$N-$polaron systems and mathematics

(with R. Frank, E. Lieb, and R. Seiringer)

Outline

- A very brief history of the polaron
- Lower bounds on the ground state energy for polarons
- Recent results on the binding of polarons
- Analysis of the bi-polaron
- Polaron effective mass

The model and some history

• H. Frölich, (1937). Model for an electron moving in an ionic crystal. Let $\mathbf{P}(x)$ be the polarization of an ionic crystal at $x \in \mathbb{R}^3$.

$$\phi(x) = \int_{\mathbb{R}^3} dy \frac{(x-y)\cdot\mathbf{P}(y)}{|x-y|^3} = \int_{\mathbb{R}^3} dy \frac{\nabla\cdot\mathbf{P}(y)}{|x-y|}$$

and neglecting transverse modes:

$$\phi(x) = \frac{1}{\sqrt{2}\pi} \int_{\mathbb{R}^3} \frac{dk}{|k|} \left(e^{ikx} a(k) + e^{-ikx} a^\dagger(k) \right).$$

Here $a(k)$ and $a^\dagger(k)$ are phonon annihilation and creation operators, momentum k, $[a(k), a^\dagger(k')] = \delta(k-k')$. Leads to **Polaron Hamiltonian** for electron moving in a phonon field,

(1) $$H^{(1)} = p^2 + \sqrt{\alpha}\phi(x) + \int a^\dagger(k) a(k)\, dk.$$

acting in $L^2(\mathbb{R}^3) \otimes$Fock space. Here, $p^2 = -\nabla^2$. Simplest example of quantized particle moving in a quantized radiation field.

A **polaron** is the electron plus its entourage of phonon–excitations of the field surrounding the electron.

Or, if N particles, N-**polaron Hamiltonian**

$$H_U^{(N)} = \sum_i^N \left(p_i^2 + \sqrt{\alpha}\phi(x_i)\right) + \int a^\dagger(k)a(k)\,dk + U \sum_{i<j}^N \frac{1}{|x_i - x_j|}.$$

• Pekar (1946), Pekar and Landau (1948). Concerned with computing the polaron ground state energy for single polaron,

$$E_0^{(1)}(\alpha) = \text{inf spectrum}\, H^{(1)}(\alpha)$$

of the polaron. Consider a product state $\psi = f(x) \times g$(phonon variables). Get

$$(2)\quad E_P(\alpha) = \inf_{\|f\|_2=1} \left\{ \int_{\mathbb{R}^3} |\nabla f|^2\, dx - \frac{\alpha}{2} \iint_{\mathbb{R}^3\times\mathbb{R}^3} \frac{|f(x)|^2\, |f(y)|^2}{|x-y|}\, dx\, dy \right\}$$

Thus, $E_P(\alpha) > E_0^{(1)}(\alpha)$ (variational calculation). Multi-electron version goes under the name Tomasevich-Pekar (1951) functional. There was "belief" emerging that the two energies, $E_P(\alpha)$ and $E_0^{(1)}(\alpha)$ were nearly equal, at least for large α.

• Lee, Low and Pines (1953). Perturbation theory (connection with meson calculations).

• Feynman (1955) wrote this ground state energy as a functional integral over (3-dimensional) Brownian motion $x(t)$: If

(3) $$Z^{(1)}(\alpha,T) = E\left[\exp\left\{\frac{\alpha}{2}\int_0^T\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x(t)-x(s)|}\right\}\right],$$

then

$$E_0^{(1)}(\alpha) = -\lim_{T\to\infty}\frac{1}{T}\ln\left(Z^{(1)}(T)\right).$$

By Jensen's inequality, Feynman got an upper bound on $E_0^{(1)}(\alpha)$ for large α close to Pekar $E_P(\alpha)$. (Both energies go like $c\alpha^2$, with the two constants within 3%–Pekar was better.)

• "Simply a mathematical problem" of computing the energy (and mass)."

For later purposes, we note that

$$Z_U^{(N)}(\alpha,T)$$
$$= E\left[\exp\left\{\sum_{i,j}^{N}\frac{\alpha}{2}\int_0^T\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x_i(t)-x_j(s)|} - U\sum_{i\neq j}^{N}\int_0^T\frac{dt}{|x_i(t)-x_j(t)|}\right\}\right].$$

• Lieb and Yamazaki (1958). Lower bound on the ground state energy.

• Donsker and Varadhan (1983). Large deviations for diffusions applied to the polaron ground state. Pekar is exact in limit, $\alpha \to \infty$. Lieb and Thomas (1997) gave estimate

$$E_P(\alpha) - E_0^{(1)}(\alpha) = \mathcal{O}(\alpha^{9/5}).$$

• Bogolubov and Bogolubov, Jr.(2000), (Time-ordered product representation, Thermodynamic states.)

• DeVreese (...). Argument over applicability to high temperature superconductivity.... (See Physics World, Oct.1998) "Superconductivity debate gets ugly" and, particularly, Chakraverty, Ranniger, and Feinberg, Phys. Rev. Letters, **81**, 433. which ruled out polarons as a player in High T_c.

From their paper:

"As concerns the question whether bipolarons could possibly play the role for such bosonic quasiparticles and their condensation, we find that such a possibility is ruled out. The tragedy of beautiful theories, Aldous Huxley once remarked, is that they are often destroyed by ugly facts. One perhaps can add that the comedy of not so beautiful theories is that they cannot even be destroyed; like figures in a cartoon they continue to enjoy the most charming existence until the celluloid runs out." (And response in PRL from Alexandrov...).

• *Remarks* Lower bounds and stability. Polaron provides a relatively simple laboratory for computations.

A lower bound on $E_U^{(N)}(\alpha)$, upper bound on $Z_U^{(N)}(\alpha, T)$.

- $(N = 1)$, a probability bound. By Clark-Ocone formula, write

$$
\begin{aligned}
&\frac{\alpha}{2}\int_0^T\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x(t)-x(s)|} \\
(4) \qquad = \quad &\frac{\alpha}{2}E\left[\int_0^T\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x(t)-x(s)|}\right] + \alpha\int_0^T \rho(x(\cdot),T)\cdot dx(s),
\end{aligned}
$$

with $\rho(,)$ a *bounded function*. Get bound

$$
E_0^{(1)}(\alpha) \geq -\frac{\alpha}{2}E\left[\int_0^\infty \frac{e^{-|t|}\,dt}{|x(t)|}\right] - \frac{3\alpha^2}{2}\|\rho\|_\infty^2
$$

Set

$$E^{(N)}_{U=0}(\alpha) = \text{inf spectrum}\, H^{(N)}_U(\alpha)$$

• $(N \geq 2)$. Since $e^{-|t|}$ is positive definite as is $1/|x|$,

$$\begin{aligned} & 2\int_0^T\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x_i(t)-x_j(s)|} \\ \text{(5)} \qquad & \leq \quad \int_0^T\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x_i(t)-x_i(s)|} + \int_0^T\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x_j(t)-x_j(s)|} \end{aligned}$$

which implies that

$$E^{(N)}_{U=0}(\alpha) \geq N E^{(1)}_0(N\alpha).$$

Recent Results on non-binding of polarons

Problems: a bowling ball–cheap mattress analogy

- For two polarons, there is an effective attraction, so that they bind. For U small, small Coulomb repulsion, there is binding of polarons, in particular

  $$E_U^{(2)}(\alpha) < 2E_0^{(1)}(\alpha)$$

  Can Coulomb repulsion be strong enough (U large) so that the polarons don't bind?

- For N polarons can the Coulomb repulsion be made large enough so that the system is stable, $E_U^{(N)}(\alpha) \geq -CN$?

  Can the repulsion be still stronger so that there are no polaron molecules or clusters?

Frank, Lieb, Seiringer, Thomas, Phys. Rev. Lett. **104** (2010); Les Pubications Mathématics de l'IHÉS **113** no.1 (2011).

- For the bi-polaron problem, $N = 2$.

Theorem 1. *There exists a constant C such that if $U \geq C\alpha$, then there is no binding for the bi-polaron system, i.e.,*

$$E_U^{(2)}(\alpha) = 2E_0^{(1)}(\alpha).$$

- Multi-polaron case, arbitrary N.

Theorem 2. *For $U \geq 2\alpha$, the multi-polaron system is stable, i.e, there is a constant $C(U,\alpha)$ such that*

$$E_U^{(N)}(\alpha) \geq -C(U,\alpha)N$$

.

Moreover, there exists a constant $C(\alpha)$ such that for $U \geq C(\alpha)$,

$$E_U^{(N)}(\alpha) = NE_0^{(1)}(\alpha).$$

In other words, there are no polaron clusters.

Analysis for bi-polaron case

• If particles were confined to separated boxes of size ℓ a distance $\geq c\ell$ then a repulsive Coulomb potential can compensate: Recall (for $N = 2$)

$$Z_U^{(2)}(\alpha) = E\left[\exp\left\{\sum_{i,j}^{2} \frac{\alpha}{2}\int_0^T\!\!\int_0^T \frac{e^{-|t-s|}\,dt\,ds}{|x_i(t)-x_j(s)|} - U\int_0^T \frac{dt}{|x_1(t)-x_2(t)|}\right\}\right].$$

The "cost" of confining the particles to such boxes is c_o/ℓ^2.

- **Localization: cost of confining**
    - As a "reverse" uncertainty principle.

Let

$$\phi(x) = \begin{cases} \cos(x\pi/2\ell) & \text{for } |x| \leq \ell, \\ 0 & \text{otherwise.} \end{cases}$$

Then

$$\sum_k \phi^2(x + k\ell) \equiv \sum_k \phi_k^2(x) \equiv 1.$$

Then for a Schrödinger state ψ, with energy expectation

$$\langle \psi, H\psi \rangle \equiv \langle \partial_x \psi, \partial_x \psi \rangle + \langle \psi, V\psi \rangle = E\langle \psi, \psi \rangle,$$

we have an identity:

$$\sum_k \left\{ \langle \partial_x \phi_k \psi, \partial_x \phi_k \psi \rangle + \langle \phi_k \psi, V\phi_k \psi \rangle - \left(\frac{\pi^2}{4\ell^2} + E\right) \langle \phi_k \psi, \phi_k \psi \rangle \right\} \equiv 0$$

At least one of the terms must be less than zero:

Lemma 1. *Let* ψ *be a state with energy expectation* $\langle\psi, H\psi\rangle = E\langle\psi,\psi\rangle$. *Then the state* ψ *can be* *localized* *in a box* $[2\ell]^d$, *call it* ψ_D, *so that*

$$\langle\psi_D, H\psi_D\rangle \le \left(E + \frac{\pi^2 d}{4\ell^2}\right)\langle\psi_D,\psi_D\rangle.$$

• A slight rewriting: Suppose the particles are confined to cubes of size ℓ, and the cubes are separated by distance $d \geq \ell$. Then for such confined particles, ($H_\ell^{(2)}$ will mean Dirichlet boundary conditions for $H_{U=0}^{(2)}(\alpha)$)

(6) $$H_\ell^{(2)} + q/|x_1 - x_2| \geq 2E_0^{(1)}$$

for $q > c\alpha$ and some constant c.

Let then $V = q/|x_1 - x_2|$, with the q as above, $c_o = 6(\pi/2)^2$, and let ℓ_0 be defined by the <u>gap</u>

$$\frac{c_0}{\ell_o^2} = 2E_0^{(1)} - E_{U=0}^{(2)}$$

Set

(7) $$W_j(x) = \frac{c_0}{2^{j-1}\ell_0^2}\chi_{[0,10\times 2^{(j-1)/2}\ell_o]}(|x|).$$

indicator function for the interval $[0, 10 \times 2^{(j-1)/2}\ell_o]$).

Claim 1. For some constant C

(8) $$\sum_{j=1}^{\infty} W_j(x) \leq \min\left\{\frac{C}{|x|^2}, 2c_o/\ell_o^2\right\}.$$

Claim 2. For any two-electron Ψ,

$$\langle \Psi, H^{(2)}_{U=0} + V + \sum_j^n W_j(x_1 - x_2)\Psi \rangle \geq -\frac{c_0}{2^n \ell_o^2} + 2E_0^{(1)}. \tag{9}$$

Proof. Induction. $n = 1$ First take cubes of size $\ell_1 = \sqrt{2}\ell_o$, so that

$$\langle \Psi, H^{(2)}_{U=0} + V + W_1(x_1 - x_2)\Psi \rangle \geq -\frac{c_0}{2\ell_o^2} + \langle \Psi_D, H^{(2)}_{\ell_1} + V + W_1(x_1 - x_2)\Psi_D \rangle$$

where the Ψ_D denotes the "confined" Ψ, with the particles in their cubes, the cubes chosen using $H^{(2)}_{U=0} + V + W_1$. That first term on the rhs is the cost of confining the two particles.

a) These cubes could be within a distance $\leq \ell_1$ of each other: If so, $\langle \Psi_D, H^{(2)}_{U=0} + W_1 \Psi_D \rangle \geq 2E_0^{(1)}$ since $W_1 = c_0/\ell_0^2$ on the support of Ψ_D and so compensates for the gap. $V \geq 0$ does no harm.

b) If the two cubes are of distance $\geq \ell_1$, we have that $\langle \Psi_D, H^{(2)}_{U=0} + V\Psi_D\rangle \geq 2E_0^{(1)}$ by Eq.(6) and even more so with W_1 in there.

Thus, in either case the expectation on the rhs is $\geq 2E_0^{(1)}$ and the induction starts. (Take $\ell_n = 2^{n/2}\ell_o$....)

Now pick another Coulomb potential which majorizes $V + \sum W$.

Question: The 1-polaron effective mass problem

Set

$$E_0^{(1)}(\alpha, p) = E_0^{(1)}(\alpha) + \frac{p^2}{2m(\alpha)}$$
$$= -\lim_{T\to\infty} \frac{1}{T} \ln \left(E \left[\exp \left\{ \frac{\alpha}{2} \int_0^T \int_0^T \frac{e^{-|t-s|}\, dt\, ds}{|x(t) - x(s) + (t-s)p|} \right\} \right] \right)$$

Here, $m(\alpha)$ is an effective mass for the polaron as a dressed particle. What's $m(\alpha)$?