Nonlinear mechanism of plasmon damping in electron gas

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At plasmon resonance, the condition of applicability of the linear response theory, which is the smallness of the oscillating field, evidently breaks down. We suggest a variant of the quadratic response theory which remains valid near and at plasma frequency and demonstrate that, as could be anticipated, the nonlinearity serves itself to restrict the amplitude of plasma oscillations, thus providing a mechanism of ''nonlinear damping.'' We apply this approach to calculate the damping of plasmon in two-dimensional electron gas below the threshold wave vector, which damping has recently been observed experimentally in the S_1 surface band of Si(111)$-\sqrt{3}\times\sqrt{3}$-Ag.

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The linear random phase approximation (RPA) picture of dynamic screening in electron gas, worked out by Lindhard[1] in three dimensions (3D) and by Stern[2] in 2D, respectively, predicts the undamped plasmon up to a critical wave vector, determined by the condition of plasmon coupling with particle-hole excitations. For inelastic electron scattering, this means that in the vicinity of the plasmon resonance the energy-loss function

$$L(\mathbf{q},\omega)=-\mathrm{Im}\frac{1}{\epsilon(\mathbf{q},\omega)}, \tag{1}$$

where ϵ is the wave vector $(\mathbf{q})$ and frequency (ω) dependent dielectric function, is Dirac's δ function, with zero width and infinite height. One mechanism which restricts the plasma resonance is exchange and correlation, the account of which leads to finite damping at all wave vectors in the linear response theory.[3] However, when applied to the case of two-dimensional electron gas, exchange-correlation broadening accounts to less than 1% of the relative width of the plasmon peak $\Delta\omega_p(\mathbf{q})/\omega_p(\mathbf{q})$.[4,5]

The purpose of this work is to demonstrate that alternative mechanism of stronger damping of plasmon peak is due to the nonlinearity of the dynamic response, even if considered within the RPA. This mechanism puts a limit on the intensity of the plasmon amplitude and, consequently, introduces the finite damping of plasmon even for the wave vectors *below* the single-particle excitation threshold.

The philosophy of our approach is that the relation of the linear response theory

$$\phi(\mathbf{q},\omega)=\frac{1}{\epsilon(\mathbf{q},\omega)}\phi_{\mathrm{ext}}(\mathbf{q},\omega), \tag{2}$$

where ϕ and ϕ_{ext} are the total and the externally applied scalar potentials of the electric field, respectively, is valid under the assumption that ϕ and ϕ_{ext} are small. This assumption evidently breaks down for ϕ at the resonance frequency $\omega_p(\mathbf{q})$, particularly so if ϵ is real and passes through zero at this frequency; then, ϕ becomes infinite.

Within the quadratic response theory one can write either

$$\phi(\mathbf{q},\omega)=\frac{1}{\epsilon(\mathbf{q},\omega)}\phi_{\mathrm{ext}}(\mathbf{q},\omega)+\int\epsilon_2^{-1}(\mathbf{q},\omega,\mathbf{k},\omega_1)\times\phi_{\mathrm{ext}}(\mathbf{q}-\mathbf{k},\omega-\omega_1)\phi_{\mathrm{ext}}(\mathbf{k},\omega_1)d\mathbf{k}d\omega_1 \tag{3}$$

or

$$\phi_{\mathrm{ext}}(\mathbf{q},\omega)=\epsilon(\mathbf{q},\omega)\phi(\mathbf{q},\omega)+\int\epsilon_2(\mathbf{q},\omega,\mathbf{k},\omega_1)\times\phi(\mathbf{q}-\mathbf{k},\omega-\omega_1)\phi(\mathbf{k},\omega_1)d\mathbf{k}d\omega_1, \tag{4}$$

where ϵ_2 and ϵ_2^{-1} are the quadratic and the inverse quadratic dielectric functions, respectively. Equations (3) and (4) are equivalent far from plasma resonance.[9] However, at the resonance they are not. Indeed, similar to the linear theory, Eq. (3) fails at the resonance, since ϕ grows infinite. However, nothing catastrophic happens with Eq. (4) near or at the resonance, and we can hope that the solution ϕ of this equation remains small as long as ϕ_{ext} is small, which is the condition of the applicability of the perturbative approach. We will see below that this supposition proves to be true.

Let us solve Eq. (4). First, it is straightforward to see that the transformation (4) leaves invariant the space of functions of the form

$$\phi(\mathbf{q},\omega)=\sum_{n=-\infty}^{\infty}B_n\delta(\mathbf{q}-n\mathbf{q}_0)\delta(\omega-n\omega_0). \tag{5}$$

Then, if

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$$\phi_{\mathrm{ext}}(\mathbf{q},\omega)=\sum_{n=-\infty}^{\infty} A_n\,\delta(\mathbf{q}-n\mathbf{q}_0)\,\delta(\omega-n\omega_0), \tag{6}$$

we have from Eq. (4)

$$A_l=\epsilon(l)B_l+\sum_{n=-\infty}^{\infty}\epsilon_2(l,n)B_nB_{l-n}, \tag{7}$$

where we have introduced the notation

$$\epsilon(l)=\epsilon(l\mathbf{q}_0,l\omega_0),$$

$$\epsilon_2(l,n)=\epsilon_2(l\mathbf{q}_0,l\omega_0,n\mathbf{q}_0,n\omega_0). \tag{8}$$

The infinite system of equations (7), the unknowns being B_l, is the one to be solved to find the total potential, when the external one is known through A_l. To ensure the potentials to be real in real space, the coefficients in Eqs. (5) and (6) obey the relations

$$A_{-n}=A_n^*,$$

$$B_{-n}=B_n^*.$$

We will assume the single-wave external potential $A_n=0$, $n\neq\pm1$. To make the system (7) solvable, we must retain only a finite number of B_n. If only $B_{\pm1}$ are kept, we are taken back to the linear case. The simplest nontrivial solution comes from retaining $B_{\pm1}$ and $B_{\pm2}$ only. In this case we have, from Eq. (7),[10]

$$B_1=B_{-1}^*=\frac{A_1}{\epsilon(1)-\dfrac{2|B_1|^2\epsilon_2(1,2)\epsilon_2(2,1)}{\epsilon(2)}}, \tag{9}$$

$$B_2=B_{-2}^*=-\frac{B_1^2\epsilon_2(2,1)}{\epsilon(2)}.$$

From Eq. (9) we also have the cubic equation for $|B_1|^2$:

$$|B_1|^2\left|\epsilon(1)-\frac{2|B_1|^2\epsilon_2(1,2)\epsilon_2(2,1)}{\epsilon(2)}\right|^2=|A_1|^2. \tag{10}$$

The above formulas are quite general; however, in view of the application we make, we will refer below to the 2D case. To second order in the total field we can write for the energy absorbed in the sheet per unit area per unit time[11]

$$Q=\langle\mathbf{j}(\mathbf{r},t)\mathbf{E}(\mathbf{r},t)\rangle$$
$$=\frac{|\omega_0||\mathbf{q}_0|}{\pi}\mathrm{Im}\sum_{n=1}^{\infty}n^2|B_n|^2\epsilon(n), \tag{11}$$

where $\mathbf{j}$ is the current density, $\mathbf{E}$ is the electric field, and $\langle\cdots\rangle$ stands for an average over the time period. Introducing the energy-loss function

$$L(\mathbf{q},\omega)=\frac{\pi Q}{|\omega_0||\mathbf{q}_0||A_1|^2}, \tag{12}$$

by use of Eqs. (9) and (10) we can write in the case of two total waves

$$L(\mathbf{q},\omega)=\frac{\mathrm{Im}\,\epsilon(1)}{\left|\epsilon(1)-\dfrac{2|B_1|^2\epsilon_2(1,2)\epsilon_2(2,1)}{\epsilon(2)}\right|^2}+4|A_1|^2\left|\frac{\epsilon_2(2,1)}{\epsilon(2)}\right|^2\frac{\mathrm{Im}\,\epsilon(2)}{\left|\epsilon(1)-\dfrac{2|B_1|^2\epsilon_2(1,2)\epsilon_2(2,1)}{\epsilon(2)}\right|^4}. \tag{13}$$

Equation (13) together with Eq. (10) solves the problem in the approximation of two total waves. They evidently generalize the linear theory equation (1), the latter being reproduced by omitting the second term in Eq. (13) and keeping only $|\epsilon(1)|^2$ in the denominator of the first one.

Let us now show what a difference Eq. (13) makes with regard to the plasmon damping as compared with Eq. (1). Let us suppose that there is no damping of plasmons in linear theory—i.e., that $\mathrm{Im}\,\epsilon(1)=0$ when $\mathrm{Re}\,\epsilon(1)=0$. The major consequence of Eq. (13) for plasmon damping is that $\epsilon(2)\equiv\epsilon(2\mathbf{q}_0,2\omega_0)$ can have the nonzero imaginary part when $\epsilon(1)$ has not.

In Fig. 1 we plot the energy-loss function of the 2D electron gas in the S_1 surface band of Si(111)-$\sqrt{3}\times\sqrt{3}$-Ag using RPA linear[2] and quadratic[6] dielectric functions of 2D electron gas. The parameters of the system are[7] $m_{\mathrm{eff}}=0.3$, $n=1.9\times10^{13}\ \mathrm{cm}^{-2}$, and the background dielectric function is taken to be $(\epsilon_{\mathrm{Si}}+1)/2=6.25$.

The two-total-wave approximation is valid as long as $\mathrm{Im}\,\epsilon(2)\neq0$ at the plasmon frequency. For parameters of our example this holds for the wave vector larger than $\sim0.55k_f$. For smaller q's a larger number of waves must be included. We could obtain the solution of Eqs. (7), analogs to Eqs. (9) and (10) in the case of three waves. Then we have the algebraic equation of the seventh order to find $|B_1|^2$, and $B_{\pm1}$, $B_{\pm2}$, and $B_{\pm3}$ are expressed by it. The formulas are, however, lengthy and we do not write them here. The general rule is obvious: the approximation of n total waves gives the finite damping of plasmon as long as $\mathrm{Im}\,\epsilon(n)\neq0$ at the plasmon frequency determined by $\mathrm{Re}\,\epsilon(1)=0$. In Fig. 2 we plot $\mathrm{Re}\,\epsilon(1)$ together with three first $\mathrm{Im}\,\epsilon(n)$ to illustrate the way the plasmon frequency falls into the interval of damping of one of the consecutive $\epsilon(n)$. For $q=0.5k_f$ the first nonzero $\mathrm{Im}\,\epsilon(n)$ at plasmon frequency is $\mathrm{Im}\,\epsilon(3)$ [Fig. 2(a)] and, correspondingly, the two- and three-wave approximations differ drastically [Fig. 2(b)]. In contrast, for $q=0.63k_f$ the plasmon falls into the interval of nonzero $\mathrm{Im}\,\epsilon(2)$ [Fig. 2(c)] and, the two- and three-wave approximations differ insignificantly [Fig. 2(d)]. When q grows, $\epsilon(n)$ with smaller n's acquire a nonzero imaginary part at plasmon frequency, so the plasmon linewidth grows, which is in the qualitative agreement with experiment in Ref. 7.

The important feature of Eqs. (13) and (10) is that the loss function depends on the amplitude of the external perturbation A_1, which evidently is not the case in the linear theory.

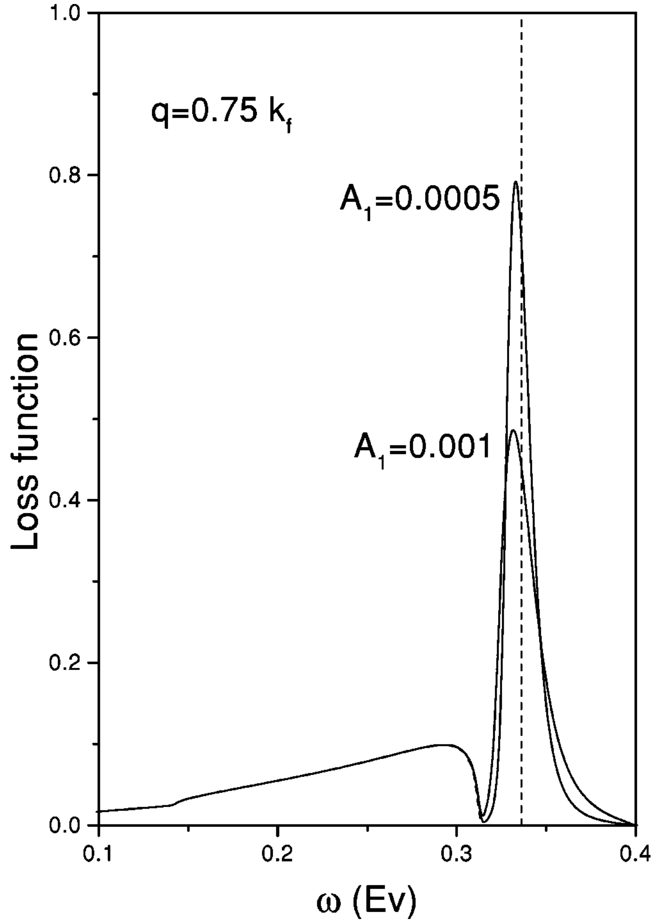

FIG. 1. Energy-loss function of 2D electron gas with parameters indicated in the text. The solid lines are the plots of Eq. (13). The vertical dashed line stands for the δ peak at a plasmon frequency for the loss function as predicted by linear theory.

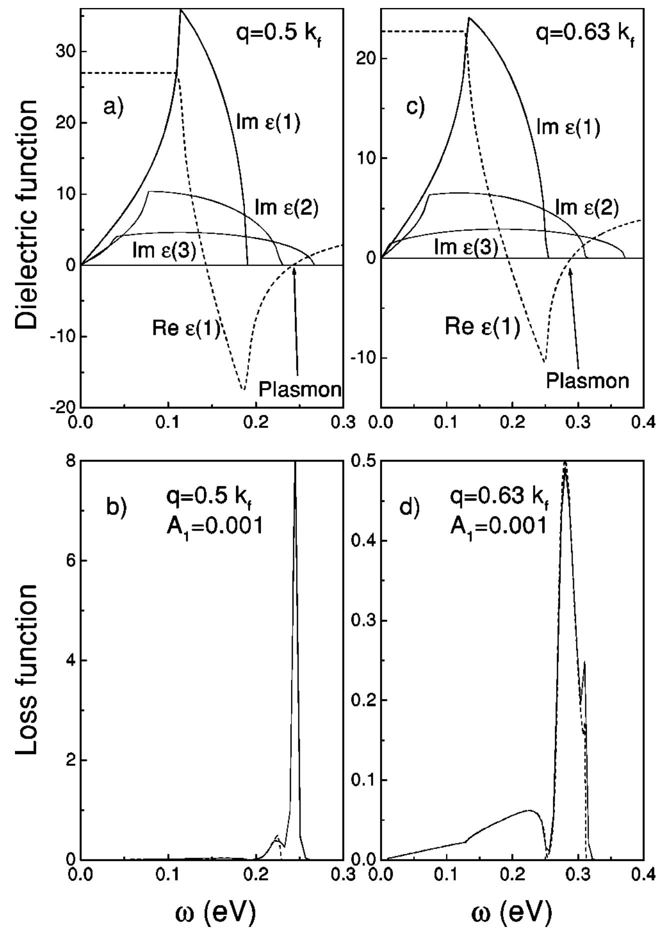

FIG. 2. The damping of plasmon at a given q is determined by the first of $\epsilon(n)$'s which have a nonzero imaginary part at plasmon frequency (see text). In (a) and (c) the solid lines are $\mathrm{Im}\,\epsilon(n)$ for $n=1,2,$ and 3. The dashed line is $\mathrm{Re}\,\epsilon(1)$. In (b) and (d) the solid (dashed) lines are loss functions in the approximation of three (two) waves, respectively.

As can be expected, the linewidth of plasmon decreases with decreasing A_1; however, in contrast with linear theory, the former never becomes zero at the finite latter. Let us add that if we plotted the absorbed energy Q (which is the physical quantity) instead of the loss function, the height of the plasmon peak would also decrease with decreasing wave vector, and no divergency occurs in the $A_1 \to 0$ limit.

Let us make a remark on the comparison of the above theory with electron-energy-loss spectroscopy (EELS) experiment. We treat here the problem of the response of electron gas to the perturbation by an external field. The interrelation of this problem with the problem of charged particles inelastic scattering has been studied in detail in Ref. 8, where the formulas relating these two tasks have been derived within the theory quadratic in the incident charge-target interaction. At present there exists no theory to relate these two tasks in the third and higher orders of the perturbation series. However, the theory based on Eq. (4) [in contrast with Eq. (3)] contains all orders of the external bare perturbation A_1. Thus we find ourselves in the position when, having solved the problem of the response to the external field, we cannot take advantage of this solution to describe quantitatively the inelastic scattering of charges by the same target. The closely related problem is that Eq. (13) depends on the amplitude of the external field, which has not a direct interpretation in the EELS setup. For all these difficulties, however, it is intuitively clear that the damping we have obtained in the response theory should manifest itself in the scattering event too.

In conclusion, we have constructed the response theory based on the relation between the external and the total field to second order in the latter. We have used this theory to study the response near and at the plasma resonance, and have demonstrated the existence of the plasmon damping completely due to the nonlinearity of the response. We have illustrated the theory by an explicit calculation in the case of 2D electron gas with the parameters corresponding to those of the S_1 surface band of Si(111)-$\sqrt{3}\times\sqrt{3}$-Ag, for which system the experiment shows the plasmon damping at wave vectors below the threshold value.

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[9] Then substituting $\phi=\phi_{\rm ext}/\epsilon$ into the quadratic term of Eq. (4), one finds

$$\epsilon_2^{-1}(\mathbf{q},\mathbf{k},\omega,\omega_1) = -\epsilon_2(\mathbf{q},\mathbf{k},\omega,\omega_1)/\epsilon(\mathbf{q},\omega)\epsilon(\mathbf{k},\omega_1)\epsilon(\mathbf{q}-\mathbf{k},\omega-\omega_1).$$

[10] We use the symmetry $\epsilon_2(n,m)=\epsilon_2(n,n-m)$ throughout (Ref. 6).

[11] Actually, it can be shown that Eq. (11) is exact to third order in the total field if $\mathrm{Im}\,\epsilon(1)=0$ (the case we are mostly interested here in). To keep the fourth-order terms in Eq. (11) would be inconsistent with having $\mathbf{j}$ to second order only.