Time-dependent current density functional theory for the linear response of weakly disordered systems

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Time-dependent density functional theory (TDFT) provides a way of calculating, in principle exactly, the linear response of interacting many-electron systems, and thus allows one to obtain their excitation energies. For extended systems, there exist excitations of a collective nature, such as bulk- and surface plasmons in metals or intersubband plasmons in doped semiconductor quantum wells. This paper develops a quantitatively accurate first-principles description for the frequency and the linewidth of such excitations in inhomogeneous weakly disordered systems. A finite linewidth in general has intrinsic and extrinsic sources. At low temperatures and outside the region where electron-phonon interaction occurs, the only intrinsic damping mechanism is provided by electron-electron interaction. This kind of intrinsic damping can be described within TDFT, but one needs to go beyond the adiabatic approximation and include retardation effects. It has been shown [G. Vignale, C. A. Ullrich, and S. Conti, Phys. Rev. Lett. 79, 4878 (1997)] that a density-functional response theory that is local in space but nonlocal in time has to be constructed in terms of the currents, rather than the density. This theory will be reviewed in the first part of this paper. For quantitatively accurate linewidths, extrinsic dissipation mechanisms, such as impurities or disorder, have to be included in the response theory. In the second part of this paper, we discuss how extrinsic dissipation can be described within the so-called memory function formalism. This formalism will first be introduced and reviewed for homogeneous systems. We will then present a synthesis of TDFT with the memory function formalism for inhomogeneous systems, which allows one to account simultaneously for intrinsic and extrinsic damping of collective excitations. As an example where both sources of dissipation are important and where high-quality experimental data is available for comparison, we discuss intersubband plasmons in a 40 nm wide GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum well.

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I. INTRODUCTION

The calculation of excitation energies and linewidths of collective excitations in extended electronic systems is one of the outstanding problems in many-body theory. Time-dependent density functional theory (TDFT) offers a powerful and elegant approach to this difficult problem. To set the stage for the developments that are to follow, we shall begin this paper with a summary of the key elements of TDFT. Let

\[ \hat{H}_0 = \sum_i \left\{ \frac{\mathbf{p}_i^2}{2m} + v_0(\mathbf{r}_i) \right\} + \frac{1}{2} \sum_{i \neq j} U(|\mathbf{r}_i - \mathbf{r}_j|) \]

be the Hamiltonian of a many-electron system, where \( \mathbf{r}_i \) and \( \mathbf{p}_i \) are the canonical coordinates and momenta of the \( i \)th electron, \( m \) is its mass, \( v_0(\mathbf{r}) \) is a static external potential, which includes contributions from randomly distributed impurities and other sources of disorder, and \( U(|\mathbf{r}_i - \mathbf{r}_j|) \) is the Coulomb interaction potential. To calculate the excitation energies, one adds to \( \hat{H}_0 \) a small time-dependent perturbation of the form

\[ \hat{H}_1(t) = \int d^3r \, v_1(\mathbf{r}, t) \hat{n}(\mathbf{r}) , \]

where \( v_1(\mathbf{r}, t) = v_1(\mathbf{r}, \omega)e^{-i\omega t} + c.c. \) is a periodic potential \( (v_1 \ll v_0) \) that couples linearly to the density operator \( \hat{n}(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i) \). One then computes the time-dependent density of the system, which, in the linear approximation, will be given by

\[ n(\mathbf{r}, t) = n_0(\mathbf{r}) + n_1(\mathbf{r}, \omega)e^{-i\omega t} + c.c. , \]

where \( n_0(\mathbf{r}) \) is the ground-state density, and \( n_1(\mathbf{r}, \omega) \) is linearly related to \( v_1(\mathbf{r}, \omega) \) via

\[ n_1(\mathbf{r}, \omega) = \int d^3r \, \chi(\mathbf{r}, \mathbf{r}', \omega)v_1(\mathbf{r}', \omega) . \]

The density-density response function \( \chi(\mathbf{r}, \mathbf{r}', \omega) \) contains the essential information about those excited states of the system that are coupled to the ground state by the perturbation \( \hat{H}_1(t) \). More specifically, in a finite system (atom or molecule) this response function has a discrete set of poles on the real frequency axis, corresponding to the discrete excitation energies of the system. In an extended system, the poles merge into a continuous branch cut along the real axis. However, isolated poles can arise in the lower half of the complex frequency plane: they correspond to collective excitations of the system, where the imaginary part of the frequency defines the characteristic lifetime of the excitation.
The basic TDFT strategy for calculating $\chi(r, r', \omega)$ is to construct a noninteracting system that has the same ground-state density $n_0(r)$, and yields the same density response $n_1(r, \omega)$ as the interacting system under study. The dynamics of this noninteracting system is controlled by an effective single-particle potential which is written as the sum of the total external potential $v_0(r) + v_1(r, t)$ plus the Hartree potential

$$v_H(r, t) = e^2 \int d^3r' \frac{n(r', t)}{|r - r'|}$$

(5)

plus a remainder, which is known as the "exchange-correlation" (xc) potential $v_{xc}(r, t)$. It is not at all obvious that such a potential $v_{xc}$ can be constructed, but, if it can, then Runge and Gross showed that it is a unique functional of the time-dependent density up to within an additive function of time. The form of the xc potential depends, in general, on the initial state of the system, but this dependence disappears if one assumes, as we do here, that the system is initially in its ground state.\footnote{The effective noninteracting Hamiltonian (also known as the Kohn-Sham Hamiltonian) that yields the exact density is then given by \( \hat{H}_{KS}(t) = \sum_i \left\{ \frac{p_i^2}{2m} + v_0(r_i) + v_{H,0}(r_i) + v_{xc,0}(r_i) \right\} + \int d^3r \left[ v_1(r, t) + v_{H,1}(r, t) + v_{xc,1}(r, t) \right] n(r) \), \( \rightarrow \) (6)

where both the Hartree and the xc potential have been written as the sum of static parts $v_{H,0}, v_{xc,0}$ associated with the ground-state density, and (small) time-dependent parts $v_{H,1}, v_{xc,1}$ associated with the time-dependent density. The static part of the Kohn-Sham Hamiltonian [first line of Eq. \( \rightarrow \)] yields the exact ground-state density in the interacting system, while the time-dependent part [the second line of Eq. \( \rightarrow \)] yields the exact density response.

In the linear response regime the xc potential can be written as

$$v_{xc}(r, t) = v_{xc,0}(r) + \int_{-\infty}^{t} dt' \int d^3r' f_{xc}(r, r', t-t')n_1(r', t'),$$

(7)

where $v_{xc,0}(r)$ depends only on the ground-state density, and $f_{xc}(r, r', t-t')$ is the retarded xc kernel, formally defined as

$$f_{xc}(r, r', t-t') = \frac{\delta v_{xc}[n(r, t)]}{\delta n(r', t')} \bigg|_{n_0(r)} .$$

(8)

Fourier transformation of both $v_{xc}$ and $f_{xc}$ with respect to time leads to the simpler relation

$$v_{xc,1}(r, \omega) = \int d^3r' f_{xc}(r, r', \omega)n_1(r', \omega) .$$

(9)

We denote by $\chi_{KS}(r, r', \omega)$ the density-density response function of the static Kohn-Sham system [the first line of Eq. \( \rightarrow \)]. The second line of the same equation can then be regarded as a time-dependent perturbation to the first. The time-dependent density response is therefore given by

$$n_1(r, \omega) = \int d^3r' \chi_{KS}(r, r', \omega) \left\{ v_1(r', \omega) + \int d^3r'' \left[ \frac{e^2}{|r' - r''|^2} + f_{xc}(r'', \omega) \right] n_1(r'', \omega) \right\},$$

(10)

where the second and the third terms in the curly brackets arise from $v_{H,1}$ and $v_{xc,1}$, respectively. Comparing Eqs. \( \rightarrow \) and \( \rightarrow \) we obtain the following integral relation between the exact density-density response function $\chi(r, r', \omega)$ and the noninteracting response function $\chi_{KS}(r, r', \omega)$:

$$\chi(r, r', \omega) = \chi_{KS}(r, r', \omega) + \int d^3x \chi_{KS}(r, x, \omega) \int d^3y \frac{e^2}{|x - y|^2} + f_{xc}(x, y, \omega) \right\} \chi(y, r', \omega) .$$

(11)

This can also be written as

$$\chi^{-1}(r, r', \omega) = \chi_{KS}^{-1}(r, r', \omega) - \frac{e^2}{|r - r'|} - f_{xc}(r, r', \omega),$$

(12)

where $\chi^{-1}$ is the matrix inverse of $\chi$.

The excitation energies are finally obtained from the poles of the linear response function, i.e., from the solution of the eigenvalue problem

$$\int d^3r' \chi^{-1}(r, r', \omega) \Xi(r', \omega) = 0 ,$$

(13)

where $\Xi(r, \omega)$ is the function that describes the spatial dependence of the density in the excited state.
assumption), the xc kernel in Eq. (1) is expected to be a small correction, which can be approximated with relative impunity. The simplest approximation is to ignore both nonlocality in space and retardation in time. This leads to the widely used adiabatic local density approximation (ALDA) in which one poses

\[ v_{xc}(r, t) = \left( \frac{d\epsilon_{xc}(n)}{dn} \right)_{n=n(r, t)}, \tag{14} \]

where \( \epsilon_{xc}(n) \) is the xc energy density of the homogeneous electron gas of density \( n \). The right-hand side of Eq. (14) is nothing but the local density approximation for the ground-state xc potential evaluated at the time-dependent density. In terms of the xc kernel, this approximation implies

\[ f_{xc}(r, r', \omega) = \frac{d^2\epsilon_{xc}(n)}{dn^2} \delta(r - r'), \tag{15} \]

which is a purely real and frequency-independent object.

The ALDA is a remarkably successful approximation, despite the fact that it entirely neglects the frequency dependence of the xc kernel, that is, the retarded dependence of the xc potential on the density at earlier times. In atoms the ALDA has yielded reasonably accurate values of the excitation energies. Most of the residual inaccuracy has been traced to the fact that the ground-state xc potential in the LDA fails at large distance from the nucleus. An optimized effective potential approach is similar in spirit to Eq. (14), but still without retardation, yields a dramatic improvement in accuracy. Applications to more complex systems (molecules, polymers) have met with similar degree of success. The essential reason seems to be that the frequency dependence of the xc kernel is rather weak, because it is controlled by multi-electron excitations, which are either very high in energy (atoms and molecules), or smoothly distributed through a spectral range (extended systems).

There are, however, some important features of the dynamical response that cannot be accounted for in any way by an instantaneous xc potential. Quite generally, the need for a dynamical theory of \( f_{xc} \) arises in the study of excitations that do not have an analogue in the Kohn-Sham system. Perhaps the clearest example of this is provided by collective excitations in extended electronic systems, such as bulk- and surface plasmons in metals or inter- and intrasubband plasmons in doped semiconductor quantum wells. In this case, the ALDA would predict resonance peaks of vanishing width, in glaring contradiction to experiment.

Attempts to go beyond the ALDA to include retardation date back to the mid-eighties. In 1985 Gross and Kohn proposed a dynamical local density approximation for \( f_{xc} \) which was designed to preserve the local relationship between \( v_{xc,1} \) and the density, while including retardation in time. Their approximation reads

\[ f_{xc}(r, r', \omega) = f_{xc}^{h}(k = 0, \omega) \delta(r - r'), \tag{16} \]

where \( f_{xc}^{h}(k, \omega) \) is the xc kernel of the uniform electron gas calculated at the local ground-state density \( n_0(r) \) (more about \( f_{xc}^{h} \) will be said in the next sections). Because \( f_{xc}(\omega) \) is complex, this approximation yields a finite linewidth for excitations that would have zero linewidth in the ALDA.

Unfortunately, the Gross-Kohn approximation suffers from several inconsistencies, such as the failure to satisfy the generalized Kohn’s theorem and related sum rules. As a consequence, it was found that within this approximation, intersubband plasmons in quantum wells may become substantially overdamped. These deficiencies were ultimately traced back to the fact that a local approximation for the dynamical xc potential in terms of the density does not exist (except at \( \omega = 0 \), in which case it is the static LDA). The reason for this startling result is that the xc kernel of a non-homogeneous system is a function of infinite range in space, or, more precisely, the spatial Fourier transform \( f_{xc}(k, k', \omega) \) diverges when \( k \to 0 \) at constant \( k' \) or vice versa. (In the homogeneous case one has \( k = k' \) and the singularity disappears.)

Vignale and Kohn (VK) and Vignale, Ullrich and Conti (VUC) showed that the nonlocality problem could be circumvented by working with the current density rather than the density as a basic variable. The idea is to perturb the system with a time-dependent vector potential \( a_1(r, t) = a_1(r, \omega) e^{-i\omega t} + c.c. \) rather than with a scalar potential. The perturbing Hamiltonian has the form

\[ \hat{H}_1(t) = \int d^3r \mathbf{a}_1(r, t) \cdot \hat{\mathbf{p}}_r(r), \tag{17} \]

where \( \hat{\mathbf{p}}_r(r) = -\frac{i}{2m} \sum_i [\hat{\mathbf{p}}_r \delta(r - r_i) + \delta(r - r_i) \hat{\mathbf{p}}_r] \) is the paramagnetic current density operator. One then calculates the current response, and determines the excitation energies from the poles of the current-current response function.

The Kohn-Sham Hamiltonian in this time-dependent current density functional theory (TCDFT) contains an xc vector potential \( a_{xc,1}(r, t) \), which is a (linear) functional of the full current density response \( j_1(r, t) = \mathbf{J}_1(r, t) + n_0(r) \mathbf{a}_1(r, t)/m \):

\[ a_{xc,1,\alpha}(r, \omega) = \sum_\beta \int d^3r' f_{xc,\alpha\beta}(r, r', \omega) j_{1,\beta}(r', \omega), \tag{18} \]

where \( f_{xc,\alpha\beta}(r, r', \omega) \) is the tensorial generalization of the usual xc kernel (here and in the following, \( \alpha, \beta \) denote Cartesian components). The static part of the Kohn-Sham Hamiltonian remains unchanged, and the ground-state density is still determined by the static xc field \( v_{xc,0} \).

It turns out that the xc vector potential \( \mathbf{a}_{xc,1} \) is local in space, retarded in time, satisfies the generalized Kohn’s
remarkable here is that the nonlocality of density necessarily be a nonlocal functional of the density. What is one sees that the longitudinal component of the current is undetermined. Thus, a local functional of density can be completely eliminated by “up-grading” TDF to a description in terms of \( a_{\text{xc,1}} \) and \( j_1 \). This is the essence of the VK and VUC theory.

Since any scalar potential can be represented by an equivalent vector potential, and since the density is easily calculated from the current [see Eq. (19)], we see that the ordinary TDF is a special case of the TCDFT formulation. An additional advantage of this formulation is that it allows one to treat the more general problem of the response of an electronic system to an electromagnetic field having both longitudinal and transverse components, whereas the original Runge-Gross formulation is limited to longitudinal fields, i.e., fields that can be expressed as the gradient of a scalar potential.

Although the VK and VUC formulations are important steps enabling the calculation of the linewidth of elementary excitations in extended systems, they are still not sufficient to achieve quantitative accuracy in cases of practical interest. For example, the calculation of the linewidth of the intersubband (ISB) plasmon in a 40-nm GaAs/Al\(_{0.33}\)Ga\(_{0.7}\)As quantum well reported in Ref. 23, based on VUC formalism, yielded a linewidth about 5 times smaller than the experimental value. The reason for this disappointing result is that the theory, as it stands, does not take into account other intrinsic and extrinsic sources of damping, such as electron-phonon interactions, electron-impurity scattering, and, in the case of quantum wells, interfacial roughness. All these interactions contribute to the linewidth and must be included in any calculation that aspires to quantitative accuracy.

In this paper we take a first step in this direction by showing how two of the most prominent contributions to the low-temperature linewidth of plasmons in quantum wells, namely electron-impurity scattering and interfacial roughness, can be built into the current-density functional formalism.

Our approach is based on the “marriage” of the TCDFT formalism with the memory function formalism described, for example, by Forster. In the homogeneous electron gas limit this approach reduces to the Belitz-Das Sarma treatment of the effect of impurities on bulk plasmons, which, in turn, can be viewed as the high-frequency extension of the Mermin relaxation-time approximation for the density-density response function of an electron gas in the presence of randomly distributed impurities. Of course, our interest lies in strongly inhomogeneous systems, such as quantum wells, which exhibit the intersubband plasmon resonance. Such resonances are of practical interest in connection with the design of infrared detector devices.

Our strategy is to derive an integral equation which relates the current response function of the disordered interacting many-electron system to that of the same system in the absence of disorder: the latter is calculated by the standard TCDFT outlined above. We shall show that this approach (despite some inevitable approximations in the treatment of disorder) meets with considerable success: the linewidth of the intersubband plasmon is considerably enhanced by disorder – in particular, by interfacial roughness – and agrees quantitatively with the measured one. More importantly, the qualitative behavior of the linewidth as a function of an external electric field that controls the shape of the quantum well is correctly reproduced.

The remainder of this paper is organized as follows: In Section II we review the main aspects of the TCDFT formalism for the linear response of many-electron systems. In Section III we review the memory function formalism and demonstrate its application to the case of a homogeneous electron gas with randomly distributed impurities. These sections are meant to make the paper self-contained and to provide the necessary background for the following more technical parts. In Section IV we combine the memory function formalism with TCDFT for inhomogeneous systems and derive the key integral equations for the current-current response functions. Finally, in Section V we demonstrate the power of the method by calculating the linewidth of the intersubband plasmon in a quantum well and comparing to recent experimental results, and in Section VI we give our conclusions.

II. TCDFT BEYOND THE ADIABATIC LDA

A. Exchange-correlation kernels in the homogeneous electron gas

For orientation, let us first consider the \( \text{xc} \) kernels of a homogeneous electron gas. Because of translational invariance, it is convenient to work with the Fourier transform \( j(k, \omega) \) of the current density. The linear response of this quantity to a vector potential \( a_1(k, \omega) \) can be written as

\[
j_{1,\alpha}(k, \omega) = \sum_\beta \chi_{\alpha,\beta}(k, \omega) a_{1,\beta}(k, \omega),
\]

where \( \chi_{\alpha,\beta}(k, \omega) \) is the current-current response tensor. Due to rotational invariance, the responses of the longi-
tudinal (parallel to k) and transverse (perpendicular to k) components of the current are completely independent, and one can write

$$j_{1,L(T)}(k,\omega) = \chi_{L(T)}(k,\omega)a_{1,L(T)}(k,\omega),$$  \hspace{1cm} (22)

where \(L(T)\) denotes the longitudinal (transverse) component, and \(\chi_{L(T)}(k,\omega)\) is the longitudinal (transverse) response function. According to the general linear response formalism, \(\chi_{L(T)}\) is given by

$$\chi_{L(T)}(k,\omega) = \frac{n}{m} + \sum_{l} \frac{1}{\omega - \omega_{l} + i\eta} \left( 1 - \frac{1}{\omega + \omega_{l} + i\eta} \right),$$  \hspace{1cm} (23)

where \(|0\rangle\) is the ground state, \(|l\rangle\) is the \(l\)-th excited state, and \(\omega_{l}\) is the excitation energy \(E_{l} - E_{0}\). The Fourier transform of the paramagnetic current operator \(\hat{j}_{p}(k)\) is given by

$$\hat{j}_{p}(k) = \frac{1}{2m} \sum_{i} [\hat{p}_{i} e^{-i\mathbf{k}\cdot\mathbf{r}_{i}} + e^{-i\mathbf{k}\cdot\mathbf{r}_{i}} \hat{p}_{i}] .$$  \hspace{1cm} (24)

Note that the total current response is the sum of the “London current” \(na_{1}/m\) and the paramagnetic current [the expectation value of (23)]. A key feature of Eq. (23) is that in the limit of \(k \to 0\) and finite \(\omega\) it has the limiting form

$$\chi_{L}(k,\omega) \to \frac{n}{m\epsilon(\omega)} + \alpha_{L}(\omega)k^{2},$$

$$\chi_{T}(k,\omega) \to \frac{n}{m} + \alpha_{T}(\omega)k^{2},$$  \hspace{1cm} (25)

where \(\alpha_{L(T)}(\omega)\) are functions of frequency only, \(\epsilon(\omega) = 1 - \lim_{k \to 0} n\nu(k)k^{2}/m\omega^{2}\) is the homogeneous dielectric function, and \(v(k)\) is the Fourier transform of the Coulomb interaction. Notice that the difference between the longitudinal and transverse results at \(k = 0\) is due to the long range of the Coulomb interaction: the difference vanishes if \(v(k)\) diverges more slowly than \(1/k^{2}\).

Translational invariance is the essential reason for the small-\(k\) behavior of the \(\chi\)'s. In the \(k \to 0\) limit the current operator reduces to the total momentum operator plus a correction that vanishes linearly with \(k\). Thus the first term on the right-hand side of Eq. (25) is the response of the center-of-mass momentum, which obeys a simple equation of motion under the action of the external force, while the second term, of order \(k^{2}\), comes from the residual part of the operator, which is linear in \(k\). There are no cross terms, since the dynamics of the center of mass is decoupled from that of the internal degrees of freedom.

Let us now turn to the xc potentials. The idea is to express the exact current response (23) as the response of a noninteracting electron gas to an effective vector potential, written as

$$a_{\text{eff},1}(k,\omega) = a_{1}(k,\omega) + a_{H,1}(k,\omega) + a_{\text{xc},1}(k,\omega).$$  \hspace{1cm} (26)

The Hartree component \(a_{H,1}\) is purely longitudinal (since it is just another way of describing the scalar Hartree potential) and is given by

$$a_{H,1}(k,\omega) = \frac{k^{2}}{\omega^{2}} v(k) j_{1,L}(k,\omega) k .$$  \hspace{1cm} (27)

The xc potential can be decomposed into its longitudinal and transverse components (with respect to the direction of \(k\)) as follows:

$$a_{\text{xc},1}(k,\omega) = \frac{k^{2}}{\omega^{2}} \left[ f_{\text{xc},L}(k,\omega) j_{1,L}(k,\omega) k + f_{\text{xc},T}(k,\omega) j_{1,T}(k,\omega) \right],$$  \hspace{1cm} (28)

The factor \(k^{2}/\omega^{2}\) has been introduced, in analogy to (27), so that the longitudinal component of the xc vector potential is equivalent to the scalar xc potential

$$v_{\text{xc},1}(k,\omega) = \frac{\omega a_{\text{xc},1,L}}{k} = f_{\text{xc},L}(k,\omega) n_{1}(k,\omega)$$  \hspace{1cm} (29)

\((j_{L} = n_{1}\omega/k)\), thus making \(f_{\text{xc},L}(k,\omega)\) identical with the usual \(f_{\text{xc},L}^{\text{KS}}(k,\omega)\) of the ordinary density functional theory.

In analogy to Eq. (12), the relationship between the interacting current-current response function and its noninteracting counterpart \(\chi_{\text{KS}}\) takes the form

$$\chi_{L(T)}^{\text{1-1}}(k,\omega) = \chi_{\text{KS},L(T)}^{\text{1-1}}(k,\omega) - \frac{k^{2}}{\omega^{2}} \left[ v_{L(T)}(k) + f_{\text{xc},L(T)}^{\text{h}}(k,\omega) \right],$$  \hspace{1cm} (30)

where we have defined \(v_{L}(k) = v(k)\) and \(v_{T}(k) = 0\).

Notice that, according to Eqs. (23), \(\chi_{L}^{\text{1-1}}(k,\omega)\) and \(\chi_{\text{KS},L}^{\text{1-1}}(k,\omega) - k^{2}v_{L}(k)/\omega^{2}\) have the same limit \(m\epsilon(\omega)/n + O(k^{2})\) for \(k \to 0\) and finite \(\omega\). Similarly, \(\chi_{T}^{\text{1-1}}(k,\omega)\) and \(\chi_{\text{KS},T}^{\text{1-1,L}}(k,\omega) - k^{2}v_{T}(k)/\omega^{2}\) have the same limit \(m/n + O(k^{2})\). Thus, we see that Eq. (30) is consistent with the limiting forms (23) if and only if the \(k \to 0\) limits of the xc kernels \(f_{\text{xc},L(T)}^{\text{h}}(k,\omega)\) are finite functions of frequency:

$$\lim_{k \to 0} f_{\text{xc},L(T)}^{\text{h}}(k,\omega) \equiv f_{\text{xc},L(T)}^{\text{h}}(\omega).$$  \hspace{1cm} (31)

Because of the central role these functions play in the developments to follow, we now describe their properties in detail.

**B. Properties of the homogeneous xc kernels**

The calculation of the xc kernels \(f_{\text{xc},L(T)}^{\text{h}}(\omega)\) is a very difficult problem in many-body theory. Approximate calculations have been done using (i) Interpolation schemes between exact high and low frequency limits (ii) Perturbation theory (iii) Mode decoupling approximations. Here we simply summarize the main results that have been established to date, and refer the reader to the original references.
1. The high-frequency limit is a purely real constant given by

\[
\lim_{\omega \to \infty} f_{xc,L(T)}^h(\omega) = \frac{1}{2n} \left[ d_{L(T)}(\langle ke \rangle - \langle ke \rangle_0) + e_{L(T)} \langle pe \rangle \right],
\]

which is also known as the thrid moment sum rule. \( \langle ke \rangle \) and \( \langle pe \rangle \) are the expectation values of the kinetic and potential energy, respectively, and \( \langle ke \rangle_0 \) is the noninteracting kinetic energy. In three dimensions, \( d_L = 4, \quad e_L = 8/15, \quad d_T = 4/3, \quad e_T = -4/15 \). In two dimensions, \( d_L = 6, \quad e_L = 5/4, \quad d_T = 2, \quad e_T = -1/4 \) (see Ref. 35).

The behavior of the imaginary part of the longitudinal xc kernel was first determined by Glick and Long (3D) and Holas and Singwi (2D), making use of second-order perturbation theory, which becomes, in all likelihood, exact in the high-frequency limit. More recently, their calculation has been confirmed and extended to the transverse kernel by a different method based on the equations of motion for the current response function. The result is

\[
\lim_{\omega \to \infty} \text{Im} f_{xc,L(T)}^h(\omega) = -a_L,T \pi^{4-D} \omega^{-D/2} \frac{\epsilon^2}{a_0^2},
\]

where \( a_0 = \hbar^2/m \) is the Bohr radius, \( \omega = \hbar a_0/e^2 \) is the dimensionless frequency, and \( D \) is the number of spatial dimensions. The coefficients are \( a_L = 23/30 \) and \( a_T = 16/15 \) in three dimensions, and \( a_L = 11/32 \) and \( a_T = 9/32 \) in two dimensions.

2. The \( \omega \to 0 \) limit was first worked out by Conti and Vignale and is subtly different from the static limit, which is obtained by setting \( \omega = 0 \) before letting \( k \to 0 \). The result for \( f_{xc,L} \) in \( D \) dimensions is

\[
\lim_{\omega \to 0} f_{xc,L}^h(\omega) = e''_s(n) + \frac{2(D-1)}{D} \lim_{\omega \to 0} f_{xc,T}^h(\omega),
\]

where \( e''_s(n) \equiv d^2 e_s(n)/dn^2 \). The second term on the right-hand side of this expression is proportional to

\[
\lim_{\omega \to 0} f_{xc,T}^h(\omega) = \frac{2E_F}{25n} \left( \frac{3F_2 - 5F_1}{3 + F_1} \right),
\]

in three dimensions, and

\[
\lim_{\omega \to 0} f_{xc,T}^h(\omega) = \frac{E_F}{2n} \left( \frac{F_2 - F_1}{2 + F_1} \right)
\]

in two dimensions, where \( F_1 \) are the conventional Landau parameters of the electron liquid. Note that the first term on the right-hand side of Eq. (34) is the usual compressibility obtained from the static limit \( \lim_{\omega \to 0} \lim_{\omega \to 0} f_{xc,L}^h(k,\omega) \). Thus, Eq. (34) vividly shows the noncommutativity of the \( k \to 0 \) and \( \omega \to 0 \) limits.

3. Due to the causality properties of the linear response functions, the xc kernels must be analytic functions of \( \omega \) in the upper half of the complex plane. This leads to the Kramers-Kronig relations, which relate the real parts of the xc kernels to their imaginary parts:

\[
\text{Re} f_{xc}^h(\omega) = f_{xc}^h(\infty) + \frac{1}{\pi} \int d\omega' \frac{\text{Im} f_{xc}^h(\omega')}{\omega' - \omega},
\]

where \( P \) denotes the “principal part” integral.

4. In a very recent development, the low-frequency behavior of the imaginary parts of the xc kernels has also been calculated exactly to leading order in the strength of the Coulomb interaction. The results are

\[
\text{Im} f_{xc,T}^h(\omega) \to -\frac{1}{(\eta a_0^D)^2} \frac{\eta}{\hbar/a_0^D} \frac{\epsilon^2}{a_0^2},
\]

and

\[
\text{Im} f_{xc,L}^h(\omega) \to \frac{2(D-1)}{D} \text{Im} f_{xc,T}^h(\omega),
\]

where the dimensionless “shear viscosity” \( \frac{\eta}{(\hbar/a_0^D)} \) is given by

\[
\frac{\eta}{(\hbar/a_0^D)} = -\frac{k_F a_0}{45 \pi^3} \left( 5 - \frac{\lambda + 5}{\lambda} \right) \tan^{-1} \lambda
\]

\[
- \frac{2}{\lambda} \sin^{-1} \left( \frac{1}{\sqrt{1 + \lambda^2}} \right)
\]

\[
+ \frac{2}{\lambda \sqrt{2 + \lambda^2}} \left[ \frac{\pi}{2} - \tan^{-1} \frac{1}{\sqrt{2 + \lambda^2}} \right]
\]

in three dimensions, and

\[
\frac{\eta}{(\hbar/a_0^D)} = \frac{1}{12 \pi^2} \left( 2 \left[ \ln(\lambda + 1) - \frac{\lambda}{1 + \lambda} \right]
\right.
\]

\[
- \int_0^1 dx \left( \ln(\lambda + 1) \lambda^2 x^{1/2 + x^2 + 1} \right)
\]

in two dimensions. In the above expressions \( \lambda \) is defined as \( \lambda = 2k_F/k_s \), where \( k_s \) is the screening wavevector, e.g., in RPA, \( k_s = (4 \pi k_F a_0)^{1/2}/\pi a_0 \) in three dimensions, and \( k_s = 2/a_0 \) in two dimensions. The derivation of these results is presented in Ref. 35.

5. Parametrized expressions. To keep our presentation self-contained, we also include the explicit parametrization for \( f_{xc,L}(\omega) \) that has been used in the calculations of Section V. This is the original Iwamoto-Gross-Kohn parametrization and has the form

\[
\text{Im} f_{xc,L}(\omega) = \frac{a(n)\omega}{[1 + b(n)\omega^{3/4}]^{5/4}}
\]

with the coefficients \( a(n) \) and \( b(n) \) determined by the compressibility and third moment sum rules, and the Kramers-Kronig dispersion relations. The real part of \( f_{xc,L}(\omega) \) is then calculated with the help of the dispersion relation. More recent analytic expressions for \( f_{xc,L}(\omega) \) and \( f_{xc,T}(\omega) \) have been obtained by Nifosi, Conti and Tosatti and Qian and Vignale (QV). These new expressions possess considerable structure in the frequency dependence due to two-plasmon excitations. The QV expression reproduces the exact perturbative limit of \( f_{xc}(\omega) \) in the limit \( \omega \to 0 \). Additional details about these expressions can be found in the original references.
C. The exchange-correlation field for a homogeneous electron gas

As a preparation for the study of inhomogeneous systems let us now examine the real-space form of the xc vector potential. It is convenient for this purpose to introduce the xc electric field

$$E_{\text{xc},1}(\mathbf{k}, \omega) = i\omega a_{\text{xc},1}(\mathbf{k}, \omega)$$

$$= -\frac{1}{i\omega} \left\{ \mathbf{k} \cdot \mathbf{j}_1(\mathbf{k}, \omega) \right\} f_{\text{xc},L}(\omega) + k^2 f_{\text{xc},T}(\omega) j_{1,T}(\mathbf{k}, \omega) \right\}. \quad (43)$$

Splitting off the familiar ALDA contribution

$$E_{\text{ALDA}}^{\text{ALDA}}(\mathbf{k}, \omega) = i\epsilon_{\text{ALDA}}^n \frac{\mathbf{k} \cdot \mathbf{j}_1}{\omega}, \quad (44)$$

we can write

$$E_{\text{xc},1}(\mathbf{k}, \omega) = E_{\text{ALDA}}^{\text{ALDA}}(\mathbf{k}, \omega)$$

$$- \frac{1}{i\omega} \left\{ \mathbf{k} \cdot \mathbf{j}_1(\mathbf{k}, \omega) \right\} \left[ f_{\text{xc},L}(\omega) - \epsilon_{\text{ALDA}}^n \right] \nabla (\nabla \cdot \mathbf{u}) + f_{\text{xc},T}(\omega) \nabla^2 \mathbf{u} \right\}. \quad (45)$$

Introducing at this point the velocity field

$$\mathbf{u} = \frac{\mathbf{j}_1}{n} \quad (46)$$

and Fourier transforming Eq. (43) to real space, we get

$$E_{\text{xc},1}(\mathbf{r}, \omega) = E_{\text{ALDA}}^{\text{ALDA}}(\mathbf{r}, \omega)$$

$$+ \frac{n}{i\omega} \left\{ \left[ f_{\text{xc},L}(\omega) - f_{\text{xc},T}(\omega) - \epsilon_{\text{ALDA}}^n \right] \nabla (\nabla \cdot \mathbf{u}) + f_{\text{xc},T}(\omega) \nabla^2 \mathbf{u} \right\}. \quad (47)$$

It is easy to verify that this expression can be re-written as

$$E_{\text{xc},1}(\mathbf{r}, \omega) = E_{\text{ALDA}}^{\text{ALDA}}(\mathbf{r}, \omega) + \frac{1}{n} \sum_{\beta} \frac{\partial \sigma_{\text{xc},\alpha\beta}(\mathbf{r}, \omega)}{\partial \mathbf{r}_\beta}, \quad (48)$$

where the xc stress tensor $\sigma_{\text{xc},\alpha\beta}$ is defined as

$$\sigma_{\text{xc},\alpha\beta} = \tilde{\eta}(n, \omega) \left( \frac{\partial u_\alpha}{\partial r_\beta} + \frac{\partial u_\beta}{\partial r_\alpha} - \frac{2}{D} \nabla \cdot \mathbf{u} \delta_{\alpha\beta} \right) + \tilde{\zeta}(n, \omega) \nabla \cdot \mathbf{u} \delta_{\alpha\beta}, \quad (49)$$

and

$$\tilde{\eta}(n, \omega) = -\frac{n^2}{i\omega} f_{\text{xc},T}(\omega) \quad (50)$$

$$\tilde{\zeta}(n, \omega) = -\frac{n^2}{i\omega} \left( f_{\text{xc},L}(\omega) - \frac{2(D-1)}{D} f_{\text{xc},T}(\omega) - \epsilon_{\text{ALDA}}^n \right) \quad (51)$$

are generalized (i.e., frequency-dependent and complex) visco-elastic constants of the electron liquid. In particular, the real parts of $\tilde{\eta}$ and $\tilde{\zeta}$ [related to the imaginary parts of $f_{\text{xc},L}(\omega)/\omega$] play the role of shear and bulk viscosities, respectively, while the imaginary parts of $\omega \tilde{\eta}$ and $\omega \tilde{\zeta}$ [related to the real parts of $f_{\text{xc},T}(\omega)$] are interpreted as post-ALDA xc contributions to frequency-dependent elastic constants $\mu$ (shear modulus) and $K$ (bulk modulus) of the electron liquid: $\mu_{\text{dyn}} = n^2 \text{Re} f_{\text{xc},T}(\omega)$ and

$$K_{\text{dyn}} = n^2 \text{Re} \left[ f_{\text{xc},T}(\omega) - \frac{2(D-1)}{D} f_{\text{xc},T}(\omega) - \epsilon_{\text{ALDA}}^n \right] \quad (52)$$

Because $\text{Im} f_{\text{xc},L}(\omega)$ vanish linearly for $\omega \to 0$ (point 4 of Section II.B), we see that the viscosity coefficients $\tilde{\eta}$ and $\tilde{\zeta}$ stay finite in the $\omega \to 0$ limit. Eqs. (38) of Section II.B imply that the $\omega \to 0$ limit of the bulk viscosity $\lim_{\omega \to 0} \tilde{\zeta}(\omega)$ is exactly zero at least to within the accuracy of our perturbative calculation. By virtue of the limiting form (54), we also see that $\lim_{\omega \to 0} K_{\text{dyn}}(\omega) = 0$, implying that the bulk modulus of the electron liquid is entirely accounted for by the ALDA contribution $\epsilon_{\text{ALDA}}^n$.

The fact that the shear modulus does not vanish for $\omega \to 0$ but tends to the finite value of Eq. (51) is perhaps surprising. One ordinarily thinks of liquids as having zero
shear modulus. The reason for this strange behavior is that we are taking the $k \to 0$ limit before the \( \omega \to 0 \) limit. Thus, the system remains “dynamical” down to zero frequency. Of course, this would not be true if the \( \omega \to 0 \) limit were taken at finite \( k \). In that limit, \( f^h_{xc,T} \) is no longer related to the shear modulus, but to the static diamagnetic susceptibility, which is extremely small. The truly static shear modulus is zero, as expected.

D. The exchange-correlation field in the inhomogeneous electron gas

The main result of the previous section, Eq. (18), is artfully written so that it can immediately be turned into a local density approximation for the xc electric field of an inhomogeneous electron liquid within the limit $n \to n_0(r)$, where $n_0(r)$ is the ground-state density of the inhomogeneous liquid. Of course, the xc kernels must also be evaluated at the local density.

An important question is this: Why should the replacement $n \to n_0(r)$ be done in Eq. (18) rather than in one of the many equivalent expressions one can generate starting from Eq. (17)? For example, why not write the second term on the right-hand side of Eq. (47) in the equivalent form

$$
\frac{1}{i \omega} \left\{ \left[ f^h_{xc,L}(\omega) - f^h_{xc,T}(\omega) - \epsilon''_{xc}(\omega) \right] \nabla (\nabla \cdot j_1) + f^h_{xc,T}(\omega) \nabla^2 j_1 \right\}
$$

before substituting $n$ by $n_0(r)$? The answer is that this and similar ambiguities are completely removed by general physical requirements which we now discuss.

First of all, because the Coulomb interaction obeys Newton’s third law, the net force exerted by the xc electric field on the system must vanish. At the local level, Newton’s third law implies that a small volume of the electron liquid cannot exert a net force on itself. Accordingly, the net force acting on an arbitrary volume element must be expressible as the integral of the external stresses exerted by the surrounding fluid on the surface of the volume element. The mathematical expression of this requirement is that the force density must be the divergence of a local stress tensor as in Eq. (18).

A similar argument can be applied to the net torque acting on a volume element of the fluid. Again, this must be expressible in terms of a surface integral, and it is not difficult to see that the condition for this to happen is that the stress tensor be a symmetric rank-2 tensor [24].

Finally, Galilean invariance requires the stress tensor to vanish identically when the fluid moves as a whole, i.e., when the velocity field is spatially uniform. It is for this reason that the stress tensor must contain derivatives of the velocity field and not of the current.

These criteria unambiguously establish Eq. (18) as the correct expression in which the substitution $n \to n_0(r)$ should be made. This expression was originally derived by Vignale and Kohn (VK) by a more laborious and apparently quite different path, which more clearly exposed the underlying approximations and the conditions for their validity. VK considered a weakly inhomogeneous electron liquid modulated by a charge-density wave of small amplitude $\gamma$ and small wave vector $q$. Both $k$ and $q$ were assumed to be small not only relative to the Fermi momentum $k_F$, but also relative to $\omega/u_F$ ($u_F$ being the Fermi velocity). The latter condition assures that the phase velocity of the density disturbance is much faster than the Fermi velocity, so that no form of static screening can occur. Under these assumptions, all the components of the tensorial kernel $f_{xc,\alpha\beta}$ could be calculated, up to first order in the amplitude of the charge density wave, and to second order in the wave vectors $k$ and $q$. The calculations were greatly facilitated by a set of sum rules which are mathematically equivalent to the zero-force and zero-torque requirements discussed above. The result of the analysis was that the diagonal matrix elements $f_{xc,\alpha\beta}(k, k, \omega)$ remain equal to $f^h_{xc}(k, \omega)$ to first order in $\gamma$, but the off-diagonal elements $f_{xc,\alpha\beta}(k + q, k, \omega)$ acquire a finite value, given by

$$
f_{xc,\alpha\beta}(k + q, k, \omega) = -\frac{\gamma}{\omega^2} \left\{ \left( \delta f^h_{xcL}(\omega) - f^h_{xcT}(\omega) \right) n\alpha q\beta + f^h_{xcT}(\omega) q^2 \delta_{\alpha\beta} - n \frac{\partial f^h_{xcT}}{\partial n} k \cdot (k + q) \delta_{\alpha\beta} \right. \\
+ A(n, \omega)(k_\alpha + q_\alpha) k_\beta - B(n, \omega) k_\alpha (k_\beta + q_\beta) \right\},
$$

(53)

where $\delta f^h_{xcL} \equiv f^h_{xcL}(\omega, n) - \epsilon''_{xc}(n)$, $A(n, \omega) \equiv \{ n(2\partial f^h_{xcT}/\partial n - \partial f^h_{xcL}/\partial n + 3f^h_{xcT} - \delta f^h_{xcL}) \}$ and $B(n, \omega) \equiv \{ n(\partial f^h_{xcT}/\partial n + 3f^h_{xcT} - \delta f^h_{xcL}) \}$. A remarkable feature of this result is that the off-diagonal matrix elements of $f_{xc,\alpha\beta}$ do not exhibit any singularity for $k$ or $q$ tending to zero in any order. This is in marked contrast with the off-diagonal elements of the scalar (density) xc kernel which, when calculated for the same system, exhibit a power singularity of the form $k \cdot q/k^2$ for $k \to 0$ at finite $q$. This is the fundamental reason why the dynamical local density approximation is possible in terms of the current, but not in terms of the density.

Eq. (53) can be translated into a real space expression for $E_{xc,1}(r, \omega)$. More details of the derivation, which is
The velocity field in the post-ALDA term is dictated by the conditions of validity of the real-space approach are \( |\nabla n_0(r)/n_0(r) | \) much smaller than \( k_F(r) \) and \( \omega/\mu_E(r) \), where \( k_F(r) \) and \( \mu_E(r) \) are the local Fermi momentum and velocity. In Ref. 28, the practical relevance of these conditions was investigated in detail. It was found that the approach could be successfully applied to describe intersubband plasmons in wide single quantum wells, but failed for narrow double quantum wells. The failure in the latter case was traced back to a strong violation of the above criteria of validity in the region of the barrier between the two wells. The physical reason is that electronic motion through tunnelling barriers implies strong internal compression of the electron liquid, which locally destroys coherence of the electron dynamics and leads to a breakdown of the simple hydrodynamical picture. In such a situation, a hybrid between the VUC and the more robust Gross-Kohn approximation (11) provides a pragmatic and practically useful remedy.

Because the occurrence of two spatial derivatives of the velocity field in the post-ALDA term is dictated by general principles, Eq. (18) is expected to remain valid even for large values of \( u \), i.e., in the nonlinear regime, provided that \( u \) and \( n \) are sufficiently slowly varying. The argument goes as follows. Suppose we tried to extend Eq. (18) into the nonlinear regime by including terms of order \( u^2 \). Because the stress tensor must depend on first derivatives of \( u \), such corrections would have to go as \( (\nabla u)^2 \). But then the force density, given by the derivative of the stress tensor, would have to involve at least three derivatives. Thus, for sufficiently small spatial variation of the density and velocity fields, the nonlinear terms can be neglected.

Since the ALDA is an intrinsically nonlinear approximation, VUC proposed that Eq. (18), written in the time domain, could provide an appropriate description of both linear and nonlinear response properties. A nonlinear, retarded expression for \( \tau_{xc,1} \) was also proposed by Dobson et al. The two approximations coincide in “one-dimensional systems” (i.e., when one has a unidirectional current density field that depends only on one coordinate), but differ in the general case.

III. MEMORY FUNCTION FORMALISM AND TCDFT FOR HOMOGENEOUS SYSTEMS

In the preceding sections, we outlined a linear response formalism within TCDFT that goes beyond the adiabatic approximation and allows one to account for intrinsic damping of collective excitations in electronic systems, caused by dynamical many-body effects. As mentioned in the Introduction, this is usually not sufficient to achieve quantitative agreement with experimentally measured linewidths. In reality, intrinsic damping is often overshadowed by strong extrinsic dissipation mechanisms, such as impurities or disorder (in this paper, we consider the low-temperature case only and limit the discussion to systems where LO phonon scattering does not occur).

Effects of impurities and disorder in the linear dynamics of a many-electron system are conveniently discussed in the language of relaxation functions, which then naturally leads to the so-called memory function formalism. In this paper, we perform a conceptually new step and unite the memory function formalism with TCDFT in the linear response regime, which will then allow us to treat both intrinsic and extrinsic damping from first principles and on an equal footing. This is necessary for an accurate description of experiments performed on very clean samples (such as the quantum well we shall discuss in section V), where intrinsic and extrinsic damping may be of comparable magnitude.

The purpose of this section is twofold: To make this paper self-contained, we first review the memory function formalism for the homogeneous case. We then make contact between relaxation and response functions, thereby integrating the memory function formalism with TCDFT. In Section V, we shall extend the approach to systems that are inhomogeneous in one spatial direction (such as quantum wells), and show how it can be applied to discuss intrinsic and extrinsic damping of collective charge-density excitations in such systems.

A. Relaxation and Linear Response

Suppose we are interested in the dynamics of a set of \( N+1 \) observables, \( \{ \hat{A}_0(r, t), \ldots, \hat{A}_N(r, t) \} \), each \( \hat{A}_i(r, t) \) coupling to a small perturbing external field \( \delta a_i(r, t) \). The Hamiltonian in the presence of these fields is give (in Schrödinger representation) by

\[
\hat{H}(t) = \hat{H}_0 + \sum_i \int d^3r \, \hat{A}_i(r) \delta a_i(r, t) .
\]

Consider now the case where the external fields are adiabatically turned on beginning at \( t = -\infty \), and then abruptly switched off at \( t = 0 \):

\[
\delta a_i(r, t) = \begin{cases} \delta a_i(r) e^{\omega t} & \text{for } t \leq 0 \\ 0 & \text{for } t > 0 . \end{cases}
\]

The system starts out from non-equilibrium at \( t = 0^+ \) and, being left to itself, relaxes back towards equilibrium. The first-order change of the non-equilibrium expectation value of an observable can then be written as

\[
\delta \langle \hat{A}_i(r, t) \rangle_{\text{noneq.}} = \sum_j \int d^3r' \tilde{C}_{ij}(r, r', t) \delta a_j(r') ,
\]

where the correlation (or Kubo) function in the presence of disorder is defined as

\[
\tilde{C}_{ij}(r, r', t) = \int_0^\beta d\beta' \langle \hat{A}_i(r, t) \hat{A}_j(r', -i\beta') \rangle_{\text{eq.}}.
\]
This may be rewritten as
\[
\tilde{C}_{ij}(r, r', t) = \left\langle \hat{A}_i(r) | e^{-iL t} | \hat{A}_j(r') \right\rangle ,
\] (58)
where \( L \) is the Liouville operator governing time evolution of the system via
\[
\dot{\hat{A}}_i(t) = iL\hat{A}_i(t) = [\hat{A}_i(t), \hat{H}]/(i\hbar),
\tag{59}
\]
and the scalar product \( \langle \ldots | \ldots \rangle \) is defined by Eq. (57). We impose normalization on the set of variables, i.e. \( \langle \hat{A}_i(r) | \hat{A}_j(r') \rangle = \delta_{ij} \). In the following, we are interested in the zero temperature limit \( \beta \rightarrow \infty \).

The correlation function (57) is related to the absorptive part of the usual quantum mechanical response functions \( \chi_{ij}(r, r', t) \) as follows:
\[
i\partial_t \tilde{C}_{ij}(r, r', t) = \chi''_{ij}(r, r', t).
\] (60)
Since relaxation occurs for \( t > 0 \), it is customary to introduce the Laplace transform of the quantities of interest, which, e.g., for the correlation function is defined as
\[
\tilde{C}_{ij}(r, r', \xi) = \int_0^\infty dt \ e^{i\xi t} \tilde{C}_{ij}(r, r', t),
\] (61)
where \( \xi \) is a complex number in the upper half of the complex plane. One then finds the following relationship between the Kubo relaxation functions and the response functions:
\[
\tilde{C}_{ij}(r, r', \xi) = [\chi_{ij}(r, r', \xi) - \chi_{ij}(r, r', i0)]/(i\xi).
\] (62)
For the remainder of this section, we assume that the system is spatially homogeneous (a more general case will be considered in Sec. [3]). Eq. (60) can then be Fourier transformed into momentum space, and one obtains
\[
\tilde{C}_{ij}(q, \xi) = \int_0^\infty dt \ e^{i\xi t} \tilde{C}_{ij}(r, r', t),
\] (63)
The Laplace transform of the correlation function (68) is then given by
\[
\tilde{C}_{ij}(q, \xi) = \left\langle \hat{A}_i(q) | \frac{i}{\xi - \mathcal{L}} | \hat{A}_j(q) \right\rangle.
\] (64)

\section{Projectors and Memory Functions}

The observables \( \{\hat{A}_0(q), \ldots, \hat{A}_N(q)\} \) can be regarded as vectors in a Hilbert space. The Liouvillian \( \mathcal{L} \) acts as a linear operator in that space, see Eq. (53). We define a projection operator \( \mathcal{P} \) onto the space spanned by \( \{\hat{A}_0(q), \ldots, \hat{A}_N(q)\} \) as
\[
\mathcal{P} = \sum_i |\hat{A}_i(q)\rangle \langle \hat{A}_i(q)|,
\] (65)
and its complement \( \mathcal{Q} = 1 - \mathcal{P} \) projects perpendicular to it. One can then formally write Eq. (64) as
\[
\tilde{C}_{ij}(q, \xi) = \left\langle \hat{A}_i(q) | \frac{i}{\xi - \mathcal{L} \mathcal{Q} - \mathcal{P}} | \hat{A}_j(q) \right\rangle.
\] (66)
Following Forster, one performs a few straightforward manipulations in Eq. (66) and finds
\[
\tilde{C}_{ij}(q, \xi) = \frac{i}{\xi} \delta_{ij} + \frac{1}{\xi} \sum_k \left\{ \left\langle \hat{A}_i(q) | \mathcal{L} | \hat{A}_k(q) \right\rangle \right\} \tilde{C}_{kj}(q, \xi).
\] (67)
Defining \( C_{ij}(q, \xi) = i\tilde{C}_{ij}(q, \xi) \), \( \Omega_{ik}(q) = \langle \hat{A}_i(q) | \mathcal{L} | \hat{A}_k(q) \rangle \) and \( M_{ik}(q, \xi) = \left\langle \hat{A}_i(q) | \frac{1}{\mathcal{Q} \mathcal{L} Q - \xi} \mathcal{Q} | \hat{A}_k(q) \right\rangle \) we rewrite Eq. (67) as follows:
\[
\sum_k \left\{ \xi \delta_{ik} - \Omega_{ik}(q) + M_{ik}(q, \xi) \right\} \tilde{C}_{kj}(q, \xi) = -\delta_{ij}.
\] (68)
\( \Omega_{ij}(q) \) can be viewed as characteristic frequency or restoring force matrix of the system. Our particular interest, however, lies in the memory function matrix \( M_{ik}(q, \xi) \), which introduces dissipation into the electron dynamics of the system. In general, dissipation originates from intrinsic as well as extrinsic scattering mechanisms. The former, caused by electron-electron interaction alone, are present even in a perfectly “clean” system. In the previous section we discussed the treatment of intrinsic dissipation in the framework of TCDFT. Now we describe how additional extrinsic dissipation (e.g. caused by scattering off disorder or charged impurities) can be taken into account simultaneously.
\( M_{ik} \) has the formal structure of a correlation function between two projected forces, \( \mathcal{Q} \hat{A}_i(q) \) and \( \mathcal{Q} \hat{A}_j(q) \). These forces act perpendicular to the vector space of variables \( \{\hat{A}_i\} \), thus providing a coupling to other degrees of freedom of the system (which effectively form a “thermal bath”). Accordingly, the frequency dynamics of \( M_{ik}(q, \xi) \) is determined by \( \mathcal{Q} \mathcal{L} Q \), where those fluctuations of the Liouville operator are projected out that occur only within the space of variables \( \{\hat{A}_i\} \), therefore describing the internal dynamics of the “bath.”

The correlation functions \( C_{ij}(q, \xi) \) are determined by a set of \( (N + 1)^2 \) coupled equations, Eq. (67), whose solution will be discussed for an example in Sec. [II]. The observables \( \hat{A}_i \) are not restricted to be scalars, but can also be vectors (or \( n \)-th rank tensors). In general, all correlation functions \( C_{ij}(k, \omega) \) as well as \( \Omega_{ij} \) and \( M_{ij} \) are tensors whose rank equals the sum of the ranks of \( A_i \) and \( A_j \).
Assuming that explicit solutions for the $C_{ij}$ have been found, the final step is then to make contact with linear response theory, which involves Fourier transforms (with frequency $\omega$) rather than Laplace transforms (with frequency $\xi$) of the associated response and correlation functions. Fortunately, the relationship between Fourier and Laplace transforms is a straightforward linear one, so that the response functions $\chi_{ij}(q,\omega)$ can be simply obtained from

$$C_{ij}(q,\omega) = \left[\chi_{ij}(q,\omega) - \chi_{ij}(q,0)\right]/\omega. \quad (72)$$

Here, $\omega$ are real frequencies, and $C_{ij}(q,\omega) \equiv C_{ij}(q,\xi = \omega + i0^+)$. 

### C. Generalized Relaxation Time Approximation

In the following, we discuss the case where there are only two observables of interest: density fluctuations $\hat{\rho}(q)$ and current density $\hat{j}(q)$. Including normalization, we have

$$\hat{A}_0(q) = \frac{\hat{\rho}(q)}{\langle \hat{\rho}(q) | \hat{\rho}(q) \rangle^{1/2}} \quad (73)$$

and

$$\hat{A}_1(q) = \frac{\hat{j}(q)}{\sqrt{n}}. \quad (74)$$

where $n$ is the uniform density of the system, and $\langle \hat{\rho}(q) | \hat{\rho}(q) \rangle = \chi(q,0)$, the static density-density response function (in the presence of disorder). Eq. (74) then describes the four associated correlation functions $C_{00}, C_{01}, C_{10}$ and $C_{11}$, i.e. $\hat{\rho}\hat{\rho}, \hat{\rho}\hat{j}, \hat{j}\hat{\rho}$ and $\hat{j}\hat{j}$.

Since we deal with the homogeneous and isotropic case, Eq. (74) decouples into two independent equations for the longitudinal ($L$) and transverse ($T$) components of the correlation functions:

$$\sum_{k=0}^1 \left[ \omega \delta_{ik} - \Omega_{ik}^L(q) + M_{ik}^L(q,\omega) \right] C_{kj}^L(q,\omega) = -\delta_{ij}, \quad (75)$$

and

$$\left[ \omega - \Omega_{11}^T(q) + M_{11}^T(q,\omega) \right] C_{ij}^T(q,\omega) = -1. \quad (76)$$

Eq. (75), with $i,j = 0,1$, represents a system of 4 equations coupling the 4 possible longitudinal correlation functions $\langle \hat{\rho}\hat{\rho}, \hat{\rho}\hat{j}, \hat{j}\hat{\rho}, \hat{j}\hat{j} \rangle$. Since there is no coupling between density and transverse currents, there is only a single transverse correlation function, $\langle \hat{j}\hat{j} \rangle$, determined by Eq. (76). Using the continuity equation,

$$\mathcal{L}\hat{\rho}(q) = -q^2\hat{j}(q), \quad (77)$$

we convince ourselves that indeed $\langle \hat{A}_0 | \hat{A}_0^\dagger \rangle = 0$. Furthermore, since $\mathcal{L}\hat{A}_0$ is proportional to $\hat{A}_0^\dagger$, the first component of the “perpendicular” force $\hat{Q}_0$ is identically zero, so that

$$M_{00}^L(q,\omega) = M_{01}^L(q,\omega) = M_{10}^L(q,\omega) = 0. \quad (78)$$

In the following, we will be concerned with the limit of weak disorder. In this limit, it is a good approximation to assume that all static correlation functions are unaffected by disorder. This means that $\Omega^{L(T)}(q)$ contains effects of Coulomb interaction only. Likewise, we assume

$$\chi_{ij}^{L(T)}(q,0) = \chi_{ij}^{L(T)c}(q,0), \quad (79)$$

where the superscript “$c$” denotes the “clean” response function. In general, static disorder effects (mainly changes of the density of state at the Fermi surface) are weak, provided $(\tau\varepsilon_F)^{-1} \ll 1$, where $\varepsilon_F$ is the Fermi energy, and $\tau$ is a characteristic disorder scattering time for the system under study.

In general, the memory functions contain both intrinsic (Coulomb interaction) and extrinsic (disorder) contributions. In the limit of weak disorder, one can separate them as follows:

$$M_{11}^{L(T)}(q,\omega) \equiv M_{11}^{L(T)in}(q,\omega) + M_{11}^{L(T)ex}(q,\omega). \quad (80)$$

We combine the intrinsic part with $\Omega_{kk}$, thereby defining a frequency-dependent, dissipative restoring force matrix $\Omega_{ik}^{L(T)in}(q,\omega)$, which contains effects of Coulomb interactions only:

$$\Omega_{ik}^{L(T)in}(q,\omega) \equiv \Omega_{ik}^{L(T)in}(q) + M_{ik}^{L(T)in}(q,\omega)\delta_{ij}\delta_{k1}. \quad (81)$$

One then obtains from Eqs. (75) and (76):

$$\sum_{k=0}^1 \left[ \omega + M_{ik}^{L(T)in}(q,\omega) \right] \delta_{ik} - \Omega_{ik}^{L(T)in}(q,\omega) \right] C_{kj}^L(q,\omega) = -\delta_{ij} + \delta_{i0}M_{ex}^L(q,\omega)C_{ij}^T(q,\omega), \quad i,j = 0,1 \quad (82)$$

and the subscript “$ex$” of $M_{ex}^L$ and $M_{ex}^T$ in the following. To solve Eqs. (82) and (83), we introduce “clean” longitudinal (transverse) reference functions $C_{ij}^{L(T)c}(q,\omega)$,
defined in the absence of dissipation (i.e., $M^L = 0 = M^T$), as follows:

\[
\sum_{k=0}^{1} \left[ \omega \delta_{ij} - \Omega_{i k}^{L} (q, \omega) \right] C_{k j}^{L} (q, \omega) = -\delta_{ij} , \quad i, j = 0, 1 , \tag{84}
\]

\[
\left[ \omega - \Omega_{11}^{L} (q, \omega) \right] C_{11}^{L} (q, \omega) = -1 . \tag{85}
\]

The desired correlation functions are then expressed in terms of these reference functions. In the longitudinal case, we find

\[
C_{ij}^{L} (q, \omega) = C_{ij}^{L} (q, \omega) + C_{0j}^{L} (q, \omega) \times M^L C_{0i}^{L} (q, \omega) , \quad i, j = 0, 1 , \tag{86}
\]

or explicitly

\[
C_{00}^{L} (q, \omega) = \frac{C_{00}^{L} C_{00}^{L}}{1 + M^L C_{00}^{L}} \tag{87}
\]

\[
C_{10}^{L} (q, \omega) = \frac{C_{10}^{L} C_{00}^{L}}{1 + M^L C_{00}^{L}} \tag{88}
\]

\[
C_{01}^{L} (q, \omega) = \frac{C_{01}^{L} C_{00}^{L}}{1 + M^L C_{00}^{L}} \tag{89}
\]

\[
C_{11}^{L} (q, \omega) = \frac{C_{11}^{L} + C_{11}^{L} M^L C_{00}^{L} - C_{10}^{L} M^L C_{01}^{L}}{1 + M^L C_{00}^{L}} . \tag{90}
\]

where all “clean” functions carry the arguments $(q, \omega + M^L)$. For the transverse case, on the other hand, we simply obtain

\[
C_{ij}^{T} (q, \omega) = C_{ij}^{L} (q, \omega + M^T) . \tag{91}
\]

We now make contact with the response functions as follows. For the case of density-density response, we have from Eq. (72):

\[
C_{00}^{L} (q, \omega) = \frac{1}{\omega} \left[ \chi_{00}^{L} (q, \omega) - \chi_{00}^{L} (q, 0) \right] / \chi_{00}^{L} (q, 0) , \tag{92}
\]

where the function $\chi_{00}^{L} (q, 0)$ in the denominator arises from the normalization of the variable $A_0$, see Eq. (73). Similar expressions are obtained for the other longitudinal and transverse correlation functions. We then find from Eq. (87):

\[
\frac{1}{\chi_{00}^{L} (q, \omega)} = \frac{1}{\omega + M^L} \frac{1}{\chi_{00}^{L} (q, \omega + M^L)} + \frac{M^L}{\omega + M^L} \frac{1}{\chi_{00}^{L} (q, 0)} . \tag{93}
\]

Eq. (23) is formally in agreement with Belitz and Das Sarma (equation (2.3) in Ref. 23). However, $\chi_{00}^{L} (q, \omega)$ here denotes the exact, fully interacting response function of the homogeneous electron gas, not just the RPA response function.

\[
\chi_{00}^{L} (q, \omega) = \frac{q}{\omega} \chi_{01}^{L} (q, \omega) = \frac{q}{\omega} \chi_{01}^{L} (q, \omega) = \frac{q^2}{\omega} \chi_{11}^{L} (q, \omega) . \tag{94}
\]

It is not difficult to show that the results for $\chi_{00}^{L} , \chi_{01}^{L}$ and $\chi_{11}^{L}$ obtained in this fashion are consistent with Eqs. (33)-(37).

In the same way one finds the transverse current-current response function from Eq. (72):

\[
\chi_{11}^{T} (q, \omega) = \frac{\omega}{(\omega + M^L)} \chi_{11}^{T} (q, \omega + M^T) + \frac{M^T}{(\omega + M^T)} \chi_{11}^{T} (q, 0) . \tag{95}
\]

Expressions that are formally similar to Eqs. (93)-(95) were recently derived by Conti and Vignale in the framework of Mermin’s relaxation time approximation.

In this formalism, the role of $M^L (q, \omega)$ is taken by a frequency- and momentum-independent phenomenological scattering rate $i/\tau$. Note that the second term on the right-hand side of (95) is absent in Ref. 36, because there the diamagnetic susceptibility of the electron gas, $\chi_{11}^{T} (q, 0)$, was implicitly taken to be zero.

Finally, explicit expressions for the memory functions $R^L (q, \omega)$ and $R^T (q, \omega)$ are obtained in the following way from Eq. (70): first, we approximately write

\[
M^L (q, \omega) = \left\langle \hat{F}^L (q) \left| \frac{1}{L - \omega} \right| \hat{F}^L (q) \right\rangle , \tag{96}
\]

and similar for $M^T$, i.e. we resort to the standard approximation of replacing the projected by the full Liouville operator by setting $Q \approx 1$ in the denominator. We thus assume that the huge amount of degrees of freedom in the thermal bath and their extremely complex time evolution are completely dominating over the small subspace of observables $A_0, A_1$ and its relatively well-controlled time evolution.

In turn, the fluctuating longitudinal forces have the form

\[
\hat{F}^L (q) = \frac{1}{\sqrt{n}} \sum_{k} \frac{q [q \cdot (k - q)]}{q^2} U(q - k) \rho(k) , \tag{97}
\]

and the transverse forces are

\[
\hat{F}^T (q) = -\frac{1}{\sqrt{n}} \sum_{k} \frac{q \times (q \times k)}{q^2} U(q - k) \rho(k) . \tag{98}
\]

Here, $U(q)$ is a random scattering potential. In the weak-disorder limit, we can perform the following decoupling, up to within corrections of higher than second order in the disorder potential:
Since the system is homogeneous, we have \( k = k' \), and we arrive at the following expression for the longitudinal memory function:

\[
M^L(q, \omega) = \frac{1}{n} \sum_k \langle U(q - k) \rangle \frac{1}{L - \omega} U(q - k') \rho(k') \approx \langle \hat{\rho}(k) \rangle \frac{1}{L - \omega} \rho(k') \langle U(q - k) U(q - k') \rangle_{\text{disorder}} .
\]

(99)

One thus needs to calculate \( M^L(q, \omega) \) and \( C_{00}^L(q, \omega) \) via self-consistent solution of Eqs. (87) and (100). The so determined \( C_{00}^L(q, \omega) \) then serves as input for the transverse memory function

\[
M^T(q, \omega) = \frac{1}{n} \sum_k \langle U(q - k) \rangle^2 \frac{[q \cdot (q - k)]^2}{q^2} C_{00}^L(q, \omega) \chi_{00}(k, 0) .
\]

(101)

which was obtained using the same decoupling approximation that led to Eq. (100) for \( M^L(q, \omega) \).

IV. MEMORY FUNCTION FORMALISM AND TCDFT FOR INHOMOGENEOUS SYSTEMS

A. Formalism

We now generalize the memory function formalism to systems that are inhomogeneous in one spatial direction, but still homogeneous in the plane perpendicular to it. The example we have in mind are quantum wells whose direction of growth is the \( z \)-axis. One can then in general no longer decouple longitudinal and transverse components of the correlation functions. A special case where this is still possible will be discussed in some detail later on.

The generalization of Eq. (71) for this inhomogeneous situation is

\[
\sum_k \int dz'' \left[ \omega \delta(z'' - z) \delta_{ik} - \Omega_{ik}(q_{||}, z, z'') + M_{ik}(q_{||}, z, z'', \omega) \right] C_{kj}(q_{||}, z'', z', \omega) = -\delta_{ij} \delta(z - z') ,
\]

(102)

where \( q_{||} \) is the in-plane wave vector. We again consider density fluctuations and current density as the only variables. Just like in the homogeneous case, by virtue of the continuity equation, all elements of the \( 2 \times 2 \) memory function matrix \( M_{ik} \) are zero except \( M_{11} \). As before, we can separate intrinsic and extrinsic contributions to the memory function in the weak-disorder limit, \( M_{11} \equiv M_{\text{in}} + M_{\text{ex}} \), and we combine the intrinsic part with \( \Omega_{ik} \), defining

\[
\Omega_{ik}^{\text{in}}(q_{||}, z, z', \omega) \equiv \Omega_{ik}(q_{||}, z, z') + M_{\text{in}}(q_{||}, z, z', \omega) \delta_{ij} \delta_{k1} .
\]

(103)

Eq. (102) thus becomes

\[
\sum_k \int dz'' \left\{ \omega \delta(z'' - z) + M_{\text{ex}}(q_{||}, z, z'', \omega) \right\} \delta_{ik} - \Omega_{ik}^{\text{in}}(q_{||}, z, z'', \omega) \right] C_{kj}(q_{||}, z'', z', \omega) = -\delta_{ij} \delta(z - z') + \delta_{ij} \int dz' \ M_{\text{ex}}(q_{||}, z, z', \omega) C_{ij}(q_{||}, z', z', \omega) , \quad i, j = 0, 1 .
\]

(104)

Similar to Sec. II C, we will solve this set of equations by introducing suitable reference functions. However, the inhomogeneity of the system prevents us from using the same trick as for the homogeneous case, where we directly expressed the correlation functions in the presence of disorder in terms of the “clean” correlation functions, with their frequency argument \( \omega \) replaced by \( \omega + M_{\text{ex}} \). Now, by contrast, the memory function \( M_{\text{ex}}(q_{||}, z, z', \omega) \) is no longer simply a number, but acts in conjunction with an integral operator, see Eq. (104). To deal with this difficulty, we first define a set of intermediate reference functions \( C_{ij}^{R_k}(q_{||}, z, z', \omega) \) that satisfy the following coupled equations:

\[
\sum_k \int dz'' \left\{ \omega \delta(z'' - z) + M_{\text{ex}}(q_{||}, z, z'', \omega) \right\} \delta_{ik} - \Omega_{ik}^{\text{in}}(q_{||}, z, z'', \omega) \right] C_{kj}^{R_k}(q_{||}, z'', z', \omega) = -\delta_{ij} \delta(z - z') , \quad i, j = 0, 1 .
\]

(105)
In terms of these reference functions, the full correlation functions are given, combining Eqs. (104) and (102), through the following Dyson-type integral equation:

\[
C_{ij}(q||, z, z', \omega) = C_{ij}^R(q||, z, z', \omega) \\
- \int dz_1 \int dz_2 \ C_{00}^R(q||, z, z_1, \omega) \ M_{ex}(q||, z_1, z_2, \omega) \ C_{0j}(q||, z_2, z', \omega). \tag{106}
\]

We now use a similar trick to obtain the reference functions \(C_{ij}^R\). Defining the “clean” response function via

\[
\sum_{k=0}^{1} \int dz'' \left( \omega \delta(z'' - z) \Omega^{in}_{kk}(q||, z, z'', \omega) \right) C_{kj}^c(q||, z'', z', \omega) = -\delta_{ij} \delta(z - z'), \tag{107}
\]

we get from Eqs. (106) and (107):

\[
C_{ij}^R(q||, z, z', \omega) = C_{ij}^c(q||, z, z', \omega) \\
+ \sum_{k=0}^{1} \int dz_1 \int dz_2 \ C_{ik}^c(q||, z, z_1, \omega) \ M_{ex}(q||, z_1, z_2, \omega) \ C_{kj}^R(q||, z_2, z', \omega). \tag{108}
\]

The density-density reference correlation function is explicitly given by

\[
C_{00}^R(q||, z, z', \omega) = C_{00}^c(q||, z, z', \omega) \\
+ \int dz_1 \int dz_2 C_{00}^c(q||, z, z_1, \omega) M_{ex}(q||, z_1, z_2, \omega) C_{00}^R(q||, z_2, z', \omega) \\
+ \int dz_1 \int dz_2 C_{01}^c(q||, z, z_1, \omega) M_{ex}(q||, z_1, z_2, \omega) C_{10}^R(q||, z_2, z', \omega). \tag{109}
\]

At this point, it is convenient to introduce the following set of auxiliary functions, which will allow us later to write the memory function \(M_{ex}\) in a more compact form (see below):

\[
\Phi(q||, z, z', \omega) \equiv \int dz'' C_{00}(q||, z, z'', \omega) \chi_c(q||, z'', z', 0) \tag{110}
\]

\[
\Phi^R(q||, z, z', \omega) \equiv \int dz'' C_{00}^R(q||, z, z'', \omega) \chi_c(q||, z'', z', 0) \tag{111}
\]

\[
\Phi^c(q||, z, z', \omega) \equiv \int dz'' C_{00}^c(q||, z, z'', \omega) \chi_c(q||, z'', z', 0). \tag{112}
\]

Again it is assumed that all extrinsic damping effects can be neglected for the static response function. In terms of these functions, Eq. (110) becomes for \(i = j = 0:\)

\[
\Phi(q||, z, z', \omega) = \Phi^R(q||, z, z', \omega) \\
- \int dz_1 \int dz_2 \int dz_3 \Phi^R(q||, z, z_1, \omega) \left[ \chi_c(q||, z_1, z_2, 0) \right]^{-1} M_{ex}(q||, z_2, z_3, \omega) \Phi(q||, z_3, z', \omega). \tag{113}
\]

In the example to be discussed in section $\gamma$, only the case \(q|| = 0\) will be of interest. Since in this case the continuity equation can be used to explicitly eliminate the current density in favor of the density, Eq. (109) can be written as

\[
\Phi^R(0, z, z', \omega) = \Phi^c(0, z, z', \omega) \\
+ \int dz_1 \int dz_2 \int dz_3 \Phi^c(0, z, z_1, \omega) \left[ \chi_c(0, z_1, z_2, 0) \right]^{-1} M_{ex}(0, z_2, z_3, \omega) \Phi^R(0, z_3, z', \omega) \\
+ \int dz_1 \int dz_2 \int dz_3 \left[ \chi_c(0, z_1, z', \omega) \frac{M_{ex}(0, z_1, z_2, \omega)}{n(z_1)n(z_2)} \right]^{\frac{z_1}{z}} dz_3 \chi_c(0, z, z', \omega). \tag{114}
\]

The desired density-density response functions are then finally obtained using

\[
\Phi(q||, z, z', \omega) = \frac{\chi(q||, z, z', \omega) - \chi^c(q||, z, z', 0)}{\omega} \tag{115}
\]

\[
\Phi^R(q||, z, z', \omega) = \frac{\chi^R(q||, z, z', \omega) - \chi^c(q||, z, z', 0)}{\omega} \tag{116}
\]

\[
\Phi^c(q||, z, z', \omega) = \frac{\chi^c(q||, z, z', \omega) - \chi^c(q||, z, z', 0)}{\omega}. \tag{117}
\]
To summarize: Eqs. (109)–(117) allow one to express the interacting density-density response function of the system in the presence of intrinsic and extrinsic dissipation, \( \chi(q, z, z', \omega) \), in terms of the interacting response function for the “clean” system, \( \chi^c(q, z, z', \omega) \), i.e. including intrinsic dissipation alone. \( \chi^c \) is calculated, in principle exactly, using the framework of TCDFT outlined in Sec. IV A. Although admittedly somewhat frightening in appearance, the integral equations (113) and (114) involve only one-dimensional integrals, and their numerical solution is therefore quite manageable, as will be shown below.

### B. Memory functions for impurity and interface roughness scattering

In general, a vector field \( \mathbf{V}(r) \) is decomposed into longitudinal and transverse components as follows: \( \mathbf{V}^L(r) = -(1/4\pi)\nabla \int d^3r' [\nabla \cdot \mathbf{V}(r')] / |r - r'| \) and \( \mathbf{V}^T(r) = (1/4\pi)\nabla \times \nabla \times \int d^3r' \mathbf{V}(r') / |r - r'| \). In our case, i.e. working in a mixed \((q, z)\)-representation, it is convenient to define the operator \( \mathbf{D}(q, z) \) as

\[
\mathbf{D}(q, z) = \left( \frac{q}{i\nabla} \right).
\]

The longitudinal fluctuating forces are then given by

\[
\hat{F}_L(q, z) = \frac{1}{\sqrt{n(z)}} \mathbf{D}(q, z) \int dz' \int \frac{d^2p||}{(2\pi)^2} \frac{e^{-q_|| z - z'}}{2q||} \left\{ \hat{\rho}(p||, z') \mathbf{D}(p|| - q, z') U(q, z') - \hat{\rho}(p||, z') \mathbf{D}(p|| - q, z') U(q, z') \right\}
\]

and the transverse fluctuating forces are

\[
\hat{F}_T(q, z) = -\frac{1}{\sqrt{n(z)}} \mathbf{D}(q, z) \times \mathbf{D}(q, z) \int dz' \int \frac{d^2p||}{(2\pi)^2} \frac{e^{-q_|| z - z'}}{2q||} \left[ \hat{\rho}(p||, z') \mathbf{D}(p|| - q, z') U(q, z') - \hat{\rho}(p||, z') \mathbf{D}(p|| - q, z') U(q, z') \right].
\]

One finds from Eqs. (113) and (120) that in the homogeneous limit (no \( z \) dependence of the density fluctuations \( \rho \) and the scattering potential \( U \)), the longitudinal and transverse forces only have in-plane components, given by the 2-D versions of Eqs. (17) and (18). In the following, we shall limit the discussion of the inhomogeneous situation to a case of special interest, namely \( q|| = 0 \). In that case, only the \( z \)-component of \( \mathbf{F}^L \) survives, and is given by

\[
\hat{F}_L(z) = \frac{i}{\sqrt{n(z)}} \int \frac{d^2p||}{(2\pi)^2} \hat{\rho}(p||, z) \nabla z U(p, z).
\]

The transverse force, in the same limit, acts in the \( x - y \) plane only:

\[
\hat{F}_T(z) = \frac{1}{\sqrt{n(z)}} \int \frac{d^2p||}{(2\pi)^2} \hat{\rho}(p||, z) U(p, z).
\]

We note that, by symmetry, in the limit \( q|| = 0 \) there is a natural decoupling of the formalism outlined above (Sec. IV A) into separate sets of equations of the type (113)–(117) determining longitudinal and transverse response functions, respectively. In other words, \( L - T \) cross correlations are absent since the associated fluctuating forces are perpendicular to each other. The longitudinal and transverse memory functions are obtained in a quite straightforward manner from Eqs. (113) and (120), using the same approximate decoupling procedure that was used for the homogeneous case in Sec. III C. The result is:

\[
M_{\text{ex}}^{L}(z, z', \omega) = \int \frac{d^2p||}{(2\pi)^2} \frac{\Phi(p||, z, z', \omega)}{\sqrt{n(z)n(z')}} \nabla z \nabla z' \left\{ U(-p, z) U(-p, z') \right\}
\]

and

\[
M_{\text{ex}}^{T}(z, z', \omega) = \int \frac{d^2p||}{(2\pi)^2} \frac{\Phi(p||, z, z', \omega)}{\sqrt{n(z)n(z')}} \rho||^2 \left\{ U(-p, z) U(-p, z') \right\}.
\]
associated with some extrinsic damping mechanism. In the following, we shall focus on two examples specific to quantum wells: damping by charged impurities and by interface roughness.

The potential associated with a single, statically screened, positively charged impurity at position \( z_1 \) is

\[
U(p||, z) = \frac{2\pi}{\varepsilon(p||)} \frac{e^{-|p|||z - z_1|}}{p||}, \tag{125}
\]

where \( \varepsilon(p||) \) is the 2D dielectric function. The longitudinal memory function for charged-impurity scattering is thus

\[
M^l(z, z', \omega) = \int \frac{d^2p||}{\varepsilon^2(p||)} \frac{\Phi(p||, z, z', \omega)}{\sqrt{n(z)n(z')}} \int d\tilde{z} n_i(\tilde{z})
\times \text{sign}(z - \tilde{z}) \text{sign}(z' - \tilde{z}) e^{-p|||z - \tilde{z}|} e^{-p|||z' - \tilde{z}|}, \tag{126}
\]

where \( n_i(z) \) is the number of impurities per volume.

Likewise, the longitudinal memory function associated with interface roughness is

\[
M^l_R(z, z', \omega) = \int \frac{d^2p||}{(2\pi)^2} \frac{\Phi(p||, z, z', \omega)}{\sqrt{n(z)n(z')}} \langle U(p||) \rangle^2
\times \text{sign}(z - z_t) \delta(z' - z_t) + \delta(z - z_r) \delta(z' - z_r), \tag{127}
\]

where \( U(p||) \) is the random roughness scattering potential, assumed for simplicity to be the same at the left and right interfaces, \( z_t \) and \( z_r \). It is common to assume a Gaussian form for the autocorrelation function of the random interface roughness which leads to

\[
\langle U(p||) \rangle^2 = \pi \mu^2 \Delta^2 \eta^2 e^{-\rho_0^2 \eta^2/4}. \tag{128}
\]

Here, \( \mu \) is the height of the potential step at the interface, and the correlation length \( \eta \) and average roughness height \( \Delta \) are controlled by material and growth conditions. In the presence of both impurity and roughness scattering, the memory functions \( M^l \) and \( M^l_R \) are additive (i.e., different extrinsic scattering mechanisms are assumed to be uncorrelated).

Some practical complications arise from the fact that the memory functions explicitly depend on \( \Phi(q||, z, z', \omega) \) at all \( q|| \), not just \( q|| = 0 \). This means that \( \Phi \) should be calculated self-consistently from Eqs. (113) for all \( q|| \), which is a very demanding computational task. Therefore, as a first approximation, we ignore self-consistency and instead use the noninteracting \( \Phi_0(q||, z, z', \omega) \) in (126) and (127), defined by replacing \( \chi \) and \( \chi_c \) with \( \chi_{KS} \) in Eq. (113). The wave vector dependence of \( \Phi_0 \) is thus known analytically (see the explicit expression for \( \chi_{KS} \) below). This is expected to be a reasonable approximation as long as plasmon damping is not too strong.

V. LINEWIDTH OF INTERSUBBAND PLASMONS IN A QUANTUM WELL

In semiconductor quantum wells, the conduction band splits up into several subbands, and electrons (supplied e.g. by remote doping) can perform collective transitions between them. These so-called intersubband (ISB) plasmons are currently of great experimental and theoretical interest being the basis of a variety of new devices operating in the terahertz regime, such as detectors[1] and quantum cascade lasers[2]. In designing these devices, the emphasis usually lies in covering a particular frequency range. However, often it is desirable that the transitions also have a narrow linewidth, to achieve better frequency resolution and larger peak absorption in detectors, and higher gain in lasers. The linewidth arises from a complicated interplay of a variety of scattering mechanisms, intrinsic (electron-electron and electron-phonon) as well as extrinsic ones (impurity, alloy-disorder and interface roughness). Many aspects of this interplay are still not well understood, in particular the relative importance of the individual mechanisms.

To disentangle the various contributions to the ISB linewidth, it is helpful to consider a situation where some of them are not effective. In a recent experiment, Williams et al. studied collective ISB transitions in an n-type 40 nm wide single GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As quantum well, with Si doping centers 100 nm away from the well. Sharp transitions were found well below the LO phonon frequency of GaAs (35.6 meV), at a temperature of 2.3 K. Thus, neither remote impurity nor phonon scattering are playing any significant role (nor is alloy-disorder scattering, as shown in Ref. [3]). The linewidth is therefore expected to be dominated by bulk impurity and interface roughness scattering, while electronic many-body effects have traditionally been neglected. However, for high-quality samples such as the one used in the experiment discussed here, this is no longer justified.

In the experiment[1] two parameters were controlled independently: the electronic sheet density \( N_e \) (from 0.05 to 1.3 x 10\(^{11}\) cm\(^{-2}\)), and a static electric field \( E \) perpendicular to the well which pushes the electrons against one of its edges. This provides an ideal tool to distinguish interface roughness from other damping effects.

We describe ISB plasmons within a one-band effective-mass approximation with parabolic subbands[3] which is a widely used and, for our purposes, sufficiently accurate method for GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As quantum wells. The case of the clean quantum well was treated in detail in Ref. [2]. The non-interacting response function reads

\[
\chi_{KS}(q||, z, z', \omega) = \sum_{\mu=1}^{N_{occ}} \sum_{\nu=1}^{\infty} F^{\mu\nu}(q||, \omega) \varphi_{\mu}(z) \varphi_{\nu}(z'), \tag{129}
\]

where

\[
F^{\mu\nu}(q||, \omega) = -2 \int \frac{dk^||}{(2\pi)^2} \left[ \frac{f(\epsilon_\mu + k^2_l/2)}{q||k^|| + a_{\mu\nu}(q||) + \omega + i\eta} + \frac{f(\epsilon_\mu + k^2_l/2)}{q||k^|| + a_{\mu\nu}(q||) - \omega - i\eta} \right], \tag{130}
\]
\[ \sigma_{\mu
u}(q_{||}) = \frac{q_{||}^2}{2} + \epsilon_\nu - \epsilon_\mu. \]

\( f \) is the Fermi function at \( T = 0 \), and \( \eta \) is a positive infinitesimal. \( \epsilon_\mu \) and \( \varphi_\mu(z) \) are the Kohn-Sham energies and wave function (in LDA) of the quantum well. For the experimental range of \( N_s \), the system under study has 9 bound levels, only the lowest being occupied \((N_{\text{occ}} = 1)\).

We consider perturbations of the form \( \mu_{\text{ext,1}}(z, \omega) = E_0 z \), corresponding to monochromatic plane electromagnetic waves of amplitude \( E_0 \) polarized along the \( z \)-axis, the direction of growth of the quantum well. Having solved the response equation \( \chi_{\text{KS}} \) the photoabsorption cross section is then obtained as \( \sigma(\omega) = -(8\pi\omega/E_0c)\text{Im} \int dz \varphi_\mu(z, \omega) \) and can be directly compared with data from photoabsorption measurements. \( \sigma(\omega) \) has a peak at the plasmon frequency \( \Omega \) with linewidth (HWHM) \( \Gamma \).

In Fig. 1, we plot the dispersions \( \Omega(q_{||}) \) of the ISB and the intrasubband (or 2D) plasmon in the clean quantum well \((N_s = 1.0 \times 10^{11} \text{ cm}^{-2})\), calculated within ALDA, see Eq. [15]. The imaginary part of the Kohn-Sham response function \( \chi_{\text{KS}} \) determines the regime of damping by single-particle excitations (Landau damping), as indicated by the shaded region in Fig. 1. Outside that region, in particular at small \( q_{||} \), the plasmons are undamped in ALDA, for which \( f_{\text{sc}} \) is frequency-independent and real.

In reality, however, the absence of momentum conservation in the \( z \)-direction and coupling via Coulomb interaction opens the possibility of plasmon decay into more complicated excitations, such as multiple electron-hole pairs, even at \( q_{||} = 0 \). To take this effect into account, one has to go beyond the ALDA and include dynamical xc effects.

In Fig. 2 we show the electric field dependence of the ISB plasmon frequencies \( \Omega(q_{||} = 0) \) for different values of \( N_s \). In the experimental data, built-in electric fields are subtracted, so that \( \Omega(E) \) exhibits a minimum for \( E = 0 \) and rises quadratically for small fields. \( \Omega(E) \) increases most rapidly for the smallest \( N_s \), since higher electronic densities tend to screen the external electric field more efficiently. At the same time, the depolarization shift increases with \( N_s \). As a consequence, the curves of \( \Omega(E) \) for different \( N_s \) are crossing each other. These features are very well reproduced by theory. Ignoring dynamical xc effects (i.e., using RPA) induces a 10% blueshift of \( \Omega \), which then compares less favorably with experiment.

To demonstrate the importance of including many-body effects in the response equation, we plot the bare subband spacings in the bottom panel of Fig. 3. The results are clearly qualitatively wrong: there is no crossing of \( \Omega(E) \) for different \( N_s \), in contradiction to experiment.
The calculated Γ for the clean quantum well (intrinsic damping through electron-electron interaction only), shown in the bottom panel of Fig. 3, lies clearly below the experimental values, which is hardly surprising. However, it can be seen that these purely electronic effects are far from negligible, at least for $N_s$ not too small, and provide an intrinsic lower limit to the linewidth of order 0.1 to 0.2 meV for $N_s \sim 10^{11} \text{cm}^{-2}$.

The middle part of Fig. 3 shows $\Gamma(E)$ calculated including electronic, impurity, and interface roughness damping, using the combined TCDFT and memory function formalisms outlined above, see Eqs. (113, 117). The results are now in very good agreement with experiment even away from flat band, as long as $E$ is not too large. We now briefly discuss the details of how the various contributions to extrinsic damping were modelled.

The presence of bulk impurities in the quantum well is mainly caused by segregation of donors from the lower delta-doped layer and diffusion along $z$ during growth. We use the functional form $n_i(z) = 1.33 \times 10^{-2}/30\text{nm} \times 10^{15} \text{cm}^{-3}$, proposed in Ref. 31 to explain in-plane mobility data, for the bulk impurity concentration in Eq. (126). We also include scattering from the upper delta-doped layer (remote impurity density $4.8 \times 10^{11} \text{cm}^{-2}$).

In Ref. 31, the in-plane mobility was found to be dominated by bulk impurity scattering. By contrast, it turns out that neither bulk nor remote impurities contribute much to the linewidth. The behavior of $\Gamma$ is instead dominated by interface roughness scattering and can in fact be qualitatively explained by it alone: Via (127), $\Gamma$ depends on the product of density fluctuations at the edges, which, for $E = 0$, have largest amplitude for highest $N_s$. For finite $E$, electrons get pushed towards one edge, but less so for higher densities due to screening of the external field. $\Gamma(E)$ thus rises more steeply for smaller $N_s$, and the curves cross.

We take a roughness scattering potential of the form $U(z) \delta(z-L/2)$. The height of the potential step for our quantum well is $\mu = 257.6 \text{meV}$. The roughness parameters are chosen as $\eta = 64.4\text{Å}$ and $\Delta = 4\text{Å}$, to give the best fit to experiment for the largest $N_s$.

Both $\eta$ and $\Delta$ are in the characteristic range found by lattice imaging techniques.

We also find that including electron-electron scattering does lead to a significant quantitative improvement for $\Gamma$, in particular for small $E$.

For $|E| \gtrsim 1 \text{mV/nm}$, the experimental linewidth saturates. This saturation can be understood as a negative feedback effect, related to the self-consistency of the memory functions (126) and (127). Roughly speaking, the plasmon linewidth comes from the imaginary part of the memory function, as in our calculations, thus means that rapidly saturates $\Gamma$. Neglect of the self-consistency of the memory function, as in our calculations, thus means that this saturation effect cannot be fully captured, as can be seen from Fig. 3.

VI. CONCLUSION

In this paper, we have dealt with a long-standing problem in the many-body theory of extended systems: the calculation of collective electronic excitations and their associated linewidths in systems that are both inho-
genuine and weakly disordered, in the sense that the random potential can be treated as a perturbation. In any real extended system, collective excitations are subject to dissipation, causing the associated coherent, plasmon-like motion to decay into many individual, incoherent degrees of freedom associated with single-particle excitations. It has been common practice in the literature to describe these processes with phenomenological assumptions of varying degrees of refinement. This paper, by contrast, presents a new formalism that allows one to calculate excitation energies and their lifetimes entirely from first principles.

Our approach begins with the basic notion that there are two classes of mechanisms that are responsible for dissipation of collective electronic dynamics. The first class is intrinsic scattering, which occurs even in a “perfect” material or device. Here we have in mind primarily systems where phonon scattering is inactive, so the only source of intrinsic dissipation are electronic many-body effects such as multiple particle-hole excitations. The second class of dissipation mechanisms is extrinsic in nature, such as scattering off impurities and disorder.

Our treatment of intrinsic scattering relies on TDFT for the linear response. Fundamental existence theorems guarantee that TDFT describes the linear dynamics of interacting many-electron systems in principle exactly, including dissipation of collective degrees of freedom. In practice, however, the success of a TDFT approach relies on the approximations used for the linearized xc potential. The most widely used approximation, the ALDA, has proved to be useful for calculating accurate excitation energies, but it produces linewidths that are strictly zero. Thus, a non-adiabatic description, which includes retardation, is required. A non-adiabatic dynamical density-functional approach which is local in space but nonlocal in time has to be formulated replacing the density with the current as basic variable (TCDFT). As a result, the linearized xc potential in the TCDFT response equation in general acquires a frequency dependence and an imaginary part, leading to finite linewidths.

To deal with extrinsic scattering, on the other hand, we make use of a powerful formal technique, the so-called memory function formalism. This approach can be traced back to the relaxation time approximation, but it replaces the simple phenomenological relaxation time $\tau$ with the memory function $M(q, \omega)$, which is defined microscopically as a correlation function between fluctuating random forces. The memory function formalism is developed in the language of Kubo relaxation functions, which are, however, intimately connected to the (current) density response functions.

The final step then consists in uniting the memory function formalism with linear response theory in TCDFT. We thus arrive at a new, self-consistent theory that expresses the response function of an interacting system in the presence of both intrinsic and extrinsic damping in terms of the “clean” interacting response function (which contains only intrinsic damping) and the memory function (which accounts only for extrinsic damping).

We finally applied the theory to describe ISB plasmons in a wide GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum well. Using reasonable values for the roughness parameters, we obtained quantitative agreement with the experimentally measured linewidth. But we also found that purely electronic damping due to dynamical exchange and correlations makes non-negligible contributions to the linewidth, especially for high electronic densities, where the effect can be as high as a few tens of percents.

A further remarkable outcome of this study is the physical insight that the ISB plasmon linewidth is primarily controlled by interfacial roughness, and only weakly affected by the concentration of bulk impurities. The opposite is true for the in-plane mobility, which is primarily controlled by bulk impurities. Thus, the correlation between ISB plasmon linewidth and in-plane mobility is rather weak, which is physically understandable since currents are flowing perpendicular to the quantum well in the former case, and parallel to it in the latter.

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