

Anomalous Behavior in an Anderson-Holstein Model for a Single Molecule Transistor

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Abstract

This lab was designed to test whether Poisson statistics can be used to model the occurrence of radio active decay. The equipment used included: a scintillation detector used to observe the count data for a radio active source, and a Geiger counter to observe the background radiation count. We get good agreement when the correct type of distribution is used.

In this paper we discuss the Kondo behavior of a single molecular transistor using the Anderson-Holstein, [1] model using the numerical renormalization group (NRG) method. Within the Anderson-Holstein model there is an effective potential between electrons defined as: $U_{eff} = U_{Coulomb} - U_{phonon}$. Here, $U_{Coulomb}$ is the Coulomb repulsion between electrons in the transistor, and U_{phonon} is a phonon-mediated attraction. A previous study [4] has revealed anomalous behavior for the particular case $U_{eff} = 0$ where the Coulomb repulsion and phono-mediated attraction should exactly cancel. Instead it has been found that for large $U_{Coulomb} = U_{phonon}$ the properties are similar to those expected for a value $U_{eff} > 0$.

We study the Anderson-Holstein Hamiltonian [1] for a single molecule transistor coupled

linearly to a phonon mode of frequency ω_0 . This can be written in three different terms:

$$H = H_m + H_e + H_{m-e} \quad (1)$$

H_m describes the molecule, H_e describes the source and drain electrodes and H_{m-e} describes the coupling between the molecule and electrodes. Written explicitly in second quantization formalism,

$$H_m = \epsilon_d n_d + U n_{d\uparrow} n_{d\downarrow} - \lambda(n_d - 1)(a + a^\dagger) + \omega_0 a^\dagger a \quad (2)$$

$$H_e = \sum_{\mathbf{k}, \sigma} \epsilon(\mathbf{k}) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} \quad (3)$$

$$H_{m-e} = \sum_{\mathbf{k}, \sigma} V_{\mathbf{k}\sigma} (d^\dagger c_{\mathbf{k}\sigma} + H.c.) \quad (4)$$

In H_m , ϵ_d is the position of the electronic molecular level relative to the Fermi level of the electrodes, $n_d = \sum_\sigma d_\sigma^\dagger d_\sigma$ counts the number of electrons on the molecule and U is the Coulomb repulsion between the electrons. The final two in H_m terms describe the phonon vibrational modes. In H_e , the summation counts the number of conduction electrons in the electrodes, while $\epsilon(\mathbf{k})$ is the dispersion of the conduction electrons. Finally H_{m-e} describes the process of electrons hopping from the leads to the molecule and the reverse process, where $V_{\mathbf{k}\sigma}$ is the tunneling matrix element for each process. In all three Hamiltonians the term $c_{\mathbf{k}\sigma}$ destroys an electron of spin σ and wave vector \mathbf{k} in a linear combination of source and drain states having energy $\epsilon(\mathbf{k})$, the Hermitian conjugate operator creates an electron in the same way. The term d destroys an electron on the molecule while d^\dagger creates an electron on the molecule.

We use the NRG method to analyze the Anderson-Holstein Hamiltonian. In this method the continuum of conduction band energies is logarithmically discretized to allow for the greatest resolution around the Fermi level. Then the conduction band Hamiltonian (eqn 3)

is mapped via a Lanczos transformation onto a chain form that can be solved iteratively by diagonalizing a sequence of approximate Hamiltonians H_n , $n = 0, 1, 2, \dots$. Successive Hamiltonians H_n and H_{n+1} are related by,

$$H_{N+1} = \sqrt{\Lambda}H_N + \sum_{\sigma} (c_{N,\sigma}^{\dagger}c_{N+1,\sigma} + c_{N+1,\sigma}^{\dagger}c_{N,\sigma}) \quad (5)$$

where Λ is the discretization parameter. [3] In this paper we set $\Lambda = 2.5$ and keep between 1000 and 4000 states, although we rarely found it necessary to go beyond 3000 states. The number of bosons was restricted to a maximum N_b that we set between 7 and 31. In this paper we also set $V_{\mathbf{k}\sigma} = 0.25$, $\omega_0 = 0.5$ and $\epsilon_d = -U_{Coulomb}/2$. All energies are given in multiples of the conduction band width D and temperatures are given in multiples of D/k_b where k_b is the Boltzmann constant and $D = 1$. Finally we find it convenient to define $\alpha = \lambda^2/\omega^2$

Given the eigenstates $|m\rangle$ and their energies E_m after iteration N , one can calculate the molecular contributions to spin and charge susceptibilities.

$$\chi_c = \frac{1}{Z} \sum_{n,m} \frac{e^{-E_n/k_B T_n} - e^{E_m/k_B T_n}}{E_m - E_n} |\langle n|N - \langle N\rangle|m\rangle|^2 \quad (6)$$

$$\chi_s = \frac{1}{Z} \sum_{n,m} \frac{e^{-E_n/k_B T_n} - e^{E_m/k_B T_n}}{E_m - E_n} |\langle n|S_z|m\rangle|^2 \quad (7)$$

Where S_z is the total spin-z operator and N is the total electron number operator and T_N is a temperature scale chosen so that $k_B T_N \sim D\Lambda^{-N/2}$, ie. the thermal energy matches the characteristic energy scale of the state. $Z_n = \sum_n e^{-E_n/k_B T}$ is the partition function at temperature T . [2]

One can also calculate the molecules contribution to the energy:

$$U = \frac{1}{Z} \sum_n E_n e^{-E_n/k_B T} - \frac{1}{Z_0} \sum_n E_n^0 e^{-E_n^0/k_B T} \quad (8)$$

Where the first term describes the system with a molecule and the second describes a system without a molecule. Thus the molecular contribution to the free energy can be calculated:

$$F = -k_B T \ln(Z/Z_0) \quad (9)$$

where Z is the partition function with the molecule and Z_0 is the partition function without the molecule. From F we can obtain the entropy $S = (U - F)/T$ and the specific heat $C = T \frac{dS}{dT}$ [2]

1 $U_{Coulomb} > 0$

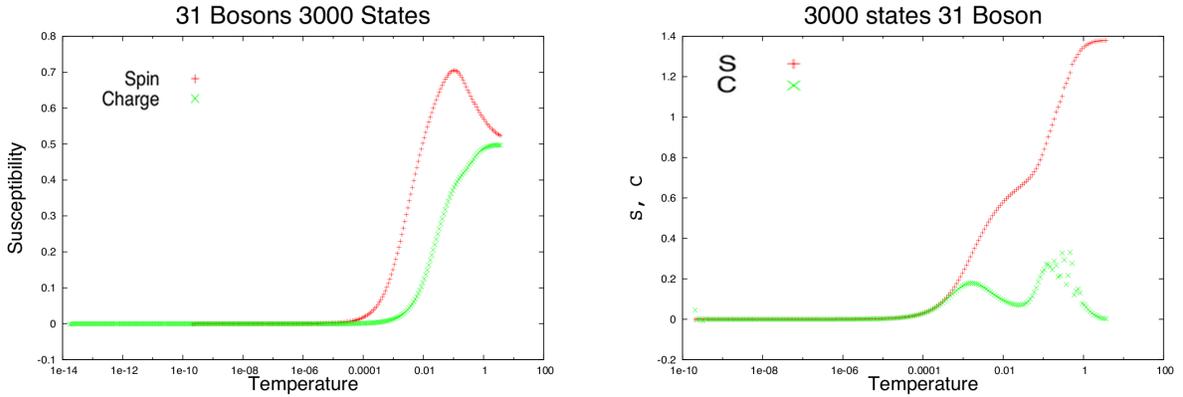


Figure 1: with $U_{Coulomb} = 1$ and $\alpha = -.3$

Although we are interested in the regime where $U_{Coulomb} = U_{phonon}$ we must first explain

what occurs in the regime where $U_{Coulomb} > U_{phonon}$ and hence $U_{eff} > 0$. Here in figure 1 we see the charge susceptibilities are rapidly suppressed, going to zero around a temperature of 10^{-2} while the spin susceptibility increases. In the entropy plot we see that the entropy plateaus at a value of $S = \log(2)$ indicating that only the spin up and spin down states are available. [4] The onsite Coulomb interaction leads to the presence of only a single electron on the molecule, flipping its spin as current flows.

2 $U_{eff} = 0$

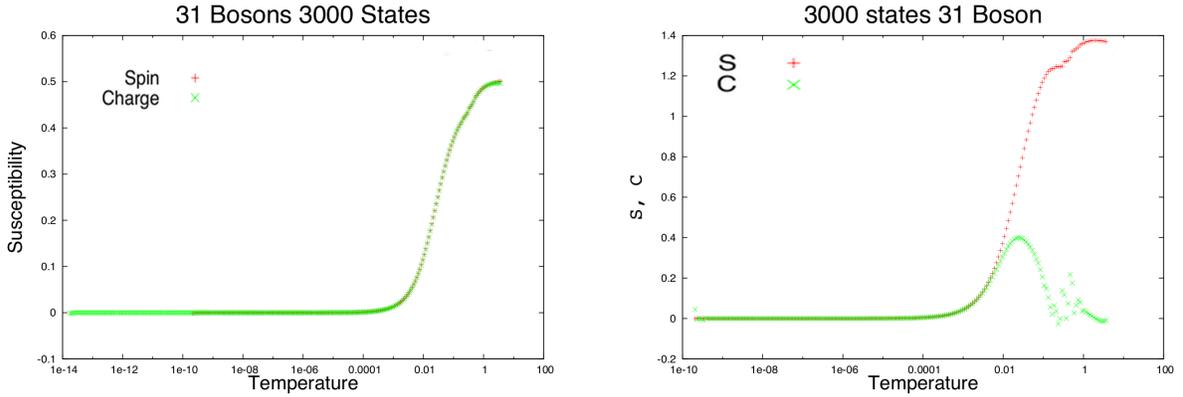


Figure 2: with $U_{Coulomb} = U_{phonon} = 1$ we see the equivalence of spin and charge susceptibilities as expected.

In studying the case $U_{eff} = 0$ we initially set $U_{Coulomb} = U_{phonon} = 1$. In this regime we naively expect an equivalence of spin and charge susceptibilities and for $U_{coulomb} = 2\alpha\omega_0 = 1$ we observe just that. In figure 2 we see that it is extremely difficult to distinguish the spin and charge susceptibilities, in addition the entropy curve has a small plateau at $S = \log(4)$ showing that all four states are available to the system. This indicates that the onsite Coulomb interaction is exactly cancelled by the phonon mediated attraction.

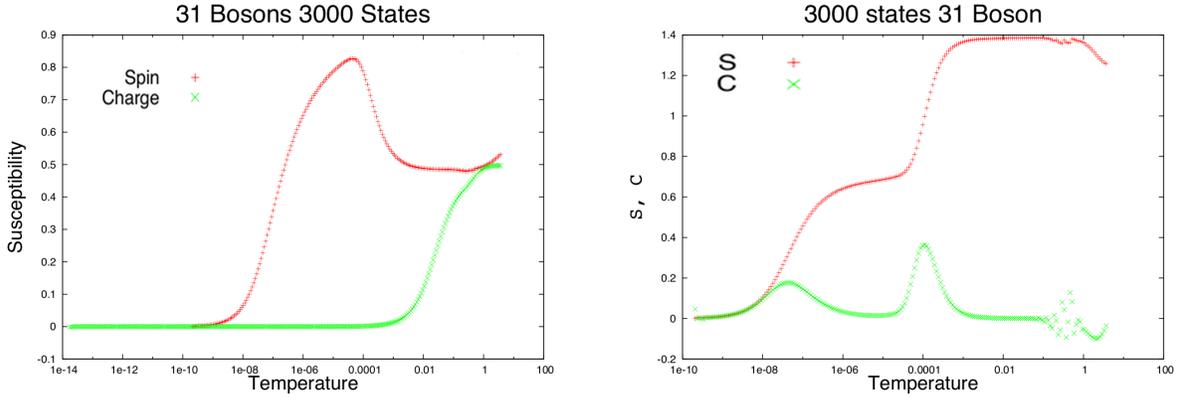


Figure 3: spin and charge susceptibility for $U_{Coulomb} = U_{phonon} = 9$

Now we set $U_{Coulomb} = 2\alpha\omega_0 = 9$. One would expect similar results to figure 2, as the onsite interaction is still zero. However we see in figure 3(a) that the charge susceptibility is suppressed while the spin susceptibility increases. The results in figure 3 appear to more closely resemble the results from the previous section. The reader should note this is odd given that the onsite interaction has been set to zero. In addition we also plot the entropy and specific heat in figure 3(b). Where we see a plateau around $S = \log(4)$ and a second where $S = \log(2)$. This, along with the suppression of the charge susceptibility, indicates that the double occupied and unoccupied states are not available at low temperature in this regime.

To investigate this further we look at the expectation of the double occupancy state, $\langle n_{\downarrow}n_{\uparrow} \rangle$, as seen in figure 4 labeled by their α value. We note that if there on site interaction were zero we'd naively expect that for all values of α that $\langle n_{\downarrow}n_{\uparrow} \rangle = 0.25$ However we see that $\langle n_{\downarrow}n_{\uparrow} \rangle$ is rapidly suppressed for all values of α thus this implies the existence of a hitherto unknown onsite interaction: $U_{residual}$. We can see that for $\alpha \leq 4$ that the expectation value isn't suppressed significantly, keeping a value above 0.2, however the value for $\alpha > 4$ is deeply suppressed with $\alpha = 9$ having $\langle n_{\uparrow}n_{\downarrow} \rangle < 0.05$. However all have the same high temperature

value of $\langle n_{\uparrow}n_{\downarrow} \rangle$ as expected.

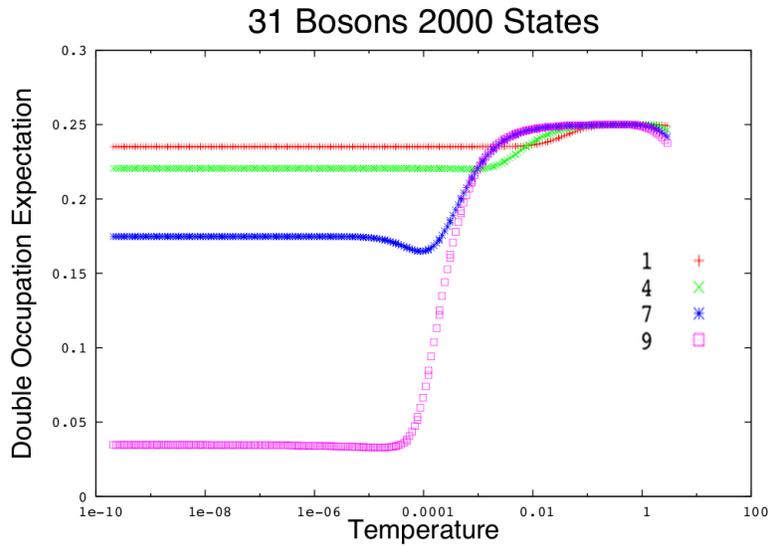


Figure 4: The double occupation for various α

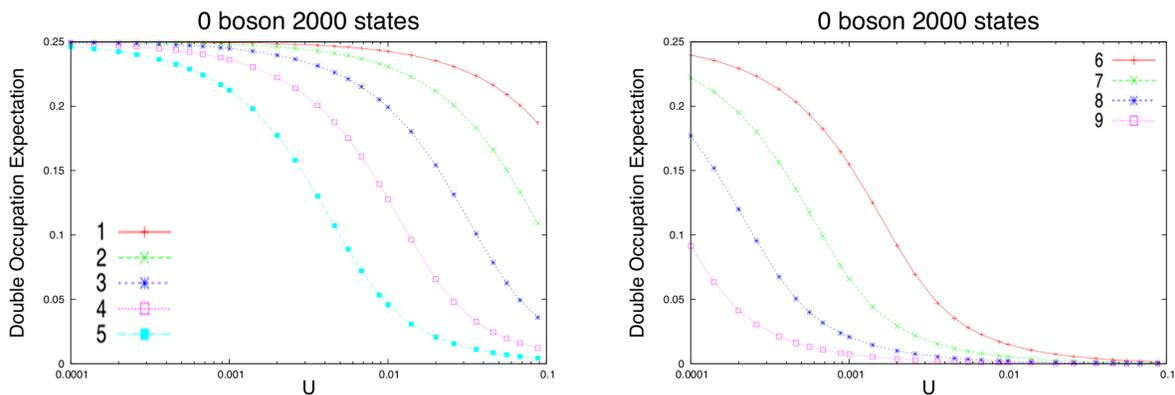


Figure 5: Shows how the double occupation expectation changes with respect to $U_{Coulomb}$. The plots are labeled for various of α . We use 2000 states due to time constraints

Once the $\langle n_{\uparrow}n_{\downarrow} \rangle$ was known we were then able to determine the value of the residual interaction $U_{residual}$. To do so we started by making figure 5; to calculate this we defined

the effective hybridization to be $V_{eff} = Ve^{-\alpha^2/2}$. Then $\langle n_{\uparrow}n_{\downarrow} \rangle$ was calculated for various values of $U_{Coulomb}$ in the zero boson limit. We can see for large α that the occupation is not close to the expected value of 0.25, even for extremely small interaction potential. This is puzzling, so we mapped the low temperature $\langle n_{\uparrow}n_{\downarrow} \rangle$ values in figure 4 to the potential values in figure 5. This produced figure 6. Figure 6 shows how this $U_{residual}$ affects the $\langle n_{\uparrow}n_{\downarrow} \rangle$

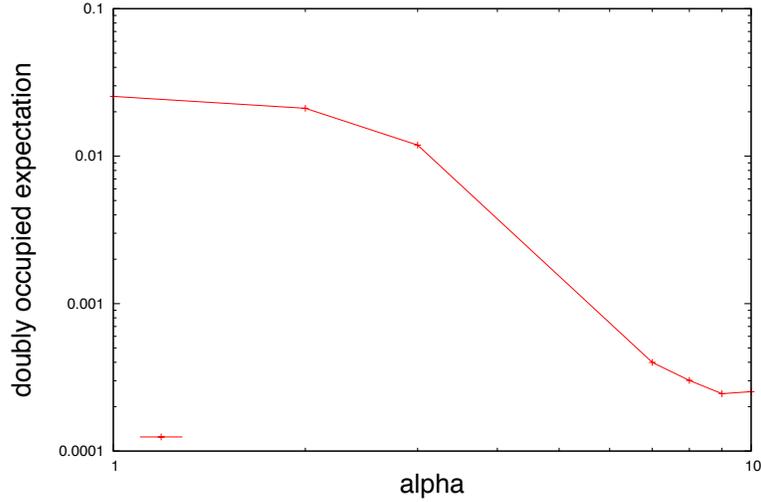


Figure 6: Shows how the double occupation expectation changes with respect to α . This shows how α affects the double occupied expectation value

values. However to gather any conclusions from figure 6 more work is required. First off there is no point for $\alpha = 0$ and the asymptotic behavior for $\alpha \rightarrow \infty$ needs to be investigated further. Hotta concludes that this interaction for $\alpha = 9$ is caused by a polaron interaction in the molecule. [4] However we feel that further behavior into figure 6 is required before such a conclusion can be drawn.

3 References

- [1] A.C. Hewson and D Meyer, J. Physics. Condensed Matter 14, 427 (2002)
- [2] K Ingersent, personal interview. June 29th 2011
- [3] T Costi, *Wilson's Numerical Renormalization group* (Chapter 1)
- [4] T Hotta, J. Phys. Soc. Jpn. 76 8 (2007)