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Phenomenological plasmon broadening and relation to the dispersion

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ABSTRACT

Pragmatic ways of including lifetime broadening of collective modes in the electron liquid are critically compared. Special focus lies on the impact of the damping parameter onto the dispersion. It is quantitatively exemplified for the two-dimensional case, for both, the charge ('sheet'-)plasmon and the spin-density plasmon. The predicted deviations fall within the resolution limits of advanced techniques.

1. Introduction

The study of plasmons, the collective oscillations of electrons, has a long and successful history [1,2]. Their coupling to light in nanostructures, known as 'plasmonics', holds high promise for revolutionary applications [3], where the performance of actual devices is crucially limited by metallic losses [4]. An undemanding inclusion of a plasmon's damping via a constant, irrespective of the losses' origin(s), is commonly achieved via a Drude type dielectric function [2]. The response of charges, however, is nonlocal, which is the more important the smaller the size of the nanoparticles is.

The random phase approximation (RPA) [2], historically a milestone, provides a dielectric function explicitly depending on both, frequency ω as well as wave vector q. Its failure to include finite lifetime effects was treated early by Mermin [5], his approach still being widely applied. Recent examples in bulk systems include calculations of the electrons' inelastic mean free path [6], stopping power [7], and a generalization to spin wave damping [8]. In layers, it has been employed, e.g., to obtain quasi-particle properties [9] of the two-dimensional electron gas (2Deg), or when accounting for inter-band-excitation losses in graphene [10].

Major techniques for studying the charge response to external perturbations are scattering experiments [11] and, for long wavelengths, optical measurements. They yield the same dispersion *only* for an undamped plasmon; for realistic lifetimes slightly different results are obtained.[1] This discrepancy in the plasmon *dispersion* $\omega_{\rm pl}(q)$ increases with its linewidth (often broadening with q). Consequently, comparing theory and high-resolution experiments needs appropriate caution.

Forefront scattering data for dispersion and damping are known for metallic monolayers [12–14] and semiconductor quantum wells [15,16]. Many 2Degs being rather dense [17], RPA predictions are sufficiently accurate, once damping effects are built in effectively. The 2Deg, our prototype, shares the vanishing of $\omega_{\rm pl}(q\to 0)$ with graphene. There, too, the plasmon was studied with optical as well as scattering methods [18–22] (plasmons in graphene are thoroughly reviewed in [23,24]).

Mermin's approach conserves the local electron number, invoking just a single additional constant η (the inverse lifetime τ). Extensions further conserving local energy and momentum were developed (and applied to a two-component plasma) by Röpke [25–27], and, independently, by Atwal and Ashcroft [28]. These sophisticated theories yield intricate response functions with a wave vector dependent linewidth, as also found in [29]. But in view of realistic materials an uncomplicated RPA extension incorporating plasmon lifetimes via a phenomenological (potentially q-dependent) parameter is preferential. Describing damping irrespective of the microscopic mechanism(s), phenomenological life times can be taken from both, experiments or published data from theories, which cannot be easily recreated.

The purpose of this work is to critically compare other simple approaches with that of Mermin, and to study the resulting plasmon dispersions. Deviations from the classical plasma frequency, ω_p , turn out larger than expected.

After briefly discussing general aspects in Section 2, we present quantitative (zero temperature) results (both in RPA and beyond) in Section 3 for a 2Deg. There, the vanishing of $\omega_{\rm pl}(q\to 0)$ implies a comparably high relative width, also in case of rather weak damping. Finally, we study the spin-density plasmon in the partially spin-polarized case in Section 4. For Fourier Transforms conventions are as in [1].

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[1] For the Drude case this was already noted in [2, p.207f.],

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2. Theoretical overview

A plasmon is conventionally obtained from the complex dielectric function $\epsilon = \epsilon_\mathrm{I} + i\,\epsilon_\mathrm{II}$ via these, closely related but not exactly equal definitions:

- as a maximum in the double differential scattering cross section [12,16],

$$\mathrm{Im}\frac{-1}{\epsilon(q,\,\omega_\mathrm{pl}^{(a)})} = \max; \tag{1a}$$

- as the vanishing of the complex ϵ for complex ω [28], determining reflection coefficients (purely oscillatory waves change to decaying ones)

$$\epsilon(q,\,\omega_\mathrm{I} + i\omega_\mathrm{II}) = 0, \quad \omega_\mathrm{pl}^{(b)} \equiv \omega_\mathrm{I}; \tag{1b}$$

- often approximated as the zero of Re ϵ with real frequencies [21],

$$\epsilon_\mathrm{I}(q,\,\omega_\mathrm{pl}^{(c)}) = 0; \tag{1c}$$

  (the approximation being justified if $\omega_\mathrm{I} \gg \omega_\mathrm{II}$, and $\omega_\mathrm{pl}^{(c)}$ is always lower than $\omega_\mathrm{pl}^{(a,b)}$ [30]),
- or, if the phase shift is not relevant, as minimal magnitude of ϵ (implying a maximal electric field),

$$|\epsilon(q,\,\omega_\mathrm{pl}^{(d)})| = \min. \tag{1d}$$

The Drude model [2] for charge carriers with a classical plasma frequency ω_p reads $\epsilon_\mathrm{Dru}(q,\,\omega) = 1 - \omega_p^2/\omega(\omega + i\eta)$. Here, $\omega_\mathrm{pl}^{(a)}$ and $\omega_\mathrm{pl}^{(b)}$ differ <1% for η even as large as ω_p, but conditions (1c–1d) yield clearly distinct values, unless η is rather small. The criterion appropriate to the setup must be chosen for cutting edge experimental resolutions. Typical values reported [12,14] are ~10 meV (roughly 10% of ω_pl), ~25…125 meV ≈ 10…50% of ω_pl [19] and ~ 100 meV [31]. By definition, (1a) yields a symmetric Lorentzian near $\omega_\mathrm{pl}^{(a)}$, whereas expansions around $\omega_\mathrm{pl}^{(b-d)}$ contain first order terms in the denominator, too:

$$\mathrm{Im}\frac{-1}{\epsilon(q,\,\omega)} \approx \begin{cases} \dfrac{\alpha}{\left(\omega-\omega_\mathrm{pl}^{(a)}\right)^2+\gamma^2} \\ \dfrac{\alpha}{\left(\omega-\omega_\mathrm{pl}^{(b-d)}\right)^2+\beta\left(\omega-\omega_\mathrm{pl}^{(b-d)}\right)+\gamma^2} \end{cases}. \tag{2}$$

Clearly, the discrepancy in differently computed ω_pl-values depends on the specific $\epsilon(q,\,\omega)$ used. Some common forms are given next.

The linear response of an electron liquid to external perturbations in RPA-type approaches reads

$$\epsilon_\mathrm{RPA}(q,\,\omega) = 1 - v(q)\chi^0(q,\,\omega); \tag{3}$$

here, $v(q)$ denotes the Coulomb interaction, and $\chi^0(q,\,\omega)$ the density–density response function of non-interacting fermions [1]. It shows the typical electron–hole (e/h) excitation band in the $(q,\,\omega)$-plane. An adiabatically turned on perturbation corresponds to $\omega \to \omega + i0^+$. This ensures causality, but yields an undamped plasmon. An obvious idea to include damping is to use $\chi^0(q,\,\tilde{\omega})$ with $\tilde{\omega} \equiv \omega + i\eta$ and inverse lifetime $\eta \equiv 1/\tau$,

$$\epsilon_\mathrm{Lin}(q,\,\omega) \equiv 1 - v(q)\chi^0(q,\,\tilde{\omega}). \tag{4}$$

This also broadens the e/h band (as $\mathrm{Im}\chi^0(q,\,\tilde{\omega})$, at any q, only vanishes when $|\omega\tau| \gg 1$). It catches the eye that Eq. (4) alters the static response

$$\epsilon_\mathrm{Lin}(q,\,0) = 1 - v(q)\chi^0(q,\,i\eta) \xrightarrow[q\to 0]{} 1 + \omega_p^2/\eta^2 \tag{5}$$

violating $\epsilon(q \to 0,\ 0) = 1 + v(q)\,N(E_\mathrm{F})$ (the perfect screening sum rule, $N(E_\mathrm{F})$ is the density of states at the Fermi energy). The correctness of this limit may be of less importance for plasmonic applications, which are far from static.

Mermin [5] corrected the deficiency. He deriv"ed

$$\epsilon_\mathrm{Me}(q,\,\omega) \equiv 1 - \frac{v(q)\chi^0(q,\,\tilde{\omega})}{1 + i\eta g_\mathrm{Me}(q,\,\tilde{\omega})}; \tag{6a}$$

$$g_\mathrm{Me}(q,\,\tilde{\omega}) \equiv \frac{1}{\tilde{\omega}}\left(\frac{\chi^0(q,\,\tilde{\omega})}{\chi^0(q,\,0)} - 1\right). \tag{6b}$$

Albeit elegant, analytical calculations with Eq. (6) quickly get cumbersome, in particular when the relations are meant as matrix equations (2×2 for spin-dependent screening or electron–hole liquids, infinite matrices in reciprocal lattice vectors of crystals). Note that (6b) does *not* yield, as it should, the classical plasmon for long wavelengths,

$$\omega_\mathrm{pl}^{(b)}(q \to 0)_\mathrm{Me} \not\rightarrow \omega_p, \tag{7}$$

neither in the bulk nor for the 2Deg [28] (there, also $\omega_\mathrm{pl}^{(a)}(q \to 0)$ shows a mismatch with the $\sqrt{q}-$ dependence, *cf.* Fig. 1).

Comparing approaches with the structure of (6a) but arbitrary ω instead of $\tilde{\omega}$ is worthwhile,

$$\epsilon_g(q,\,\omega) \equiv 1 - \frac{v(q)\chi^0(q,\,\omega)}{1 + i\eta g(q,\,\omega)}. \tag{8}$$

The elementary choice $g_1 \equiv \mathrm{sgn}(\omega_\mathrm{I})\hbar/E_\mathrm{F}$ (i.e. simply adding a constant to the RPA's susceptibility denominator, with a sign function for proper symmetry), serves to enhance a long-lived plasmon's visibility in graphical representations. The dielectric function with $g_\mathrm{D} \equiv 1/\omega$ reduces to the Drude model for $\omega \gg \hbar q^2/2m$ (m is the effective electron mass). For the optical conductivity σ this implies

$$\sigma(\omega) = \frac{ne^2\tau/m}{\omega(g(0,\,\omega) - i\tau)} \xrightarrow[g=g_\mathrm{D}]{} \frac{ne^2\tau/m}{1 - i\omega\tau}. \tag{9}$$

To achieve an interpolation between static RPA screening and the Drude case we propose the form

$$g_{E_\mathrm{F}}(\omega) \equiv \frac{\omega}{\omega^2 + E_\mathrm{F}^2/\hbar^2}. \tag{10}$$

The main deficiency of this ansatz is to violate the *f*-sum rule (due to additional poles at $\hbar\omega = \pm\, iE_\mathrm{F}$),

$$\int_0^\infty \frac{d\omega}{\pi}\,\omega\,\mathrm{Im}\frac{-1}{\epsilon(q,\,\omega)} = \frac{\omega_p^2}{2}. \tag{11}$$

When the focus lies on plasmon properties (e.g. the q-dependence), this can be acceptable: No large frequency range (often inaccessible anyhow [32]) needs to be measured for comparing peak positions and widths.

Both, ϵ_Me and ϵ_Lin fulfill Eq. (11), for the latter occasionally reported otherwise [7]. The contour for the integration (11) in the complex $(\omega_\mathrm{I},\,\omega_\mathrm{II})$-plane is taken along the quarter circle enclosing the first quadrant. For a purely real integration kernel on the ω_II-axis only the arc contributes, provided Im ϵ^{-1} is analytic in the upper half plane (as it should) [1]. The RPA response function obeys these conditions. From its high frequency expansion,

$$\epsilon_\mathrm{RPA}(q \to 0,\,|\omega| \to \infty) \approx 1 - \omega_p^2/|\omega|^2, \tag{12}$$

it follows that ϵ_Lin also fulfills the *f*-sum rule. While state-of-the-art approaches [25–28] satisfy the widest set of sum rules and treat electron correlations properly, impurity scattering still has to be accounted for phenomenologically.

Note that the static structure factors $S(q)$ obtained from the above dielectric functions via

$$\int_0^\infty \frac{d(\hbar\omega)}{\pi}\mathrm{Im}\frac{-1}{\epsilon(q,\,\omega)} = v(q)S(q) \tag{13}$$

differ for each approach. For a meaningful comparison of scattering intensities, their normalization by $S(q \to 0)$ appears advisable.