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Published in:
Physica E-Low-Dimensional Systems & Nanostructures

DOI:
10.1016/j.physe.2009.06.069

2010

Citation for published version (APA):

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Modeling of cotunneling in quantum dot systems

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Transport through nanosystems is treated within the second order von Neumann approach. This approach bridges the gap between rate equations which neglect level broadening and cotunneling, and the transmission formalism, which is essentially based on the single-particle picture thereby treating many-particle interactions on an approximate level. Here we provide an alternative presentation of the method in order to clarify the underlying structure. Furthermore we apply it to the problem of cotunneling. It is shown that both elastic and inelastic cotunneling can be described quantitatively, while the transmission approach with a mean-field treatment of the interaction provides an artificial bistability.

PACS numbers: 73.23.Hk, 73.63.-b

I. INTRODUCTION

Nanostructure technology allows for the fabrication of small structures, such as quantum dots [1], nanowires [2], carbon nanotubes [3], as well as molecular structures, whose electronic properties are dominated by a small number of electrons. The insertion into electrical circuits exhibits current-bias (IV) relations, which strongly differ from conventional resistors. Typical features are the suppression of current for low bias due to Coulomb blockade and pronounced current steps, whose position describes the leads, where $\sigma = \uparrow, \downarrow$ denotes the spin, $k$ labels the spatial wave functions of the contact states, and $\ell$ denotes the lead (typically $\ell = L/R$ for the left and right lead, respectively). In most works a thermal occupation of the leads

$$
\langle c_{k}\sigma \ell \rangle = f_{k}\sigma \ell - \frac{1}{e(E_{k}-\mu_{\ell})/k_{B}T+1},
$$

with chemical potential $\mu_{\ell}$ is assumed, where the bias $\epsilon V_{\text{bias}} = \mu_{L} - \mu_{R}$ drives the current in a two terminal system. Conventionally noninteracting leads are assumed. Finally, $H_{T}$ describes the coupling between the leads and the quantum dot.

If $H_{D}$ does not contain many-particle interactions, it is straightforward to derive a transmission formula (see [6] and references therein) yielding a particle current from lead $\ell$ into the quantum dot

$$
J_{\ell} = \frac{1}{2\pi\hbar} \int dE T_{\ell\rightarrow d}(E) [f_{d}(E) - f_{\ell}(E)].
$$

Here the transmission function is determined by the single-particle spectrum of $H_{D}$ in connection with the

\[\text{FIG. 1: Schematic current-bias relation for a quantum dot, showing the qualitative influence of the key energy scales.}\]
coupling $H_T$, which essentially broaden the levels. A formal tool is the nonequilibrium Green function formalism [7]. An inclusion of many-particle effects within the mean-field level is straightforward and exchange-correlation effects can be taken into account by more involved mean-field potentials [8]. Also time-dependent simulations are possible using time-dependent density functional theory [4]. However, hardly any results exist, which go beyond mean-field (a rather old example is [10]). Thus this approach works for arbitrary temperatures if correlation effects due to the Coulomb interaction in the system are small.

Another approach is to start from the von-Neumann equation

$$i\hbar \frac{d}{dt} \rho = -[\rho, H]$$

(4)

treating the time evolution of the density operator $\rho$ containing both the system and the leads. As only the time-evolution of the system is of interest, one can trace-out the lead degree of freedom in order to obtain the reduced dot density operator

$$\rho_{\text{dot}}(t) = \text{Tr}_{\text{leads}} \{ \rho(t) \} .$$

(5)

The key task is to find an approximate equation of motion for $\rho_{\text{dot}}(t)$ taking into account the coupling $H_T$. This provides (generalized) master equations [11].

To lowest order in $H_T$ electron transitions between the quantum dot and the leads are treated independently from each other (here called first order following the number of correlated transitions). Restricting to diagonal elements of $\rho_{\text{dot}}(t)$, one obtains to lowest order the Pauli master equations [12, 38]. Furthermore, coherences, represented by nondiagonal matrix elements, can be treated in different ways (frequently also denoted quantum master equations). The traditional approach is the Wangness-Bloch Redfield kinetics [14, 15] which has been applied to quantum dots in [16, 17]. While reasonable results are typically found, there is a fundamental problem due to the occurrence of negative occupations as the equations are not of Lindblad type [18]. Lindblad-type kinetic equations were derived in [19, 20], but only for the high-bias limit. All first-order approaches entirely neglect broadening effects but can treat the interactions in the system exactly by a diagonalization of the isolated dot Hamiltonian [21]

$$H_D = \sum_a E_a |a\rangle \langle a|,$$

(6)

where the states $|a\rangle$ are many-particle states, which may be highly correlated.

In order to treat both broadening and interaction, we introduced a second order approach based on the von Neumann equation [21] which considers both the dynamics of $\rho_{\text{dot}}(t)$ and higher-order tunneling transitions [22]. This second order von Neumann approach (2vN) bridges the gap between the transmission formalism, which is fully reproduced for systems without interactions at arbitrary temperatures, and the first order generalized master equation schemes, which are recovered in the limits of high temperature or large bias.

In this paper we provide a slightly different presentation of our 2vN method in Sec. III in order to highlight its structure. Its range of validity and previous results are summarized in Sec. III. In Sections IV-VI we show new results demonstrating its applicability to the cases of elastic and inelastic cotunneling, respectively. The failure of mean-field models, providing a fictitious bistability is addressed in Sec. V as well.

II. THE SECOND ORDER VON NEUMANN APPROACH

Using $H_D$ in its diagonal representation [4], the tunneling between the states in the leads and the dot reads (see Appendix A of [22])

$$H_T = \sum_{k\sigma, ab} T_{ba}(k\sigma\ell)|b\rangle \langle a| c^\dagger_{k\sigma\ell} + \text{h.c.}$$

(7)

The matrix element $T_{ba}(k\sigma\ell)$ is the scattering amplitude for an electron in the state $k\sigma\ell$ tunneling from the lead onto the dot, thereby changing the dot state from the state $|a\rangle$ to the state $|b\rangle$. Note that this amplitude vanishes unless the number of electrons in state $|b\rangle$, $N_b$, equals $N_a + 1$. Here we use the convention that the particle number increases with the positon in the alphabet of the denoting letter.

A general state vector for the entire system is written as $|ag\rangle = |a\rangle \otimes |g\rangle$, with $|g\rangle = |\{N_{k\sigma\ell}\rangle$ denoting the state of both leads, where $N_{k\sigma\ell} \in \{0, 1\}$. To ensure the anti-commutator rules of the operators we use the following notation $|g - k\sigma\ell\rangle \equiv |c_{k\sigma\ell};g\rangle$ and $|g + k\sigma\ell\rangle \equiv |c^\dagger_{k\sigma\ell};g\rangle$. The order of indices is opposite to the order of the operators. E.g. $|g - k'\sigma'\ell' + k\sigma\ell\rangle = c^\dagger_{k\sigma\ell} c^\dagger_{k'\sigma'\ell'} |g\rangle = -c_{k'\sigma'\ell'} c^\dagger_{k\sigma\ell} |g\rangle = -|g + k\sigma\ell - k'\sigma'\ell'\rangle$, taking into account the anti-commutation rules of the operators. To simplify the notation, $\sigma\ell$ is only attached to $k$ the first time the index $k$ appears in the equation, and in the following it is implicitly assumed to be connected with $k$. Furthermore, we apply the convention that $\sum_{k\sigma\ell}$ means summing over $k$ and $\sigma$ with a fixed $\ell$ being connected to $k$ in this sum.

The density matrix elements are defined as

$$\rho^{[n]}_{ag, bg'} = \langle ag|\rho|bg'\rangle$$

(8)

where the label $n$ provides the number of electron or hole excitations needed to transform $g$ into $g'$. Examples are $\rho^{[1]}_{bg - k;ag}$ and $\rho^{[2]}_{cg - k;ag + k'}$. We denote the elements as $n$-chix elements in the following.
The von Neumann equation (11) gives equations of motion of the type

\[ i\hbar \frac{d}{dt} \rho_{bg;b'g}^{[0]} = (E_b - E_{b'}) \rho_{bg;b'g}^{[0]} + \sum_{a,\kappa\ell} T_{ba}(k) \rho_{ag+k;b'g}^{[1]} + \sum_{c,\kappa\ell} T_{cb}(k) \rho_{cg-k;b'g}^{[1]} - \sum_{c,\kappa\ell} \rho_{bg;c \ell}^{[1]} T_{cb}(k) - \sum_{a,\kappa\ell} \rho_{bg;ag+k}^{[1]} T_{ba}^{*}(k), \]

and

\[ i\hbar \frac{d}{dt} \rho_{cg-k;\ell}^{[1]} = (E_c - E_{c'}) \rho_{cg-k;\ell}^{[1]} + \sum_{b'} T_{cb'}(k) \rho_{tg-k;bg}^{[0]} - \sum_{c'} \rho_{cg-k;\ell}^{[2]} T_{cb'}(k) - \sum_{a} \rho_{cg-k;ag+k}^{[2]} T_{ba}^{*}(k'). \]

(9)

(10)

Thus the \( \Phi_{ba}^{[1]}(k\ell) \) terms describe current amplitudes. Note that all 1-ehx terms can be described by \( \Phi_{ba}^{[1]}(k\ell) \) as \( \rho_{ag+k;bg}^{[1]} = (\rho_{bg-k;ag})^* \) and \( \sum_{g} \rho_{ag+k;bg}^{[1]} = \sum_{g'} \rho_{ag+k';bg'}^{[1]} - k' \).

Performing the sum \( \sum_{g} \) to the time evolution (10) some terms do not directly obtain the form \( \Phi_{ba}^{[0]} \). Here we approximate

\[ \sum_{g} \delta_{N_k1} \rho_{bg;bg}^{[0]} \approx f_k \sum_{g} \rho_{bg;bg}^{[0]} = f_k \Phi_{b,b}^{[0]}, \]

\[ \sum_{g} \rho_{ag+k;bg}^{[0]} = \sum_{g'} \delta_{N_k0} \rho_{ag+k';g'}^{[0]} \approx (1 - f_k) \Phi_{c,c}^{[0]}. \]

Similar approximations are done for the \( \rho_{bg-k;ag}^{[1]} \) elements appearing in the equation for the 2-ehx terms. This approximation is a factorization of the lead occupations, which are assumed not to be affected by the transition processes. The result is a closed set of differential equations for the reduced density matrix \( \Phi_{b,b}^{[0]} \), the current elements \( \Phi_{ba}^{[1]}(k\ell) \) and the similarly defined 2-ehx terms \( \Phi_{b,c}^{[2]}(-k\ell + k'\ell';0), \Phi_{c,c}^{[2]}(-k\ell - k'\ell';0) \).

Defining a discrete set of \( k \)-states, one can set up a column vector consisting of all the elements of the density-matrix \( \Phi = (\Phi_{[0]} \Phi_{[1]} \Phi_{[2]}) \), where the sub-vectors contain all the elements of the density-matrix with a specific \( n \)-value, as well as the complex conjugates of the complex elements. The equation of motion for the vector \( \Phi \) can be cast on a matrix form

\[ i\hbar \frac{d}{dt} \Phi = \begin{pmatrix} E_{00} & M_{01} & 0 \\ M_{10} & E_{11} & M_{12} \\ 0 & M_{21} & E_{22} \end{pmatrix} \Phi = M \Phi. \]

(14)

The submatrices \( E_{nn} \) are diagonal and contain the energy differences between the states involved.

Now we consider the \( \Phi_{[2]} \) terms in a stationary approximation, yielding

\[ \Phi_{[2]} = (-E_{22} + i0^+)^{-1} M_{21} \Phi_{[1]}, \]

where the \( i0^+ \) ensures causality, corresponding to the Markov limit for the highest-order elements.

Inserting the result into Eq. (14) leads to the matrix equation

\[ i\hbar \frac{d}{dt} \Phi_{[0]} = \begin{pmatrix} E_{00} & M_{01} & 0 \\ M_{10} & E_{11} & M_{12} \\ 0 & M_{21} & E_{22} \end{pmatrix} \Phi_{[0]} \]

(15)

where \( M_{11} = E_{11} + M_{12} (-E_{22} + i0^+)^{-1} M_{21} \) is not diagonal. Due to the \( k \)-dependence it is very laborious to express \( \Phi_{[1]} \) solely in terms of \( \Phi_{[0]} \) thereby reducing the problem to a generalized master equation. The explicit, and completely general, expressions for the equation of motion for \( \Phi_{[0]} \) and \( \Phi_{[1]} \) are given in Eqs. (10,11) of [22] and is the main result of that paper. [39]

The sub-matrices \( M_{10} \) and \( M_{01} \) only contain elements proportional to the tunneling amplitude \( T_{ba} \), while the matrix \( M_{11} \) involves terms proportional to \( T_{ba}^{*} \). Thus the
stationary solution of Eq. (15), together with the normalization \( \sum_{n} \Phi_{bb}^{[0]} = 1 \), will contain all powers of \( T_{11} \), and so will the stationary occupations and coherences. That is, the approach does not provide a systematic expansion in powers of the tunneling coupling. For the current, which is proportional to \( T_{11} \Phi^{[1]} \), all terms up to order \( T_{11} \) as well as a class of higher-order terms are taken into account.

Summarizing, the 2vN approach considers the reduced density matrix (11) and the current amplitudes (12) as variables, which are determined by the closed set of dynamical equations (15). In deriving these, three approximations are applied: (i) only coherent processes involving transitions of at most two different \( k \)-states are considered, i.e. all \( n \)-ehx terms with \( n \geq 3 \) are assumed to vanish, (ii) the time-dependence of terms generating 2-ehx terms is neglected which corresponds to the Markov limit, (iii) the level occupations in the leads, \( f_{k\sigma} \), is unaffected by the couplings to the dot, i.e. the densities in the leads and on the dot can be factorized.

### III. RANGE OF VALIDITY AND COMPARISON WITH OTHER METHODS

For noninteracting systems, Eq. (3) gives the correct result for arbitrary temperature and bias. In [22] we demonstrated analytically that using the 2vN approach, Eq. (3) is fully recovered for a single-level system. Numerically, we also found full agreement for all ranges of parameters, including double dots [23] and the ferromagnetic Anderson model with an applied magnetic field [24], where coherences are of central importance. Thus we have strong indications that the 2vN method is able to treat transport correctly for noninteracting systems over the full temperature and bias ranges.

For interacting systems and temperatures \( k_{B}T \gg \Gamma \) or in the high bias limit, the 2vN reproduces the well-known results of the methods presented in [22][22]. This demonstrates the correct treatment of charging effects in interacting systems.

For the Anderson model with infinite Coulomb repulsion, the 2vN equations could be solved analytically, see [22], and the result agree with the diagrammatic real-time transport theory in the resonant tunneling approximation [22], where the onset of Kondo physics is observed (see also Eq. (2) of [24]). However, in the Kondo limit itself, the model misses the unitary limit and unphysical results are found, as the strong correlations between the dot and the leads are not properly reflected.

The validity of the 2vN approach for time-dependent problems has not been carefully investigated. As the Markov approximation is invoked, it might not be valid for strongly time-dependent systems, where non-Markovian effects are important due to memory effects, which are also relevant when evaluating higher-order moments, as e.g., the noise [28][29]. As an example, the current through a single spinless level was presented in [22] which does not exhibit the oscillations found from a time-dependent Green function approach [30].

In a recent paper, Jin et al. also consider quantum transport in the same spirit as in the 2vN approach by keeping correlations between the leads and the dot and performing an expansion in the tunneling Hamiltonian [31]. They report a proof that they obtain the 2vN approach as a second-order expansion.

### IV. ELASTIC COTUNNELING

As an example we apply the 2vN method in the elastic cotunneling regime for a two-level spinless system. We show that the 2vN results agree with a mean-field solution and with a scattering result.

The system is described by the Hamiltonian

\[
H = \sum_{k,\ell=L,R} E_{k\ell} c_{k\ell}^\dagger c_{k\ell} + \sum_{kmn} V_{kmn} c_{k+}^\dagger c_{m-} d_n + h.c.
\]

\[
+ \sum_{n} E_n d_n^\dagger d_n + U d_1^\dagger d_1 d_2^\dagger d_2,
\]

with \( n = 1, 2 \) denoting the two dot states. The first term is the Hamiltonian of the leads, and the last two terms are the two single-particle states of the dot and the interaction between them. The second term is the tunnel Hamiltonian with the tunneling amplitudes \( V_{kmn} \). Below it is assumed that \( V_{kmn} = x_{kn} t_k \), i.e. the couplings between both dot states and the lead states have a fixed phase factor \( x_{kn} \), and \( t_k \) is assumed to be a real number. The coupling constants are defined as \( \Gamma_{kn}(E) = 2\pi \sum_k |V_{kmn}|^2 \delta(E - E_{kmn}) = |x_{kn}|^2 \Gamma(E) \). For the 2vN calculations a constant value \( \Gamma \) for \( |E| \leq 0.95W \) is used, and it is assumed that \( \Gamma(E) = 0 \) for \( |E| > W \). For \( 0.95W < |E| < W \) an elliptic interpolation is applied.

Using the Hamiltonian above, the system can be in four different many-particle states, denoted \( |0\rangle, |1\rangle, |2\rangle, |d\rangle = d_1^\dagger d_2^\dagger |0\rangle \), with energies \( 0, E_1, E_2, E_d \), respectively.

The transport is calculated in a setup with \( E_1 \ll \mu_e \) and \( E_2 + U \gg \mu_e \) such that the state \( |1\rangle \) is the ground state. Sequential tunneling processes are blocked due to the Coulomb interaction between the electrons, but a leakage current due to elastic cotunneling processes can occur. Here, the 2vN results are compared with a mean-field solution embedded in a nonequilibrium Green function framework, and a scattering formalism.

In the mean-field solution, the interaction term in the Hamiltonian is replaced with

\[
U d_1^\dagger d_1 d_2^\dagger d_2 \rightarrow U \left\{ \left[ d_1^\dagger d_1 \left( d_2^\dagger d_2 + d_2^\dagger d_2 \right) \right] - \left[ d_1^\dagger d_2 \left( d_2^\dagger d_1 + d_1^\dagger d_1 \right) \right] \right\},
\]

where the first \([...]-\)bracket is the Hartree term and the second the Fock term. The occupations are calculated self-consistently whereafter the current can be evaluated [41].
For the scattering result, the elastic second-order scattering rate, \( \gamma_{11}^{RL} \), is the sum over all processes where an electron labelled \( k' \) has been transferred from the left contact to the state \( kR \) in the right contact, leaving the dot state unchanged. This can be realized in two different ways, \( |1 \rangle \rightarrow |0 \rangle \rightarrow |1 \rangle \) and \( |1 \rangle \rightarrow |d \rangle \rightarrow |1 \rangle \), where the amplitudes for these two processes are added coherently.

The rate is calculated as follows \([32]\): Assume that initially the dot is in state \( |1 \rangle \) and the leads are in a state \( |\nu_L \nu_R \rangle \), i.e. the final state is \( |\nu_L \nu_R \rangle \). The probability for the leads to be in the state \( |\nu_L \nu_R \rangle \) is given by \( W_{\nu_L \nu_R} = W_{\nu_L} W_{\nu_R} \), as the leads are assumed uncorrelated. In the final state an electron \( k' \) has been transferred from left to right ending up in the state \( kR \), i.e. the final state is \( |f(k'k') = c_{kR} \epsilon_{k'k} \rangle \) with energy \( E_i - E_{k'k} + \epsilon_{kR} \). According to the \( T \)-matrix formalism the second-order scattering rates are

\[
\gamma_{11}^{RL} = 2\pi \sum_{kk'} \sum_{\nu_L \nu_R} W_{\nu_L \nu_R} \left\langle f_{kk'}^{R} \left| \frac{1}{E_i - H_0} \right. \right| H_T \left| k \right. \rangle \delta(E_f - E_i),
\]

and after some algebra we arrive at

\[
\gamma_{11}^{RL} = \frac{1}{2\pi} \int dE \left| \sum_{kk'} \frac{x_{kk'}^{R} x_{kk'}^{L}}{E_1 - E + \frac{E_2 + U - E}{2} \times n_F (E - \mu_L) [1 - n_F (E - \mu_R)]} \right|^2, \tag{19}
\]

where \( n_F(E) = \left[ 1 + e^{E/k_B T} \right]^{-1} \) is the Fermi function, and energy-independent coupling constants are assumed (the Wide-Band Limit). The rate \( \gamma_{11}^{RL} \) is found by interchanging \( L \leftrightarrow R \). Below we only calculate the integral for \( k_B T = 0 \), but note that for finite temperatures the integral diverges and a regularization procedure is needed \([33]\).

For \( k_B T = 0, \mu_R = eV_{bias} > 0 \) and \( \mu_R = 0 \), we obtain

\[
\gamma_{11}^{RL} = \frac{1}{2\pi} \int_0^{eV_{bias}} \int dE \left| \sum_{kk'} \frac{x_{kk'}^{R} x_{kk'}^{L}}{E_1 - E + \frac{E_2 + U - E}{2}} \right|^2, \tag{20}
\]

and \( \gamma_{11}^{LR} = 0 \). Assuming the state \( |1 \rangle \) to be almost completely occupied (i.e. \( |E_1| / \Gamma \ll 1 \) and \( (E_2 + U) / \Gamma \gg 1 \)), the second-order elastic cotunneling current is \( I_{el.cotun}^{\Gamma} = \frac{1}{\pi} \gamma_{11}^{RL} \).

In Fig. 2 the current versus bias voltage is calculated in the elastic cotunneling regime using the 2\( v \)N method, the mean-field approximation in a Green function framework, and the second-order scattering method. For the latter, the calculation is for vanishing temperature, while the other calculations are for \( k_B T = \Gamma / 10 \). For \( E_1 = -3\Gamma \) almost perfect agreement between all three methods is found, while deviations between the scattering method and the others occur for \( E_1 = -2\Gamma \), which is most likely due to the fact that the state \( |1 \rangle \) is not fully occupied in this case as assumed in the derivation of the second-order scattering rate \([19]\).

In summary, from a comparison with both a mean-field solution and a scattering formalism, we have shown that the 2\( v \)N method is able to quantitatively describe elastic cotunneling processes even for temperatures much lower than the energy scale set by the coupling to leads, \( k_B T \ll \Gamma \).

\section{V. INELASTIC COTUNNELING}

For higher bias voltages the agreement between the 2\( v \)N approach and the mean-field solution is assumed to be less perfect, as the latter can lead to bistable solutions \([34]\), which are not present in a generalized master equation approach. Fig. 3 shows a comparison between the 2\( v \)N approach and the mean-field solution over the full bias range for \( E_1 = -2\Gamma \), with the rest of the parameters as in Fig. 2.

Considering first the 2\( v \)N result, the curve shows increased current when the bias matches the energy difference between the levels, \( eV_{bias} = |E_1 - E_2| = 18\Gamma \). This is due to the onset of inelastic cotunneling \([32]\), which leads to a population of the excited state, \( |2 \rangle \). After the inelastic cotunneling process, additional cotunneling-assisted sequential tunneling processes can occur \([35,36]\). Finally, at \( eV_{bias} = E_2 + U = 36\Gamma \) sequential tunneling through the upper level becomes possible. The value for the current is consistent with the master equation result \((3\Gamma / 8\hbar)\) plus additional cotunneling through the lower level.
In contrast, the mean-field solution misses the onset of inelastic cotunneling and instead a bistable behaviour is observed, and it also overestimates the current after the onset of sequential tunneling. Both is due to an insufficient description of the Coulomb interaction and emphasizes the need for a method which can describe both higher-order tunneling processes (as elastic and inelastic cotunneling) and many-particle interactions.

VI. SUMMARY

The second order von Neumann approach provides a quantitative description of transport through nanostructures for arbitrary bias and temperatures above the Kondo temperature. In the low-bias regime, the elastic cotunneling current is in very good agreement with both the second-order scattering result and the mean-field solution, even for temperatures much lower than the energy scale set by the coupling to leads. Inelastic cotunneling is also well captured by the 2vN approach, whereas a mean-field solution within a nonequilibrium Green functions framework shows an artificial bistability in this regime.


[37] A finite lifetime of the lead states has recently been addressed in [8].

[38] A higher-order expansion can be done within the diagrammatic real-time transport theory [13].

[39] In [22], $\Phi^{0}_{\beta / \beta}$ and $\Phi^{1}_{\alpha \alpha}(k \sigma \ell)$ are denoted $w_{\beta / \beta}$ and $\phi_{\alpha \alpha}(k \sigma \ell)$, respectively.

[40] For practical reasons only two-level systems have been studied so far.

[41] The calculations are analogous to those presented in [24].