) + \hbar\omega_0, E(\alpha, B, 0)]$$
(33)

corresponding to either scattering states with one real phonon of energy $\hbar\omega_0$ present or delocalized electronic states in the z direction ($\omega_0 > 0$ is the minimum of the phonon dispersion, see eq. (5)).

In the finite temperature case the formal free energy is analytical in α , B, λ and T, too ($\alpha \ge 0$, B, λ , T > 0). We omit the proof, but refer again to Ref. [15].

One may propose also more extended Hamiltonians to describe other situations. For example, Chen, Ding and Lin [20] describe an interface electron in a half-infinite crystal in a mirror-image potential which interacts both with bulk LO phonons and with surface optical phonons. Our qualitative methods are readily transferred to this case, too. In particular, there exists no phase transition as function of the coupling parameter, potential strength, magnetic field strength or the temperature.

In conclusion, we have proven the analyticity of the physical quantities of magneto-polaron like systems in parameters like magnetic field strength, electron-phonon coupling parameter, temperature etc. This implies that all changes in the polaron state are smooth. Nonanalyticities predicted by variational calculations are artifacts of the approximation.

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