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Relativistic Velocities in Nanomaterials: Analysis of the Velocities Correlation Function with a New Analytical Model

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Abstract **– In this paper it is presented an interesting analysis of the behaviour of the velocities correlation function in the hypothesis of relativistic velocities inside a nanostructure. A new analytical model is considered, which has a wide scale range of applicability; in this contest the nanoscale will be considered, but the model holds from sub-pico-level to macro-level. The theoretical framework is performed, so as examples of application.**

Keywords **– Drude-Lorentz-Like Models, Nano-Bio-Technology, Relativistic Velocity, Theoretical Modelling, Transport Processes.**

I. Introduction

The charge transport is one of the most important aspects at nanoscale; it can be influenced by particles dimensions and presents different characteristics with respect to those of bulk. In mesoscopic systems the mean free path of charges, related to scattering phenomena, can become larger than the particle dimensions; the transport depends therefore by dimensions and, in principle, corrections of the transport bulk theories are possible. Also in a thin film, the smallest nanostructure dimension can be less than the free displacement, requiring therefore modifications to existing theoretical transport models. At theoretical level, various techniques have been utilized for the knowledge of transport phenomena, in particular analytical descriptions based on transport equations and numerical approaches, as classical and quantum Monte Carlo simulations.

Recently a new analytical model has been performed, based on the complete Fourier transform of the frequency-dependent complex conductivity of the studied system [1,2], able to present analytical expressions of the three most important transport functions, i.e. the velocities correlation function $<\vec{v}(t)\cdot\vec{v}(0)>_T$, the mean square deviation of position $R^2(t)$ and the diffusion coefficient $D(t)=1/2\,(dR^2(t)/dt)$.

At experimental level, one of the most important technique for the study of the frequency-dependent complex-valued far-infrared photoconductivity $\sigma(\omega)$ is the Time-resolved THz Spectroscopy (TRTS), an ultrafast non-contact optical probe; experimental data are usually fitted via Drude-Lorentz, Drude-Smith [3] and Effective Medium Models [4].

The new considered model fits very well with existing data and contains the previous indicated models as subsets. In the following of paper a brief overview of the fundamental utilized models will be illustrated; starting by the Drude model, its most important extensions are considered, coming to results of this new appeared model, which involves the quantum-relativistic reality.

II. Past and Recent Transport Models

In relation to metals, the past scientists thought in terms of models in which electrons are relatively free and move under the influence of electric fields. Historically two models of the elementary metals theory were created:
a) the "Drude model", published in 1900 and based on the kinetic theory of an electron gas in a solid [5,6]. Electrons were considered to have the same average kinetic energy E_m;
b) a variation of the Drude model, integrated with the foundations of quantum mechanics, called "Sommerfeld model" [7].

In the Drude model the assumption of a mechanism of collisions among ions and electrons allows the thermal equilibrium for the electrons and implies the application of the kinetic theory of gases. The free electrons have only kinetic energy, therefore the average energy is $E_m=(3/2)\,k_B\,T$, where k_B is the Boltzmann's constant. The correlation with an average quadratic velocity v_m is possible through the relation $E_m=(3/2)\,k_B\,T=m\,v_m^2/2$, where m is the free electron mass. At environment temperature v_m is of order of 10^7 cm/s and represents the average thermal electrons velocity. It is also assumed that the time of diffraction is very small with respect to every other considered time. Through such collisions, electrons acquire a thermal equilibrium corresponding to the temperature T of the metal. The possible presence of a constant electric field determines an extra average velocity (the "drift velocity") given by $v_d=-(e\,E/m)\,t$. The relaxation time τ is defined as the average time between two collisions, and get a mean free path $l_{mfp}=v_m\,\tau$. The current density is given by $\vec{J}=\sigma_{cond}\,\vec{E}$, where σ_{cond} is the electric conductivity. This result has been an important goal of the classic theory for the metals conduction.

The Lorentz model, published in 1905, is a refining of the Drude model with statistical aspects [8]. Electrons are considered as free charges, with charge "*-e* ", and described by a maxwellian velocity distribution. An electron gas in a spatial region with a constant electric field has a constant drift velocity, corresponding to a current density $\vec{J}$, proportional to the applied field $\vec{J}=\sigma_0\,\vec{E}$, with $\sigma_0=n\,e^2\,\tau/m$ (n is the electron density). Estimating the relaxation time τ, Drude and Lorentz obtained conductivity values in good accordance with experiments. In presence of an electric field of the form $E(t)=E_0\, e^{-i\omega t}$, the complex conductibility is writable as $\sigma_\omega=\sigma_0/(1-i\omega\tau)$. Such model, known as "Drude-Lorentz model", received some success, but underlined also serious difficulties.

Starting by the Drude-Lorentz model, it is possible to obtain the velocities correlation function, the quadratic average distance crossed by the charges as a function of time and to examine directly the possible compatibility with the Einstein relation $D/\mu=k_B T/e$, where D is the diffusion coefficient and μ the mobility [8].

Considerable variations of the Drude-Lorentz model appeared in the following years; the most utilized are:

c) the "Maxwell-Garnett model" (MG): in this model the dielectric function is given by aDrude term with an additional "vibrational" contribution at a finite frequency ω_o, which leads to a dielectric function of the form:

$$\varepsilon_{//}(\omega)=1-\frac{\omega_p^2}{\omega(\omega+i/\tau)}+\frac{\omega_s^2}{\omega_0^2-\omega^2-i\gamma\omega}, \qquad (1)$$

where the amplitude ω_s, the resonant frequency ω_0 and the damping constant γ are material-dependent constants. The MG model is used for describingan isotropic matrix containing spherical isolated inclusions, such as metal particles dispersed in a surrounding host matrix [9,10].

d) The "Effective Medium Theories" (EMTs): in this case the electromagnetic interactions between pure materials and host matrices are approximately taken into account. The commonly used EMTs include the MG model and the "Bruggeman model" (BR), this last as particular variation of the MG model [11,12].

In the THz regime, the dielectric function $\varepsilon_m(\omega)$ consists normally of contributions of the high-frequency dielectric constant, conduction free electrons and lattice vibration:

$$\varepsilon_m(\omega)=\varepsilon_\infty-\frac{\omega_p{}^2}{\omega^2+i\gamma\omega}+\sum_j\frac{\varepsilon_{st_j}\,\omega_{TO_j}{}^2}{\omega_{TO_j}{}^2-\omega^2-i\Gamma_j\,\omega}. \qquad (2)$$

In Eq. (2), ε_∞ is the high-frequency dielectric constant, the second term describes the contribution of free electrons or plasmons and the last term is related to optical phonons.

If the response originated mainly by the contribution offree electrons or plasmons, the Drude model is usually adopted, in the form:

$$\varepsilon_m(\omega)=\varepsilon_\infty-\frac{\omega_p{}^2}{\omega^2+i\gamma\omega}, \qquad (3)$$

and describes with good approximation the dielectric properties of metals and semiconductors.If the interaction of a radiation field with the fundamental lattice vibration plays a dominant role and results in absorption of electromagnetic wave, due to the creation or annihilation of lattice vibration, the dielectric function $\varepsilon_m(\omega)$ consists mainly of the contributions of the lattice vibrations, expressed by the classical pseudo-harmonic phonon model in the first approximation:

$$\varepsilon_m(\omega)=\varepsilon_\infty+\frac{\varepsilon_{st}\,\omega_{TO}{}^2}{\omega_{TO}{}^2-\omega^2-i\gamma\omega} \qquad (4)$$

e) The "Smith model": Smith started by the response theory for the optical conductivity, considering an electric field impulse applied to a system, in order to examine the answer with respect to the current [3,13].The real part of $\sigma(\omega)$ related to it is:

$$\int_0^\infty \mathrm{Re}\,\sigma(\omega)\,d\omega=\frac{\pi}{2}\,j(0)=\frac{\omega_p^2}{8} \qquad (5)$$

If the initial current decays exponentially to its initial value with relaxation time τ, it is possible to write:

$$j(t)/j(0)=\exp(-t/\tau), \qquad (6)$$

from which the standard Drude formula is obtainable:

$$\sigma(\omega)=(ne^2\tau/m)/(1-i\omega\tau) \qquad (7)$$

Eq. (7) can be considered as the first term of a series of the form:

$$j(t)/j(0)=\exp(-t/\tau)\left[1+\sum_{n=1}^{\infty}c_n\,(t/\tau)^n/n!\right] \qquad (8)$$

The c_n factors hold into account of the original electrons velocity, remained after the n-th collision. The analytical form of the complex conductivity is:

$$\sigma(\omega)=\frac{ne^2\tau}{m(1-i\omega\tau)}\left[1+\sum_{n=1}^{\infty}\frac{c_n}{(1-i\omega\tau)^n}\right] \qquad (9)$$

III. A New Appeared Analytical Model

A recent theoretical analytical formulation showed to fit very well with experimental scientific data and offers interesting new predictions of various peculiarities in nanostructures [1,2,14-23]. The model contains a gauge factor, which permits its use to study the dynamics of reality processes by sub-pico-level to macro-level, presenting oscillations in time, so as diffusivity characteristics in time [24].

It is based on the complete Fourier transform of the frequency-dependent complex conductivity $\sigma(\omega)$ of the system, which can be deduced from linear response theory (Green-Kubo formula) [25,26]:

$$\sigma_{\beta\alpha}(\omega)=(e^2/\hbar V)\int_0^\infty dt e^{i\omega t}\times\int_0^\beta d\lambda<\vec{v}^\alpha(t-i\lambda)\vec{v}^\beta(0)> \qquad (10)$$

By inversion of Eq. (10), it is possible to obtain the expression of the velocities correlation function $<\vec{v}(t)\cdot\vec{v}(0)>_T$. The presence of an integration from 0 to ∞ is however a problem for the analytical inversion, but it can be overcame evaluating the integral on the entire time axis (-∞, +∞). Considering the real part of the complex conductivity in Eq. (10), the extension to the entire time axis is possible and a complete Fourier transform can be performed. The integral can be resolved in the complex plane considering a Cauchy integration; the

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velocities correlation function is evaluated exactly by the residue theorem [27]. With the analytical expression of $<\vec{v}(t)\cdot\vec{v}(0)>_T$ it is possible to obtain also the analytical form of $R^2(t)$ and $D(t)$.

The real part of $\sigma(\omega)$ results:

$$\mathrm{Re}\,\sigma_{\beta\alpha}(\omega)=\frac{e^2}{2Vk_BT}\int_{-\infty}^{+\infty}dt\left\langle\vec{v}^{\alpha}(0)\vec{v}^{\beta}(t)\right\rangle_T e^{-i\omega t} \tag{11}$$

The integral in Eq. (11) spans the entire *t*-axis, so we can perform the complete inverse Fourier transform. It gives:

$$<\vec{v}^{\alpha}(0)\,\vec{v}^{\beta}(t)>_T=\frac{k_BTV}{\pi e^2}\int_{-\infty}^{+\infty}d\omega\,\mathrm{Re}\,\sigma_{\beta\alpha}(\omega)e^{i\omega t} \tag{12}$$

The new introduced key idea is the possibility to perform a complete inversion of Eq. (12) on temporal scale, i.e. considering the entire time axis (-∞, +∞), not the half time axis (0, +∞), as usually considered in literature [28]. The classical and the quantum version of the indicated model has been performed [1,2,14]; currently the focus is on the relativistic version [29].

IV. Relativistic Motion in Nanostructures

The starting point is the motion equation of a particle travelling in a nanostructure. If we consider relativistic velocity, the considered dynamics law is:

$$\frac{d}{dt}(m_{part}\,\vec{v})=\sum_i\vec{F}_i \tag{13}$$

We studied the condition of relativistic variation of the mass along the *x*-axis and in the fixed ground reference frame; about the forces acting on the carrier (electrons in this case), it has been considered an outer passive elastic-type force of the form $F_{el}=Kx$, a passive friction-type force of the form $F_{fr}=\lambda\dot{x}$, depending by velocity and with $\lambda=m_{part}/\tau$, and an outer oscillating electric field $E=eE_0e^{-i\omega t}$.

For Eq. (13) solutions of the form:

$x=x_0e^{-i\omega t}$ (14)

have been considered. After analytical calculation, the real part of the complex conductivity results:

$$\mathrm{Re}\,\sigma=\frac{Ne^2\tau\gamma}{m_0}\left(\frac{\omega^2}{\omega^2\gamma^2+\tau^2(\omega_0^2-\gamma(1+\beta^2\gamma^2)\omega^2)^2}\right) \tag{15}$$

with $\beta=v/c$ and $\gamma=1\big/\sqrt{1-\beta^2}$.

With the procedure used in the classical case [2], searching values of ω which vanish the denominator of Eq. (15), the solutions are of the form:

$$\omega_{(R/I)_{rel}}=\frac{1}{2\tau\rho}\left(i\pm\sqrt{\frac{4\rho\omega_0^2\tau^2}{\gamma}-1}\right) \tag{16}$$

with $\rho=1+\beta^2\gamma^2=\gamma^2$. We have three cases in relation to the sign of the quantity $\Delta_{rel}=\frac{4\rho\omega_0^2\tau^2}{\gamma}-1$: $\Delta_{rel}>0$, $\Delta_{rel}=0$, $\Delta_{rel}<0$.

The velocities correlation function $\left\langle\vec{v}(0)\cdot\vec{v}(t)\right\rangle$ has therefore the following analytical form:

$\Delta_{rel}>0$

$$\left\langle\vec{v}(0)\cdot\vec{v}(t)\right\rangle=\left(\frac{k_BT}{m_0}\right)\left(\frac{1}{\gamma\rho}\right)\exp\left(-\frac{t}{2\tau\rho}\right)\times\left(\cos\left(\frac{\alpha_{R_{rel}}}{2\rho}\frac{t}{\tau}\right)-\frac{1}{\alpha_{R_{rel}}}\sin\left(\frac{\alpha_{R_{rel}}}{2\rho}\frac{t}{\tau}\right)\right) \tag{17}$$

with $\alpha_{R_{rel}}=\sqrt{\frac{4\rho\omega_0^2\tau^2}{\gamma}-1}\in\mathbb{R}^+$;

$\Delta_{rel}<0$

$$\left\langle\vec{v}(0)\cdot\vec{v}(t)\right\rangle=\frac{1}{2}\left(\frac{k_BT}{m_0}\right)\left(\frac{1}{\gamma\rho}\right)\left(\frac{1}{\alpha_{I_{rel}}}\right)\times\left(\left(1+\alpha_{I_{rel}}\right)\exp\left(-\frac{(1+\alpha_{iI})}{2\rho}\frac{t}{\tau}\right)-\left(1-\alpha_{I_{rel}}\right)\exp\left(-\frac{(1-\alpha_{iI})}{2\rho}\frac{t}{\tau}\right)\right) \tag{18}$$

with $\alpha_{I_{rel}}=\sqrt{1-\frac{4\rho\omega_0^2\tau^2}{\gamma}}\in(0,1)\subset\mathbb{R}$.

The case $\Delta_{rel}=0$ reduces to relativistic Drude model.

V. Examples of Application

As example of application it has considered the motion of electrons, at different velocities, in a nanostructure of ZnO[30-32]. We underline that the model holds for charged particles in general, not necessarily electrons. Changing the nanomaterial, it needs to consider the right effective mass and relaxation time. Data to be implemented in Eqs (17,18) are resumed in Table 1.

Table 1: Data related to the variation of the carrier velocity v inside the nanostructure.

$v(cm/s)$	v/c	$1/\rho$	$\frac{1+\alpha_{I_{Rel}}}{2\rho}$ (a)	$\frac{1+\alpha_{I_{Rel}}}{2\rho}$ (b)	$\frac{\alpha_{R_{Rel}}}{2\rho}$ (c)	$\frac{\alpha_{R_{Rel}}}{2\rho}$ (d)
10^7	$0.334\cdot10^{-3}$	0.999	0.749	0.549	2.497	9.99
10^8	$0.334\cdot10^{-2}$	0.999	0.749	0.549	2.497	9.99
10^9	$0.334\cdot10^{-1}$	0.998	0.748	0.549	2.495	9.98
10^{10}	0.334	0.888	0.666	0.488	2.22	8.88
$2\cdot10^{10}$	0.834	0.304	0.228	0.167	0.76	3.04

(a): $\alpha_{I_{Rel}}=0.5$; (b): $\alpha_{I_{Rel}}=0.1$; (c): $\alpha_{R_{rel}}=5$; (d): $\alpha_{R_{rel}}=20$.

Fig. 1 represents the evolution of $\langle \vec{v}(0)\cdot\vec{v}(t)\rangle$ in time for a fixed value of $\alpha_{R_{rel}}$ in relation to three different velocities of electrons, with $\tau=0.84\cdot10^{-13}\,s$ and $T=300\,K$ [30-33]. The classical "Drude" velocity $v=10^{7}\,cm/s$ implies a negligible variation in mass for the electrons. We note that the increase in velocity tends to raise the wavelength of the damped oscillation, reducing its amplitude.

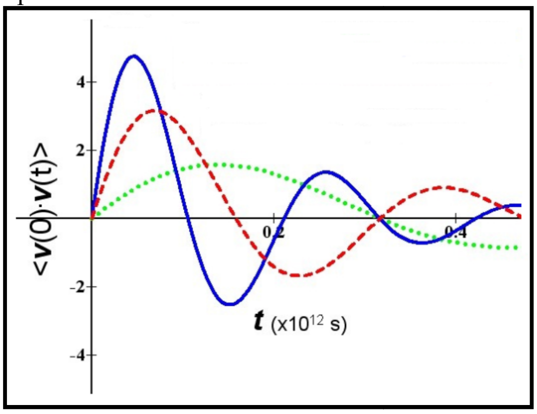

Fig.1. $\langle \vec{v}(0)\cdot\vec{v}(t)\rangle$ vs$t$ for fixed value $\alpha_{R_{rel}}=5$; the considered velocity of the carrier is $v=10^{7}\,cm/s$ (blue solid line), $v=10^{10}\,cm/s$ (red dashed line) and $v=2\cdot10^{10}\,cm/s$ (green dots line).

In Fig. 2 the same situation is presented, but with a different value of the parameter $\alpha_{R_{rel}}$. The initial more marked compression of the curve (blue solid line) obeys to the same variation indicated in the previous case.

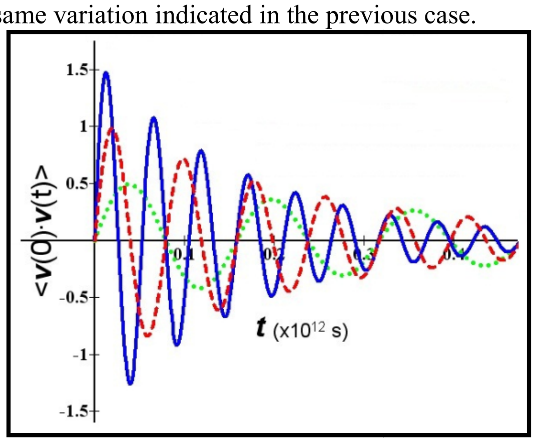

Fig.2. $\langle \vec{v}(0)\cdot\vec{v}(t)\rangle$ vs$t$ for fixed value $\alpha_{R_{rel}}=20$; the considered velocity of the carrier is $v=10^{7}\,cm/s$ (blue solid line), $v=10^{10}\,cm/s$ (red dashed line) and $v=2\cdot10^{10}\,cm/s$ (green dots line).

In Figs 3 and 4 the parameter $\alpha_{I_{rel}}$ has been considered. We note as the typical Smith behaviour of $\langle \vec{v}(0)\cdot\vec{v}(t)\rangle$ tends to become negative in longer times with respect to the classical case and the curves approach the x-axis when the velocity of the carrier increases.

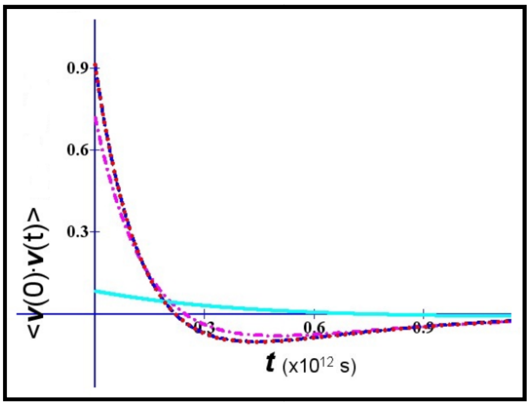

Fig.3. $\langle \vec{v}(0)\cdot\vec{v}(t)\rangle$ vs$t$ for fixed value $\alpha_{I_{rel}}=0.5$; the considered velocity of the carrier is $v=10^{7}\,cm/s$ (blue dashed line) (superposed to the red dots line, representing the classical case), $v=10^{10}\,cm/s$ (violet dot-dashed line) and $v=2.5\cdot10^{10}\,cm/s$ (clear blue solid line).

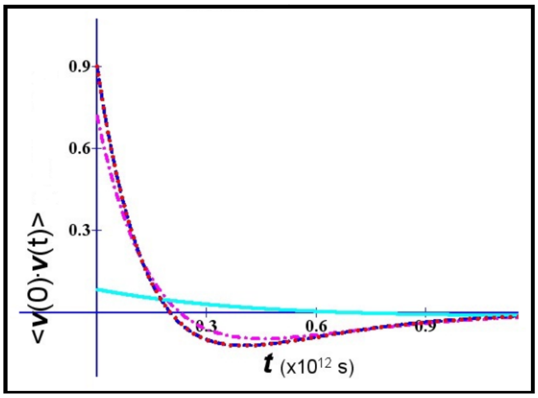

Fig.4. $\langle \vec{v}(0)\cdot\vec{v}(t)\rangle$ vs$t$ for fixed value $\alpha_{I_{rel}}=0.1$; the considered velocity of the carrier is $v=10^{7}\,cm/s$ (blue dashed line) (superposed to the red dots line, representing the classical case), $v=10^{10}\,cm/s$ (violet dot-dashed line) and $v=2.5\cdot10^{10}\,cm/s$ (clear blue solid line).

VI. Conclusion

In this work it has been the effect of relativistic velocities of carriers inside a nanostructure in relation to the variation of mass, utilizing a new interesting analytical model, appeared both in classical and in quantum form, and tested in last years with good accordance with experimental existing data [1,2,14-23,34]; new results regarding the velocities correlation function have been presented. The core of the model regards the possibility to obtain analytical expression of the most important quantities concerning the dynamics of a system, through the complete Fourier transform of the real part of the frequency-dependent complex conductivity $\sigma(\omega)$, extending the integration over time on the entire time axis (-∞, +∞). This extension is mathematically very elegant, because of the analytical approach, and gives new information's about the dynamics of systems through the elaboration of experimental data.

At applied level, we suggest the possibility to a fast/ultrafast injection of carriers in a nanostructure for possible practical needs of raising the wavelength of the damped oscillation for the velocities correlation function and reducing its amplitude.

The complete development of the relativistic calculation will provide interesting peculiarities and new results, like the found time oscillations in velocity at beginning of process for the quantum non-relativistic version [1,14,16,34], which could be appropriately tested through experimental time-resolved techniques, like TRTS [35-37].

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