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 simultaneously account for intrinsic and extrinsic damping of collective excitations. As an example where both sources of dissipation are important and where high-quality experimental data are 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)[1–6] 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 some of the key elements of TDFT. Let

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

be the Hamiltonian of a many-electron system, where $\mathbf{r}_i$ and $\mathbf{p}_i$ are the canonical coordinates and momenta of the ith 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,[2,5] one adds to $\hat{H}_0$ a small time-dependent perturbation[7] of the form

$$\hat{H}_1(t) = \int d^3r v_1(\mathbf{r},t)\hat{n}(\mathbf{r}), \qquad (2)$$

where $v_1(\mathbf{r},t) = v_1(\mathbf{r},\omega)e^{-i\omega t} + \text{c.c.}$ 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

$$n(\mathbf{r},t) = n_0(\mathbf{r}) + n_1(\mathbf{r},\omega)e^{-i\omega t} + \text{c.c.}, \qquad (3)$$

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

$$n_1(\mathbf{r},\omega) = \int d^3r \chi(\mathbf{r},\mathbf{r}',\omega)v_1(\mathbf{r}',\omega). \qquad (4)$$

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 $\chi(\mathbf{r},\mathbf{r}',\omega)$ is to construct a *noninteracting* system that has the same ground-state density $n_0(\mathbf{r})$, and yields the same density response $n_1(\mathbf{r},\omega)$ as the interacting system under study. The dynamics of this noninteracting system is controlled by an effective single-particle potential that is written as the sum of the total external potential $v_0(\mathbf{r})+v_1(\mathbf{r},t)$ plus the Hartree potential

$$v_{\rm H}(\mathbf{r},t)=e^2\int d^3r'\frac{n(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|} \tag{5}$$

plus a remainder, which is known as the ''exchange-correlation'' (xc) potential $v_{\rm xc}(\mathbf{r},t)$. It is not at all obvious that such a potential $v_{\rm xc}$ can be constructed, but, if it can, then Runge and Gross[1] 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.[8] The effective noninteracting Hamiltonian (also known as the Kohn-Sham Hamiltonian) that yields the exact density is then given by

$$\hat{H}_{\rm KS}(t)=\sum_i\left\{\frac{p_i^2}{2m}+v_0(\mathbf{r}_i)+v_{\rm H,0}(\mathbf{r}_i)+v_{\rm xc,0}(\mathbf{r}_i)\right\} + \int d^3r[v_1(\mathbf{r},t)+v_{\rm H,1}(\mathbf{r},t)+v_{\rm xc,1}(\mathbf{r},t)]\hat{n}(\mathbf{r}), \tag{6}$$

where both the Hartree and the xc potentials have been written as the sum of static parts $v_{\rm H,0}, v_{\rm xc,0}$ associated with the ground-state density, and (small) time-dependent parts $v_{\rm H,1}, v_{\rm xc,1}$ associated with the time-dependent density. The static part of the Kohn-Sham Hamiltonian [first line of Eq. (6)] yields the exact ground-state density in the interacting system, while the time-dependent part [second line of Eq. (6)] yields the exact density response.

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

$$v_{\rm xc}(\mathbf{r},t)=v_{\rm xc,0}(\mathbf{r})+\int_{-\infty}^{t}dt'\int d^3r' f_{\rm xc}(\mathbf{r},\mathbf{r}',t-t')n_1(\mathbf{r}',t'), \tag{7}$$

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

$$f_{\rm xc}(\mathbf{r},\mathbf{r}',t-t')=\left.\frac{\delta v_{\rm xc}[n](\mathbf{r},t)}{\delta n(\mathbf{r}',t')}\right|_{n_0(\mathbf{r})}. \tag{8}$$

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

$$v_{\rm xc,1}(\mathbf{r},\omega)=\int d^3r' f_{\rm xc}(\mathbf{r},\mathbf{r}',\omega)n_1(\mathbf{r}',\omega). \tag{9}$$

We denote by $\chi_{\rm KS}(\mathbf{r},\mathbf{r}',\omega)$ the density-density response function of the *static* Kohn-Sham system [the first line of Eq. (6)]. 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

$$n_1(\mathbf{r},\omega)=\int d^3r'\chi_{\rm KS}(\mathbf{r},\mathbf{r}',\omega)\left\{v_1(\mathbf{r}',\omega) + \int d^3r''\left[\frac{e^2}{|\mathbf{r}'-\mathbf{r}''|}+f_{\rm xc}(\mathbf{r}',\mathbf{r}'',\omega)\right]n_1(\mathbf{r}'',\omega)\right\}, \tag{10}$$

where the second and the third terms in the curly brackets arise from $v_{\rm H,1}$ and $v_{\rm xc,1}$, respectively. Comparing Eqs. (4) and (10) we obtain the following integral relation between the exact density-density response function $\chi(\mathbf{r},\mathbf{r}',\omega)$ and the noninteracting response function $\chi_{\rm KS}(\mathbf{r},\mathbf{r}',\omega)$:

$$\chi(\mathbf{r},\mathbf{r}',\omega)=\chi_{\rm KS}(\mathbf{r},\mathbf{r}',\omega)+\int d^3x\chi_{\rm KS}(\mathbf{r},\mathbf{x},\omega) \times\int d^3y\left[\frac{e^2}{|\mathbf{x}-\mathbf{y}|}+f_{\rm xc}(\mathbf{x},\mathbf{y},\omega)\right]\chi(\mathbf{y},\mathbf{r}',\omega). \tag{11}$$

This can also be written as

$$\chi^{-1}(\mathbf{r},\mathbf{r}',\omega)=\chi_{\rm KS}^{-1}(\mathbf{r},\mathbf{r}',\omega)-\frac{e^2}{|\mathbf{r}-\mathbf{r}'|}-f_{\rm xc}(\mathbf{r},\mathbf{r}',\omega), \tag{12}$$

where χ^{-1} is the matrix inverse of χ.

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

$$\int d^3r'\chi^{-1}(\mathbf{r},\mathbf{r}',\omega)\Xi(\mathbf{r}',\omega)=0, \tag{13}$$

where $\Xi(\mathbf{r},\omega)$ is the function that describes the spatial dependence of the density in the excited state.

Equation (11) is the main formal result in the TDFT approach to the calculation of excitation energies. From a fundamental point of view, calculating $f_{\rm xc}$ is of course no easier than calculating χ. The main advantage of recasting linear response theory within TDFT is of a more practical nature: as long as the exact excitations of the interacting system are in qualitative correspondence to those of the Kohn-Sham system (a kind of ''Fermi liquid'' assumption), the xc kernel in Eq. (11) is expected to be a small correction, which can be approximated with relative impunity.[11] The simplest approximation is to ignore both nonlocality in space and retardation in time. This leads to the widely used *adiabatic local-density approximation*[12] (ALDA) in which one poses

$$v_{\rm xc}(\mathbf{r},t)=\left.\frac{d\epsilon_{\rm xc}(n)}{dn}\right|_{n=n(\mathbf{r},t)}, \tag{14}$$

where $\epsilon_{\rm xc}(n)$ is the xc energy density[13] of the homogeneous electron gas of density n. The right-hand side of Eq. (14) is nothing but the local-density approximation (LDA) for the *ground-state* xc potential evaluated at the time-dependent density. In terms of the xc kernel, this approximation implies

$$f_{\rm xc}(\mathbf{r},\mathbf{r}',\omega)=\delta(\mathbf{r}-\mathbf{r}')\frac{d^2\epsilon_{\rm xc}(n)}{dn^2}\bigg|_{n=n_0(\mathbf{r})}, \qquad (15)$$

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.[5,14] 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,[15] similar in spirit to Eq. (14) (i.e., still without retardation), yields a dramatic improvement in accuracy.[4] Applications to more complex systems[16–18] (molecules, polymers) have met with similar degrees of success. The essential reason seems to be that the frequency dependence of the xc kernel is rather weak, because it is controlled by multielectron 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 $f_{\rm xc}$ arises in the study of excitations that do not have an analog 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 intersubband 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[2] proposed a dynamical local-density approximation for $f_{\rm xc}$, which was designed to preserve the local relationship between $v_{\rm xc,1}$ and the density, while including retardation in time. Their approximation reads

$$f_{\rm xc}(\mathbf{r},\mathbf{r}',\omega)=f_{\rm xc}^h(k=0,\omega)\,\delta(\mathbf{r}-\mathbf{r}'), \qquad (16)$$

where $f_{\rm xc}^h(k,\omega)$ is the xc kernel of the uniform electron gas calculated at the local ground-state density $n_0(\mathbf{r})$ (more about $f_{\rm xc}^h$ will be said in the next sections). Because $f_{\rm xc}(\omega)$ is complex, this approximation yields a finite linewidth for excitations that would have zero linewidth in the ALDA.[19,20]

Unfortunately, the Gross-Kohn approximation (16) suffers from several inconsistencies, such as the failure to satisfy the generalized Kohn's theorem[21] and related sum rules.[22,23] As a consequence, it was found[24] 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 nonhomogeneous system is a function of infinite range in space, or, more precisely, the spatial Fourier transform $f_{\rm xc}(\mathbf{k},\mathbf{k}',\omega)$ diverges when $k\to 0$ at constant k' or vice versa. (In the homogeneous case one has $\mathbf{k}=\mathbf{k}'$ and the singularity disappears).

Vignale and Kohn[23] (VK) and Vignale, Ullrich, and Conti[25] (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 (1) with a time-dependent *vector potential* $\mathbf{a}_1(\mathbf{r},t)=\mathbf{a}_1(\mathbf{r},\omega)e^{-i\omega t}+$c.c. rather than with a scalar potential. The perturbing Hamiltonian has the form

$$\hat{H}_1(t)=\int d^3r\,\mathbf{a}_1(\mathbf{r},t)\cdot\hat{\mathbf{j}}_p(\mathbf{r}), \qquad (17)$$

where $\hat{\mathbf{j}}_p(\mathbf{r})=(1/2m)\Sigma_i[\hat{\mathbf{p}}_i\delta(\mathbf{r}-\mathbf{r}_i)+\delta(\mathbf{r}-\mathbf{r}_i)\hat{\mathbf{p}}_i]$ is the *paramagnetic* current density operator.[26] 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 $\mathbf{a}_{\rm xc,1}(\mathbf{r},t)$, which is a (linear) functional of the full current density response $\mathbf{j}_1(\mathbf{r},t)=\mathbf{j}_{p1}(\mathbf{r},t)+n_0(\mathbf{r})\mathbf{a}_1(\mathbf{r},t)/m$:

$$a_{{\rm xc},1,\alpha}(\mathbf{r},\omega)=\sum_\beta\int d^3r'\,f_{{\rm xc},\alpha\beta}(\mathbf{r},\mathbf{r}',\omega)j_{1,\beta}(\mathbf{r}',\omega), \qquad (18)$$

where $f_{{\rm xc},\alpha\beta}(\mathbf{r},\mathbf{r}',\omega)$ is the tensorial generalization of the usual xc kernel (here and in the following, α,β 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 $v_{\rm xc,0}$.

It turns out that the xc vector potential *does* admit a local approximation in terms of the current density: as we shall see in the next section, the form of this approximation is essentially determined by symmetry considerations and can be expressed in terms of an xc stress tensor.[25] The resulting expression for $\mathbf{a}_{\rm xc,1}$ is local in space, retarded in time, satisfies the generalized Kohn's theorem,[21,22] and allows a consistent calculation of the linewidth of elementary excitations, at least the part of it that arises from intrinsic many-body effects. The fundamental reason why all this is possible is that the relationship between the longitudinal current and the density is nonlocal. From the continuity equation

$$\frac{\partial n_1(\mathbf{r},t)}{\partial t}=-\boldsymbol{\nabla}\cdot\mathbf{j}_1(\mathbf{r},t) \qquad (19)$$

one sees that the longitudinal component of the current is given by

$$j_{1,L}(\mathbf{r},t)=\frac{1}{4\pi}\int d^3r'\frac{n_1(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|}, \qquad (20)$$

while the transverse component of the current remains undetermined. Thus, a local functional of $j_{1,L}$ will necessarily be a nonlocal functional of the density. What is remarkable here is that the nonlocality of $v_{\rm xc,1}$ as a functional of the density

can be completely eliminated by ''upgrading'' TDFT to a description in terms of $\mathbf{a}_{\mathrm{xc},1}$ and $\mathbf{j}_1$. This is the essence of the VK and VUC theories.

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 TDFT is a special case of the TCDFT formulation.[27] 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 plasmon in a 40-nm $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well reported in Ref. 24, based on VUC formalism, yielded a linewidth about five 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 achieve 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.[28] In the homogeneous electron gas limit this approach reduces to the Belitz–Das Sarma[29] 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[30] 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,[31,32] 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 Sec. II we review the main aspects of the TCDFT formalism for the linear response of many-electron systems. In Sec. 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 Sec. 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 Sec. 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 Sec. 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 xc kernels of a *homogeneous* electron gas. Because of translational invariance, it is convenient to work with the Fourier transform $\mathbf{j}(\mathbf{k},\omega)$ of the current density. The linear response of this quantity to a vector potential $\mathbf{a}_1(\mathbf{k},\omega)$ can be written as

$$j_{1,\alpha}(\mathbf{k},\omega)=\sum_{\beta}\chi_{\alpha\beta}(\mathbf{k},\omega)a_{1,\beta}(\mathbf{k},\omega), \tag{21}$$

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

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

where $L(T)$ denotes the longitudinal (transverse) component, and $\chi_{L(T)}(\mathbf{k},\omega)$ is the longitudinal (transverse) response function. According to the general linear response formalism,[33] $\chi_{L(T)}$ is given by

$$\chi_{L(T)}(\mathbf{k},\omega)=\frac{n}{m}+\sum_{l}|\langle l|\hat{j}_{p,L(T)}(\mathbf{k})|0\rangle|^2 \times\left\{\frac{1}{\omega-\omega_{l0}+i\eta}-\frac{1}{\omega+\omega_{l0}+i\eta}\right\}, \tag{23}$$

where $|0\rangle$ is the ground state, $|l\rangle$ is the lth excited state, and ω_{l0} is the excitation energy E_l-E_0. The Fourier transform of the paramagnetic current operator $\hat{\mathbf{j}}_p(\mathbf{k})$ is given by

$$\hat{\mathbf{j}}_p(\mathbf{k})=\frac{1}{2m}\sum_i\left[\hat{\mathbf{p}}_i e^{-i\mathbf{k}\cdot\mathbf{r}_i}+e^{-i\mathbf{k}\cdot\mathbf{r}_i}\hat{\mathbf{p}}_i\right]. \tag{24}$$

Note that the total current response is the sum of the ''London current'' $n\mathbf{a}_1/m$ and the paramagnetic current [the ex-

pectation value of Eq. (24)]. A key feature of Eq. (23) is that in the limit of $k\to 0$ and finite ω it approaches the form

$$\chi_L(k,\omega)\to \frac{n}{m\epsilon(\omega)}+\alpha_L(\omega)k^2,$$
$$\chi_T(k,\omega)\to \frac{n}{m}+\alpha_T(\omega)k^2, \tag{25}$$

where $\alpha_{L(T)}(\omega)$ are functions of frequency only, $\epsilon(\omega)=1-\lim_{k\to 0} nv(k)k^2/m\omega^2$ 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 χ'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 Eqs. (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 (22) as the response of a noninteracting electron gas to an effective vector potential, written as

$$\mathbf{a}_{\mathrm{eff},1}(\mathbf{k},\omega)=\mathbf{a}_1(\mathbf{k},\omega)+\mathbf{a}_{\mathrm{H},1}(\mathbf{k},\omega)+\mathbf{a}_{\mathrm{xc},1}(\mathbf{k},\omega). \tag{26}$$

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

$$\mathbf{a}_{\mathrm{H},1}(\mathbf{k},\omega)=\frac{k^2}{\omega^2}v(k)j_{1,L}(\mathbf{k},\omega)\hat{\mathbf{k}}, \tag{27}$$

where $\hat{\mathbf{k}}=\mathbf{k}/k$. The xc potential can be decomposed into its longitudinal and transverse components (with respect to the direction of $\mathbf{k}$) as follows:

$$\mathbf{a}_{\mathrm{xc},1}(\mathbf{k},\omega)=\frac{k^2}{\omega^2}[f^h_{\mathrm{xc},L}(k,\omega)j_{1,L}(\mathbf{k},\omega)\hat{\mathbf{k}} + f^h_{\mathrm{xc},T}(k,\omega)\mathbf{j}_{1,T}(\mathbf{k},\omega)]. \tag{28}$$

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

$$v_{\mathrm{xc},1}(\mathbf{k},\omega)=\frac{\omega a_{\mathrm{xc},1,L}}{k}=f^h_{\mathrm{xc},L}(k,\omega)n_1(\mathbf{k},\omega) \tag{29}$$

($j_L=n_1\omega/k$), thus making $f^h_{\mathrm{xc},L}(k,\omega)$ identical with the usual $f^h_{\mathrm{xc}}(k,\omega)$ of the ordinary TDFT.[2]

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

$$\chi^{-1}_{L(T)}(k,\omega)=\chi^{-1}_{\mathrm{KS},L(T)}(k,\omega)-\frac{k^2}{\omega^2}[v_{L(T)}(k)+f^h_{\mathrm{xc},L(T)}(k,\omega)], \tag{30}$$

where we have defined $v_L(k)=v(k)$ and $v_T(k)=0$.

Notice that, according to Eqs. (25), $\chi_L^{-1}(k,\omega)$ and $\chi^{-1}_{\mathrm{KS},L}(k,\omega)-k^2v_L(k)/\omega^2$ have the same limit $m\epsilon(\omega)/n+O(k^2)$ for $k\to 0$ and finite ω. Similarly, $\chi_T^{-1}(k,\omega)$ and $\chi^{-1}_{\mathrm{KS},T}(k,\omega)-k^2v_T(k)/\omega^2$ have the same limit $m/n+O(k^2)$. Thus, we see that Eq. (30) is consistent with the limiting forms (25) if and only if the $k\to 0$ limits of the xc kernels $f^h_{\mathrm{xc},L(T)}$ are finite functions of frequency,

$$\lim_{k\to 0} f^h_{\mathrm{xc},L(T)}(k,\omega)\equiv f^h_{\mathrm{xc},L(T)}(\omega). \tag{31}$$

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 $f^h_{\mathrm{xc},L(T)}(\omega)$ 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,[2] (ii) Perturbation theory,[34,35] and (iii) Mode-decoupling approximations.[36] 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

$$\lim_{\omega\to\infty} f^h_{\mathrm{xc},L(T)}(\omega)=\frac{1}{2n}[d_{L(T)}(\langle ke\rangle-\langle ke\rangle_0)+e_{L(T)}\langle pe\rangle], \tag{32}$$

which is also known as the *third-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, $d_L=4$, $e_L=8/15$, $d_T=4/3$, $e_T=-4/15$. In two dimensions, $d_L=6$, $e_L=5/4$, $d_T=2$, $e_T=-1/4$ (see Ref. 36).

The behavior of the imaginary part of the longitudinal xc kernel was first determined by Glick and Long[34] [three-dimensional (3D) case] and Holas and Singwi[35] (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[36] by a different method based on the equations of motion for the current response function. The result is

$$\lim_{\omega\to\infty} \mathrm{Im}\, f^h_{\mathrm{xc},L(T)}(\omega)=-a_{L,T}\pi^{4-D}\omega^{-D/2}\frac{e^2}{a_0^{1-D}}, \tag{33}$$

where $a_0=\hbar^2/me^2$ is the Bohr radius, $\bar{\omega}=\hbar\omega a_0/e^2$ 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,[37] and is subtly different from the static limit, which is obtained by setting $\omega=0$ *before* letting $k\to 0$. The result for $f_{\mathrm{xc},L}$ in D dimensions is

$$\lim_{\omega\to 0} f^h_{\mathrm{xc},L}(\omega)=\epsilon''_{\mathrm{xc}}(n)+\frac{2(D-1)}{D}\lim_{\omega\to 0} f^h_{\mathrm{xc},T}(\omega), \qquad (34)$$

where $\epsilon''_{\mathrm{xc}}(n)=d^2\epsilon_{\mathrm{xc}}(n)/dn^2$. The second term on the right-hand side of this expression is proportional to

$$\lim_{\omega\to 0} f^h_{\mathrm{xc},T}(\omega)=\frac{2\varepsilon_F}{25n}\left(\frac{3F_2-5F_1}{3+F_1}\right), \qquad (35)$$

in three dimensions, and

$$\lim_{\omega\to 0} f^h_{\mathrm{xc},T}(\omega)=\frac{\varepsilon_F}{2n}\left(\frac{F_2-F_1}{2+F_1}\right) \qquad (36)$$

in two dimensions, where ε_F is the Fermi energy, and F_l are the conventional Landau parameters of the electron liquid.[33] Note that the first term on the right-hand side of Eq. (34) is the usual compressibility obtained from the static limit $\lim_{k\to 0}\lim_{\omega\to 0} f^h_{\mathrm{xc},L}(k,\omega)$. 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 ω 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:

$$\mathrm{Re}\, f^h_{\mathrm{xc}}(\omega)=f^h_{\mathrm{xc}}(\infty)+\frac{1}{\pi}P\int d\omega'\frac{\mathrm{Im}\, f^h_{\mathrm{xc}}(\omega')}{\omega'-\omega}, \qquad (37)$$

where P denotes the ''principal part'' integral.

4. In a very recent development[38] 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

$$\mathrm{Im}\, f^h_{\mathrm{xc},T}(\omega)\to -\frac{1}{(na_0^D)^2}\frac{\eta}{(\hbar/a_0^D)}\frac{-e^2}{\omega a_0^{1-D}}, \qquad (38)$$

and

$$\mathrm{Im}\, f^h_{\mathrm{xc},L}(\omega)\to \frac{2(D-1)}{D}\mathrm{Im}\, f^h_{\mathrm{xc},T}(\omega), \qquad (39)$$

where the dimensionless ''shear viscosity'' $\eta/(\hbar/a_0^D)$ is given by

$$\frac{\eta}{(\hbar/a_0^D)}=-\frac{k_Fa_0}{45\pi^3}\left\{5-\left(\lambda+\frac{5}{\lambda}\right)\tan^{-1}\lambda-\frac{2}{\lambda}\sin^{-1}\frac{\lambda}{\sqrt{1+\lambda^2}}+\frac{2}{\lambda\sqrt{2+\lambda^2}}\left[\frac{\pi}{2}-\tan^{-1}\frac{1}{\lambda\sqrt{2+\lambda^2}}\right]\right\} \qquad (40)$$

in three dimensions, and

$$\frac{\eta}{(\hbar/a_0^D)}=\frac{1}{12\pi^2}\left\{2\left[\ln(\lambda+1)-\frac{\lambda}{1+\lambda}\right]-\int_0^1 dx\frac{\lambda^2x}{(\lambda x+1)(\lambda\sqrt{1-x^2}+1)}\right\} \qquad (41)$$

in two dimensions. In the above expressions λ is defined as $\lambda=2k_F/k_s$, where k_F is the Fermi wave vector, and k_s is the screening wave vector: in random-phase approximation (RPA), for example, $k_s=(4\pi k_Fa_0)^{1/2}/\pi a_0$ in three dimensions, and $k_s=2/a_0$ in two dimensions. The derivation of these results is presented in Ref. 38.

5. Parametrized expressions. To keep our presentation self-contained, we also include the explicit parametrization for $f^h_{\mathrm{xc},L}(\omega)$ that has been used in the calculations of Sec. V. This is the original Iwamoto-Gross-Kohn parametrization,[2,39] and has the form

$$\mathrm{Im}\, f^h_{\mathrm{xc},L}(\omega)=\frac{a(n)\omega}{[1+b(n)\omega^2]^{5/4}} \qquad (42)$$

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 $f^h_{\mathrm{xc},L}(\omega)$ is then calculated with the help of the dispersion relation (37). More recent analytic expressions for $f^h_{\mathrm{xc},L}(\omega)$ and $f^h_{\mathrm{xc},T}(\omega)$ have been obtained by Nifosi, Conti, and Tosi[36] and Qian and Vignale (QV).[38] These new expressions possess considerable structure in the frequency dependence due to two-plasmon excitations. The QV expression reproduces the exact perturbative limit of $f^h_{\mathrm{xc}}(\omega)$ 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

$$\mathbf{E}_{\mathrm{xc},1}(\mathbf{k},\omega)=i\omega\mathbf{a}_{\mathrm{xc},1}(\mathbf{k},\omega)=-\frac{1}{i\omega}\{\mathbf{k}[\mathbf{k}\cdot\mathbf{j}_1(\mathbf{k},\omega)]f^h_{\mathrm{xc},L}(\omega)+k^2f^h_{\mathrm{xc},T}(\omega)\mathbf{j}_{1,T}(\mathbf{k},\omega)\}. \qquad (43)$$

Splitting off the familiar ALDA contribution

$$\mathbf{E}^{\mathrm{ALDA}}_{\mathrm{xc},1}(\mathbf{k},\omega)=i\mathbf{k}\epsilon''_{\mathrm{xc}}\frac{\mathbf{k}\cdot\mathbf{j}_1}{\omega}, \qquad (44)$$

we can write

$$\mathbf{E}_{\mathrm{xc},1}(\mathbf{k},\omega)=\mathbf{E}_{\mathrm{xc},1}^{\mathrm{ALDA}}(\mathbf{k},\omega)-\frac{1}{i\omega}\{\mathbf{k}[\mathbf{k}\cdot\mathbf{j}_1(\mathbf{k},\omega)]\times[f_{\mathrm{xc},L}^h(\omega)-\epsilon_{\mathrm{xc}}'']+k^2 f_{\mathrm{xc},T}^h(\omega)\mathbf{j}_{1,T}(\mathbf{k},\omega)\}. \tag{45}$$

Introducing at this point the velocity field

$$\mathbf{u}=\frac{\mathbf{j}_1}{n} \tag{46}$$

and Fourier transforming Eq. (45) to real space, we get

$$\mathbf{E}_{\mathrm{xc},1}(\mathbf{r},\omega)=\mathbf{E}_{\mathrm{xc},1}^{\mathrm{ALDA}}(\mathbf{r},\omega)+\frac{n}{i\omega}\{[f_{\mathrm{xc},L}^h(\omega)-f_{\mathrm{xc},T}^h(\omega)-\epsilon_{\mathrm{xc}}'']\boldsymbol{\nabla}(\boldsymbol{\nabla}\cdot\mathbf{u})+f_{\mathrm{xc},T}^h(\omega)\nabla^2\mathbf{u}\}. \tag{47}$$

It is easy to verify that this expression can be rewritten as

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

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

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

and

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

$$\tilde{\zeta}(n,\omega)=-\frac{n^2}{i\omega}\left(f_{\mathrm{xc},L}^h(\omega)-\frac{2(D-1)}{D}f_{\mathrm{xc},T}^h(\omega)-\epsilon_{\mathrm{xc}}''\right) \tag{51}$$

are generalized (i.e., frequency-dependent and complex) viscoelastic constants of the electron liquid.[37] In particular, the *real* parts of $\tilde{\eta}$ and $\tilde{\zeta}$ [related to the imaginary parts of $f_{\mathrm{xc},L(T)}^h(\omega)/\omega$] 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 $f_{\mathrm{xc},L(T)}^h(\omega)$] are interpreted as post-ALDA xc contributions to frequency-dependent elastic constants μ (shear modulus) and K (bulk modulus) of the electron liquid: $\mu_{\mathrm{dyn}}=n^2\mathrm{Re}\,f_{\mathrm{xc},T}^h(\omega)$ and $K_{\mathrm{dyn}}=n^2\mathrm{Re}\{f_{\mathrm{xc},T}^h(\omega)-[2(D-1)/D]f_{\mathrm{xc},T}^h(\omega)-\epsilon_{\mathrm{xc}}''\}$ (see Ref. 37 for details; the full elastic constants μ and K given there also include the kinetic and the ALDA part of the xc contribution).

Because $\mathrm{Im}\,f_{\mathrm{xc},L(T)}^h(\omega)$ vanish linearly for $\omega\to0$ (point 4 of Sec. II B), we see that the viscosity coefficients $\tilde{\eta}$ and $\tilde{\zeta}$ stay finite in the $\omega\to0$ limit. Equations (38), (39) of Sec. II B imply that the $\omega\to0$ limit of the bulk viscosity $\lim_{\omega\to0}\tilde{\zeta}(\omega)$ is exactly zero at least to within the accuracy of our perturbative calculation. By virtue of the limiting form (34), we also see that $\lim_{\omega\to0}K_{\mathrm{dyn}}(\omega)=0$, implying that the bulk modulus of the electron liquid is entirely accounted for by the ALDA contribution ϵ_{xc}''.

The fact that the shear modulus does not vanish for $\omega\to0$ but tends to the finite value of Eq. (34) 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 $k\to0$ limit *before* the $\omega\to0$ limit. Thus, the system remains ''dynamical'' down to zero frequency. Of course, this would not be true if the $\omega\to0$ limit were taken at finite k. In that limit, $f_{\mathrm{xc},T}^h$ 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. (48), 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 through the replacement $n\to n_0(\mathbf{r})$, where $n_0(\mathbf{r})$ 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 $n\to n_0(\mathbf{r})$ be done in Eq. (48) rather than in one of the many equivalent expressions one can generate starting from Eq. (45)? For example, why not write the second term on the right-hand side of Eq. (47) in the equivalent form

$$\frac{1}{i\omega}\{[f_{\mathrm{xc},L}^h(\omega)-f_{\mathrm{xc},T}^h(\omega)-\epsilon_{\mathrm{xc}}'']\boldsymbol{\nabla}(\boldsymbol{\nabla}\cdot\mathbf{j}_1)+f_{\mathrm{xc},T}^h(\omega)\nabla^2\mathbf{j}_1\} \tag{52}$$

before substituting n by $n_0(\mathbf{r})$? 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. (48).

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.[40]

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. (48) as the correct expression in which the substitution $n \to n_0(\mathbf{r})$ should be made. This expression was originally derived by VK on 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 γ and small wave vector $\mathbf{q}$. Both k and q were assumed to be small not only relative to the Fermi momentum k_F but also relative to ω/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 $f_{\mathrm{xc},\alpha\beta}$ 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 that are mathematically equivalent to the zero-force and zero-torque requirements discussed above. The result of the analysis was that the diagonal matrix elements $f_{\mathrm{xc},\alpha\beta}(\mathbf{k},\mathbf{k},\omega)$ remain equal to $f^h_{\mathrm{xc}}(k,\omega)$ to first order in γ, but the off-diagonal elements $f_{\mathrm{xc},\alpha\beta}(\mathbf{k}+\mathbf{q},\mathbf{k},\omega)$ acquire a finite value, given by[23]

$$
\begin{aligned}
&f_{\mathrm{xc},\alpha\beta}(\mathbf{k}+\mathbf{q},\mathbf{k},\omega) \\
&\quad = -\frac{\gamma}{\omega^2}\Bigg\{ (\delta f^h_{\mathrm{xc},L} - f^h_{\mathrm{xc},T}) q_\alpha q_\beta + f^h_{\mathrm{xc},T} q^2 \delta_{\alpha\beta} \\
&\qquad - n \frac{\partial f^h_{\mathrm{xc},T}}{\partial n} \mathbf{k}\cdot(\mathbf{k}+\mathbf{q})\,\delta_{\alpha\beta} + \mathcal{A}(n,\omega) \\
&\qquad \times (k_\alpha + q_\alpha) k_\beta - \mathcal{B}(n,\omega) k_\alpha (k_\beta + q_\beta) \Bigg\}, \quad (53)
\end{aligned}
$$

where $\delta f^h_{\mathrm{xc},L} \equiv f^h_{\mathrm{xc},L}(\omega,n) - \epsilon''_{xc}(n)$, $\mathcal{A}(n,\omega) \equiv [n(2\partial f^h_{\mathrm{xc},T}/\partial n - \partial f^h_{\mathrm{xc},L}/\partial n + 3f^h_{\mathrm{xc},T} - \delta f^h_{\mathrm{xc},L}]$ and $\mathcal{B}(n,\omega) \equiv [n\partial f^h_{\mathrm{xc},T}/\partial n + 3f^h_{\mathrm{xc},T} - \delta f^h_{\mathrm{xc},L}]$. A remarkable feature of this result is that the off-diagonal matrix elements of $f_{\mathrm{xc},\alpha\beta}$ 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 $\mathbf{k}\cdot\mathbf{q}/k^2$ 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.

Equation (53) can be translated into a real-space expression for $\mathbf{E}_{\mathrm{xc},1}(\mathbf{r},\omega)$. More details of the derivation, which is quite laborious, are given in Ref. 23. Finally, the resulting expression can be rearranged[25] in the elegant form of Eq. (48). The conditions of validity of the real-space approach are $|\boldsymbol{\nabla} n_0(\mathbf{r})|/n_0(\mathbf{r})$ much smaller than $k_F(\mathbf{r})$ and $\omega/u_F(\mathbf{r})$, where $k_F(\mathbf{r})$ and $u_F(\mathbf{r})$ are the local Fermi momentum and velocity. In Ref. 24, 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 tunneling 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 (16) provides a pragmatic and practically useful remedy.[24]

Because the occurrence of two spatial derivatives of the velocity field in the post-ALDA term is dictated by general principles, Eq. (48) 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. (48) 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. (48), written in the time domain, could provide an appropriate description of both linear and nonlinear response properties. A nonlinear, retarded expression for $v_{\mathrm{xc},1}$ was also proposed by Dobson *et al.*[41] 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 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,[42] which then naturally leads to the so-called memory function formalism.[28,43] 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 Sec. 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 formal-

ism for the homogeneous case. We then make contact between relaxation and response functions, thereby integrating the memory function formalism with TCDFT. In Sec. IV 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, $\{\hat{A}_0(\mathbf{r},t), \ldots, \hat{A}_N(\mathbf{r},t)\}$, each $\hat{A}_i(\mathbf{r},t)$ coupling to a small perturbing external field $\delta a_i(\mathbf{r},t)$. The Hamiltonian in the presence of these fields is given (in Schrödinger representation, i.e., $\hat{A}_i$ independent of t) by

$$\hat{H}(t)=\hat{H}_0+\sum_i \int d^3r \hat{A}_i(\mathbf{r})\,\delta a_i(\mathbf{r},t). \tag{54}$$

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

$$\delta a_i(\mathbf{r},t)=\begin{cases}\delta a_i(\mathbf{r})e^{\eta t} & \text{for } t\leqslant 0\\ 0 & \text{for } t>0.\end{cases} \tag{55}$$

The system starts out from nonequilibrium at $t=0^+$ and, being left to itself, relaxes back towards equilibrium. The first-order change of the nonequilibrium expectation value of an observable can then be written as[28,42–44]

$$\delta\langle\hat{A}_i(\mathbf{r},t)\rangle_{\text{noneq.}}=\sum_j \int d^3r'\,\tilde{C}_{ij}(\mathbf{r},\mathbf{r}',t)\,\delta a_j(\mathbf{r}'), \tag{56}$$

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

$$\tilde{C}_{ij}(\mathbf{r},\mathbf{r}',t)=\Big\langle \int_0^\beta d\beta' [\langle\hat{A}_i(\mathbf{r},t)\hat{A}_j(\mathbf{r}',-i\beta')\rangle_{\text{eq.}} - \langle\hat{A}_i(\mathbf{r},t)\rangle_{\text{eq.}}\langle\hat{A}_j(\mathbf{r}',-i\beta')\rangle_{\text{eq.}}]\Big\rangle_{\text{disorder}}. \tag{57}$$

This may be rewritten as

$$\tilde{C}_{ij}(\mathbf{r},\mathbf{r}',t)=\langle\hat{A}_i(\mathbf{r})|e^{-i\mathcal{L}t}|\hat{A}_j(\mathbf{r}')\rangle, \tag{58}$$

where $\mathcal{L}$ is the Liouville operator governing time evolution of the system via

$$\dot{\hat{A}}_i(t)=i\mathcal{L}\hat{A}_i(t)=[\hat{A}_i(t),\hat{H}]/(i\hbar), \tag{59}$$

and the scalar product $\langle\cdots|\cdots\rangle$ is defined by Eq. (57). We impose normalization on the set of variables, i.e., $\langle\hat{A}_i(\mathbf{r})|\hat{A}_j(\mathbf{r}')\rangle=\delta_{ij}$. In the following, we are interested in the zero temperature limit ($\beta\to\infty$).

The correlation function (57) is related to the dissipative part of the usual quantum mechanical response functions $\chi_{ij}(\mathbf{r},\mathbf{r}',t)$ as follows:[28,42]

$$i\partial_t\tilde{C}_{ij}(\mathbf{r},\mathbf{r}',t)=2\chi''_{ij}(\mathbf{r},\mathbf{r}',t), \tag{60}$$

where $\chi''_{ij}(\mathbf{r},\mathbf{r}',t)$ denotes the inverse Fourier transform of $\text{Im}\,\chi_{ij}(\mathbf{r},\mathbf{r}',\omega)$.

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

$$\tilde{C}_{ij}(\mathbf{r},\mathbf{r}',\xi)=\int_0^\infty dt e^{i\xi t}\tilde{C}_{ij}(\mathbf{r},\mathbf{r}',t), \tag{61}$$

where ξ 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:

$$\tilde{C}_{ij}(\mathbf{r},\mathbf{r}',\xi)=[\chi_{ij}(\mathbf{r},\mathbf{r}',\xi)-\chi_{ij}(\mathbf{r},\mathbf{r}',i0)]/(i\xi). \tag{62}$$

For the remainder of this section, we assume that the system is spatially homogeneous (a more general case will be considered in Sec. IV). Equation (56) can then be Fourier transformed into momentum space, and one obtains

$$\delta\langle\hat{A}_i(\mathbf{q},\xi)\rangle_{\text{noneq.}}=\sum_j \tilde{C}_{ij}(\mathbf{q},\xi)\,\delta a_j(\mathbf{q}). \tag{63}$$

The Laplace transform of the correlation function (58) is then given by

$$\tilde{C}_{ij}(\mathbf{q},\xi)=\langle\hat{A}_i(\mathbf{q})|\frac{i}{\xi-\mathcal{L}}|\hat{A}_j(\mathbf{q})\rangle. \tag{64}$$

B. Projectors and memory functions

The observables $\{\hat{A}_0(\mathbf{q}), \ldots, \hat{A}_N(\mathbf{q})\}$ can be regarded as vectors in a Hilbert space. The Liouvillian $\mathcal{L}$ acts as a linear operator in that space, see Eq. (59). We define a projection operator $\mathcal{P}$ onto the space spanned by $\{\hat{A}_0(\mathbf{q}), \ldots, \hat{A}_N(\mathbf{q})\}$ as

$$\mathcal{P}=\sum_i |\hat{A}_i(\mathbf{q})\rangle\langle\hat{A}_i(\mathbf{q})|, \tag{65}$$

and its complement $\mathcal{Q}\equiv 1-\mathcal{P}$ projects perpendicular to it. One can then formally write Eq. (64) as

$$\tilde{C}_{ij}(\mathbf{q},\xi)=\langle\hat{A}_i(\mathbf{q})|\frac{i}{\xi-\mathcal{L}\mathcal{Q}-\mathcal{L}\mathcal{P}}|\hat{A}_j(\mathbf{q})\rangle. \tag{66}$$

Following Forster,[28] one performs a few straightforward manipulations in Eq. (66) and finds

$$\tilde{C}_{ij}(\mathbf{q},\xi)=\frac{i}{\xi}\delta_{ij}+\frac{1}{\xi}\sum_k \Big\{\langle\hat{A}_i(\mathbf{q})|\mathcal{L}|\hat{A}_k(\mathbf{q})\rangle - \langle\dot{\hat{A}}_i(\mathbf{q})|\mathcal{Q}\frac{1}{\mathcal{Q}\mathcal{L}\mathcal{Q}-\xi}\mathcal{Q}|\dot{\hat{A}}_k(\mathbf{q})\rangle\Big\}\tilde{C}_{kj}(\mathbf{q},\xi). \tag{67}$$

Defining

$$C_{ij}(\mathbf{q},\xi) = i\tilde{C}_{ij}(\mathbf{q},\xi), \tag{68}$$

$$\Omega_{ik}(\mathbf{q}) = \langle \hat{A}_i(\mathbf{q}) | \mathcal{L} | \hat{A}_k(\mathbf{q}) \rangle, \tag{69}$$

$$M_{ik}(\mathbf{q},\xi) = \langle \dot{\hat{A}}_i(\mathbf{q}) | \mathcal{Q} \frac{1}{\mathcal{Q}\mathcal{L}\mathcal{Q} - \xi} \mathcal{Q} | \dot{\hat{A}}_k(\mathbf{q}) \rangle, \tag{70}$$

we rewrite Eq. (67) as follows:

$$\sum_k \{\xi \delta_{ik} - \Omega_{ik}(\mathbf{q}) + M_{ik}(\mathbf{q},\xi)\} C_{kj}(\mathbf{q},\xi) = -\delta_{ij}. \tag{71}$$

$\Omega_{ij}(\mathbf{q})$ can be viewed as characteristic frequency or restoring force matrix of the system. Our particular interest, however, lies in the memory function matrix $M_{ik}(\mathbf{q},\xi)$, 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.[45] 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, $\mathcal{Q}\dot{\hat{A}}_i(\mathbf{q})$ and $\mathcal{Q}\dot{\hat{A}}_j(\mathbf{q})$. 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 $M_{ik}(\mathbf{q},\xi)$ is determined by $\mathcal{Q}\mathcal{L}\mathcal{Q}$, 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 $C_{ij}(\mathbf{q},\xi)$ are determined by a set of $(N+1)^2$ coupled equations, Eq. (71), whose solution will be discussed for an example in Sec. III C. The observables $\hat{A}_i$ are not restricted to be scalars, but can also be vectors (or nth-rank tensors). In general, all correlation functions $C_{ij}(k,\omega)$ as well as Ω_{ij} and M_{ij} are tensors whose rank equals the sum of the ranks of $\hat{A}_i$ and $\hat{A}_j$.

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 ω) rather than Laplace transforms (with frequency ξ) 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 $\chi_{ij}(\mathbf{q},\omega)$ can be simply obtained from

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

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

C. Generalized relaxation time approximation

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

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

and

$$\hat{A}_1(\mathbf{q}) = \frac{\hat{\mathbf{j}}(\mathbf{q})}{\sqrt{n}}, \tag{74}$$

where n is the uniform density of the system, and $\langle \hat{\rho}(\mathbf{q}) | \hat{\rho}(\mathbf{q}) \rangle = \chi(\mathbf{q},0)$, the static density-density response function (in the presence of disorder). Equation (71) then describes the four associated correlation functions C_{00}, C_{01}, C_{10}, and C_{11}, i.e., $\hat{\rho}$-$\hat{\rho}$, $\hat{\rho}$-$\hat{\mathbf{j}}$, $\hat{\mathbf{j}}$-$\hat{\rho}$, and $\hat{\mathbf{j}}$-$\hat{\mathbf{j}}$.

Since we deal with the homogeneous and isotropic case, Eq. (71) decouples into two independent equations for the longitudinal (L) and transverse (T) components of the correlation functions,

$$\sum_{k=0}^{1} [\omega \delta_{ik} - \Omega^L_{ik}(\mathbf{q}) + M^L_{ik}(\mathbf{q},\omega)] C^L_{kj}(\mathbf{q},\omega) = -\delta_{ij}, \tag{75}$$

$$[\omega - \Omega^T_{11}(\mathbf{q}) + M^T_{11}(\mathbf{q},\omega)] C^T_{11}(\mathbf{q},\omega) = -1. \tag{76}$$

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

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

we convince ourselves that indeed $\langle \hat{A}_0 | \hat{A}^L_1 \rangle = 0$. Furthermore, since $\mathcal{L}\hat{A}_0$ is proportional to $\hat{A}^L_1$, the first component of the ''perpendicular'' force $\mathcal{Q}\dot{\hat{A}}_0$ is identically zero, so that

$$M^L_{00}(\mathbf{q},\omega) = M^L_{01}(\mathbf{q},\omega) = M^L_{10}(\mathbf{q},\omega) = 0. \tag{78}$$

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 not affected by disorder. This means that $\Omega^{L(T)}(\mathbf{q})$ contains effects of Coulomb interaction only. Likewise, we assume

$$\chi^{L(T)}_{ij}(\mathbf{q},0) = \chi^{Lc(Tc)}_{ij}(\mathbf{q},0), \tag{79}$$

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 τ 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:

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

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

$$\Omega_{ik}^{{\rm in},L(T)}(\mathbf{q},\omega) \equiv \Omega_{ik}^{L(T)}(\mathbf{q}) + M_{\rm in}^{L(T)}(\mathbf{q},\omega)\,\delta_{i1}\delta_{k1}. \tag{81}$$

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

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

$$[\omega + M_{\rm ex}^{T}(\mathbf{q},\omega) - \Omega_{11}^{{\rm in},T}(\mathbf{q},\omega)] C_{11}^{T}(\mathbf{q},\omega) = -1. \tag{83}$$

For notational brevity, we suppress the $(\mathbf{q},\omega)$ dependence and the subscript "ex" of $M_{\rm ex}^{L}$ and $M_{\rm ex}^{T}$ in the following. To solve Eqs. (82) and (83), we introduce "clean" longitudinal (transverse) reference functions $C_{ij}^{Lc(Tc)}(\mathbf{q},\omega)$, defined in the absence of dissipation (i.e., $M^{L}=0=M^{T}$), as follows:

$$\sum_{k=0}^{1} [\omega\,\delta_{ik} - \Omega_{ik}^{{\rm in},L}(\mathbf{q},\omega)] C_{kj}^{Lc}(\mathbf{q},\omega) = -\delta_{ij}, \quad i,j=0,1, \tag{84}$$

$$[\omega - \Omega_{11}^{{\rm in},T}(\mathbf{q},\omega)] C_{11}^{Tc}(\mathbf{q},\omega) = -1. \tag{85}$$

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

$$C_{ij}^{L}(\mathbf{q},\omega) = C_{ij}^{Lc}(\mathbf{q},\omega + M^{L}) - C_{i0}^{Lc}(\mathbf{q},\omega + M^{L}) \times M^{L} C_{0j}^{L}(\mathbf{q},\omega), \quad i,j=0,1, \tag{86}$$

or explicitly

$$C_{00}^{L}(\mathbf{q},\omega) = \frac{C_{00}^{Lc}}{1 + M^{L} C_{00}^{Lc}}, \tag{87}$$

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

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

$$C_{11}^{L}(\mathbf{q},\omega) = \frac{C_{11}^{Lc} + C_{11}^{Lc} M^{L} C_{00}^{Lc} - C_{10}^{Lc} M^{L} C_{01}^{Lc}}{1 + M^{L} C_{00}^{Lc}}, \tag{90}$$

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

$$C_{11}^{T}(\mathbf{q},\omega) = C_{11}^{Tc}(\mathbf{q},\omega + M^{T}). \tag{91}$$

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

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

where the function $\chi_{00}^{L}(\mathbf{q},0)$ in the denominator arises from the normalization of the variable $\hat{A}_0$, see Eq. (73). Similar expressions are obtained for the other longitudinal and transverse correlation functions. We then find from Eq. (87):

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

Equation (93) is formally in agreement with Belitz and Das Sarma [Eq. (2.3) in Ref. 29]. However, $\chi_{00}^{Lc}(\mathbf{q},\omega)$ here denotes the *exact*, fully interacting longitudinal response function of the homogeneous electron gas, not just the RPA response function.

From Eq. (93), one easily derives explicit expressions for longitudinal density-current and current-current response functions using

$$\chi_{00}^{L}(\mathbf{q},\omega) = \frac{q}{\omega}\chi_{10}^{L}(\mathbf{q},\omega) = \frac{q}{\omega}\chi_{01}^{L}(\mathbf{q},\omega) = \frac{q^2}{\omega^2}\chi_{11}^{L}(\mathbf{q},\omega). \tag{94}$$

It is not difficult to show that the results for χ_{10}^{L}, χ_{01}^{L} and χ_{11}^{L} obtained in this fashion are consistent with Eqs. (88)–(90).

In the same way one finds the transverse current-current response function from Eq. (91):

$$\chi_{11}^{T}(\mathbf{q},\omega) = \frac{\omega}{(\omega + M^{T})}\chi_{11}^{Tc}(\mathbf{q},\omega + M^{T}) + \frac{M^{T}}{(\omega + M^{T})}\chi_{11}^{Tc}(\mathbf{q},0). \tag{95}$$

Expressions that are formally similar to Eqs. (93)–(95) were recently derived by Conti and Vignale[37] in the framework of Mermin's relaxation time approximation.[30] In this formalism, the role of $M(\mathbf{q},\omega)$ is taken by a frequency- and momentum-independent phenomenological scattering rate i/τ. Note that the second term on the right-hand side of Eq. (95) is absent in Ref. 37, because there the diamagnetic susceptibility of the electron gas, $\chi_{11}^{Tc}(\mathbf{q},0)$, was implicitly taken to be zero.

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

$$M^{L}(\mathbf{q},\omega) = \langle \hat{\mathbf{F}}^{L}(\mathbf{q})| \frac{1}{\mathcal{L} - \omega} |\hat{\mathbf{F}}^{L}(\mathbf{q})\rangle, \tag{96}$$

and similar for M^{T}, i.e. we resort to the standard approximation of replacing the projected by the full Liouville operator by setting $\mathcal{Q} \approx 1$ 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 $\hat{A}_0, \hat{A}_1$ and its relatively well-controlled time evolution.

In turn, the fluctuating longitudinal forces have the form

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

and the transverse forces are

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

Here, $U(\mathbf{q})$ 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:

$$\begin{aligned} &\langle U(\mathbf{q}-\mathbf{k})\hat{\rho}(\mathbf{k})|\frac{1}{\mathcal{L}-\omega}|U(\mathbf{q}-\mathbf{k}')\rho(\mathbf{k}')\rangle \\ &\approx \langle\hat{\rho}(\mathbf{k})|\frac{1}{\mathcal{L}-\omega}|\rho(\mathbf{k}')\rangle\langle U(\mathbf{q}-\mathbf{k})U(\mathbf{q}-\mathbf{k}')\rangle_{\text{disorder}}. \end{aligned} \tag{99}$$

Since the system is homogeneous, we have $\mathbf{k}=\mathbf{k}'$, and we arrive at the following expression for the longitudinal memory function:

$$\begin{aligned} M^L(\mathbf{q},\omega) = \frac{1}{n}\sum_{\mathbf{k}} \langle U(\mathbf{q}-\mathbf{k})\rangle^2 \frac{[\mathbf{q}\cdot(\mathbf{q}-\mathbf{k})]^2}{q^2} \\ \times C^L_{00}(\mathbf{k},\omega)\chi^{Lc}_{00}(\mathbf{k},0). \end{aligned} \tag{100}$$

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

$$\begin{aligned} M^T(\mathbf{q},\omega) = \frac{1}{n}\sum_{\mathbf{k}} \langle U(\mathbf{q}-\mathbf{k})\rangle^2 \frac{[\mathbf{q}\times(\mathbf{q}\times\mathbf{k})]^2}{q^4} \\ \times C^L_{00}(\mathbf{k},\omega)\chi^{Lc}_{00}(\mathbf{k},0), \end{aligned} \tag{101}$$

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

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

$$\begin{aligned} \sum_k \int dz''[\omega\delta(z''-z)\delta_{ik} - \Omega_{ik}(\mathbf{q}_\parallel,z,z'') \\ + M_{ik}(\mathbf{q}_\parallel,z,z'',\omega)]C_{kj}(\mathbf{q}_\parallel,z'',z',\omega) = -\delta_{ij}\delta(z-z'), \end{aligned} \tag{102}$$

where $\mathbf{q}_\parallel$ 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 2×2 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 Ω_{ik}, defining

$$\Omega^{\text{in}}_{ik}(\mathbf{q}_\parallel,z,z',\omega) \equiv \Omega_{ik}(\mathbf{q}_\parallel,z,z') + M_{\text{in}}(\mathbf{q}_\parallel,z,z',\omega)\delta_{i1}\delta_{k1}. \tag{103}$$

Equation (102) thus becomes

$$\begin{aligned} \sum_{k=0}^{1} \int dz''\{[\omega\delta(z''-z) + M_{\text{ex}}(\mathbf{q}_\parallel,z,z'',\omega)]\delta_{ik} \\ - \Omega^{\text{in}}_{ik}(\mathbf{q}_\parallel,z,z'',\omega)\}C_{kj}(\mathbf{q}_\parallel,z'',z',\omega) \\ = -\delta_{ij}\delta(z-z') + \delta_{i0}\int dz'' M_{\text{ex}}(\mathbf{q}_\parallel,z,z'',\omega) \\ \times C_{ij}(\mathbf{q}_\parallel,z'',z',\omega), \quad i,j=0,1. \end{aligned} \tag{104}$$

Similar to Sec. III 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 ω replaced by $\omega + M_{\text{ex}}$. Now, by contrast, the memory function $M_{\text{ex}}(\mathbf{q}_\parallel,z,z',\omega)$ 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 $C^R_{ij}(\mathbf{q}_\parallel,z,z',\omega)$ that satisfy the following coupled equations:

$$\begin{aligned} \sum_{k=0}^{1} \int dz''\{[\omega\delta(z''-z) + M_{\text{ex}}(\mathbf{q}_\parallel,z,z'',\omega)]\delta_{ik} \\ - \Omega^{\text{in}}_{ik}(\mathbf{q}_\parallel,z,z'',\omega)\}C^R_{kj}(\mathbf{q}_\parallel,z'',z',\omega) \\ = -\delta_{ij}\delta(z-z'), \quad i,j=0,1. \end{aligned} \tag{105}$$

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

$$\begin{aligned} C_{ij}(\mathbf{q}_\parallel,z,z',\omega) = C^R_{ij}(\mathbf{q}_\parallel,z,z',\omega) \\ - \int dz_1 \int dz_2 C^R_{i0}(\mathbf{q}_\parallel,z,z_1,\omega) \\ \times M_{\text{ex}}(\mathbf{q}_\parallel,z_1,z_2,\omega)C_{0j}(\mathbf{q}_\parallel,z_2,z',\omega). \end{aligned} \tag{106}$$

We now use a similar trick to obtain the reference functions C_{ij}^R. Defining the ''clean'' response function via

$$\sum_{k=0}^{1}\int dz''\{\omega\delta(z''-z)\delta_{ik} - \Omega_{ik}^{\mathrm{in}}(\mathbf{q}_\parallel,z,z'',\omega)\}C_{kj}^c(\mathbf{q}_\parallel,z'',z',\omega) = -\delta_{ij}\delta(z-z'), \tag{107}$$

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

$$C_{ij}^R(\mathbf{q}_\parallel,z,z',\omega) = C_{ij}^c(\mathbf{q}_\parallel,z,z',\omega) + \sum_{k=0}^{1}\int dz_1\int dz_2 C_{ik}^c(\mathbf{q}_\parallel,z,z_1,\omega) \times M_{\mathrm{ex}}(\mathbf{q}_\parallel,z_1,z_2,\omega)C_{kj}^R(\mathbf{q}_\parallel,z_2,z',\omega). \tag{108}$$

The density-density reference correlation function is explicitly given by

$$C_{00}^R(\mathbf{q}_\parallel,z,z',\omega) = C_{00}^c(\mathbf{q}_\parallel,z,z',\omega) + \int dz_1\int dz_2 C_{00}^c(\mathbf{q}_\parallel,z,z_1,\omega) \times M_{\mathrm{ex}}(\mathbf{q}_\parallel,z_1,z_2,\omega)C_{00}^R(\mathbf{q}_\parallel,z_2,z',\omega) + \int dz_1\int dz_2 C_{01}^c(\mathbf{q}_\parallel,z,z_1,\omega) \times M_{\mathrm{ex}}(\mathbf{q}_\parallel,z_1,z_2,\omega)C_{10}^R(\mathbf{q}_\parallel,z_2,z',\omega). \tag{109}$$

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):

$$\Phi(\mathbf{q}_\parallel,z,z',\omega) \equiv \int dz'' C_{00}(\mathbf{q}_\parallel,z,z'',\omega)\chi^c(\mathbf{q}_\parallel,z'',z',0), \tag{110}$$

$$\Phi^R(\mathbf{q}_\parallel,z,z',\omega) \equiv \int dz'' C_{00}^R(\mathbf{q}_\parallel,z,z'',\omega)\chi^c(\mathbf{q}_\parallel,z'',z',0), \tag{111}$$

$$\Phi^c(\mathbf{q}_\parallel,z,z',\omega) \equiv \int dz'' C_{00}^c(\mathbf{q}_\parallel,z,z'',\omega)\chi^c(\mathbf{q}_\parallel,z'',z',0). \tag{112}$$

Again it is assumed that all extrinsic damping effects can be neglected for the static response function. In terms of these functions, Eq. (106) becomes for $i=j=0$:

$$\Phi(\mathbf{q}_\parallel,z,z',\omega) = \Phi^R(\mathbf{q}_\parallel,z,z',\omega) - \int dz_1\int dz_2\int dz_3\Phi^R(\mathbf{q}_\parallel,z,z_1,\omega) \times[\chi^c(\mathbf{q}_\parallel,z_1,z_2,0)]^{-1}M_{\mathrm{ex}}(\mathbf{q}_\parallel,z_2,z_3,\omega) \times\Phi(\mathbf{q}_\parallel,z_3,z',\omega). \tag{113}$$

In the example to be discussed in Sec. V, only the case $\mathbf{q}_\parallel = 0$ 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

$$\Phi^R(0,z,z',\omega) = \Phi^c(0,z,z',\omega) + \int dz_1\int dz_2\int dz_3\Phi^c(0,z,z_1,\omega)[\chi^c(0,z_1,z_2,0)]^{-1}M_{\mathrm{ex}}(0,z_2,z_3,\omega)\Phi^R(0,z_3,z',\omega) + \int dz_1\int dz_2\int_{-\infty}^{z_1}d\tilde{z}_1\chi^c(0,z,\tilde{z}_1,\omega)\frac{M_{\mathrm{ex}}(0,z_1,z_2,\omega)}{\sqrt{n(z_1)n(z_2)}}\int_{-\infty}^{z_2}d\tilde{z}_2[\omega\Phi^R(0,\tilde{z}_2,z',\omega)-\chi(0,\tilde{z}_2,z',0)]. \tag{114}$$

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

$$\Phi(\mathbf{q}_\parallel,z,z',\omega) = [\chi(\mathbf{q}_\parallel,z,z',\omega)-\chi^c(\mathbf{q}_\parallel,z,z',0)]/\omega, \tag{115}$$

$$\Phi^R(\mathbf{q}_\parallel,z,z',\omega) = [\chi^R(\mathbf{q}_\parallel,z,z',\omega)-\chi^c(\mathbf{q}_\parallel,z,z',0)]/\omega, \tag{116}$$

$$\Phi^c(\mathbf{q}_\parallel,z,z',\omega) = [\chi^c(\mathbf{q}_\parallel,z,z',\omega)-\chi^c(\mathbf{q}_\parallel,z,z',0)]/\omega. \tag{117}$$

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, $\chi(\mathbf{q}_\parallel,z,z',\omega)$, in terms of the interacting response function for the ''clean'' system, $\chi^c(\mathbf{q}_\parallel,z,z',\omega)$, i.e., including intrinsic dissipation alone. χ^c is calculated, in principle exactly, using the framework of TCDFT outlined in Sec. II. 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 $\mathbf{V}(\mathbf{r})$ is decomposed into longitudinal and transverse components as follows: $\mathbf{V}^L(\mathbf{r}) = -(1/4\pi)\boldsymbol{\nabla}\int d^3r'[\boldsymbol{\nabla}'\cdot\mathbf{V}(\mathbf{r}')]/|\mathbf{r}-\mathbf{r}'|$ and $\mathbf{V}^T(\mathbf{r}) = (1/4\pi)\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\int d^3r'\mathbf{V}(\mathbf{r}')/|\mathbf{r}-\mathbf{r}'|$. In our case, i.e., working in a mixed $(\mathbf{q}_\parallel,z)$ representation, it is convenient to define the operator $\mathbf{D}(\mathbf{q}_\parallel,z)$ as

$$\mathbf{D}(\mathbf{q}_\parallel,z)\equiv\begin{pmatrix}\mathbf{q}_\parallel\\ i\boldsymbol{\nabla}_z\end{pmatrix}. \tag{118}$$

The longitudinal fluctuating forces are then given by

$$\hat{\mathbf{F}}^L(\mathbf{q}_\parallel,z)=\frac{1}{\sqrt{n(z)}}\mathbf{D}(\mathbf{q}_\parallel,z)\int dz'\int\frac{d^2p_\parallel}{(2\pi)^2}\frac{e^{-q_\parallel|z-z'|}}{2q_\parallel}\times\{\mathbf{D}(\mathbf{q}_\parallel,z')\cdot[\hat{\rho}(\mathbf{p}_\parallel,z')\times\mathbf{D}(\mathbf{p}_\parallel-\mathbf{q}_\parallel,z')U(\mathbf{q}_\parallel-\mathbf{p}_\parallel,z')]\} \tag{119}$$

and the transverse fluctuating forces are

$$\hat{\mathbf{F}}^T(\mathbf{q}_\parallel,z)=-\frac{1}{\sqrt{n(z)}}\mathbf{D}(\mathbf{q}_\parallel,z)\times\mathbf{D}(\mathbf{q}_\parallel,z)\times\int dz'\int\frac{d^2p_\parallel}{(2\pi)^2}\frac{e^{-q_\parallel|z-z'|}}{2q_\parallel}[\hat{\rho}(\mathbf{p}_\parallel,z')\times\mathbf{D}(\mathbf{p}_\parallel-\mathbf{q}_\parallel,z')U(\mathbf{q}_\parallel-\mathbf{p}_\parallel,z')]. \tag{120}$$

One finds from Eqs. (119) and (120) that in the *homogeneous* limit (no z dependence of the density fluctuations ρ and the scattering potential U), the longitudinal and transverse forces only have in-plane components, given by the 2D versions of Eqs. (97) and (98). In the following, we shall limit the discussion of the *inhomogeneous* situation to a case of special interest, namely, $\mathbf{q}_\parallel=0$. In that case, only the z component of $\mathbf{F}^L$ survives, and is given by

$$\hat{F}_z^L(z)=\frac{i}{\sqrt{n(z)}}\int\frac{d^2p_\parallel}{(2\pi)^2}\hat{\rho}(\mathbf{p}_\parallel,z)\nabla_z U(-\mathbf{p}_\parallel,z). \tag{121}$$

The transverse force, in the same limit, acts in the x-y plane only,

$$\hat{\mathbf{F}}_\parallel^T(z)=\frac{1}{\sqrt{n(z)}}\int\frac{d^2p_\parallel}{(2\pi)^2}\mathbf{p}_\parallel\hat{\rho}(\mathbf{p}_\parallel,z)U(-\mathbf{p}_\parallel,z). \tag{122}$$

We note that, by symmetry, in the limit $\mathbf{q}_\parallel=0$ 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. (119) and (120), using the same approximate decoupling procedure that was used for the homogeneous case in Sec. III C. The result is

$$M_{\rm ex}^L(z,z',\omega)=\int\frac{d^2p_\parallel}{(2\pi)^2}\frac{\Phi(\mathbf{p}_\parallel,z,z',\omega)}{\sqrt{n(z)n(z')}}\times\nabla_z\nabla_{z'}\langle U(-\mathbf{p}_\parallel,z)U(-\mathbf{p}_\parallel,z')\rangle \tag{123}$$

and

$$M_{\rm ex}^T(z,z',\omega)=\int\frac{d^2p_\parallel}{(2\pi)^2}\frac{\Phi(\mathbf{p}_\parallel,z,z',\omega)}{\sqrt{n(z)n(z')}}\times p_\parallel^2\langle U(-\mathbf{p}_\parallel,z)U(-\mathbf{p}_\parallel,z')\rangle. \tag{124}$$

The next step consists in finding explicit forms for the disorder-averaged random scattering potential $U(\mathbf{p}_\parallel,z)$, 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

$$U(\mathbf{p}_\parallel,z)=\frac{2\pi}{\varepsilon(p_\parallel)}\frac{e^{-p_\parallel|z-z_1|}}{p_\parallel}, \tag{125}$$

where $\varepsilon(p_\parallel)$ is the 2D dielectric function.[46] The longitudinal memory function for charged-impurity scattering is thus

$$M_I^L(z,z',\omega)=\int\frac{d^2p_\parallel}{\varepsilon^2(p_\parallel)}\frac{\Phi(\mathbf{p}_\parallel,z,z',\omega)}{\sqrt{n(z)n(z')}}\int d\tilde{z}\,n_i(\tilde{z})\times\mathrm{sgn}(z-\tilde{z})\mathrm{sgn}(z'-\tilde{z})e^{-p_\parallel|z-\tilde{z}|}e^{-p_\parallel|z'-\tilde{z}|}, \tag{126}$$

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

Likewise, the longitudinal memory function associated with interface roughness is

$$M_R^L(z,z',\omega)=\int\frac{d^2p_\parallel}{(2\pi)^2}\frac{\Phi(\mathbf{p}_\parallel,z,z',\omega)}{\sqrt{n(z)n(z')}}\langle U(p_\parallel)^2\rangle\times\nabla_z\nabla_{z'}[\delta(z-z_l)\delta(z'-z_l)+\delta(z-z_r)\delta(z'-z_r)], \tag{127}$$

where $U(p_\parallel)$ is the random roughness scattering potential, assumed for simplicity to be the same at the left and right interfaces, z_l and z_r. It is common to assume a Gaussian form for the autocorrelation function of the random interface roughness,[46,47] which leads to

$$\langle U(p_\parallel)^2\rangle=\pi\mu^2\Delta^2\eta^2e^{-p_\parallel^2\eta^2/4}. \tag{128}$$

Here, μ is the height of the potential step at the interface, and the correlation length η and average roughness height Δ are controlled by material and growth conditions. In the presence of both impurity and roughness scattering, the memory functions M_I and M_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 $\Phi(\mathbf{q}_\parallel,z,z',\omega)$ at *all*

$\mathbf{q}_\parallel$, not just $\mathbf{q}_\parallel = 0$. This means that Φ should be calculated self-consistently from Eqs. (113) for *all* $\mathbf{q}_\parallel$, which is a very demanding computational task. Therefore, as a first approximation, we ignore self-consistency and instead use the *noninteracting* $\Phi_0(\mathbf{q}_\parallel, z, z', \omega)$ in Eqs. (126) and (127), defined by replacing χ and χ_c with $\chi_{\rm KS}$ in Eq. (115). The wave-vector dependence of Φ_0 is thus known analytically (see the explicit expression for $\chi_{\rm KS}$ 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,[48] being the basis of a variety of new devices operating in the terahertz regime, such as detectors[49] and quantum cascade lasers.[50] 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.[51]

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.*[32] 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. 52). 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,[32] two parameters were controlled independently: the electronic sheet density N_s (from 0.05 to 1.3 $\times 10^{11}$ 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,[53] 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. 24. The

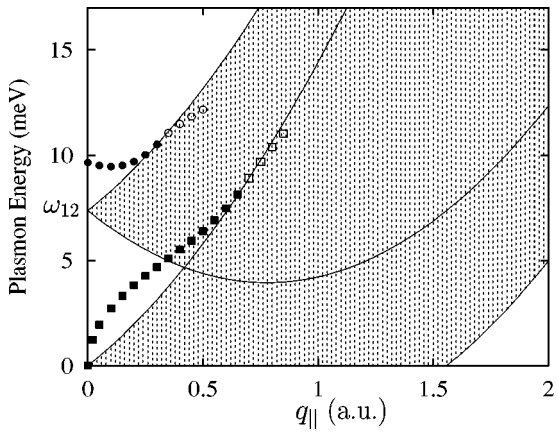

FIG. 1. Dispersions of ISB (circles) and 2D plasmon (squares) frequencies $\Omega(q_\parallel)$ in a clean quantum well. ω_{12} is the difference between the two lowest bare subband levels. Inside the shaded regions, the plasmons are subject to strong Landau damping and rapidly die off (open symbols). The experiment by Williams *et al.*[32] measures the ISB plasmon frequency and linewidth at $q_\parallel = 0$.

noninteracting response function reads

$$\chi_{\rm KS}(\mathbf{q}_\parallel, z, z', \omega) = \sum_{\mu=1}^{N_{\rm occ}} \sum_{\nu=1}^{\infty} F_{\mu\nu}(q_\parallel, \omega)\varphi_\mu(z)\varphi_\mu(z')\varphi_\nu(z)\varphi_\nu(z'), \tag{129}$$

where

$$F_{\mu\nu}(q_\parallel, \omega) = -2\int \frac{d^2k_\parallel}{(2\pi)^2}\left\{\frac{f(\epsilon_\mu + k_\parallel^2/2)}{\mathbf{q}_\parallel\mathbf{k}_\parallel + a_{\mu\nu}(q_\parallel) + \omega + i\eta} + \frac{f(\epsilon_\mu + k_\parallel^2/2)}{\mathbf{q}_\parallel\mathbf{k}_\parallel + a_{\mu\nu}(q_\parallel) - \omega - i\eta}\right\}, \tag{130}$$

$a_{\mu\nu}(q_\parallel) = q_\parallel^2/2 + \epsilon_\nu - \epsilon_\mu$, f is the Fermi function at $T=0$, and η is a positive infinitesimal. ϵ_μ 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 nine bound levels, only the lowest being occupied ($N_{\rm occ}=1$).[24]

We consider perturbations of the form $v_1(z,\omega) = E_0 z$, 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 (10), the photoabsorption cross section is then obtained as $\sigma(\omega) = -(8\pi\omega/E_0 c)\,{\rm Im}\int dz\, z\, n_1(z,\omega)$ and can be directly compared with data from photoabsorption measurements. $\sigma(\omega)$ has a peak at the plasmon frequency Ω with linewidth (half width at half maximum) Γ.

In Fig. 1 we plot the dispersions $\Omega(q_\parallel)$ of the ISB and the intrasubband (or 2D) plasmon in the clean quantum well ($N_s = 1.0\times 10^{11}$ cm^{-2}), calculated within ALDA, see Eq. (15). The imaginary part of the Kohn-Sham response function $\chi_{\rm KS}$ 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 $q_\parallel$, the plasmons are undamped in ALDA, for which $f_{\rm xc}$ is

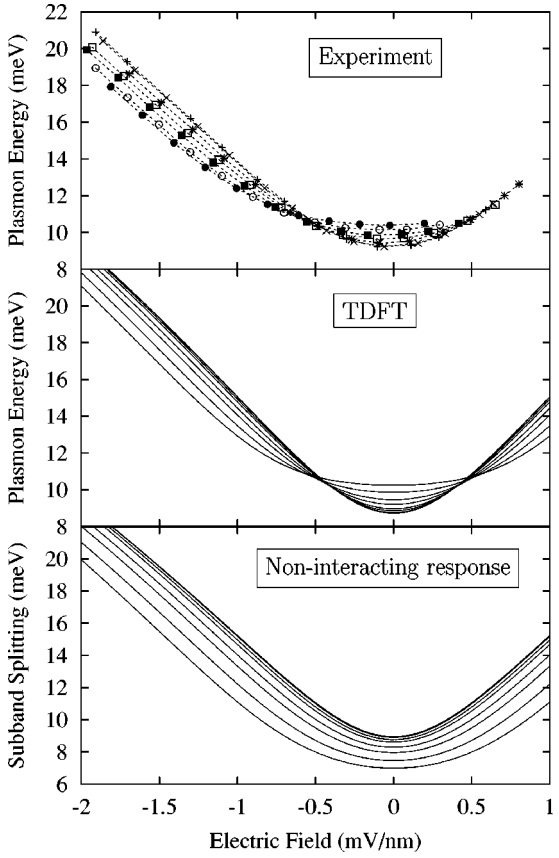

FIG. 2. ISB plasmon frequency Ω, at $q_\parallel = 0$, versus electric field E. Top: experimental data from Ref. 23. Middle: calculated results for the clean quantum well, using TDFT [Eq. (10)]. Bottom: bare LDA subband splitting. The individual curves are associated with different electronic sheet densities ($N_s = 0.05$, 0.1, 0.2, 0.3, 0.5, 0.7, 1.0, and 1.3×10^{11} cm^{-2}). The lower N_s, the steeper $\Omega(E)$ around $E = 0$.

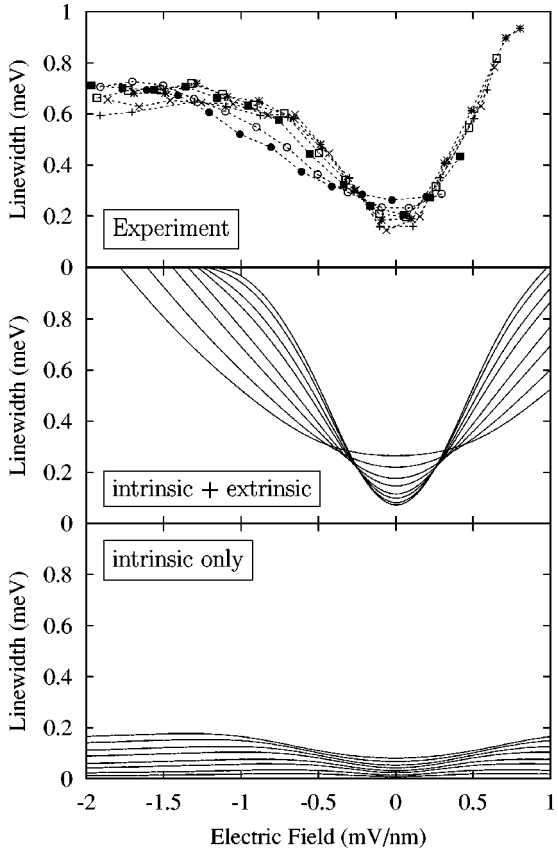

FIG. 3. ISB plasmon linewidth Γ, at $q_\parallel = 0$, versus electric field E. Top: experimental data from Ref. 23. Middle: calculated results including extrinsic (impurity and interface roughness) and intrinsic (electron-electron interaction) damping. Bottom: $\Gamma(E)$ for a clean quantum well (intrinsic damping only). The individual curves correspond to different electronic sheet densities (see Fig. 2). At $E = 0$, the lowest N_s causes the smallest Γ.

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 $q_\parallel = 0$. To take this effect into account, we go beyond the ALDA and in the following include dynamical xc effects, see Eqs. (48), (49).

In Fig. 2 we show the electric field dependence of the ISB plasmon frequencies $\Omega(q_\parallel = 0)$ 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 xc effects in Eq. (10) (i.e., using RPA) induces a 10% blueshift of Ω, 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. 2. The results are clearly qualitatively wrong: there is no crossing of $\Omega(E)$ for different N_s, in contradiction to experiment.

Figure 3 shows the ISB plasmon linewidth $\Gamma(E)$, for different N_s. For small E, the experimental data again exhibit a quadratic behavior, and $\Gamma(E)$ rises faster for smaller N_s. For large negative E, Γ saturates around 0.7 meV. For positive E (i.e., pointing in the direction of sample growth), Γ rises somewhat higher. The asymmetry of $\Gamma(E)$ is likely to be due to slightly different roughnesses of the interfaces.

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 $N_s \sim 10^{11}$ 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 modeled.

The presence of bulk impurities in the quantum well is mainly caused by segregation of donors from the lower δ-doped layer and diffusion along z during growth. We use the functional form $n_i(z)=1.33e^{-z/30\ \mathrm{nm}}\times 10^{15}\ \mathrm{cm}^{-3}$, proposed in Ref. 32 to explain in-plane mobility data, for the bulk impurity concentration in Eq. (126). We also include scattering from the upper δ-doped layer (remote impurity density $4.8\times 10^{11}\ \mathrm{cm}^{-2}$).

In Ref. 32 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 Γ is instead dominated by interface roughness scattering and can in fact be qualitatively explained by it alone: Via (127), Γ 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 (128). The height of the potential step for our quantum well is $\mu=257.6$ meV. The roughness parameters are chosen as $\eta=64.4$ Å and $\Delta=4$ Å, to give the best fit to experiment for the largest N_s. Both η and Δ are in the characteristic range found by lattice imaging techniques.[54]

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

For $|E|\gtrsim 1$ 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 M^L, which in turn depends on the imaginary part of Φ. Broadening of the plasmon resonance means that $\mathrm{Im}\Phi$ is peaked around Ω over some frequency range of width Γ. But, due to the constraint of the *f*-sum rule, *increasing* Γ means that the *height* of the peak of $\mathrm{Im}\Phi$ must *decrease*. This, in turn, limits the growth of the memory function and rapidly saturates Γ. 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 inhomogeneous 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 nonadiabatic description, which includes retardation, is required. A nonadiabatic 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 τ with the memory function $M(\mathbf{q},\omega)$, 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 describing ISB plasmons in a wide $\mathrm{GaAs/Al_{0.3}Ga_{0.7}As}$ 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 correlation 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.[32] 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.

ACKNOWLEDGMENTS

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