Some calculational improvements in applying the Hubbard model to nanomaterials

Vladan Celebonovic
Institute of Physics, University of Belgrade, Pregrevica 118, 11080 Belgrade, Serbia
E-mail: vladan@ipb.ac.rs

Abstract. John Hubbard proposed "his model" (HM) nearly sixty years ago, and one could be tempted to expect that everything (or almost so) about it is well known. Quite to the contrary, the HM is still arising interest and its applicability seems to be rising. The aim of this paper is to discuss several examples of new or already existing but improved results concerning the 2D HM and its applicability to nanomaterials. The following issues will be discussed: increasing the number of terms in various expressions used in calculating the conductivity; introducing the strain in these expressions (which is important for experiments under high pressure); defining the pressure in a 2D system and calculating the reflectivity but taking into account the imaginary part of the conductivity.

1. Introduction
Towards the middle of the last century, one of the outstanding problems of condensed matter physics was the metal to insulator transition (MI). John Hubbard tried to contribute to finding an explanation of the MI transition by proposing what later became known as the Hubbard model - HM [1]. In spite of the time elapsed since Hubbard's proposal, the HM is still attracting considerable interest.

The publication of the HM greatly helped launch the study of correlated electron systems. There exists a semi-documented detail, illustrating this point: Mr. Hubbard was sitting and reading a book on many body theory, when his mother in law passed by and asked him what was the book about. To which Hubbard replied "A murder mystery" [2].

It is common knowledge that the behavior of a physical system is determined by its Hamiltonian \( H \) from which all the thermodynamical functions can be derived. The Hamiltonian of the HM is apparently simple. It contains only two terms, one describing the hopping of electrons within the lattice, and the other the interactions of a pair of electrons with opposite spins on a lattice node. The equations are available in the literature, for example [3]. The fact that the HM is only apparently simple becomes clearly visible when attempting to solve it. The HM was solved in the 1D case [4], while work on solving the 2D case is ongoing within the Density matrix renormalisation group (DMRG) approach [5]. This kind of work is of course extremely interesting from the point of view of mathematical physics, but the obvious question is the applicability of 1D and 2D HM in material science.

In the years around 1980 and for some time after if was thought that the so called Bechgaard salts were materials which could be described by the 1D HM and that they were the simplest cases of strongly correlated electron systems. Applying the HM to these materials gave results
agreeing with experiments, but they have not been as helpful as expected in understanding the high $T_c$ materials. See, for example, [6].

Nanomaterials in 2D present a much wider field of possibilities for the application of the $HM$. To name just a few examples, these include organic electronics [7], stretchable electronic materials [8], photothermal cancer therapy [9].

There exist numerous publications on various aspects of the $HM$, including some by the present author [10]. What is the aim of another paper on the $HM$? The main aim of the present paper is improving the results obtained in previous papers. In practical terms, this means increasing the number of terms taken into account in various developments into series, and taking into account the fact that the imaginary conductivity is different from zero. It will be attempted to introduce a sitable definition of pressure in 2D systems. In that respect, it will be a distinct improvement over previous results. A detailed comparison of the experimental results with the predictions of these ”improved” calculations will be performed in the future.

1.1. The method

The calculations to be discussed in the remainder of this paper were performed using the so called memory function method. It uses the idea, known in statistical mechanics, that knowledge of the Hamiltonian of a system gives the possibility of calculating its electrical conductivity. For a recent review see, for example, [11]. The essential equations of the method are

\[
\chi_{AB}(\omega) = \langle A; B \rangle = -i \int_0^\infty \exp i\omega t < [A(t), B(0)] > dt \quad (1)
\]

where $A = B = [j, H]$, $j$ denotes the current operator, $H$ the Hamiltonian and

\[
\sigma(\omega) = i \frac{\omega_P^2}{4\pi z} \left[ 1 - \frac{\chi(z)}{\chi(0)} \right] \quad (2)
\]

where $\sigma$ denotes the electrical conductivity, $z$ is the complex frequency, $\omega_P$ is the plasma frequency, $\chi(\omega)$ is the frequency dependent susceptibility and $\chi(0)$ the static susceptibility. As $z$ is the complex frequency, it can be expressed as $z = z_1 + iz_2 = z_1 + i\alpha z_1$, where $\alpha > 0$. It can be shown that

\[
\sigma_R = \frac{\omega_P^2 \chi I}{4\pi z_1 \chi_0} \quad (3)
\]

and

\[
\sigma_I = \frac{\omega_P^2}{4\pi z_1} (1 - \frac{\chi_R}{\chi_0}) \quad (4)
\]

2. Calculation of the electrical conductivity

2.1. Existing results

The Hamiltonian of the $HM$ in 1D, expressed in the formalism of second quantisation, has the following form

\[
H = -t \sum_{l,\sigma} (c_{l,\sigma}^+ c_{l+1,\sigma} + c_{l+1,\sigma}^+ c_{l,\sigma}) + U \sum_j n_{j,\uparrow} n_{j,\downarrow} \quad (5)
\]

where all the symbols have their standard meanings, and the current operator is

\[
j = -it \sum_{l,\sigma} (c_{l,\sigma}^+ c_{l+1,\sigma} - c_{l+1,\sigma}^+ c_{l,\sigma}) \quad (6)
\]
Using these two expressions it becomes possible to calculate the commutators \( A = B = [j, H] \), to take into account their time dependence and obtain an expression for the current-current correlation function. All these steps are discussed in [12], and the final result is

\[
\chi(z) = \sum_{p,g,k,q} (32i/(1 + \exp[\beta(-\mu - 2t \cos[g])])(1 + \exp[\beta(-\mu - 2t \cos[k])]) - 1/(1 + \exp[\beta(-\mu - 2t \cos[p])])(1 + \exp[\beta(-\mu - 2t \cos[q]])] \\
(Ut)^2(\alpha z_1 + i(z_1 + 2t(\cos[q] + \cos[p] - \cos[g] - \cos[k]))) \\
(\cos(p + g)/2)(\cos((q + k)/2))(\cosh(g - p) - 1)/ \\
(N^4((\alpha z_1^2 + (z_1 + 2t(\cos[q] + \cos[p] - \cos[g] - \cos[k]))^2))
\]

(7)

The symbol \( N \) denotes the number of lattice sites, \( \mu \) is the chemical potential and the summations are limited to the first Brioullin zone. The chemical potential \( \mu \) of the electron gas on a 1D lattice is given by [13]

\[
\mu = \frac{(\beta t)^6(na - 1)|t|}{1.1029 + 0.1694(\beta t)^2 + 0.0654(\beta t)^4}
\]

where \( \beta \) denotes the inverse temperature, \( a \) the lattice constant and \( n \) the band filling. Summation of eq.(5) taking into account the smallest possible number of terms has been published in [12]. The result was an expression for the electrical conductivity of a 1D system of correlated electrons, which was applied to the Bechgaard salts, and the results obtained were in reasonable agreement with experiments.

The purely mathematical aim of the present calculation is to increase the number of terms taken into account in eq.(7), and in the subsequent calculation of the electrical conductivity. On the physical side, it will be attempted to take into account the dependence of the conductivity on the strain. This result will have applications in studies of nano materials under high external pressure.

2.2. New results for the conductivity

The starting point for this calculation is the summation in eq.(7). In order to take into account the influence of the external pressure (that is, the variability of \( a \)), one has at first to insert \( a > 1 \) in eq.(7), and after that introduce a change of variables in this expression. This has to be of the form \( k \rightarrow x/a; \ g \rightarrow y/a; \ p \rightarrow r/a ; \ q \rightarrow w/a. \) Introducing all these changes in eq.(7), and summing over the variables \( (k,y,r,w) \in (-\pi, \pi) \), one gets the following expression for the susceptibility

\[
\chi \approx \frac{32i}{(1 + \exp[\beta(2t - \mu)])(1 + \exp(\beta << 1 >>))}(tUt)^2 \\
(1 + \exp(\beta << 1 >>)(1 + \exp(\beta << 1 >>)))(\alpha z_1 + i(z_1 + 2t(\cos[1] - \cos[2]))) \\
\cos[1/2(1 - \pi/a)] + (2 - \pi/a)a \\
(-1 + \cosh[(1 - \pi/a) + (2 - \pi/a)a])/(N^4((z_1\alpha^2 + (z_1 + 2t(\cos[1] - \cos[2]))^2)) \\
+ 32i << 6 >> (1 + \cosh(<< 1 >>)) + << 3736 >>)
\]

(9)

Numbers in parenthesis << .. >> denote the number of terms omitted. This long expression for \( \chi \) is impractical in any real calculation.
An applicable expression can be obtained by a relatively simple procedure. Multiply out the products and powers in eq.(9), and express the result as a sum. This is a well defined operation, the end result of which is a huge sum of more than 30000 terms, which is far from applicable.

However, take the first 10 terms of this huge sum, expand it and take only the real part of he frequency into account (as done in [12] but for a small number of terms). One thus gets an expression for the real part of the susceptibility of a 1\text{D} HM. So, what is the improvement of this result compared to [12]? The number of terms taken into account in various developments into series has been doubled. The result for $\chi_R$, taking into account only the first 7 terms is

$$\chi_R \approx \frac{192 \exp[3\beta \mu + 2t \beta U^2 \cos[1/2]]}{N^4(\omega + 4t(\sin[1/2]))^2} \times \frac{1}{(\exp[\beta \mu] + \exp[2t \beta])^2 + 128 \exp[3\beta \mu + 2t \beta U^2 \omega \cos[1/2]]}{(\exp[\beta \mu] + \exp[2t \beta])^2} \times \frac{1}{N^4(\omega + 4t(\sin[1/2]))^2} + \ldots$$

(10)

Inserting eq.(10) into expression (4) one can obtain the imaginary part of the electrical conductivity. Using the Kramers-Kronig relations [12],[14] it becomes possible to determine the imaginary part of the susceptibility as the following integral.

$$\chi_I(\omega_0) = -2\frac{\omega_0}{\pi} P \int_0^\infty \frac{\chi_R(\omega)d\omega}{(\omega^2 - \omega_0^2)}$$

(11)

where $P$ is the principal value of the integral. The final result for the electrical conductivity of a 1\text{D} HM is

$$\sigma_R \approx \frac{\omega_0^2}{4\pi \omega_0} \times \frac{1536 \exp[2t \beta + 2\beta \mu U^2 \omega_0^3 \sin[1/2] \sin[2]]}{(\exp[2t \beta] + \exp[\mu \beta])} \times \frac{1}{(\omega_0^2 - 16t^2(\sin[1/2]))^4} + \ldots$$

(12)

The advantage of this expression, compared to the one previously presented in [12] is in the increased number of terms taken into account in various developments into series. This should allow a more precise comparison with experimental data on the conductivity.

A more complicated case is the 2\text{D} lattice, the simplest of which is a square lattice of side length $a$ which can be treated as an extension of these results. If the lattice sides are denoted by $x$ and $y$, the electric current flowing through such a system can be expressed as

$$\vec{j} = j_x \vec{e}_x + j_y \vec{e}_y$$

(13)

where $\vec{e}_x$ and $\vec{e}_y$ are unitary vectors of the two lattice axes. The total current is given by

$$j^2 = j_x^2 + j_y^2$$

(14)

By definition $j = \sigma E$ where $\sigma$ is the electrical conductivity and $E$ the electric field, and assuming that $E_x = E_y = E$, it finally follows that

$$\sigma_{2D}^2 = \sigma_x^2 + \sigma_y^2$$

(15)

The conductivities along the two axes are assumed to be mutually independent.
3. Compressing and stretching a material

The calculation of the electrical conductivity of the HM discussed in the preceding section refers to standard pressure. However, in nature, applications and various experiments, situations occur in which a material is subjected to variable pressure. The obvious effect which increased pressure has on a material is a decrease of its characteristic dimensions and of all the characteristics of the material which depend on it. Therefore the question is how one can take into account the influence of external pressure on the conductivity of the HM. The consequences of stretching on the conductivity of a material, within the HM, have been discussed to some extent in [3].

3.1. The 1D HM

Mathematically speaking, in the case of a 1D material, the function characterizing the influence of high pressure on the conductivity would be the ratio

$$\frac{\partial \sigma}{\partial a}$$

(16)

where $\sigma$ is the conductivity and $a$ the lattice constant. The influence of external pressure on a material is characterized by the strain $\epsilon$. The strain applied to an object of initial length $l_0$ so that it achieves length $l < l_0$ is defined by $\epsilon = (l_0 - l)/l_0$. Therefore, eq.(16) can be transformed in the form

$$\frac{\partial \sigma}{\partial a} = \frac{\partial \sigma}{\partial \epsilon} \left( \frac{\partial a}{\partial \epsilon} \right)^{-1}$$

(17)

The conductivity depends on the strain only indirectly, through the chemical potential, so that

$$\frac{\partial \sigma}{\partial a} = \frac{\partial \sigma}{\partial \epsilon} \left( \frac{\partial a}{\partial \epsilon} \right)^{-1} = \frac{\partial \sigma}{\partial \mu} \frac{\partial \mu}{\partial \epsilon} \left( \frac{\partial a}{\partial \epsilon} \right)^{-1}$$

(18)

Using expression (8) for the chemical potential and the definition of the strain, it turns out that the product $(\partial \mu/\partial \epsilon)(\partial \epsilon/\partial a)$ does not depend on $a$, which means that it does not depend on external pressure. However, the conductivity itself depends (among other factors) on the external pressure. It can be shown, performing some algebra on eq.(12), that

$$\frac{\partial \sigma}{\partial \mu} \approx \frac{1152 \exp[2\beta(t + \mu)]t^8U^2\beta\omega_0\omega_0^2\beta...}{N^4 \pi^4 \chi_0 \omega(\exp[2\beta t] + \exp[\beta \mu])^2...}$$

(19)

Inserting eqs.(8) and (19) in full form into eq.(18), one could obtain an explicit expression for the strain dependence of the conductivity.

3.2. The simplest 2D case

A more complex, and more interesting case is the 2D HM. The simplest possible case of a 2D lattice is a rectangular lattice, with sides of length $a$ and $b$. If the external pressure is acting within the plane of the lattice, its effect will be to reduce the surface of the elementary lattice cell. Denoting this surface by $S$, it follows that the influence of external pressure on the conductivity will be characterized by the derivative $\partial \sigma/\partial S$.

Changes of the dimensions of the lattice cell under pressure enter the calculation through the change of its sides. In the case of a rectangular lattice, the surface of the elementary lattice cell is given by

$$S = ab$$

(20)

As $a = a_0(1 - \epsilon)$ and $b = b_0(1 - \epsilon)$, where $\epsilon$ is the strain, it follows that

$$S = S_0(1 - \epsilon)^2$$

(21)
It follows that
\[
\frac{\partial \sigma}{\partial S} = \frac{\partial \sigma}{\partial \epsilon} \frac{\partial \epsilon}{\partial S} = \frac{\partial \sigma}{\partial \mu} \frac{\partial \mu}{\partial \epsilon} \left( \frac{\partial S}{\partial \epsilon} \right)^{-1}
\] (22)

Clearly \(\frac{\partial S}{\partial \epsilon} = -2S_0(1 - \epsilon)\), so
\[
\frac{\partial \sigma}{\partial S} = -\frac{1}{2S_0(1 - \epsilon)} \frac{\partial \sigma}{\partial \mu} \frac{\partial \mu}{\partial \epsilon}
\] (23)

How do we approximate the chemical potential of the electron gas on a 2D rectangular lattice? This was solved in [15], where it was shown that
\[
n = \frac{1}{2ab} \left[ 1 - \exp[-\mu/T] I_0(\frac{2t_x}{T}) I_0(\frac{2t_y}{T}) \right]
\] (24)

where \(n\) is the filling factor and \(I_0\) the modified Bessel function of the first kind of order 0. Developing into series, taking \(t_x\) and \(t_y\) as small parameters, and limiting the development to lowest order terms, one gets the following result
\[
\exp[-\mu/T] = \frac{1 - 2nab}{1 + (\frac{t_x^2 + t_y^2}{2T^2}) + (\frac{t_x^2 t_y^2}{T^4})}
\] (25)

Taking the natural logarithm of both sides, developing and retaining only the lowest order terms, one gets the following approximate result for the chemical potential of the electron gas on a 2D rectangular lattice:
\[
\mu \approx k_B T(2abn + 2(abn)^2 + \left(\frac{8}{3}\right)(abn)^3 + \left(\frac{t_x t_y}{(k_B T)^2}\right)^2 - \frac{t_x^2}{(k_B T)^2})^4 + ...]
\] (26)

Introducing the strain \(\epsilon\) in eq.(26) by \(a = a_0(1 - \epsilon)\) and \(b = b_0(1 - \epsilon)\), it becomes possible to determine the derivative \(\partial \mu / \partial \epsilon\). The derivative \(\partial \sigma / \partial \epsilon\) can (in principle) be calculated from the previously defined and calculated conductivity of the 2D rectangular lattice. Combining those partial results, one gets the final approximation for \(\partial \sigma / \partial S\), which describes the influence of high pressure on the conductivity of this particular 2D lattice.

Equation (24) gives the possibility of determining the region (or point) in phase space in which the chemical potential of a 2D electron gas becomes approximately equal to zero. In 1D problems, the point \(\mu = 0\) corresponds to a half-filled band, and experimentally to a "clean" specimen. Inserting \(\mu = 0\) into eq.(24) it follows that
\[
1 - I_0(\frac{2t_x}{T}) I_0(\frac{2t_y}{T}) - 2S_0n(1 - \epsilon)^2 = 0
\] (27)

which can be solved to give
\[
n = \frac{1 - I_0(\frac{2t_x}{T}) I_0(\frac{2t_y}{T})}{2S_0(1 - \epsilon)^2}
\] (28)

This is the value of the filling factor for which \(\mu = 0\), expressed as a function of various lattice parameters and the strain provoked by the application of pressure.

It would be interesting mathematically to determine if a point for which \(\partial \sigma / \partial S = 0\) exists in principle, or for some particular combination of material parameters. From the experimental point of view, the existence of such a point would imply that a material is not sensitive to applied pressure.
4. Optics, invisibility and the HM

The electrical conductivity of a material is linked to its reflectivity. This theoretical result is known for decades and it has very interesting experimental consequences. Namely, the existence of this link opens up the possibility of calculating the conductivity from the measured values of the reflectivity. The only "ingredient" of such a calculation is some theoretical model of the material under study; in this paper we shall discuss to some extent the HM. Full details of this calculation have recently been discussed in [10], but some possibilities for improvement shall be presented here.

Two parameters characterize the propagation of an electromagnetic wave through an anisotropic material. These are the dielectric function $\varepsilon(\omega)$ and the refractive index $N(\omega)$, with $N(\omega) = \sqrt{\varepsilon(\omega)}$ [10]. It is known from theory of optics that

$$\varepsilon = \varepsilon_R(\omega) + i\varepsilon_I(\omega)$$

and

$$N(\omega) = n(\omega) + iK(\omega)$$

(29)

$K(\omega)$ denotes the extinction coefficient. Reflectivity of a material is defined by the following quotient [10]

$$R(\omega) = \left(\frac{n}{n+1}\right)^2 + \frac{K^2}{(n+1)^2}$$

(30)

Algebraic manipulation, discussed in [10], leads to

$$K^4 + K^2 \times \left[1 - \frac{4\pi\sigma_I}{\omega}\right] - \left(\frac{2\pi\sigma_R}{\omega}\right)^2 = 0$$

(31)

The final expression for the reflectivity is [10]

$$R(\omega) = \frac{(2\pi\sigma_R)^2 - K\omega \times [4\pi\sigma_R - K\omega(1 + K^2)]}{(2\pi\sigma_R)^2 + K\omega \times [4\pi\sigma_R - K\omega(1 + K^2)]}$$

(32)

This is the expression linking the reflectivity with the real and imaginary parts of the electrical conductivity of a material. Inserting the appropriate expressions for $\sigma_R$ and $\sigma_I$ would lead to a long equation expressing the reflectivity as a function of various material parameters, which are experimentally measurable. This equation would be the "bridge" between the transport and optical parameters of a material. In general form, without making any reference to the HM, this equation is

$$R \approx \frac{-4\pi(\sigma_f^2 + \sigma_R^2) + \omega[\sigma_I - \sigma_K/\ldots]}{4\pi\sigma_f^2 - \sigma_I\omega + \ldots}$$

(33)

Equations (4),(10),(11),(12) and (15) lead to the functions $\sigma_R$ and $\sigma_I$ for the case of a 2D rectangular lattice. Inserting these results into the full expression for $R$, would give the dependence of the reflectivity on various measurable parameters of a material.

This opens up the possibility of calculating the value of the reflectivity for various values of material parameters. Turning the reasoning "upside down", it also gives the opportunity of finding the values of material parameters for which reflectivity will have any given value. The final implication is that it thus becomes possible to find values of material parameters for which reflectivity will become zero, or have any arbitrarily small value.

The importance of this conclusion is clear. We see (by microscope, telescope or any other instrument) non-radiating objects due to the fact that they reflect light (from some source)
which is incoming on their surfaces. Logically, if the reflectivity is small, the object in question will be hardly visible, and ultimately will become invisible. The conclusion is that invisibility, or visibility with difficulty of an object, can be achieved by a careful choice of the parameters of its material, and not only by various forms of cloaking.

The experimental parameter which can be controlled most easily is the temperature of the material. This fact opens another possibility of theoretically reaching the situation of \( R \cong 0 \). It is clear that the following expression holds:

\[
R(T) \cong R(T_0) + \left( \frac{\partial R}{\partial T} \right)(T - T_0)
\]

with

\[
\frac{\partial R}{\partial T} = \frac{\partial R}{\partial \sigma_R} \frac{\partial \sigma_R}{\partial T}
\]

All the functions in this equation can be calculated using results presented in this paper. The reflectivity becomes zero at the temperature given by

\[
T = T_0 - \left( \frac{\partial R}{\partial T} \right)^{-1} R(T_0)
\]

5. Conclusions
The preparation of this paper was started with a double aim: to present improvements in calculations on the \( HM \) performed by the present author (reviewed recently in [10]), and to apply those improved results to some 2D nano materials. In the course of writing, this plan was somewhat changed, to "just" presenting several mathematical improvements to previously performed calculations.

The improvements discussed are as follows: In the calculation of the electrical conductivity, the number of terms which intervene in various developments into series was drastically increased compared to [12]. This was made possible by improvements in computing technology compared to what was available back in 1997. The physical motivation for this calculation was enabling the comparison between the experimental data and the theoretical predictions with an increased precision. The calculation of the conductivity of a 2D rectangular lattice was made possible by a simple change of variables.

The influence of external pressure on the conductivity of this 2D lattice was characterized by the derivative \( \partial \sigma / \partial S \). This function was calculated, and the mechanical strain was introduced.

Finally, the link between the \( HM \) and optics was discussed to some extent. In this discussion we have touched the problem of invisibility, and pointed out that it can be achieved without cloaking.

It is hoped that in the future all these improvements will be used in real calculations pertaining to nano materials.

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