Proof of the nonexistence of (formal) phase transitions in polaron systems. I

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In recent literature it was frequently claimed that free polarons should undergo a (formal) phase transition from a mobile to a localized state, if the electron-phonon coupling constant α exceeds some critical value α_c. For $\alpha=\alpha_c$, the formal free energy should be nonanalytic in α. In this paper we prove that for optical polarons no such transition exists for finite temperatures. To do so, we use functional-integration techniques and discuss perturbation theory in α to infinite order.

I. INTRODUCTION

In this paper and in a following one we discuss analytical properties of the (formal) free energy of a polaron as a function of coupling parameters, temperature, and external fields.

The standard polaron model is defined by the well-known Hamiltonian H, proposed by Fröhlich, Pelzer, and Zienau.[1] H reads as follows:

$$H=\frac{\mathbf{P}^2}{2m}+\sum_{\mathbf{k},j}\hbar\omega_{\mathbf{k}j}a^\dagger_{\mathbf{k}j}a_{\mathbf{k}j}+\frac{1}{\sqrt{V}}\sum_{\mathbf{k},j}(g_{\mathbf{k},j}e^{i\mathbf{k}\cdot\mathbf{Q}}a_{\mathbf{k}j}+\text{H.c.})\ . \tag{1}$$

Here, m, $\mathbf{Q}$, and $\mathbf{P}$ are the mass, the position, and momentum operator of the electron. $\mathbf{k}$, j, $\omega_{\mathbf{k}j}$, $a_{\mathbf{k}j}$, and $a^\dagger_{\mathbf{k}j}$ are the wave vector, branch index, frequency, annihilation, and creation operator of phonons. Finally, V is the quantization volume, $g_{\mathbf{k}j}$ the electron-phonon coupling. It is useful to extract the dimensionless electron phonon coupling parameter α_j by defining

$$g_{\mathbf{k}j}=:\sqrt{\alpha_j}\tilde{g}_{\mathbf{k}j}\ . \tag{2}$$

We note that the structure of (1) remains as it is, if we consider systems in one, two, or three dimensions (of course the functional dependence of $g_{\mathbf{k}j}$ from $\mathbf{k}$ changes). Moreover, (1) is readily generalized, if external fields exist.

Spectral properties of H can conveniently be deduced from the diagonal elements of the reduced density matrix ρ, depending on the inverse (formal) temperature β and the set $\boldsymbol{\alpha}:=(\alpha_j)$ of coupling parameters,

$$\rho(\boldsymbol{\alpha},\beta):=\langle\mathbf{x}\,|\,\text{tr}_{\text{Ph}}e^{-\beta H}\,|\,\mathbf{x}\rangle\ . \tag{3}$$

In (3), tr_{Ph} indicates the trace operation concerning phononic degrees of freedom, $|\,\mathbf{x}\rangle$ is an eigenstate of $\mathbf{Q}$. As H is translation invariant, ρ does not depend on $\mathbf{x}$; $\mathbf{x}$ may be chosen as zero.

It will prove profitable to relate $\rho(\boldsymbol{\alpha},\beta)$ to the well-known expression $\rho(\mathbf{0},\beta)$ of the uncoupled electron-phonon system. Let us introduce

$$Z(\boldsymbol{\alpha},\beta):=\rho(\boldsymbol{\alpha},\beta)/\rho(\mathbf{0},\beta)\ . \tag{4}$$

Z in turn is connected to the formal free energy $F(\boldsymbol{\alpha},\beta)$ by

$$Z(\boldsymbol{\alpha},\beta)=\exp\{-\beta[F(\boldsymbol{\alpha},\beta)-F(\mathbf{0},\beta)]\}\ . \tag{5}$$

From the very beginning of the polaron story, it was a controversially discussed question, whether $F(\boldsymbol{\alpha},\beta)$ is a real analytic function of α_j (provided we insert for $g_{\mathbf{k}j}$ the familiar couplings). The entire discussion was probably initiated by Landau's early idea of self-trapping (see, e.g., Ref. 2) and Feynman's highly significant paper on optical polarons[3] in 1955. Feynman's concept is crucial for our approach to $F(\boldsymbol{\alpha},\beta)$. Therefore, we concentrate for a while on optical polarons and stress some important conceptual aspects. Feynman proved that all phonon effects can exactly be incorporated into a translation-invariant, noninstantaneous self-energy functional S_I for the electron. As S_I cannot be treated rigorously, it has to be approximated. A possible choice for an approximation to S_I could be a one-particle potential U containing free parameters. One may choose a variational procedure to fix these parameters and to produce upper bounds on $F(\alpha,\beta)$. For examples, see Feynman,[3] Luttinger and Lu,[4] and Manka and Suffczynski.[5] Intuitively, one expects that $U\to 0$ for $\alpha\to 0$, whereas strong binding should occur for $\alpha\to\infty$. In the latter case the electron should be trapped at a certain point in space. It may even happen that the variational principle forces one to put $U\equiv 0$ for $\alpha<\alpha_C$ and $U\neq 0$ for $\alpha>\alpha_C$, where α_C is some positive number (again, see Refs. 3 and 5). Thus the variational bound on $F(\alpha,\beta)$ exhibits a nonanalyticity. In fact, results of this kind may have caused the first conjectures, that $F(\alpha,\beta)$ itself might not be real analytic for all α,β.

It seems to be widely unknown (in fact, it was unknown to the present authors until recently) that a complete discussion of the ground-state energy $E_0(\alpha)$ was available as early as 1974 (see the paper of Fröhlich[6] and for detailed comments, those of Spohn[7,8]). If only one branch of phonons couples to the electron and

$$\omega_{\mathbf{k}}\geq\omega_0>0\ , \tag{6}$$

$$\int d^3k\ |g_{\mathbf{k}}|^2/(k_0^2+k^2)<\infty\ , \tag{7}$$

for some ω_0 and $k_0 > 0$, then $E_0(\alpha)$ is real analytic in α. Obviously, Fröhlich's important result applies to the case of optical polarons and disproves the claim of many authors, that $E_0(\alpha)$ be nonanalytic. The general shortcoming of their "proof" is that they all use variational arguments. We mention the work of Gross,[9] Manka,[10,11] Manka and Suffczynski,[5] Lepine and Matz,[12] Lepine,[13] Shoji and Tokuda,[14] and Lu and Shen.[15] It should be added however, that the lowest available bounds on $E_0(\alpha)$ do not exhibit any nonanalyticity (again, see Feynman[3] and the work of Adamowski, Gerlach, and Leschke[16] as well as that of Peeters and Devreese[17]).

The main purpose of this paper is to generalize the result for $E_0(\alpha)$ to $F(\alpha,\beta)$. There seems to be a widespread belief that a free energy $F(\mathbf{A},\beta)$, being a function of some parameters $\mathbf{A}$ and β, should generally behave more smoothly in $\mathbf{A}$ for $\beta < \infty$ than for $\beta = \infty$. To the best of our knowledge, no rigorous result supports this belief (see, e.g., Ruelle,[18] in particular, Chap. 5). Moreover, some recent results (see, e.g., Coleman[19]) cast considerable doubt on the smoothness assertion as such.

Consequently, we prove from the very beginning that $F(\alpha,\beta) - F(0,\beta)$ is real analytic in α,β for $0 \le \alpha$, $0 < \beta < \infty$. This is in a marked contrast to the work of Manka[11] and Manka and Suffczynski.[5]

To show the analyticity of $E_0(\alpha)$ Fröhlich[6] uses an operator theory. His proof cannot be generalized to arbitrary β. Our method differs totally from the one of Fröhlich and gives valuable insight into the structure of high-order perturbation theory. Furthermore it can be easily generalized. In fact, in a following paper we do generalize our discussion to the case of acoustical coupling, mixed and anisotropic couplings, lower spatial dimensions, as well as additional external fields.

II. STATEMENT OF THE PROBLEM IN TERMS OF FUNCTIONAL INTEGRALS

We start from Hamiltonian (1) and the corresponding expression (3) for $\rho(\boldsymbol{\alpha},\beta)$. In doing so, we consider free polarons in three dimensions.

It is well known from Feynman's work[3] that $\rho(\boldsymbol{\alpha},\beta)$ can be represented by a functional integral as follows:

$$\rho(\boldsymbol{\alpha},\beta) = Z_{\mathrm{Ph}} \int \delta^3 R \, e^{-S_0[\mathbf{R}] - S_I[\mathbf{R}]} , \tag{8}$$

where

$$S_0[\mathbf{R}] := \int_0^\beta d\tau \frac{m}{2\hbar^2} \mathbf{R}'^2(\tau) , \tag{9}$$

$$S_I[\mathbf{R}] := -\int_0^\beta d\tau \int_0^\beta d\tau' \frac{1}{V} \sum_{j,\mathbf{k}} \alpha_j \, |\tilde{g}_{\mathbf{k},j}|^2 e^{i\mathbf{K}\cdot[\mathbf{R}(\tau)-\mathbf{R}(\tau')]} G(\hbar\omega_{\mathbf{k},j}, \tau - \tau') . \tag{10}$$

In (8), Z_{Ph} is the partition function of free phonons, $\int \delta^3 R$ indicates Wiener-integration over three-dimensional, closed, real paths $\mathbf{R}(\tau)$ with starting- and ending-point $\mathbf{x}$. In (10), $G(\hbar\omega,\tau)$ is the temperature-dependent Green function of a harmonic oscillator,

$$G(\hbar\omega,\tau) := \cosh[\hbar\omega(|\tau| - \beta/2)] \Big/ \left[2\sinh\left[\frac{\beta\hbar\omega}{2}\right]\right] . \tag{11}$$

Every term in (10) is negative, therefore $S_1(\mathbf{R})$ is negative (see Adamowski, Gerlach, and Leschke[20]). We obtain from (8) and (4)

$$Z(\boldsymbol{\alpha},\beta) = \frac{\int \delta^3 R \, e^{-S_0[\mathbf{R}]} e^{-S_I[\mathbf{R}]}}{\int \delta^3 R \, e^{-S_0[\mathbf{R}]}} =: \langle e^{-S_I[\mathbf{R}]} \rangle_{S_0} . \tag{12}$$

The last equation defines the expectation value of $\exp[-S_I(\mathbf{R})]$ with respect to S_0. Because of $S_I(\mathbf{R}) \le 0$, a theorem of Beppo-Levi[21] assures us that

$$Z(\boldsymbol{\alpha},\beta) = \sum_{n=0}^{\infty} \frac{1}{n!} \langle (-S_I[\mathbf{R}])^n \rangle_{S_0} . \tag{13}$$

Now S_I is proportional to the coupling constants α_j. Consequently, (13) is a power series in all α_j. Every question concerning analytical properties of $Z(\boldsymbol{\alpha},\beta)$ or $F(\boldsymbol{\alpha},\beta)$ can be incorporated into the two key questions: Does the power series (13) converge for all α_j? Are the coefficients analytic functions in β?

III. OPTICAL POLARONS: RESULTS

In polar semiconductors, the interaction of electrons with LO phonons is dominant. Neglecting all other couplings, we have (see, e.g., Ref. 3)

$$g_{\mathbf{k}} = -i\hbar\omega^4 \sqrt{\hbar/2m\omega} \sqrt{4\pi\alpha}/k . \tag{14}$$

Under these circumstances, we prove the following.

Statement 1. The power series (13) for $Z(\alpha,\beta)$ exists for all complex α, the coefficients being analytical functions in β for $0 < \mathrm{Re}\beta < \infty$. Consequently, $Z(\alpha,\beta)$ is a holomorphic function in the specified domain.

For real α and β, $0 \le \alpha$, $0 < \beta < \infty$, and the quantity $Z(\alpha,\beta)$ is positive. Consequently, we can immediately deduce from statement 1:

Statement 2. $F(\alpha,\beta) - F(0,\beta)$ is a real analytic function in α,β for $\alpha \ge 0$, $0 < \beta < \infty$.

IV. OPTICAL POLARONS: PROOF OF STATEMENT 1

Our starting point is Eq. (13), which can be rewritten as follows:

$$Z(\alpha,\beta)=\sum_{n=0}^{\infty} Z_n(\beta)\alpha^n \, , \tag{15}$$

$$Z_n(\beta):=\frac{1}{n!}\left\langle \left[-\frac{1}{\alpha}S_I[\mathbf{R}]\right]^n\right\rangle_{S_0} . \tag{16}$$

According to statement 1 we have to prove that the right-hand side of (15) converges for all complex α, $Z_n(\beta)$ being an analytic function in β for $0<\mathrm{Re}\beta<\infty$.

To begin with, we reformulate (9) and (10) in an appropriate way: Firstly, we introduce $\hbar\omega$ and $\sqrt{\hbar/m\omega}$ as units of energy and length. This leads us to dimensionless variables, τ, $\mathbf{k}$, $\mathbf{R}(\tau)$. Secondly, we perform a scaling transformation

$$\mathbf{R}(\tau)\to\sqrt{\beta\hbar\omega}\,\mathbf{R}\left[\frac{\tau}{\beta\hbar\omega}\right] . \tag{17}$$

Then one finds from (9)—(11),

$$S_0[\mathbf{R}]=\int_0^1 d\tau\tfrac{1}{2}\mathbf{R}'^2(\tau) \, , \tag{18}$$

$$S_I[\mathbf{R}]=-\frac{\sqrt{2}\alpha}{4\pi^2}(\beta\hbar\omega)^{3/2}\int_0^1 d\tau\int_0^1 d\tau'\, G(\tau-\tau')\int\frac{d^3k}{k^2}e^{i\mathbf{k}\cdot[\mathbf{R}(\tau)-\mathbf{R}(\tau')]} \, , \tag{19}$$

$$G(\tau):=\cosh[\beta\hbar\omega(|\tau|-\tfrac{1}{2})]\Big/\left[2\sinh\left[\frac{\beta\hbar\omega}{2}\right]\right] . \tag{20}$$

Proceeding from (10) to (19), we have additionally replaced the $\mathbf{k}$ summation by an integration. Inserting (19) into (16) and interchanging the Wiener integration with the $\mathbf{k}$ and τ integration,[22] we obtain

$$Z_n(\beta)=\frac{1}{n!}\left[\frac{\sqrt{2}(\beta\hbar\omega)^{3/2}}{4\pi^2}\right]^n\int_0^1\cdots\int_0^1 d\tau_1\cdots d\tau_{2n}\, G(\tau_1-\tau_2)\cdots G(\tau_{2n-1}-\tau_{2n})$$
$$\times\int\cdots\int\frac{d^3k_1}{k_1^2}\cdots\frac{d^3k_n}{k_n^2}\left\langle\exp\left[i\sum_{j=1}^{n}\mathbf{k}_j[\mathbf{R}(\tau_{2j-1})-\mathbf{R}(\tau_{2j})]\right]\right\rangle_{S_0} , \tag{21}$$

for $n\geq 1$. Clearly, $Z_0(\beta)=1$.

The β dependence of $Z_n(\beta)$ is simple; it is contained in the prefactor and the functions $G(\tau)$. Consequently, the right-hand side of (21) defines an analytic function $Z_n(\beta)$ for $0<\mathrm{Re}\beta<\infty$. This proves the minor part of statement 1.

The expectation value in (21) can be calculated by standard methods (see, e.g., Adamowski, Gerlach, and Leschke[23]). One finds

$$Z_n(\beta)=\frac{1}{n!}\left[\frac{\sqrt{2}(\beta\hbar\omega)^{3/2}}{4\pi^2}\right]^n\int_0^1\cdots\int_0^1 d\tau_1\cdots d\tau_{2n}\, G(\tau_1-\tau_2)\cdots G(\tau_{2n-1}-\tau_{2n})$$
$$\times\int\cdots\int\frac{d^3k_1}{k_1^2}\cdots\frac{d^3k_n}{k_n^2}\exp\left[-\sum_{j,l=1}^{n}A_{jl}(\tau_1,\ldots,\tau_{2n})\mathbf{k}_j\cdot\mathbf{k}_l\right] , \tag{22}$$

where

$$A_{jl}(\tau_1,\ldots,\tau_{2n}):=-\tfrac{1}{2}(\tau_{2j-1}-\tau_{2j})(\tau_{2l-1}-\tau_{2l})$$
$$-\tfrac{1}{4}(|\tau_{2j-1}-\tau_{2l-1}|+|\tau_{2j}-\tau_{2l}|-|\tau_{2j-1}-\tau_{2l}|-|\tau_{2j}-\tau_{2l-1}|) . \tag{23}$$

It is well known from an early paper of Krivoglaz and Pekar[24] that

$$Z_1(\beta)=\frac{\sqrt{\pi}(\beta\hbar\omega)^{3/2}}{2\sinh(\beta\hbar\omega/2)}I_0\left[\frac{\beta\hbar\omega}{2}\right] , \tag{24}$$

where $I_0(z)$ is a modified Bessel function of first kind. To the best of our knowledge, $Z_n(\beta)$ cannot be evaluated analytically for $n\geq 2$. However, it is easily derived from (22), that

$$Z_2(\beta)\leq\frac{\pi}{4}Z_1^2(\beta) \tag{25}$$

holds true. We are now going to generalize this inequality. In a first step, we calculate upper bounds for the $\mathbf{k}$ integrals, in a second one for the τ integrals. In doing so, we assume that $A=(A_{jl})$ is a positive definite matrix almost everywhere and for any $n\geq 1$. We prove this property at the end of this section [Eqs. (35). . .]. Let us now consider the $\mathbf{k}$ integration. We use polar coordinates, defined by

$$\mathbf{k}_i=k_i(\sin\theta_i\cos\phi_i,\sin\theta_i\sin\phi_i,\cos\theta_i) . \tag{26}$$

Introducing a matrix $\widetilde{A}=(\widetilde{A}_{jl})$ by

$$\tilde{A}_{jl} := A_{jl}[\sin\theta_j\sin\theta_l(\cos\phi_j\cos\phi_l+\sin\phi_j\sin\phi_l)+\cos\theta_j\cos\theta_l] =: B_{jl}^1+B_{jl}^2+B_{jl}^3 , \quad (27)$$

we obtain

$$\sum_{j,l} A_{jl}\mathbf{k}_j\cdot\mathbf{k}_l=\sum_{j,l}\tilde{A}_{jl}k_jk_l . \quad (28)$$

As A is positive definite almost everywhere and for any $n\geq 1$, the same holds true for B^1, B^2, B^3, and $\tilde{A}$. As for B^i, this is guaranteed by the familiar determinant criterion; $\tilde{A}$ is positive as a sum of positive matrices. Therefore

$$Y_n := \int\cdots\int \frac{d^3k_1}{k_1^2}\cdots\frac{d^3k_n}{k_n^2}\exp\left[-\sum_{j,l=1}^{n}A_{jl}\mathbf{k}_j\cdot\mathbf{k}_l\right] \leq \frac{\pi^{n/2}}{2}\prod_{j=1}^{n}\left[\int_0^{2\pi}d\phi_j\int_0^{\pi}d\theta_j\sin\theta_j\right]\frac{1}{\sqrt{\det\tilde{A}}} . \quad (29)$$

(Notice that we enlarged the integration domain to $-\infty\leq k_i\leq\infty$. Consequently, we arrive at an inequality.) To proceed, we apply an inequality due to Minkowski:[25]

$$\left[\det\left[\sum_{j=1}^{3}B^i\right]\right]^{1/n}\geq\sum_{j=1}^{3}(\det B^i)^{1/n} , \quad (30)$$

for positive matrices B^i. Applying this to $\tilde{A}$ according to (27), we find

$$\det\tilde{A}\geq(\det A)\left[\left[\prod_{j=1}^{n}\cos^2\theta_j\right]^{1/n}+\left[\prod_{j=1}^{n}\sin^2\theta_j\cos^2\phi_j\right]^{1/n}+\left[\prod_{j=1}^{n}\sin^2\theta_j\sin^2\phi_j\right]^{1/n}\right]^n . \quad (31)$$

If, in addition, the inequality $a+b+c\geq 3\sqrt[3]{abc}$ (a,b,c positive) is used for the bracket in (31), all integrations in (29) can be done. This leaves us with

$$Y_n\leq\frac{[2\sqrt{\pi/3}\Gamma^3(\frac{1}{3})]^n}{2\sqrt{\det A}} . \quad (32)$$

We insert (32) into (22). Furthermore, we use

$$G(\tau)\leq G(0)=\tfrac{1}{2}\coth(\beta\hbar\omega/2) . \quad (33)$$

This leads to

$$Z_n(\beta)\leq\frac{1}{2(n!)}\left[(2\pi)^{-3/2}3^{-1/2}\Gamma^3(\tfrac{1}{3})(\beta\hbar\omega)^{3/2}\coth\left[\frac{\beta\hbar\omega}{2}\right]\right]^n \times\sum_{\phi}\int_0^1\cdots\int_0^1 d\tau_{\phi(1)}\cdots d\tau_{\phi(2n)}\frac{\Theta(\tau_{\phi(2n)}-\tau_{\phi(2n-1)})\cdots\Theta(\tau_{\phi(2)}-\tau_{\phi(1)})}{\sqrt{\det A(\tau_1,\ldots,\tau_{2n})}} , \quad (34)$$

where ϕ indicates a permutation of the numbers $1,\ldots,2n$. It is obvious that we finally need an upper bound on the τ integration. It is exactly this point that brings us back to the assumed positivity of the matrix A. The decisive idea to prove this is to rewrite $(\det A)^{-1/2}$ as functional integral in *one* spatial dimension; the corresponding component of the path is $R_1(\tau)$. Then

$$\frac{1}{[\det A(\tau_1,\ldots,\tau_{2n})]^{1/2}}=\pi^{-n/2}\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}dk_1\cdots dk_n\left\langle\exp\left[i\sum_{j=1}^{n}k_j[R_1(\tau_{2j-1})-R_1(\tau_{2j})]\right]\right\rangle_{S_0} . \quad (35)$$

The expectation value in (35) depends only on $2n$ τ arguments. Expressions of such a type may be rewritten as a Gaussian integral (see, e.g., Yeh[26]). We obtain for the τ ordering, prescribed in (34),

$$\frac{1}{\sqrt{\det A}}=\frac{\pi^{-n/2}}{(2\pi)^n}\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}dk_1\cdots dk_n\,dx_1\cdots dx_{2n}\frac{\exp\left[i\sum_{j=1}^{n}k_j(x_{\phi^{-1}(2j-1)}-x_{\phi^{-1}(2j)})\right]}{[\tau_{\phi(1)}(\tau_{\phi(2)}-\tau_{\phi(1)})\ldots(1-\tau_{\phi(2n)})]^{1/2}} \times\exp\left[-\frac{1}{2}\left[\frac{x_1^2}{\tau_{\phi(1)}}+\frac{(x_2-x_1)^2}{\tau_{\phi(2)}-\tau_{\phi(1)}}+\cdots+\frac{(x_{2n}-x_{2n-1})^2}{\tau_{\phi(2n)}-\tau_{\phi(2n-1)}}+\frac{x_{2n}^2}{1-\tau_{\phi(2n)}}\right]\right] . \quad (36)$$

The n k integrals yield n δ functions. Therefore, n x integrations can easily be done. We are left with (at least) $n+1$ Gaussian factors, which have to be integrated. As we are only interested in an upper bound, we can omit so many of them, that exactly n independent Gaussian factors remain. Without loss of generality, $\exp(-x_1^2/2\tau_{\phi(1)})$ should survive. Performing the last n x integrations, we arrive at

$$\frac{1}{[\det A(\tau_1,\ldots,\tau_{2n})]^{1/2}} \le \frac{2^{n/2}}{[\Delta(\tau_1,\ldots,\tau_{2n})]^{1/2}}\ , \tag{37}$$

where $\Delta(\tau_1,\ldots,\tau_{2n})$ is a product of $n+1$ different factors; each factor in turn is a τ difference, being positive almost everywhere [within the admitted τ domain in Eq. (34)].

According to the determinant criterion, (37) proves, that $A(\tau_1,\ldots,\tau_{2n})$ is positive almost everywhere and for any $n \ge 1$.

Inserting (37) into (34), we find

$$Z_n(\beta) \le \frac{(2n)!}{2(n!)} \left[\frac{\pi^{-3/2}}{2} 3^{-1/2}\Gamma^3(\tfrac{1}{3})(\beta\hbar\omega)^{3/2}\coth\frac{\beta\hbar\omega}{2} \right]^n$$

$$\times \max_{\phi} \left\{ \int_0^1 \cdots \int_0^1 d\tau_{\phi(1)} \cdots d\tau_{\phi(2n)} \frac{\Theta(\tau_{\phi(2n)}-\tau_{\phi(2n-1)}) \cdots \Theta(\tau_{\phi(2)}-\tau_{\phi(1)})}{[\Delta(\tau_1,\ldots,\tau_{2n})]^{1/2}} \right\} . \tag{38}$$

The value of the τ integral could depend on the kind and sequence of the τ-differences in $\Delta(\tau_1,\ldots,\tau_{2n})$. However, the inspection of two successive integrations shows, that this is not the case. Therefore, we replace $\Delta(\tau_1,\ldots,\tau_{2n})$ by $(\tau_{\phi(2)}-\tau_{\phi(1)})\cdots(\tau_{\phi(n+2)}-\tau_{\phi(n+1)})$ and obtain

$$Z_n(\beta) \le \frac{(2n)!}{2(n!)} \left[\frac{\pi^{-3/2}}{2} 3^{-1/2}\Gamma^3(\tfrac{1}{3})(\beta\hbar\omega)^{3/2}\coth\frac{\beta\hbar\omega}{2} \right]^n \frac{\pi^{(n+1)/2}}{\Gamma((3n+1)/2)}\ . \tag{39}$$

Making use of Stirling's formula, we finally arrive at

$$Z_n(\beta) < \frac{C_1\sqrt[4]{n}}{\sqrt{n!}} \left[C_2(\beta\hbar\omega)^{3/2}\coth\frac{\beta\hbar\omega}{2} \right]^n \tag{40}$$

for $n \ge 2$. In (40), $C_1 = 0.415$, $C_2 = 3.85$.

Making use of (40) and (24), Eq. (15) shows that $Z(\alpha,\beta)$ converges for all complex α. Thus our proof of statement 1 is complete.

We add a final remark: Every upper bound on $Z(\alpha,\beta)$ yields a lower bound on $F(\alpha,\beta)-F(0,\beta)$. Combining (15), (25), and (40), we obtain the following lower bound:

$$F(\alpha,\beta)-F(0,\beta) \ge -\frac{1}{\beta}\ln \Bigg[\{[1+2C(\beta)\alpha+2C^2(\beta)\alpha^2+2C^3(\beta)\alpha^3]\exp[C^2(\beta)\alpha^2]\}^{1/2}$$

$$- [C(\beta)-Z_1(\beta)]\alpha - \left[\frac{C^2(\beta)}{\sqrt{2}} - \frac{\pi}{4}Z_1^2(\beta) \right]\alpha^2 \Bigg]\ , \tag{41}$$

where $Z_1(\beta)$ is given explicitly by (24) and $C(\beta)$ is defined by

$$C(\beta) := 3.9(\beta\hbar\omega)^{3/2}\coth\left[\frac{\beta\hbar\omega}{2}\right] . \tag{42}$$

We note that the lower bound (41) is asymptotically exact both for small α and for small β.

For $\alpha \to \infty$ our lower bound exhibits the correct qualitative behavior (see Adamowski, Gerlach, and Leschke[27]):

$$\lim_{\alpha\to\infty} \frac{F(\alpha,\beta)-F(0,\beta)}{\alpha^2} > -\infty\ . \tag{43}$$

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[21]See, e.g., E. J. McShane, *Unified Integration* (Academic, New York, 1983); the theorem of Beppo-Levi is also known as monotone convergence theorem.
[22]It should be noticed that the **k** integration in (19) is not well defined in a Lebesgue sense. This can be repaired by inserting a convergence factor $\exp(-\epsilon k)$, $\epsilon > 0$. Then the interchange of integrations is fully justified. Having performed the functional integration [Eq. (22)], all expressions exist even for $\epsilon \to 0$.
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