

## 1 Title of the project

First principle calculation of many-electron transport systems

## 2 Applicant(s)

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## 3 FOM-researchgroup

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## 5 Abstract

This proposal concerns theoretical research within the fields of many-particle physics and quantum chemistry. The goal of this research is to describe the time-dependent current through a molecule between two leads. A lot of research has already been done in this field, but only within the static limit. Within this limit it is impossible to get a correct evolution of the system in time. Time-dependent effects such as memory are not taken into account. It is not even sure if the steady state current will be the same as in the static limit. To give a good description of the evolution, the exchange correlation potential  $v_{xc}$  has to be history dependent. This renders the usual exchange-correlation potentials such as the Local density Approximation (LDA) and the Generalized Gradient Approximation (GGA) useless. A new exchange correlation  $v_{xc}$  term will have to be developed that is not local in time

anymore. such a  $v_{xc}$  can be found using non-equilibrium Green functions. None of this has ever been done before and will be crucial to understand the dynamics of time dependent effects in correlated systems.

## **6 Duration of the project**

The project should start in october 2005 and finish in september 2009

## **7 Personnel**

### **7.1 Senior-scientists**

Prof. Dr. M. Filatov (RUG) will act as the promotor. This will consume about 0% of his working hours.

Dr. R. van Leeuwen (FOM) will act as copromotor and will supervise the project. It will take about 10% of his working hours.

### **7.2 Junior-scientists and technicians**

AIO K.J.H. Giesbertz (FOM) will perform the research. This will take about 90% of his working hours.

## **8 Cost estimates**

### **8.1 Personnel positions**

One PhD student

### **8.2 Running budget**

5 k€ per year will be required for one PhD student for travel expenses for visits to summer schools and conferences.

### **8.3 Equipment**

A new computer will be bought for the PhD student. This will cost 2 k€.

### **8.4 Other support**

There are no applications for other support.

## 8.5 Budget summary

personnel (positions):	2005	2006	2007	2008	2009	TOTAL
PhD students	10	41	41	41	31	164
postdocs	0	0	0	0	0	0
technicians	0	0	0	0	0	0
guests	0	0	0	0	0	0
personnel (costs)						164
running budget	1	5	5	5	4	20
equipment FOM-part	2	0	0	0	0	2
TOTAL (requested from FOM)	13	46	46	46	35	186

## 9 Research programme

### 9.1 Introduction

Since the current through single molecules has been measured, a lot of effort is put into calculations of these systems. Unfortunately no one knows how important electron-electron interactions or electron-phonon interactions are for the evolution of these systems. Until now no method is available to incorporate these effects in a correct way. It is possible to take these effects into account using Density Functional Theory (DFT), but the current has only been calculated in the static regime. However, the electron-electron and electron-phonon interactions cause dissipative and memory effects. Due to dissipative effects and especially memory effects, the system can develop a final current that has a significant different value as the steady state current that is calculated in the static regime. To understand the physics of these systems it will be very important to include these effects.

### 9.2 Preliminary work

The problem has already been studied for non-interacting electrons in a 1D system. As a simplified system a box with infinite walls filled with electrons was used. To separate the electrons a repulsive potential was placed in the middle of the box. The two areas with electrons are regarded to simulate the electrodes. For this system the ground state was calculated which was used as the equilibrium state from which the system evolved. Then at one side of the box the potential was raised by a small amount. This caused electrons to flow to the other side of the box from which a current was calculated.

The problem was studied in a 3D box as well. The potential to separate the electrons had the form of a slab to create again two regions that could be identified with the electrodes. In addition a local potential in the form of a screened Coulomb potential, also known as a Yukawa potential,  $\lambda e^{-\alpha r}/r$ , was put in the middle of the box to simulate an atom between the two leads. Again the ground state was used as the initial state to propagate the system and current could be calculated as well.

### 9.3 Existing theory

The electron-electron and electron-phonon interactions can be incorporated by using DFT. DFT provides an exact description of interacting many-particle systems in terms of a one-particle frame-work. This makes numerical calculations much more tractable. The hamiltonian in the non-interacting frame-work  $\hat{H}_s$  is defined as:

$$\hat{H}_s = -\frac{1}{2}\nabla^2 + v(\mathbf{r}) + \int \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' + v_{xc}(\rho; \mathbf{r}),$$

where  $v(\mathbf{r})$  is the external potential and  $v_{xc}(\rho; \mathbf{r})$  is the exchange-correlation potential which describes all the contributions due to particle interactions except for the Coulomb potential. DFT states that such a potential exists, but not how it should look like. People have approximated  $v_{xc}$  with potentials such as the Local Density Approximation (LDA) and the Generalized Gradient Approximation (GGA). Although the approximation are crude, DFT described a lot of systems remarkably well.

### 9.4 Innovation

There exists also a time dependent version of DFT: Time-Dependent DFT (TDDFT). However, the approximations that exist for  $v_{xc}$  are local in time, so they would not be able to describe electron-electron interactions and electron-phonon interactions in a such a way that dissipation effects and memory effects can be described. To describe these features, a new  $v_{xc}$  will have to be developed that is not local in time anymore:

$$v_{xc}(t) = v_{t_0} + \int_{t_0}^t K(t, t') dt'$$

Due to the relation between the exchange-correlation potential and non-equilibrium Green functions, a  $v_{xc}$  of this form can be found studying these Green functions [1, 2]. Non-equilibrium Green functions can be calculated

from Keldysh diagram expansions [3, 4]. The diagram technique shows the connection between correlation effects and memory, so they can be used to construct a better approximation for  $v_{xc}$ .

## 9.5 Plan of work

The research will be done at the Theoretical Chemistry group at the Rijksuniversiteit Groningen. The group has two of the world's leading experts on TDDFT and non-equilibrium Green functions (Dr. Robert van Leeuwen, FOM Springplank Fellow, and Dr. ig. Paul L. de Boeij, NWO-VIDI researcher). The group is also part of the MSC which is a leading research school in material science. At the MSC transport through single molecules is also studied experimentally in the group of Prof. B.J. van Wees [5].

**Years 1,2:** I expect that the development of new exchange-correlation functionals will take two years.

**Year 3:** One year should be sufficient to test the new functionals.

**Year 4:** I plan to use the last year to apply the new theory to the molecular transport problem.

Developing a new exchange-correlation potential that is non-local in time is very innovative and will certainly result in several papers of high scientific importance.

## 10 Infrastructure

The theoretical chemistry group in Groningen has already a cluster that can support the required calculations. Apart from the workstation no other equipment has to be bought.

## 11 Application perspective in industry, other disciplines or society

The development of a temporal non-local exchange-correlation potential has never been done before. It will be very important to understand time-dependent processes in correlated systems such as molecular devices.

## References

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