

The pinning transition of a bound large polaron

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A polaron, bound in an external attractive short-range potential, exhibits a pinning transition, which is connected with a change from a delocalized to a localized polaron ground state, as the potential strength increases. We describe this transition in more detail, determining the associated critical exponents. For a screened Coulomb potential, we calculate the critical potential strength as a function of the electron-phonon coupling parameter. The result is compared with the effective mass description of the problem. Furthermore, experimental consequences of the pinning transition are discussed.

I. Introduction

Recently, the question whether or not a polaron system, described by a generalized Fröhlich Hamiltonian, exhibits ‘phase transitions’ gained new interest. Basic works of the rigorous study of this question were done by Spohn [1, 2] and by Gerlach and the author [3, 4], who found that for a free large polaron no ‘phase transition’ does occur. The same negative answer was obtained for a small polaron [5] and a magneto-polaron [6]. Spohn [1] also pointed out the possibility of a pinning transition for a polaron bound in a short range potential, a mathematical proof of the existence of this transition was given in [7].

Summarizing these results, we state that the only possibility of a polaron ‘phase transition’ is the pinning transition for a polaron bound in an attractive short range potential. This transition really is an intrinsic property of the Fröhlich Hamiltonian. We also refer to [8] for a review of the ‘phase transition’ problem.

Interestingly enough, up to now no calculation of the pinning transition was presented, although a related system, namely a polaron bound in a Coulomb potential, is a well studied problem since the early work of Platzman [9].

The present paper is devoted to a calculation and discussion of the pinning transition. We start in Sect. II, giving an introductory physical description of the pinning transition. We found that it is a second order

transition, accompanied by a potential assisted change of localization of the polaron ground state wave function. The inverse of the ground state expectation value of the squared electronic position operator is a suitable order parameter and the associated critical exponent is determined. In Sect. III, we calculate the critical potential strength in the case of a screened Coulomb potential, using a variational method of Adamowski [10], and compare the results with the effective mass approximation. Finally in Sect. IV, we conclude our results and discuss physical and experimental consequences and applications of the pinning transition.

II. Description of the pinning transition

II.1 Pinning transition in ordinary quantum mechanics

The effect of the pinning transition is well-known from the theory of bound states in ordinary quantum mechanics, for a compilation of the literature see Spohn [1]. However, its name ‘pinning’ originates from an analogy to statistical mechanics of surfaces. Let us briefly recall the basic facts.

In three spatial dimensions, the existence of a bound ground state of a one-particle Hamiltonian

$$H_e = \mathbf{p}^2/(2m) + \beta \cdot V(\mathbf{r}) \quad (1)$$

$V(\mathbf{r})$ being an attractive short range potential, depends sensitively on the potential strength β . For a more precise definition of short range, see again the literature quoted in [1]. It turns out that for $\beta < \beta_c$ the ground state is delocalized, its associated energy $E(\beta)$ is zero, being the lower edge of the continuous spectrum. On the other hand, for $\beta > \beta_c$ the Hamiltonian H_e possesses a bound localized ground state with strictly negative energy $E(\beta)$. Consequently, at $\beta = \beta_c$ the ground state energy $E(\beta)$ exhibits a nonanalyticity.

Looking for a suitable order parameter $M(\beta)$, which describes the pinning transition, it is natural to take the inverse of the ground state expectation value of r^2 , i.e.

$$M(\beta) = 1/\langle r^2 \rangle \quad (2)$$

Then, $M(\beta) = 0$ for $\beta < \beta_c$ and $M(\beta) \geq 0$ for $\beta \geq \beta_c$.

The next question of interest concerns the critical exponents associated with the pinning transition. Here, two different exponents γ and δ are relevant. Firstly, γ is defined by

$$E(\beta) \sim (\beta - \beta_c)^\gamma, \quad \beta \rightarrow \beta_c^+ \quad (3)$$

determining the order of the pinning transition. Secondly, we define δ as

$$M(\beta) \sim (\beta - \beta_c)^\delta, \quad \beta \rightarrow \beta_c^+ \quad (4)$$

In order to determine γ and δ let us be more specific and consider cutoff-potentials of type

$$\beta \cdot V(\mathbf{r}) = \begin{cases} \beta \cdot v(r) & \text{for } r < L \\ 0 & \text{otherwise} \end{cases} \quad (5)$$

Since γ and δ are expected to reflect universal features of the pinning transition, the critical behaviour should not change, if one considers more general short range potentials.

In the case [5], for $\beta > \beta_c$, γ and δ are readily calculated. Obviously, the ground state wave function $\psi(r)$ fulfills (see Messiah [11]):

$$\psi(r) \sim \exp(-(-2mE(\beta))^{\frac{1}{2}}r/\hbar)/r \quad \text{for } r > L \quad (6)$$

The continuity of the logarithmic derivative at $r=L$ yields

$$(-2mE(\beta))^{\frac{1}{2}}/\hbar = d/dr(r\psi(r))/(r\psi(r))|_{r=L} \quad (7)$$

Now, we assume that the right hand side of (7) is analytical in $(\beta - \beta_c)$ for $\beta > \beta_c$, i.e. it is proportional to $(\beta - \beta_c)$ as $\beta \rightarrow \beta_c^+$. This assumption can explicitly be verified e.g. for a spherical square well but it should also be valid for more general potentials. However, it is an essential input in our determination of the critical exponents. By this assumption, we obtain $\gamma = 2$

and inserting $E(\beta)$ in (6) and evaluating M it turns out that also $\delta = 2$. Therefore the pinning transition is a second order transition.

II.2 Pinning transition for the polaron

It is an interesting task to study the influence of the electron-phonon interaction on the pinning transition. The problem we are dealing with is a conduction electron bound in a short range impurity potential and interacting with the LO-phonons. Such a system is described by Fröhlich's Hamiltonian [12]

$$H_F = H_e + H_{\text{oph}} + H_I \quad (8)$$

where

$$H_{\text{oph}} = \int d^3k \hbar \omega(\mathbf{k}) a^\dagger(\mathbf{k}) a(\mathbf{k}) \quad (9)$$

$$H_I = \alpha^{\frac{1}{2}} \int d^3k [g(\mathbf{k}) \exp(i\mathbf{k}\mathbf{r}) a(\mathbf{k}) + h.c.] \quad (10)$$

Here, we have used standard notation. $a^\dagger(\mathbf{k})$, $a(\mathbf{k})$ are the creation resp. annihilation operator of the phonons, α denoting the dimensionless electron-phonon coupling parameter. The phonon frequency $\omega(\mathbf{k})$ and the coupling function $g(\mathbf{k})$ are given by

$$\omega(\mathbf{k}) = \omega \quad (11)$$

$$g(\mathbf{k}) = \hbar \omega (\hbar/(2m\omega))^{\frac{1}{4}} \cdot (2\pi^2)^{-\frac{1}{2}} \cdot 1/k \quad (12)$$

In [7], it was proved that the pinning transition, which was well-known to exist for $\alpha = 0$, survives for $\alpha > 0$. The ground state energy $E(\alpha, \beta)$ is nonanalytic in β at $\beta = \beta_c(\alpha)$ where $0 \leq \beta_c(\alpha) \leq \beta_c(0)$ and $\beta_c(\alpha)$ is continuous in α . For $\beta < \beta_c(\alpha)$, $E(\alpha, \beta)$ is identical to the polaron self-energy $E(\alpha, 0)$, which is the position of the continuum edge.

Therefore the influence of the phonons is to shift the critical potential strength to smaller values. This can be qualitatively be understood as a consequence of the enhanced phonon-induced effective mass of the polaron.

The definition (2)–(4) of the order parameter M and the critical exponents γ , δ are readily transferred to the polaron case. However, as indicated by Spohn [1], it is not expected that the phonons change the critical exponents γ and δ .

In order to show this more explicitly, we propose the following intuitive argument, which works for the case (5) of cutoff potentials with cutoff L large compared to the polaron radius and which is a generalization of the discussion in Sect. II.1.

Performing a Lee-Low-Pines transformation [13], we obtain a new Hamiltonian H'_F which is unitarily equivalent to H_F :

$$H'_F = (\mathbf{p} - \mathbf{P}_{\text{ph}})^2/(2m) + H_{\text{oph}} + \beta \cdot V(\mathbf{r}) + H'_I \quad (13)$$

with

$$H'_I = \alpha^2 \int d^3 k g(\mathbf{k}) (a(\mathbf{k}) + a^\dagger(\mathbf{k})) \quad (14)$$

where $\mathbf{P}_{\text{ph}} = \int d^3 k \mathbf{k} a^\dagger(\mathbf{k}) a(\mathbf{k})$ is the usual phonon momentum operator. We look for a solution of $H'_F \Phi = E(\alpha, \beta) \Phi$ for $\beta > \beta_c$ and $r > L$ with $E(\alpha, \beta) < E(\alpha, 0)$. Since $r > L$, H'_F does not depend on \mathbf{r} any longer and the eigenstates of H'_F are linear combinations of free polaron like product states. Since L is large compared to the polaron radius and since we are only interested in studying the critical regime $\beta \gtrsim \beta_c$, only large values of r are relevant. A reasonable form of the ground state wave function Φ is as follows:

$$\Phi = C \int d^3 q a^2 e^{i\mathbf{q} \cdot \mathbf{r}/\hbar} / ((\kappa^2 + q^2)(a^2 + q^2)) \cdot \psi(\mathbf{q}), \quad a \gg \kappa, \quad \kappa \gtrsim 0 \quad (15)$$

Here, C is the obvious normalization factor. For $a \rightarrow \infty$ the first part of the right hand side of (15) just corresponds to the Fourier transform of (6). We choose a very large but $a < \infty$, so that the integrand vanishes sufficiently rapidly as $q \rightarrow \infty$. Note that, since r is large, only small q 's contribute to the integral in (15). Finally, $\psi(\mathbf{q})$ describes the phononic part, being a normalized eigenfunction of $(\mathbf{p} - \mathbf{P}_{\text{ph}})^2 / (2m) + H_{\text{oph}} + H'_I$; its associated energy eigenvalue is

$$E_q(\alpha) = E(\alpha, 0) + q^2 / (2m(\alpha)) + O(q^4) \\ = E(\alpha, 0) \cdot \cos(q(-2m(\alpha)E(\alpha, 0))^{-1/2}) + O(q^4) \quad (16)$$

$m(\alpha)$ being the polaron mass.

Assuming $\Phi = \bar{\Phi}$ and $\psi(-\mathbf{q}) = \overline{\psi(\mathbf{q})}$, we may rewrite Φ as follows:

$$\Phi = \frac{1}{2} C \cdot a^2 \int d\Omega \int_{-\infty}^{\infty} dq q^2 \cdot e^{i\mathbf{q}r \cdot \cos\theta/\hbar} / ((\kappa^2 + q^2)(a^2 + q^2)) \cdot \psi(\mathbf{q}) \quad (17)$$

where we have chosen polar coordinates defined by

$$q = q\mathbf{e} = q(\sin\Theta \cdot \cos\varphi, \sin\Theta \cdot \sin\varphi, \cos\Theta), \\ d\Omega = \sin\Theta d\Theta d\varphi$$

Now the q -integration can be evaluated by residuum techniques yielding

$$\Phi \approx -\pi\kappa \cdot \frac{1}{2} C \cdot a^2 \int d\Omega e^{-\kappa r \cdot |\cos\theta|/\hbar} / (a^2 - \kappa^2) \cdot \psi(i\kappa\mathbf{e}) \quad (18)$$

Here, we omitted the contribution resulting from the residuum at $q = \pm ia$, since it leads to an exponential decay in r which is much faster than (18) and is therefore not relevant for large r . We have also assumed that $\psi(\mathbf{q})$ has such a form that the integrand of (17) vanishes either for $Imq \rightarrow \infty$ or for $Imq \rightarrow -\infty$. Furthermore residuum contributions from $\psi(\mathbf{q})$ are neglected.

Now we calculate

$$H'_F \Phi = C \int d^3 q a^2 e^{i\mathbf{q} \cdot \mathbf{r}/\hbar} / ((\kappa^2 + q^2)(a^2 + q^2)) \cdot E_q(\alpha) \cdot \psi(\mathbf{q}) \quad (19)$$

Since only small q 's are relevant, we take $E_q(\alpha)$ as given by the last expression in (16). Calculating again the q -integral as described above, we obtain

$$H'_F \Phi \approx E(\alpha, 0) \cdot \cos(\kappa(-2m(\alpha)E(\alpha, 0))^{-1/2}) \cdot \Phi \\ = (E(\alpha, 0) - \kappa^2 / (2m(\alpha)) + O(\kappa^4)) \cdot \Phi \quad (20)$$

Hence $\kappa \sim (E(\alpha, 0) - E(\alpha, \beta))^{1/2}$ for $\beta \rightarrow \beta_c^+$. Now we proceed in analogy to (7) to determine γ . A physical condition in order to obtain a connection between the ground state energy and $\beta - \beta_c(\alpha)$ is the continuity of $(\partial\Phi/\partial r)/\Phi$ at $r=L$. For $r \rightarrow L^+$, inspection of (18) shows that $(\partial\Phi/\partial r)/\Phi$ is proportional to κ , i.e. proportional to $(E(\alpha, 0) - E(\alpha, \beta))^{1/2}$. Assuming that $(\partial\Phi/\partial r)/\Phi|_{r=L^-}$ is proportional to $\beta - \beta_c(\alpha)$ for $\beta \rightarrow \beta_c(\alpha)^+$, we obtain $\gamma=2$. δ is found by evaluating $M = (\langle \Phi | \mathbf{r}^2 | \Phi \rangle)^{-1}$. It turns out that for small κ , $M \sim \kappa^2 \sim (\beta - \beta_c(\alpha))^2$. Hence, $\delta=2$ and our somewhat intuitive determination of the critical exponents is finished.

The fundamental quantity which describes the influence of the phonons on the pinning transition is the function $\beta_c(\alpha)$. The next chapter is devoted to a discussion of the critical line in the $\alpha\beta$ -plane.

III. Calculation of the critical line for a screened Coulomb potential

Calculating the critical line $\beta_c(\alpha)$, one may use variational methods well-known from the polaron bound in a Coulomb potential, see Adamowski [10] for a review. Three aspects have to be taken into account. Firstly the numerical procedure should be capable to treat the purely electronic problem ($\alpha=0$) exactly, i.e. up to any desired accuracy. This is a one-particle quantum mechanical problem, which is easily solved using a sufficiently large number of electronic variational parameters.

Secondly, the polaron self energy ($\beta=0$) should be rather accurate, because the pinning transition is of second order and therefore $\beta_c(\alpha)$ depends sensitively on the position of the continuum edge.

As a third aspect, correlation effects between the potential and the phonon cloud around the electron are relevant resulting in a shift of $\beta_c(\alpha)$ to smaller values compared to $\beta_c(0)$.

Unfortunately, there exists no numerical procedure which takes into account all three aspects satisfactorily. On the one hand, path integral methods (see Platzman [9] and Matsuura [14]) yield a polaron

self energy by Feynman's method [15] which is very accurate. However, one has to approximate the potential $V(\mathbf{r})$ by a Gaussian potential and therefore a systematic error in calculating $\beta_c(\alpha)$ is made. On the other hand, other variational procedures treat the electronic problem exactly, but lead to a polaron self energy which is not as accurate as Feynman's result for intermediate α .

We use some of the latter method, namely the method of optimized canonical transformation proposed by Adamowski [10]. This method seems also to describe the correlation between the potential and the phonons well.

Adamowski approximates the polaron problem by a one particle problem; apart from the potential $\beta \cdot V(\mathbf{r})$ the associated Hamiltonian additionally contains two screened Coulomb potentials, an exponential potential and a self energy term. Furthermore the Hamiltonian depends on five phononic variational parameters. To calculate the ground state energy, we use an electronic trial function of the form

$$\Psi(\mathbf{r}) = \sum_{j=1}^4 c_j \exp(-\gamma_j r) \quad (21)$$

which depends on seven electronic parameters $c_2, c_3, c_4, \gamma_1, \dots, \gamma_4$. c_1 is obtained from the normalization condition.

To the end of this chapter we use polaron units of energy and length, $\hbar\omega$ and $(\hbar/(2m\omega))^{1/2}$ respectively. Note that then the kinetic term is p^2 not $p^2/2$.

As potential $\beta \cdot V(\mathbf{r})$ we take a screened Coulomb potential

$$V(\mathbf{r}) = -e^{-\mu r}/r \quad (22)$$

and set $\mu=1$, i.e. the screening length is comparable to the polaron radius. A physical more realistic potential is a dipole potential (see Sect. IV), but, as the pinning effects are not expected to depend on the explicit form of the potential sensitively, we choose here the simple screened Coulomb potential.

For $\alpha=0$, the critical potential strength is well known (see e.g. Kesarwani and Varshni [16]: $\beta_c(0) = 1.67981\dots$). With our ansatz we can reproduce this result with an error of 0.35%.

Our results are summarized in Fig. 1. Here three different lines are shown. The first solid line represents an upper bound on the exact $\beta_c(\alpha)$ and was calculated as follows: Of course the variational procedures yield upper bounds on the exact ground state energy. For α fixed and decreasing β , the value of β where the calculated energy equals the exact polaron self energy is an upper bound on $\beta_c(\alpha)$. For the polaron self energy we take the values of Larsen [17] for small α and for intermediate α we use the results of Adamowski, Gerlach and Leschke [18]. Of course, in a strict sense,

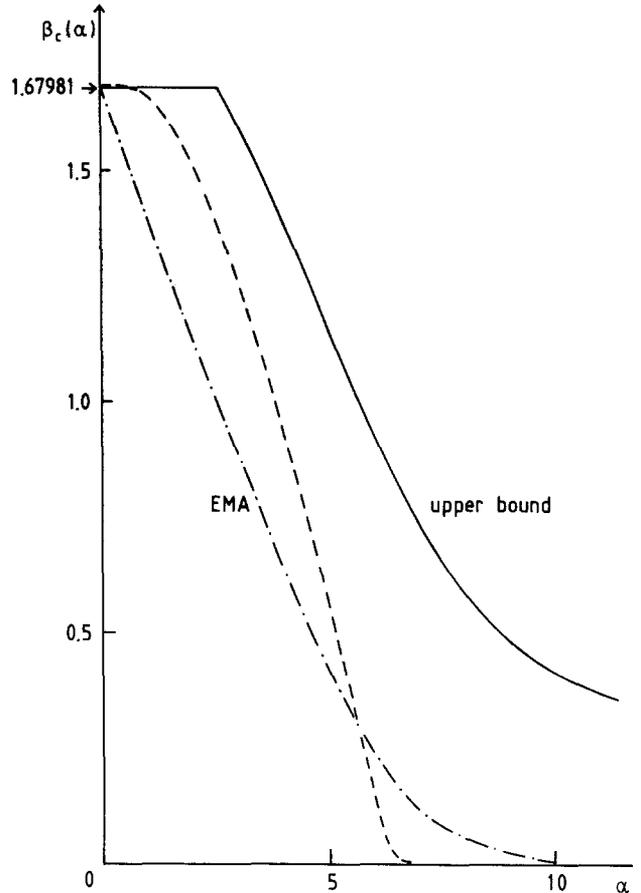


Fig. 1. Critical potential strength as function of the electron phonon coupling parameter α for a screened Coulomb potential. Upper bound (solid line), variational calculation (dashed line) and effective mass approximation (EMA)

these energies are upper bounds, too. However, the deviation from the exact energies is negligible for our purposes. By that way, we obtain an upper bound on $\beta_c(\alpha)$. Unfortunately, for $\alpha \gtrsim 2.6$, this upper bound is larger than $\beta_c(0)$. Therefore in this case the bound $\beta_c(\alpha) \leq \beta_c(0)$ known from [7] is better. Since the polaron self energy is not reproduced with sufficient accuracy by the variational method of Adamowski, our upper bound is a rather crude one.

To get a reasonable $\beta_c(\alpha)$ by our method, it seems helpful to calculate the continuum energy $E(\alpha, 0)$, too, and to take $\beta_c(\alpha)$ as that potential strength, where the calculated ground state energy abandons the calculated continuum energy. By this way, a cancellation effect of the errors in the polaron self energy occurs. The result is the dashed line; it is monotone decreasing in α . Of course, it is not any longer a definite bound on $\beta_c(\alpha)$.

The third line represents the effective mass approximation (EMA). The EMA is obtained by replacing the free polaron Hamiltonian by the effective one

particle Hamiltonian

$$H_{\text{eff}} = p^2/m(\alpha) + E(\alpha, 0) \quad (23)$$

Thus only the ground states of the momentum decomposed Hamiltonian with small total momentum are taken into account by H_{eff} . The excitation spectrum of one and more real phonons is completely neglected. In the neighbourhood of the pinning transition, however, just the ground states with small total momentum are relevant. Therefore it might be conjectured that the EMA describes the function $\beta_c(\alpha)$ correctly. At least, it should represent a *lower* bound on $\beta_c(\alpha)$ for a small α . This belief is based on a work of Mason and das Sarma [19]. They study a polaron bound in a Coulomb potential and compare the ground state energy shifts for fixed small α and varied β between the perturbational solution and the EMA. It turns out that the EMA yields an overestimation of the energy shifts.

Let $\beta_c^{\text{eff}}(\alpha)$ denote the critical potential strength of the EMA. Then performing simple scaling arguments for the effective mass Hamiltonian $p^2/m(\alpha) + E(\alpha, 0) - \beta \cdot V(\mathbf{r})$ we obtain

$$\beta_c^{\text{eff}}(\alpha) = \beta_c(0)/m(\alpha) \quad (24)$$

The polaron mass $m(\alpha)$ is well known, we take values from [20]. The limiting cases are $\beta_c^{\text{eff}}(\alpha) = \beta_c(0)(1 - \alpha/6)$ for small α and $\beta_c^{\text{eff}}(\alpha) = 44.05 \cdot \beta_c(0) \cdot \alpha^{-4}$ for large α . As a final remark, we state that the EMA reflects a universal behaviour, for the α -dependence of $\beta_c^{\text{eff}}(\alpha)$ doesn't depend on the explicit form of the potential.

Our results show that the EMA is lower than the other lines for small and intermediate α . For strong α , however, the result obtained by the method of Adamowski is lower.

IV. Experimental consequences and generalizations

To begin with, let us summarize the theoretical predictions concerning the pinning transition. Consider a conduction electron interacting with phonons and moving in an attractive short range potential. For increasing potential strength, the electron suddenly becomes localized at a critical potential strength $\beta_c(\alpha)$. For increasing electron-phonon coupling α , the critical potential strength shifts to smaller values. Near the pinning transition point there exist a localized ground state and delocalized continuum electronic states which are very close in energy.

There are several possibilities to produce a short ranged impurity potential in a crystal. One way to

do this is to consider isoelectronic impurities. In this case, however, the rangeness of the potential frequently is comparable to the lattice constant and therefore one should describe the electron-phonon interaction by a Holstein model. In fact, the analogon to the pinning transition in Holstein systems (called extrinsic self-trapping) was theoretically examined by Shinozuka and Toyozawa [21] in order to explain experimental data of Takahei and Kobayashi [22], which were performed at $\text{TlCl}_{1-c}\text{Br}_c$ systems.

The Fröhlich model, on the other hand, which we studied in this paper, is an adequate description if the rangeness of the potential is large compared to the lattice constant. A physical realization of such a potential is a dipole potential, for example, by substituting a Cl^- in NaCl by OH^- or a Br^- in KBr by CN^- . For sufficient large dipole potential strength, the existence of a bound electronic state is well known experimentally (see e.g. the review article of Narayanamurti and Pohl [23]), whereas for small dipole strength there is no such evidence for a bound state. The neighbourhood of the pinning transition ($\beta \approx \beta_c$) and its α -dependence, however, has not yet been examined systematically. One should examine several crystals with different electron-phonon coupling α and with different dipolar impurities. At a critical (α -dependent) potential strength of the dipolar impurity, the electron is trapped in the attractive 'club' of the dipole potential. This has direct consequences on the excitation spectra and the conductivity. In particular, the pinning transition is connected with a transition from a weakly conducting state ($\beta < \beta_c$) to an isolated state.

However, finite temperature casts serious problems on such a simple measurement. On the one hand, the temperature has to be low enough so that a one particle description of the conduction electron is adequate (i.e. many body effects should be negligible). On the other hand, finite temperature is needed to produce conduction electrons in a semiconductor, but it destroys the abruptness of the pinning transition.

Also spectroscopical measurements should be useful in detecting the pinning transition. Dmochowski et al. [24] have measured the vibronic coupling strength of a bound polaron ground state in CdF_2 : In as a fingerprint of its localization. Their result apply to Toyozawa's model of impurity self trapping of a small polaron but their experimental techniques should also be useful for Fröhlich polarons. Particularly, they study the phononic part of the wave function which also becomes localized as $\beta \rightarrow \beta_c^-$.

Another application is the pinning transition for excited bound states in a quantum well. A polaron in a quantum well is described by adding a triangle potential $\beta \cdot V(z)$, $\mathbf{r} = (x, y, z)$ to the free polaron

Fröhlich Hamiltonian, the x and y components of the total momentum Q_x, Q_y are conserved. Restricting oneself to fixed total momentum $Q_x = Q_y = 0$, the number of bound states in the z -direction depends on the potential strength and on the electron phonon coupling. The ground state, however, is always localized for $\alpha \geq 0$ and $\beta > 0$, as $V(z)$ is a one dimensional potential. Since the explicit form of $V(z)$ can be varied, e.g. by varying the thickness of the layers, one can be optimistic to verify a pinning effect spectroscopically.

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