Single-particle Relaxation Time in Doped Semiconductors beyond the Born Approximation

Gionni Marchetti

National Institute of Chemical Physics and Biophysics,
Rävala 10, 15042, Tallinn, Estonia

(Dated: November 16, 2018)

We compare the magnitudes of the single-particle relaxation time accurately computed by the variable phase approach are with those computed in the first Born approximation for doped semiconductors such as Si and GaAs, assuming that the Coulomb impurities are randomly distributed centers. We find that for typical dopant concentrations in Si the Born approximation can overestimate the single-particle relaxation time by roughly 40% and underestimate it by roughly 30%. It is shown that in the case of GaAs the discrepancies are typically less severe. Our analysis shows that in general these large discrepancies in Si arise from strong violations of the Friedel sum rule. This breakdown occurs in a range of doping densities for which the random phase approximation starts to break down. Moreover, our results suggest that a multi-ion correction to the electron-impurity scattering is needed for high dopant concentrations.

I. INTRODUCTION

The first Born approximation (B1) [1], hereinafter simply referred to as Born approximation, is commonly used within condensed matter theory for computing scattering-related properties, e.g. the carrier mobility or the momentum relaxation time. In combination with the random phase approximation (RPA) [2], the B1 provides the theoretical framework for understanding most of quantum processes which occur in bulk semiconductors [3]. While nothing can be stated a priori about the validity of the Born approximation for the low energy processes, it can be certainly assumed good enough for high-energy collisions. Thus, for scattering processes in solid-state systems, in principle one should calculate several terms of the Born series to make sure it converges, thereby showing that its first term (which defines the B1) is sufficiently accurate for the problem at hand. This difficult task can be accomplished by the standard computation of scattering phase shifts. Unfortunately such calculations can be very tedious and prone to errors [4].

In this work, we show that we can easily depart from B1 for the case of the single-particle relaxation time in bulk semiconductors by means of the phase variable method (VPM) [5]. This is an alternative approach to the computation of the phase shifts, whereby one casts the Schrödinger equation for the scattering problem into a first order nonlinear equation. This approach allows for fast and accurate computations of the phase shifts at low computational cost.

Our computations of the single-particle relaxation times for n-type Si and GaAs semiconductors improve on those based on the Born approximation for the doping densities under scrutiny, showing that the inaccuracies can be of about 30% - 40% for certain dopant concentrations in Si, while they prove to be less severe for GaAs, where they are smaller than about 20% . In doing so, we also gain some important physical insights about the validity of the screened electron-impurity interaction as typically accounted for at RPA level. In fact, we find that the large inaccuracies arise where the random phase approximation starts to break down or where the the electron-impurity scattering probability needs to be corrected in order to include the contribution of the coherent scattering from pairs of distinct impurity centers as proposed by Moore [6, 7].

This paper is organized as follows. In Section II we briefly discuss of the relation between the single-particle relaxation time and the Born approximation. In Section III we present the variable phase method in a pedagogical way as it is not very well-known among condensed matter theorists. Finally, in Section IV our findings for the single-particle relaxation time for n-type Si and GaAs bulk semiconductors are exhaustively discussed within the homogeneous electron gas paradigm (jellium model).

II. THE SINGLE-PARTICLE RELAXATION TIME AND BORN APPROXIMATION

In normal metals and degenerate doped semiconductors, the scattering by static impurities is characterized by two different (electronic) momentum relaxation times: the scattering time $\tau_s$ and the single-particle relaxation time $\tau_{rs}$ [8]. The latter is also commonly known as the quantum lifetime and in semiconductor physics is denoted by $\tau_{ns}$ where $n$ and $k$ are the band index and the Bloch electron wave vector in the Brillouin zone (BZ) respectively. This quantity is inversely proportional to the imaginary part of the self-energy $\text{Im}\Sigma$, i.e., $1/\tau_{rs} = (2/h)\text{Im}\Sigma$ [8].

Assuming that in the bulk of solids the impurities are Coulomb centers randomly distributed, $\tau_{rs}$ can be computed by the perturbation theoretic approach using the one-electron Green’s function for the coupled electron-
impurity system. By this approach, however, the single-particle relaxation time $\tau_s^{-1}$ is commonly obtained in B1, and reads  

$$\frac{1}{\tau_s} = \frac{2\pi m^* n_i}{\hbar^3} \int \frac{d^3k'}{(2\pi)^3} |V_{ei}(q)|^2 \frac{\delta(k' - k_F)}{k'}, \quad (1)$$

where $n_i$ and $m^*$ are the dopant concentration and the electron effective mass respectively. The Fourier component of the electron-impurity interaction potential $V_{ei}(q)$ is computed at the wave vector transfer $q = 2k_F \sin (\theta/2)$ where $k_F$ [10] $\theta$ are the Fermi wave vector and the scattering angle respectively, and according to this physical transport model, the carriers with the Fermi energy $E_F$ scatter off impurities elastically. We note that the matrix element $V_{ei}(q)$ clearly implies that the related scattering amplitude is computed in B1. In fact, the scattering amplitude $f_{B1}$ in Born approximation, is given by  

$$f_{B1}(q) = \frac{-m^*}{2\pi \hbar^2} \int d^3r e^{iqr} V_{ei}(r), \quad (2)$$

which is clearly proportional to the Fourier $q$-component of the interaction potential $V_{ei}(q)$. Note that Eq. (2) requires that the incident and scattered waves are plane. This can occur only if $V_{ei}$ is sufficiently weak that the incident electron wave function is slightly distorted in its amplitude is computed in B1. In fact, the scattering amplitude $f_{B1}$ in Born approximation, is given by [4]

$$f_{B1}(q) = \frac{-m^*}{2\pi \hbar^2} \int d^3r e^{iqr} V_{ei}(r), \quad (2)$$

which is clearly proportional to the Fourier $q$-component of the interaction potential $V_{ei}(q)$. Note that Eq. (2) requires that the incident and scattered waves are plane. This can occur only if $V_{ei}$ is sufficiently weak that the incident electron wave function is slightly distorted in its presence. As a consequence, the differential cross-section $\sigma^1$ in B1 can be obtained by $\sigma^1(q) = |f_{B1}(q)|^2$.

Now, solving the integral given by Eq. (1) one finds that $1/\tau_s$ is proportional to the electron-impurity total cross-section $\sigma^1$ in B1, i.e. $1/\tau_s \propto \sigma^1$. Therefore, in order to depart from B1, one needs to replace $\sigma^1$ by the exact total cross-section $\sigma$. The latter can be obtained through the numerical phase shifts $\delta_l$. In the following Section, we recall how this task can be successfully achieved by means of the variable phase method [11,12,13,14].

### III. THE VARIABLE PHASE METHOD

Due to the cylindrical symmetry of the scattering problem, one can expand the electron wave function $\psi(r)$ by the functions $u_l(r)$ which are the solutions of the radial Schrödinger equation. The latter, setting $2m^*$ and to $\hbar$ unity, reads

$$u''_l(r) + \left[ k^2 - l(l + 1)/r^2 - V_{ei}(r) \right] u_l(r) = 0. \quad (3)$$

The scattering potential $V_{ei}$ is responsible for the presence of $\delta_l$ in the asymptotic behavior of $u_l(r)$, i.e. $u_l(r) \sim \sin (kr - l\pi/2 - \delta_l)$ for $r \to \infty$. Thus, solving the radial Schrödinger equation for a scattering problem is equivalent to compute the $\delta_l$, thereby one can obtain the scattering total cross-section $\sigma$, and hence the exact single-particle relaxation time $\tau_s$.

The variable phase approach is an alternative method to the integration of Eq. (1) which directly yields the exact phase shifts except for very small numerical errors. In VPM one obtains $\delta_l$ by integrating the phase equation, a first order nonlinear equation which is a generalized Riccati equation. The phase equation reads [5]

$$\delta'_l(r) = -k^{-1}V_{ei}(r) \left[ \cos \delta_l(r) \hat{j}_l(kr) - \sin \delta_l(r) \hat{n}_l(kr) \right]^2, \quad (4)$$

where $\hat{j}_l$, $\hat{n}_l$ are the Riccati-Bessel functions and the boundary condition at the origin is given by $\delta_l(0) = 0$. The $\delta_l$ numerical values are then defined by the limit

$$\lim_{r \to \infty} \delta_l(r) = \delta_l, \quad (5)$$

where $r$ is the inter-particle distance, that is, taking the asymptotic $\delta_l$ values far away from the impurity center.

In order to compute the $\delta_l$ by Eq. (4) one needs to provide the electron-impurity interaction potential to the radial Schrödinger equation. This scattering potential should be short-range and regular enough as typically required in scattering theory, see Refs. [5,10]. In normal metals and doped semiconductors, when the linear screening is accounted for by the Thomas-Fermi wave vector $q_{TF}$ and the screened electron-impurity interaction is computed in RPA, $V_{ei}$ takes a Yukawa form, see Refs. [8,13,14]. Throughout this work we shall assume that this is the case, referring the reader to Refs. [2,15,16] for a detailed discussion of its validity.

It is fortunate that the Yukawa interaction potential belongs to a class of regular potentials for which one can compute the $\delta_l$ exactly by VPM. In the following along with Ref. [14] we shall consider $n$-type Si with $n_i = 10^{17} - 2.5 \times 10^{20}$ cm$^{-3}$ taking the longitudinal effective mass $m^* = 0.89m_e$ and the transverse effective mass $m^* = 0.19m_e$. [14,17], $m_e$ being the bare electron mass, and $\varepsilon = 12.0\varepsilon_0$ the dielectric constant where $\varepsilon_0$ is the vacuum permittivity. Moreover, we shall consider single-ionized impurities ($Z = 1$) and the electron density $n$ will be set to $n = n_i$. [14].

In Fig. 1 it is shown how the $\delta_l$ for $l = 1,2$ are computed in the variable phase approach for a doped Si semiconductor with $n_i = 10^{17}$ cm$^{-3}$ (top panel (a)) and $n_i = 10^{20}$ cm$^{-3}$ (bottom panel (b)). First, we note that all the phase shifts are positive due to the attractive interaction potential $V_{ei}$. Second, the $\delta_l$ correctly decrease with increasing scattering energy $E_F = h^2k_F^2/2m^*$ where $k_F \sim n^{1/3}$. Thus the phase shifts $\delta_l$ on the bottom panel (b) whose values are $\delta_0 = 0.36$ and $\delta_1 = 0.14$ in radians are smaller compared to those shown on the top panel (a) where $\delta_0 = 2.15$ and $\delta_1 = 0.17$. Indeed the carriers scatter off the impurities with higher energy increasing the dopant concentration. We also note that for a given scattering energy, $\delta_l \ll \delta_0$ due to the repulsive effect of the centrifugal potential $l(l+1)\hbar^2/2m^*r^2$ which becomes stronger for higher $l$ values. The phase variable method provides the numerical values of the $\delta_l$, as shown in Fig. 1 from the asymptotes of the $\delta_l(r)$ curves and at the same time removes their mod($\pi$) issue thus allowing
the computation of scattering phase shifts in a unique and unambiguous way [5].

IV. RESULTS AND DISCUSSION

Along with Ref. [8] we define the dimensionless quantity \( y = k_F/q_{TF} \) as we wish to compare the ratio \( \tau_s^1/\tau_s \) against \( y \). To this end, we note that \( \tau_s^1/\tau_s = \sigma/\sigma^1 \) where the exact total cross-section \( \sigma \) is given by [1]

\[
\sigma (k_F) = \frac{4\pi}{k_F} \sum_{l=0}^{l_{max}} (2l+1) \sin^2 \delta_l, \quad (6)
\]

\( l_{max} \) being the maximum of set \( \{0, 1 \cdots l - 1, l\} \) for a given \( l \), which is expected to be a small integer. In fact, even on the basis of a semi-classical analysis of scattering it can be shown that only a few partial waves will actually contribute to the total cross-section in low energy collisions [18]. Note that if the phase shifts \( \delta_l \equiv \delta_l^1 \) were computed in B1 for a potential of Yukawa form, and after we insert them into Eq. [6] one would obtain a a nice simple analytical expression for the total cross-section [4].

In Fig. 2 (top panel (a)) we plotted the ratio \( \tau_s^1/\tau_s \) against \( y \) for many different \( l_{max} \) values for a doped Si semiconductor [19]. Not surprisingly the major contributions come from \( l_{max} = 0 \) (s-wave, \( l = 0 \)) and \( l_{max} = 1 \) which accounts for s-wave and p-wave (\( l = 1 \)) together. Their contributions are substantial for \( y \ll 1 \), the interval which defines the low-energy limit of scattering theory for short-range potentials. The maxima of the curves happen roughly at the cross-over region between low-energy and high-energy carrier-impurity collisions, the latter region being defined for \( y \gg 1 \). The curves clearly show that in B1 the single-particle relaxation can be underestimated by roughly by 30% and overestimated by roughly 40% for \( y \lesssim 1 \). Such large discrepancies for the single-particle relaxation times can be understood looking at the relative exact phase shifts computed in the phase variable approach. In Fig. 2 (bottom panel (b)) the relative phase shifts \( \delta_0, \delta_1, \delta_2 \) are plotted against \( y \). Clearly, \( \delta_0 \) is too large for the Born approximation to hold, in fact its validity would require that \( \delta_l \) be small compared to...
$\pi/2$ \cite{11, 20, 21}. Note that this criterion is equivalent to say that the scattering potential is sufficiently weak. However, a better agreement between $\tau^4_s$ and $\tau_s$ is found in the limit $y \to 2$ where $\tau^4_s \approx \tau_s$. This is consistent with the fact that $\delta_l$, and the other phase shifts as well, decrease monotonically as the carriers’ energy increases, thus improving the Born approximation.

In order to understand the semiconductor many-body effects on $\tau_s$, we performed similar computations for a n-type GaAs semiconductor in the same range of doping concentrations taking the following material parameters: $m^* = 0.067m_e$ and $\varepsilon = 12.9\varepsilon_0$ \cite{13}.

In Fig. 3 (top panel (a)) we show our results for $\tau^4_s/\tau_s$ computed via the VPM. The ratio curves now show a good agreement for a large interval of $y$ values, see top panel (a). We found that $\tau^4_s$ can be overestimated by roughly 30% for $y \approx 1$ and underestimated by roughly $20\%$ in the limit $y \to 4$. For $1.8 \lesssim y \lesssim 2.2$ the ratio discrepancy is less than about $3\%$. However, here we need to make an important observation about our results for large $y$ values. It can been shown that for $y \gg 1$ the Born approximation would produce the same results as a bare Coulomb potential (Rutherford scattering) for scattering angles $\theta \gg 1/y$, see Ref. \cite{22} for an extensive analysis, thus disregarding the screening effects altogether. Therefore in this case, it is not clear whether or not the Born approximation can be still considered suitable for modeling screened electron-impurity collisions. In particular, this would certainly affect the physics of GaAs for most of the dopant concentrations under scrutiny.

Nevertheless, the nicer behavior of $\tau^4_s$ observed in GaAs can be clearly understood from the bottom panel (b) of Fig. 3 which shows that all phase shifts are now much smaller than $\pi/2$, thus improving the accuracy of the Born approximation. Note that carriers in GaAs have much more energy available in the center of mass, hence making $\delta_l$ smaller in comparison to those computed for Si. This is a direct consequence of a much smaller effective mass of the carriers in GaAs.

Until now, our findings rely on the validity of the screened Coulomb potential for modeling the electron-impurity interaction in the bulk of doped semiconductors. Within the present formalism, we can address its relationship with the random phase approximation linking the $\delta_l$ to the Fermi sphere through the Friedel sum rule (FSR) \cite{23}. The FSR states that the impurity charge must be completely neutralized by the carriers, and at the same time the extra electrons required to this end should fill the levels up to the Fermi energy of an ideal crystal. In mathematical terms the FSR reads

$$Z = \frac{2}{\pi} \sum_{l=0}^{\infty} (2l + 1) \delta_l (E_F) .$$

By the variable phase approach we computed the scattering phase shifts with the following cut-offs: $l_{\text{max}} = 5$ and $l_{\text{max}} = 6$ for Si and GaAs respectively. In Fig. 4 we plot the curves (solid line, Si) and (dashed line, GaAs) relative to random phase approximation. Whether or not RPA is applicable it depends on the smallness of the Wigner-Seitz parameter $r_s$ \cite{2}. It is expected that RPA works well for $r_s < 1$ \cite{2}. In the inset of Fig. 4 the $r_s$ curves (solid line, Si) and (dashed line, GaAs) in the same range of $y$ values are shown. Remarkably, the strong violation of Friedel sum rule observed in Si can be linked to the non-applicability of RPA ($r_s \gg 1$) for roughly the same $y$ values. Hence, in that region, it may be necessary to account for the short-range exchange and correlation effects in carriers’ dynamics in Si, which are not present in the RPA. Furthermore we observe that for large $y \to 2$, see Fig. 4 some large violations of the FSR for Si and GaAs as well would start to happen again. Indeed, according to the present physical model, there is no way to prevent $\delta_l$ from decreasing for increasing $y$ values. Thus, if we wish to continue working in the RPA, i.e. keeping the screened Coulomb potential, due to the corresponding smallness of $r_s$, we would need to include the coherent scattering from pairs of distinct impurity centers as found by Moore in his attempt to go beyond the B1 for the electron-impurity scattering \cite{5,7}. This seems physically sound due to the reduction of the average distance between the Coulomb impurity centers in that region.

We conclude by noting that in the present work we limited ourselves to Thomas-Fermi screening for the homogeneous electron gas \cite{13}. Within the random phase approximation, a different screened electron-impurity potential emerges when the many-body effects are taken into account by Lindhard screening. In this case, the scattering potential, whose analytical form is not given, shows a tail with an oscillatory behavior far away from the impurity center \cite{13}. For this reason, we cannot employ the VPM in such a case.

![Figure 4](image-url)

**FIG. 4.** The curves (solid line, Si and dashed line, GaAs) versus $y$ obtained by computing $\delta_l$ from VPM and using Eq. 7 and assuming $Z = 1$. In the inset the relative curves of the dimensionless Wigner-Seitz parameter $r_s$ versus $y$ are shown.
In summary, we showed that the VPM outperforms the Born approximation when it comes to compute $\tau_s$. From a practical point of view, these accurate numerical values may be employed as input to other condensed matter models and/or to applications of the density functional theory (DFT) \[24\][26]. From a theoretical point of view, we gained some important physical insights of the many-body dynamics within the homogeneous electron gas model. Finally, our approach restores the unitarity (probability conservation) which is manifestly violated by the Born approximation as it fails to satisfy the optical theorem.

ACKNOWLEDGMENTS

We are grateful to Fabio Caruso and Giustino Feliciano for some useful comments. We are indebted to Marco Patriarca and Sean Fraser for reading a preliminary version of the manuscript. This work was supported by institutional research funding IUT (IUT39-1) of the Estonian Ministry of Education and Research, by the Estonian Research Council grant PUT (PUT1356).

[1] J. R. Taylor, *Scattering Theory: The Quantum Theory of Nonrelativistic Collisions* (John Wiley & Sons, Inc., New York, 1972).
[2] G. Giuliani and G. Vignale, *Quantum Theory of Electron Liquids* (Cambridge University Press, Cambridge, UK, 2005).
[3] B. K. Ridley, *Quantum Processes in Semiconductors*, 5th ed. (Oxford University Press, Oxford, 2013).
[4] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill, Singapore, 1968).
[5] F. Calogero, *Variable Phase Approach to Potential Scattering* (Academic Press, 1967).
[6] E. J. Moore, *Phys. Rev.* **160**, 607 (1967).
[7] E. J. Moore, *Phys. Rev.* **160**, 618 (1967).
[8] S. Das Sarma and F. Stern, *Phys. Rev. B* **32**, 8442 (1985).
[9] A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Englewood Cliffs, NJ, 1963).
[10] Assuming that the incident wave and the scattered wave have wave vectors $\mathbf{k}$ and $\mathbf{k'}$ respectively, then $\mathbf{q} = \mathbf{k} - \mathbf{k'}$. Note that $q = |\mathbf{q}|$.
[11] F. Calogero, *Nuovo Cimento* **27**, 745 (1963).
[12] The total cross-section $\sigma$ is given by the standard formula $\sigma(k) = \frac{2\pi}{k^2} \sum (2l + 1) \sin^2 \delta_l$ [11].
[13] N. W. Ashcroft and M. D. Mermin, *Solid State Physics* (Saunders College, Philadelphia, 1976).
[14] F. Caruso and F. Giustino, *Phys. Rev. B* **94**, 115208 (2016).
[15] G. Marchetti, *Journal of Physics: Condensed Matter* **30**, 475701 (2018).
[16] G. Marchetti, (2018), arXiv:1801.02210.
[17] The overall effective mass is $m^* = \frac{1}{2/m_{\parallel} + 1/m_{\perp}}$, i.e. $m^* \approx 0.25m_e$. Note that in Ref. [27] $m_{\parallel} = 0.91m_e$, however this would make a negligible difference.
[18] G. Marchetti and I. D’Amico, *physica status solidi (b)* **254**, 1600806 (2017).
[19] The number of computed waves is chosen to allow the convergence of $\tau_s$ curves in the range of doping densities under scrutiny.
[20] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Springer-Verlag, Berlin Göttingen Heidelberg, 1957).
[21] P. M. Morse and W. P. Allis, *Phys. Rev.* **44**, 269 (1933).
[22] D. ter Haar, *Problems in Quantum Mechanics*, 3rd ed. (Dover Publications, Inc., New York, U. S. A, 2014).
[23] G. D. Mahan, *Many-Particle Physics* (Kluwer Academic., New York, U. S. A, 2000).
[24] P. Hohenberg and W. Kohn, *Phys. Rev.* **136**, B864 (1964).
[25] W. Kohn and L. J. Sham, *Phys. Rev.* **140**, A1133 (1965).
[26] K. Capelle, *Brazilian Journal of Physics* **36**, 1318 (2006).
[27] O. Madelung, *Semiconductors Data Handbook*, 3rd ed. (Springer-Verlag, Berlin Heidelberg, 2004).