Appendix A

Temporal Bell Inequalities

A.1 An Introduction to Temporal Bell Inequalities:

In a paper provocatively entitled "Quantum Mechanics versus Macroscopic Realism: Is the Flux There when Nobody Looks? A. J. Leggett and A. Garg[1] proposed a way to determine whether the magnetic flux of a SQUID (superconducting quantum interference device) was compatible with the postulates:

(A1) *Macroscopic Realism*: "A macroscopic system with two or more macroscopically distinct states available to it will at all times *be* in one or the other of these states."

(A2) *Noninvasive Measurability*: "It is possible, in principle, to determine the state of the system with arbitrary small perturbation on its subsequent dynamics."

The interest in SQUIDS arises because they may perhaps be prepared in "macroscopic" current superpositions, thus testing whether Quantum Mechanics(QM) holds

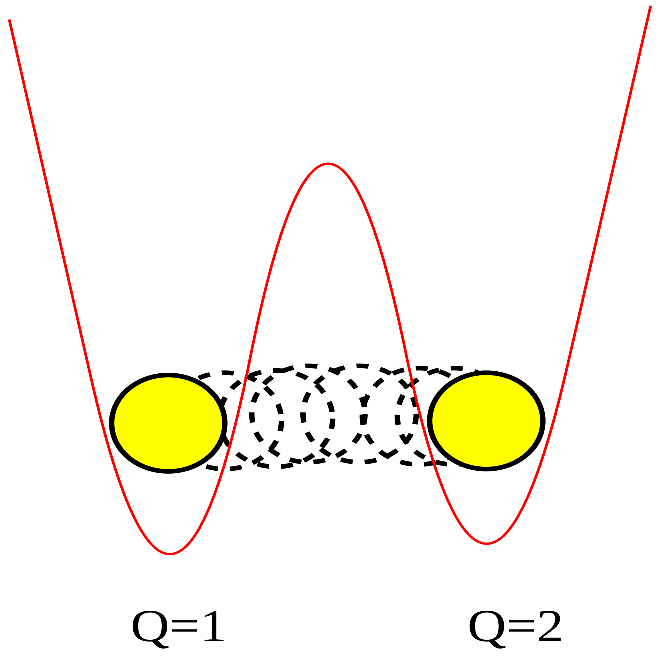

Figure A.1: **double-well potential for a tunneling particle**
A particle tunnels from one well to another. The variable Q measures in which well the particle is situated at a given time t_i.

on the macroscopic level. Let us consider a particle in a double well such as the one depicted in figure A.1 as a simple model.

Define an operator $\hat{Q}(t)$ which takes on the value $+(-)1$ when the particle is measured in the left(right) well. It then follows that for a set of times t_1, t_2, t_3, t_4, the (hypothetical) measurement values (eigenvalues) of the $Q's$ obey the relation:

$$-2 \leq Q(t_1)Q(t_2) + Q(t_2)Q(t_3) + Q(t_3)Q(t_4) - Q(t_1)Q(t_4) \leq 2 \qquad \text{(A.1)}$$

, where $Q(t)$ is a value of the operator $\hat{Q}(t)$. If one assumes *Macroscopic Realism* (A1), then Q(t) takes the meaning of a history or trajectory in some generalized coordinate space, at least if these histories exist in the sense that at any time the particle is one well, the histories Q(t) are an “element of reality”. Armed with this, define correlation functions by averaging over an ensemble of systems following different histories. The two-time correlation functions would be:

$$K_{12} = K(t_1, t_2) = \int [DQ] P[Q(t)] Q(t_1) Q(t_2). \tag{A.2}$$

The integral is over all histories, and $P[Q(t)]$ is the probability of obtaining a certain history in the given ensemble. By averaging the relation between the $Q(t)$'s of equation (A.1) over a given ensemble, we obtain a *Temporal Bell Inequality*:

$$|K_{12} + K_{23} + K_{34} - K_{14}| \leq 2, \tag{A.3}$$

as suggested by Leggett and Garg.

To measure the correlation functions of eq(A.2) one needs the assumption **(A2)** of *Noninvasive Measurability* because for a given history, $Q(t_2)$ is taken to be independent of what the measurement of $Q(t_1)$ gave. In other words, one wants a *conditionally independent representation* of the joint probability distributions. Thus the first analogy with the usual spatial Bell inequalities is that here, noninvasive measurability is similar to locality in space. In other words, the temporal Bell inequalities assume a "locality in time". The time correlation functions K_{ij} are similar to the spatial correlation functions used in the usual Bell inequalities.

But, in quantum theory one cannot assign a definite value to an observable until it has been measured (unless it is in an eigenstate of the measuring apparatus). Thus one cannot consistently define a probability for a given set of histories for time inequalities, and the temporal Bell inequalities can be violated. As a specific example of quantum dynamics consider the double well system of figure 1, but now in the case where it can be accurately approximated by the two-state Hamiltonian[2]:

$$H = -\frac{\hbar}{2} \Delta \sigma_{\mathbf{x}}. \tag{A.4}$$

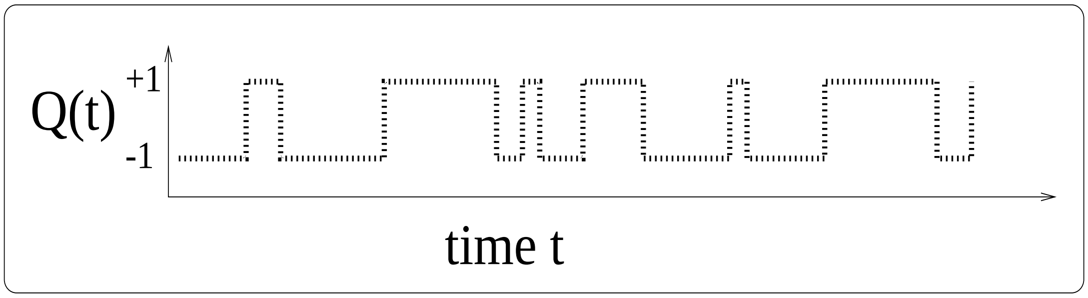

Figure A.2: **A classical history for a particle tunneling between two wells** The operator $\hat{Q}(t)$ takes on the definite value $Q(t) = +(-)1$ when the particle is measured in the left(right) well. Such classical "telegraph noise" histories do not exist for quantum coherent tunneling of the particle.

If the system is prepared in an eigenstate of the σ_z operator, i.e. it is in a definite $Q(t)$ state at time $t = t_1$, then the time correlation is given by:

$$K_{12} = cos(\Delta(t_1 - t_2)) \tag{A.5}$$

so that the time $(t_1 - t_2)$ is analogous to the angle between the polarization measured on each part of an EPR pair. It is not hard to find a set $\{t_1, t_2, t_3, t_4\}$ that violates the inequality (A.3)[1]. A set of histories such as is depicted in figure A.2 does not exist. Here one sees an immediate contrast to the spatial EPR which is usually formulated in terms of one state (the singlet state for spins or polarizations). In the temporal case, the measurement of any operator which does not commute with H can be made to violate certain inequalities for any initial state. But of course EPR can be formulated for many different states as well, it just has to stay in that state.

[1] Note that the measurements refer to independent measurements of members of the same ensemble with the same initial preparation at t_1 They are not subsequent measurements on the same system

A.2 Difficulties in Measuring the $\mathrm{K_{ij}}$

The practical measurement of these time correlation functions is complicated by the general rule that an act of measurement in quantum mechanics affects the subsequent dynamics by back-action effects etc... In contrast to the spatial EPR case where special relativity can be invoked to ensure locality in space, no such principle exist to ensure locality in time. Another difficulty arises from the fact that to measure $\hat{Q}(t)$ at a precise time t one must assume an instantaneous "von Neumann" type collapse process, an approximation which may not be valid at all. The K_{ij} are also ensemble averaged, which could be either a large number of identical systems, or one system prepared in the same initial state each time. These difficulties led to some criticism shortly after the Leggett/Garg paper came out.

The critique by L. Ballentine

The first (at least in PRL) to criticize the measuring scheme of Leggett and Garg was Ballentine, who claimed that the contradiction with QM is produced solely by the assumption (A2) of noninvasive measurability[3]. His argument was based on an explicit measurement scheme that showed non violation of the Bell inequalities. But, his analysis was actually totally irrelevant, as he assumed that each system in an ensemble was measured at multiple times, say t_1, t_2, t_3 and t_4. This was then repeated to get an ensemble average. Even in QM the $Q(t_i)$ are taken to be real "elements of reality" when they're actually measured, so that dissipative measuring at say three subsequent times for the same system will give a third order distribution function that will trivially return the second order marginals which in turn determine the correlation functions. The latter will no longer violate the Bell inequalities[4].

The critique by A. Peres

Peres gave a different critique of the specific double well scenario with a semi-classical argument based on the uncertainty principle. It can easily be boiled down to the following:

To measure which well the particle is in, one needs to determine its coordinate with an accuracy $\Delta q \leq Q_0$, where Q_0 is the half the distance between the two wells. Together with the position momentum uncertainty principle $\Delta q \Delta p \geq \hbar$ this gives:

$$\Delta p \geq \hbar / Q_0. \tag{A.6}$$

But to get an appreciable tunneling amplitude:

$$\hbar \Delta \sim \exp - \int_{Q_0}^{Q_0} \frac{p(q)}{\hbar} dq \tag{A.7}$$

the integral in the exponent must not be much smaller than one. Peres approximates the integral by $p_0 Q_0 / \hbar$, where p_0 is the classical momentum corresponding to the particle being at the bottom of a well so that one finds the second condition:

$$p_0 \leq \hbar / Q_0, \tag{A.8}$$

from which follows:

$$\Delta p \geq p_0, \tag{A.9}$$

and a measurement of sufficient accuracy to determine the position of the particle is seen to disturb it with enough momentum to kick it over the barrier, i.e. the measurement cannot be approximated by a simple "von Neumann" collapse of the wave function. However, a more careful analysis gives a window where $\Delta p < p_0$[6], so that Peres' argument doesn't hold under closer scrutiny; his approximations were too crude.

Ideal Negative Result Experiment

Both Peres and Ballentine fell prey to the idea that one must measure the particle position at times t_i and t_j separately to determine the joint probability densities. This works fine in the spatial EPR experiments which measure coincidence rates, but is not really necessary. The solution to the measurement problem is simply to measure the joint probability directly. This can be done by "Ideal Negative Result Experiments", an idea Leggett and Garg mentioned in their original paper, and which was worked out in great detail for SQUIDS by Tesche[7]. The basic idea is this: set up a measuring apparatus so it only measures (dissipatively) only one of the two well states (say $Q = 1$) at time t_1, and does a normal dissipative measurement at time t_2. Then one can measure $K_{12}^{-} = K(-, t_1; Q, t_2)$ by only counting those runs in which the first measurement apparatus gave a null result (and, one assumes, didn't affect the system) which corresponds to $Q(t_1) = -1$. The same can be repeated with the opposite arrangement for t_1 to give K_{12}^{+} (but the same ensemble), from which one can easily extract the full correlation function $K_{12} = K_{12}^{-} + K_{12}^{+}$, and see if it violates the Bell inequalities, and thus also the condition (A1) of *Macrorealism*, But even in this scheme, there is one unresolved difficulty, as in general there will be but one system on which to measure repeatedly, and it will be hard to get it into exactly the same initial state each time to insure one is measuring the same ensemble!

A.3 Conclusions

The temporal Bell inequalities are thus shown to be demonstrable in principle, and they have many direct analogies with spatial Bell inequalities. When they are violated as predicted by QM, one cannot assign consistent probabilities to the histories, and the idea of *Macrorealism* is no longer valid. There is nothing conceptually new in this, but the violation of the Bell inequalities has a nice interpretation in that one cannot assign a consistent history (i.e. one cannot represent the position of the particle by telegraph noise such as is depicted in figure A.2), and as such this provocatively demonstrates how classical ideas of reality break down at the quantum level. An additional point of interest would be to measure the violation of these inequalities in SQUIDS where the flux variable can be regarded as macroscopic in itself, but still obey quantum mechanics for its dynamics. However, the condition of "locality in time" is much more difficult to ensure than is "locality in space" due to the difficulties in the measurement problem, and because there is nothing analogous to special relativity to separate the time processes. It may be interesting to generalize the temporal Bell inequalities to the more modern (and spectacular) GHZ type[8, 9] relations, thus avoiding the problem of ensemble preparation. While it is not hard to see how to measure products of more than two observables with the two state system, one cannot measure the analogon of more than one plane of EPR polarization so it is not immediately obvious what system is needed to study GHZ correlations.

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[4] This follows from the existence of the third order distribution function. See for example N.D. Mermin, Lecture Notes I, Cornell, Fall '95.

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Appendix B

Coupling of the tunneling atom to Raleigh surface phonons: Ohmic dissipation

Ohmic dissipation occurs when the spectral density $J(\omega)$ is linear in the frequency ω[1], i.e.:

$$J(\omega) = \sum_{\alpha} \frac{(\lambda_\alpha)^2}{m\omega_\alpha} \delta(\omega - \omega_\alpha) \sim \omega. \tag{B.1}$$

A given λ_α is the coupling to the linear coordinate of the environment oscillator (phonon) α in the total Hamiltonian.

In the case of coupling to bulk modes with an external (*monopole*) force[2][1], the coupling goes as:

$$\lambda_k \sim \frac{1}{\sqrt{N}} \tag{B.2}$$

[1] The use of the word force is an unfortunate misnomer: force is a purely classical concept. Instead this should be the gradient of the potential. However, in keeping with common conventions in the literature, we continue to use the rather akward word force.

where N is the number of atoms. In the case of the bulk waves it is thus independent of k, which gives ohmic dissipation.

For Rayleigh modes there is a penetration depth of $1/\kappa$, where

$$\kappa = \sqrt{k_{\|}^2 - \omega^2/c^2} = (\sqrt{1-\xi^2})k_{\|}. \tag{B.3}$$

The function ξ can be found in Landau and Lifshitz[3] section (**24**) and depends on one elastic constant only. The momentum parallel to the surface is denoted by $k_{\|}$. The number of atoms N that participate in an oscillation are proportional to the number of surface atoms N_s times the penetration depth $1/\kappa$ so that for a given parallel momentum $k \parallel$ the coupling constant goes as:

$$\lambda_k \sim \frac{1}{\sqrt{N_s/\kappa}} \sim \sqrt{k_{\|}}. \tag{B.4}$$

When this form of λ_k is plugged into the spectral density (eq B.1) the extra factor k compensates for the 2-dimensional density of states which is proportional to ω.

All this can be put on a firmer footing by writing the displacement field as:

$$\vec{u}(\vec{r}) = \sum_s \sum_k \frac{1}{\sqrt{C}} \sqrt{\frac{\hbar}{2m\omega(\vec{k}, s)}} \left(a_{\mathbf{ks}} + a_{\mathbf{ks}}^{\dagger}\right) \exp\left(i\vec{k}_{\|} \cdot \vec{r}\right) \exp\left(\kappa z\right)\right) \exp\left(-i\omega t\right) \tag{B.5}$$

Here $\mathbf{k}$ is the wave vector, and s is a polarization index.

By demanding that each mode α contributes an elastic energy $\hbar\omega a_\alpha^\dagger a_\alpha$, and then by equating two times the kinetic energy to the total energy :

$$2E_{kin} = \int \rho \mid \dot{u} \mid^2 = \sum_\alpha \hbar\omega a_\alpha^\dagger a_\alpha \tag{B.6}$$

one finds the normalization constants C. The coupling term in the Hamiltonian to first approximation is:

$$H_{int} = \vec{F} \cdot \vec{u}(\vec{0}) \tag{B.7}$$

from which the coupling constants $\{\lambda_k\}$ directly follow for the different kinds of modes.

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Appendix C

Finite Temperature Hohenberg-Kohn-Mermin Theorem for Multi-Component Charged Systems

C.1 Introduction

Two versions of the H-K-M theorem for multi-component charged systems are proven. One for a neutral canonical ensemble, and one for a grand-canonical ensemble. Neither proof needs a Born-Oppenheimer approximation.

C.2 Neutral Canonical Ensemble

Consider a multicomponent system of charged particles at constant volume, and particle numbers, and in contact with an external heat bath - i.e. at constant temperature T. The particle numbers of each component are chosen so that the overall system is neutral.

The equilibrium properties of such a system are governed by the canonical statistical density operator:

$$\hat{\rho}_0 = \exp[-\beta \hat{H}]/Tr\left[\exp[-\beta \hat{H}]\right] \tag{C.1}$$

where the Hamiltonian operator $\hat{H}$ has the form:

$$\hat{H} = \hat{T} + \hat{U} + \hat{V}, \tag{C.2}$$

$\hat{V}$ is a series of one-body terms of the form:

$$\hat{V} = \sum_i \int \hat{n}_i(\vec{r}) v_i(\vec{r}) d\vec{r} \tag{C.3}$$

where the $\{v_i(\vec{r})\}$ are the external potentials and the $\{\hat{n}_i(\vec{r})\}$ satisfy the following neutrality constraint on their integrated ensemble averages:

$$\sum_i Z_i \int n_i(\vec{r}) d\vec{r} = 0. \tag{C.4}$$

The $\{Z_i\}$ are the charges of each constituent and the one particle densities $\{n_i(\vec{r})\}$ are given by:[1]

$$n_i(\vec{r}) = Tr\left[\hat{\rho}_0 \hat{n}_i(\vec{r})\right]/Tr\hat{\rho}_0, \tag{C.5}$$

where $\hat{\rho}_0$ is the equilibrium density matrix of eq (C.1). Now define the functional:

$$F[\hat{\rho}] = Tr\left[\hat{\rho}\left(\hat{H} + \frac{1}{\beta}\ln\hat{\rho}\right)\right] \tag{C.6}$$

[1] For a canonical system the trace is only over states with the particle number fixed beforehand, so the constraint (C.4) holds at the level of the *operators* $\hat{n}(\vec{r})$ also.

for an arbitrary density operator $\hat{\rho}$ with unit trace. Mermin has shown that at equilibrium, this functional satisfies a minimum property [2]

$$F[\hat{\rho}] > F[\hat{\rho}_0] \quad \text{for} \quad \hat{\rho} \neq \hat{\rho}_0. \tag{C.7}$$

By equating the minimum of $F[\hat{\rho}]$ with the Helmholtz free energy F:

$$F[\hat{\rho}_0] = -\frac{1}{\beta} \ln Tr \left[\exp \left[-\beta \hat{H} \right] \right] = F, \tag{C.8}$$

contact is made with Thermodynamics.

For a given $\hat{T}$ and $\hat{U}$, the external potential $\hat{V}$ determines the equilibrium density $\hat{\rho}$ - giving the surjective map A:

$$A: \quad \hat{V} \quad \to \hat{\rho}. \tag{C.9}$$

A given density matrix $\hat{\rho}$ determines the densities $\{\hat{n}_i(\vec{r})\}$ by equation(C.5), giving the map B:

$$B: \quad \hat{\rho} \quad \to \{n_i(\vec{r}\}, \tag{C.10}$$

which is also surjective.

The H-K-M proof then goes as follows - first we show that both A and B are injective (one to one) and thus bijective (fully invertible)[4].

For a one-component system the injectivity of A can easily be shown, but the statement : a given set $\{v_i(\vec{r})\}$ leads to a unique equilibrium density matrix $\hat{\rho}_0$, while plausible, can't be rigorously proven for multi-component systems such as the

[2] In the Appendix of his paper[1], Mermin shows the minimum principle for a grand-canonical ensemble. The proof carries through identically for a canonical ensemble. Only very general properties of the density matrix are used (such as positive definiteness), so that the proof also carries over to multi-component systems with no change, except that the density matrix is now understood to carry the information of the full multi-component system

one under consideration, or for that matter, spin-polarized systems with an external magnetic field[4]. We will assume it is true; a proof is lacking.

The injectivity of the map B can be shown by the following argument:

Assume that two different external potential sets $\{v_i(\vec{r})\}$, and $\{v_i'(\vec{r})\}$[3] with equilibrium densities $\hat{\rho}_0$, and $\hat{\rho}_0'$ give the same set of external densities $\{n_i(\vec{r})\}$. Then:

$$\begin{aligned} F' &= Tr\left[\hat{\rho}_0'\left(H' + \frac{1}{\beta}\ln\hat{\rho}_0'\right)\right] \\ &< Tr\left[\hat{\rho}_0\left(H' + \frac{1}{\beta}\ln\hat{\rho}_0\right)\right] \\ &< F + Tr\left[\hat{\rho}_0\left(V' - V\right)\right] \end{aligned} \tag{C.11}$$

so that:

$$\begin{aligned} F' &< F + Tr\left[\hat{\rho}_0 \sum_i \int d\vec{r}\left(v_i'(\vec{r})\hat{n}_i(\vec{r}) - v_i(\vec{r})\hat{n}_i(\vec{r})\right)\right] \\ &= F + \sum_i \int \left(v_i'(\vec{r}) - v_i(\vec{r})\right) n_i(\vec{r}). \end{aligned} \tag{C.12}$$

Simply interchanging the primes (reversing the designation of the two systems) gives:

$$F < F' + \sum_i \int \left(v_i(\vec{r}) - v_i'(\vec{r})\right) n_i(\vec{r}). \tag{C.13}$$

Adding (eq C.12) and (eq C.13) gives:

$$F + F' < F' + F, \tag{C.14}$$

an evident contradiction, from which it follows that a given density matrix and external potential set $\{v_i(\vec{r})\}$ uniquely determines the density set $\{n_i(\vec{r})\}$, so that the map B is indeed injective.

[3] Different means more than a constant difference for canonical systems, this is not necessary for a grand-canonical ensemble

Thus for a given $\hat{T}$ and $\hat{U}$ in the original Hamiltonian of equation (C.2) the density matrix is a unique functional of the set of external densities, and the same is true of the free energy functional $F[\{n\}]$, which obeys the minimum principle that the Helmholtz Free energy is given by the set of $\{n_i(\vec{r})\}$ which minimize the functional F. (With the understanding that the $\{n_i(\vec{r})\}$ satisfy the neutrality constraint)

C.3 Grand Canonical Ensemble

The proof for a multi-component charged Grand-Canonical ensemble is quite similar. Simply replace F with Ω, and V with $V - \mu N$, where μ is really a set of chemical potentials. The neutrality constraint (C.4) now holds for the averages only, and no longer at the level of the operators, but that was never necessary for the proof.

One possible subtlety would be if one were to do a constrained trace, i.e. a trace over neutral states. Then the states one traces over will depend on the particular V, and so perhaps different ρ_0's could give identical $\{n_i(\vec{r})\}$'s just because the states change. However, this problem is circumvented by the realization that the constrained trace is equal to the full trace, which is independent of the external potential used (the Lieb-Lebowitz result[5]).

C.4 Some Comments

- Firstly, the proof only goes for a set, so a given one-body density $n_i(\vec{r})$ may be produced by two different sets of external potentials, but the sets $\{n_i(\vec{r})\}$ will be different.

- Secondly, notice that no Born-Oppenheimer assumptions are needed. In the usual electronic H-K theorem, the assumption is that all the $\{n_i(\vec{r})\}$ are externally fixed except the electronic density $n_e(\vec{r})$. This is basically the B-O theorem, the electrons relax while the ions are frozen. Only the potential that couples to the electrons is varied. From the multi-component proof we did it is clear that if all external potentials save one are fixed, the one particle density of that one component must change if it is external potential changes, but there is still a freedom for the other one-particle densities to change. The usual H-K theorem assumes they stay fixed, and are part of the $\hat{T}+\hat{U}$ terms.

Similarly in the B-O approximation for classical ions, the ions now have some effective (many-body) interaction $U_{eff}(\{r_i\})$ which may depend on all the ionic coordinates, but as long as it is expressible as a potential, the usual H-K-M theorem goes through. All electronic effects are put into the effective interaction beforehand.

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Appendix D

Non-linear response functions

In a homogeneous electron gas with a rigid neutralizing background, a perturbing potential $\lambda v(q)$ induces a charge density:

$$\rho^{ind}(k) = \lambda\chi_1(k)v(k) + \lambda^2 \sum_{\vec{k}_1}\sum_{\vec{k}_2} \chi_2(k, k_1, k_2) v^{ps}(k_1) v^{ps}(k_2) + ..., \tag{D.1}$$

where $\rho^{ind}(k)$ is measured w.r.t. the homogeneous density ρ_0. From the Hohenberg-Kohn-Sham formulation[1, 2] the identical problem can be set-up in terms of the response of non-interacting electrons to the effective potential $\Gamma(q)$:

$$\rho^{ind}(k) = \chi_1^0(k)\Gamma(k) + \sum_{\vec{k}_1}\sum_{\vec{k}_2} \chi_2^0(k, k_1, k_2)\Gamma(k_1)\Gamma(k_2), \tag{D.2}$$

where the effective potential is given by:

$$\Gamma(k) = \lambda v(k) + \frac{8\pi}{k^2}\rho^{ind}(k) + \mu_{xc}(k, [\rho]), \tag{D.3}$$

and the $\chi_i^0(..)$ denote the non-interacting response-functions, which are properties of the *homogeneous* electron gas. The induced density can be expanded in powers of λ:

$$\rho^{ind}(k) = \lambda\rho_1(k) + \lambda^2\rho_2(k) + \lambda^3\rho_3(k) + ... \tag{D.4}$$

and the exchange-correlation potential $\mu_{xc}(q,[\rho])$ can be expanded in powers of the induced density $\rho^{ind}(k)$:

$$\mu(k,[\rho^{ind}]) = \mu_1(k)\rho^{ind}(k) + \sum_{\vec{k}_1}\sum_{\vec{k}_2}\mu_2(k,k_1,k_2)\rho^{ind}(k_1)\rho^{ind}(k_2) + ... \qquad (D.5)$$

The full interacting response functions are found by equating powers of λ in (D.1) and (D.2). The first order interacting response function is:

$$\chi_1(k) = \frac{\chi_1^0(k)}{\epsilon(k)}, \qquad (D.6)$$

and the second order interacting response function is:

$$\chi_2(k_1,k_2,k_3) = \frac{\chi_2^0(k_1,k_2,k_3) + \frac{1}{2}\mu_2(k_1,k_2,k_3)\chi_1^0(k_1)\chi_1^0(k_2)\chi_1^0(k_3)}{\epsilon(k_1)\epsilon(k_2)\epsilon(k_3)}. \qquad (D.7)$$

The third order interacting response function (neglecting the μ_i (i.e. setting them to 0) is

$$\begin{aligned}\chi_3(k_1,k_2,k_3,k_4) &= \Big\{\chi_3^0(k_1,k_2,k_3,k_4) \\ &\quad +2\chi_2^0(k_1,k_2,k_3+k_4)v_c(k_3+k_4)\chi_2^0(k_3+k_4,k_3,k_4)\Big\}\Big/ \\ &\quad \{\epsilon(k_1)\epsilon(k_2)\epsilon(k_3)\epsilon(k_4)\}, \qquad (D.8)\end{aligned}$$

and so forth. The dielectric function $\epsilon(k)$ is given by:

$$\epsilon(k) = 1 - \left(\frac{8\pi}{k^2} + \mu_1(k)\right)\chi_0^1(k). \qquad (D.9)$$

Expressions for the first and second order non-interacting response functions are known: the first order was first derived by Lindhard in 1954[3]:

$$\chi_1^0(k) = \frac{-k_F}{2\pi^2}\left(\frac{1}{2} + \frac{4k_F^2-k^2}{8k_Fk}\ln\left|\frac{k+2k_F}{k-2k_F}\right|\right), \qquad (D.10)$$

the second order was first derived by Lloyd and Sholl in 1968[4], and has a much more complex form[1]. We plot the non-interacting, exchange-correlation, and full

[1] The full form is too long to put in this thesis, readers can consult reference [4] or alternatively, access the following: "http://www.lassp.cornell.edu/~ardlouis/Thesis/Lloyd_Sholl.html" .

response in figures (D.1 -D.3).

For a given exchange-correlation potential $\mu_{xc}(r,[\rho])$, the functions $\mu_n(r)$ are the functional derivatives :

$$\mu_n(r;r_1..r_j) = \left.\frac{\delta^j \mu_{xc}(r)}{\delta\rho(r_1)..\delta\rho(r_j)}\right|_{\{\rho(r_1)...\rho(r_j)\}\to\{\rho_0...\rho_0\}} . \tag{D.11}$$

When the LDA approximation to the exchange-correlation potential is employed, the $\mu_i(..)$ reduce to a simple functions of ρ in the homogeneous limit; there is no q dependence.

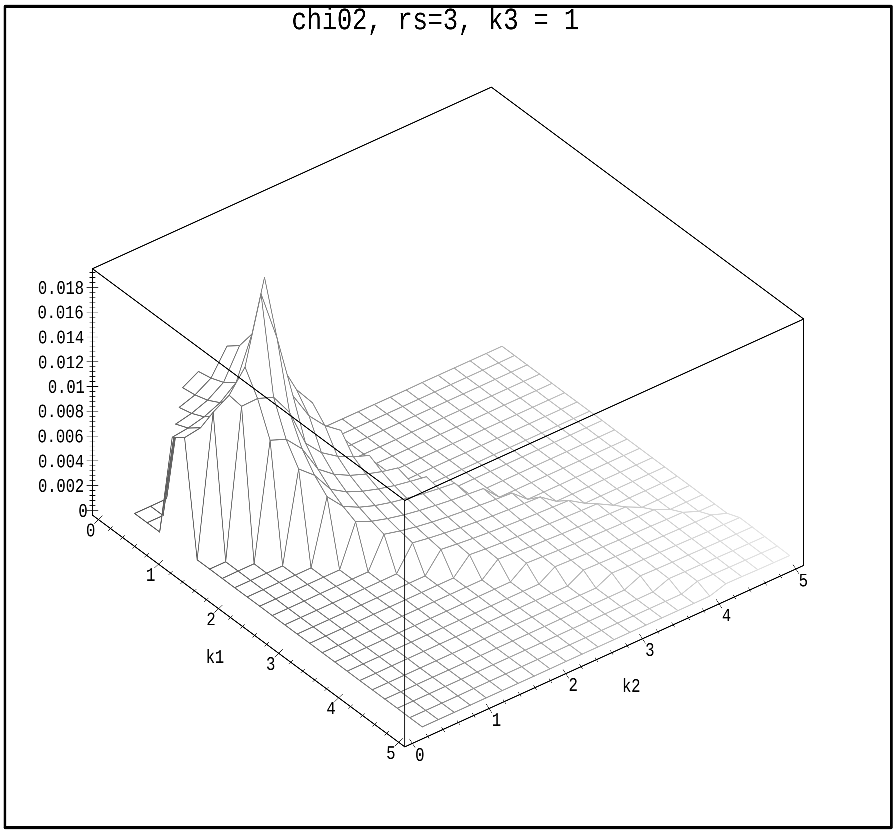

Figure D.1: **Non-interacting $\chi_2^0(k_1, k_2, k_3 = 1)$ for $rs = 3$ and fixed k_3.** The second-order response function $\chi_2^0(k_1, k_2, k_3)$ for non-interacting electrons was taken from reference[4]. As the fixed variable k_3 changes, so does $\chi_2^0(k_1, k_2, k_3)$, but the function remains highly peaked around $k_i = 2k_F$. The function volume is restricted by the limits $(k_3 < k_1 + k_2)$, $(k_1 < k_2 + k_3)$ and $(k_2 < k_3 + k_1)$, which stem from the delta-function constraint 4.15 and must be simultaneously satisfied. Here, $2k_f = 1.28$ a.u.

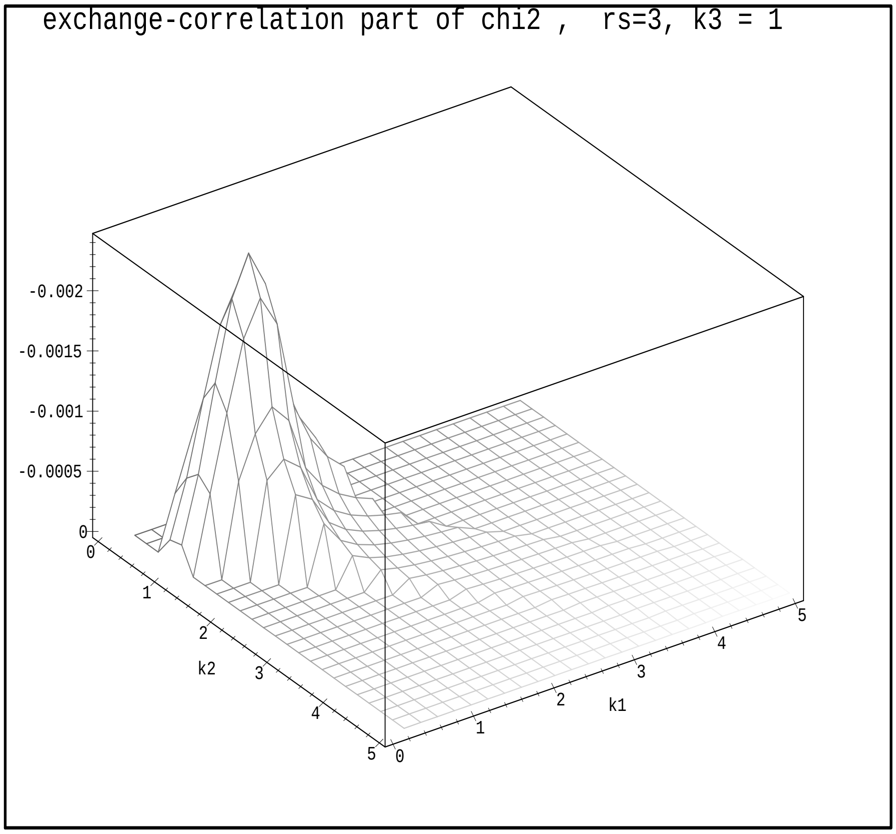

Figure D.2: **LDA exchange-correlation contribution to response for $rs = 3$ and fixed k_3.**
The exchange-correlation contribution to the second-order response function (D.7) has the form:

$$\chi_2^{xc}(k_1, k_2, k_3) = \frac{1}{2}\frac{\mu_2(k_1, k_2, k_3)\chi_1^0(k_1)\chi_1^0(k_2)\chi_1^0(k_3)}{\epsilon(k_1)\epsilon(k_2)\epsilon(k_3)}.$$

We plot it for an LDA form of $\mu_2(k_1, k_2, k_3)$, which has no k dependence; it is a function of density only. Note the inverted axes: this contribution *weakens* the response. The function volume is restricted by the limits $(k_3 < k_1+k_2)$, $(k_1 < k_2+k_3)$ and $(k_2 < k_3+k_1)$, which stem from the delta-function constraint 4.15 and must be simultaneously satisfied. Here, $2k_f = 1.28$ a.u.

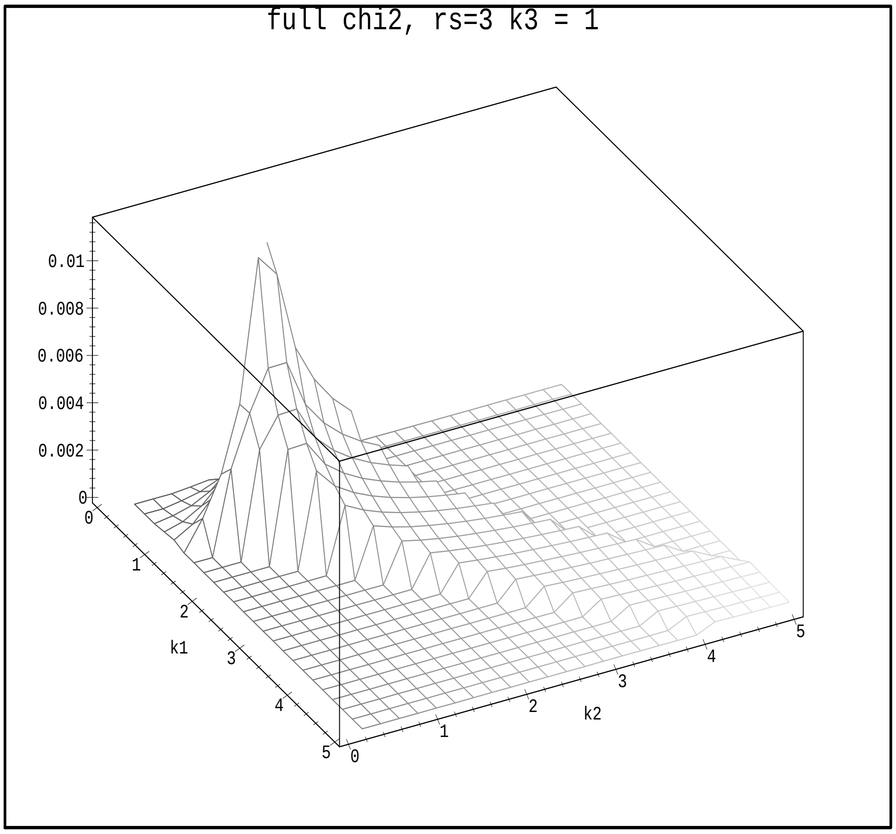

Figure D.3: **Full $\chi^2(k_1, k_2, k_3 = 1)$ from (D.7) for $rs = 3$ and fixed k_3.** As the fixed variable k_3 changes, so does the second order response function $\chi_2(k_1, k_2, k_3)$, but the function remains highly peaked around $k_i = 2k_F$. The function volume is restricted by the limits $(k_2 < k_1+k_2)$, $(k_1 < k_2+k_3)$ and $(k_2 < k_3+k_1)$, which stem from the delta-function constraint 4.15 and must be simultaneously satisfied. Here, $2k_f = 1.28$ a.u.

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