PHYSICAL REVIEW A VOLUME 20, NUMBER 6 DECEMBER 1979

Correlational correction to plasmon dispersion

G. Kalman
Department of Physics, Boston College, Chestnut Hill, Massachusetts 02167

K. I. Golden
Department of Electrical Engineering, Northeastern University, Boston, Massachusetts 02115
(Received 18 December 1978)

The authors question the suggestion that plasmon dispersion *increases* for small values of the coupling over its random-phase-approximation value, and conclude that, contrary to what has been stated in the literature, it does not: high-frequency-moment sum-rule and Kramers-Kronig arguments, when properly treated, do not entail such a consequence.

The plasmon dispersion relation in a one-component plasma (ocp) for $k \to 0$

$$\omega^2(k) = \omega_0^2(1 + sk^2/\kappa^2)\,,$$
$$\kappa^2 = 4\pi e^2 n\beta\,, \quad \omega_0^2 = 4\pi e^2 n/m\,, \tag{1}$$

in the zero-coupling ($\gamma \equiv \kappa^3/4\pi n = 0$) limit exhibits the well-known Bohm-Gross behavior with

$$s(\gamma = 0) = 3\,. \tag{2}$$

For finite γ, however, the precise character of the function $s(\gamma)$ is unknown, although a few points have been obtained[1] through molecular-dynamics simulation:

$$s(\gamma = 1.71) = 4.96\,, \quad s(\gamma = 52.3) = 0.00\,,$$
$$s(\gamma = 2009) = -17.3\,. \tag{3}$$

Elsewhere[2] we have argued that a consistent small-γ expansion of the first and second equations of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy, as given by Coste,[3] leads to

$$\left.\frac{ds}{d\gamma}\right|_{\gamma=0} < 0\,.$$

At the same time, Baus[4] has found

$$\left.\frac{ds}{d\gamma}\right|_{\gamma=0} > 0$$

as a result of his approximate treatment of the BBGKY hierarchy. The positiveness of $ds/d\gamma$ is believed to be further corroborated through a work by Ichimaru, Tange, Totsuji and Pines[5] (ITTP), who argue that model-independent considerations require $ds/d\gamma > 0$ on quite general grounds. The arguments of ITTP rest on the high-frequency-moment sum rules pertaining to $\epsilon(\vec{k}, \omega)$, the plasma dielectric function, and on the Kramers-Kronig relations, relating its real and imaginary parts, $\epsilon'(\vec{k}, \omega)$ and $\epsilon''(\vec{k}, \omega)$, to each other. In this work we examine what consequences these two, indeed quite general, constraints entail as to the behavior of the plasmon dispersion curve. We will conclude that the proofs of ITTP are incorrect and that there is no *a priori* reason to accept $ds/d\gamma > 0$. We will also comment on the correct high-frequency behavior of $\epsilon(\vec{k}, \omega)$ for small values of γ.

The plasmon dispersion function is determined from the dispersion relation

$$\epsilon(\vec{k}, \omega) = 0 \tag{4}$$

where, to lowest order in γ,

$$\begin{aligned}\epsilon(\vec{k}, \omega) &= 1 + \alpha_0(\vec{k}, \omega) + \gamma\alpha_1(\vec{k}, \omega)\\ &= 1 + \alpha_0(\vec{k}, \omega)[1 + \gamma v(\vec{k}, \omega)]\end{aligned} \tag{5}$$

with $\alpha_0(\vec{k}, \omega)$ being the random-phase approximation (RPA) polarizability. Evidently, if we set

$$\omega(k) = \omega_0(k) + \delta\omega(k) \tag{6}$$

where $\omega_0(k)$ is the RPA plasmon solution,

$$\delta\omega(k) = -\gamma\,\frac{\alpha_0(\vec{k}, \omega = \omega_0(k))}{\dfrac{d}{d\omega}\alpha_0(\vec{k}, \omega = \omega_0(k))}\,v(\vec{k}, \omega = \omega_0(k))\,. \tag{7}$$

Momentum conservation requires that in an ocp

$$\lim_{k\to 0} v(\vec{k}, \omega) = -(k^2/\kappa^2)(\omega_0^2/\omega^2)\mathcal{V}(\omega) \tag{8}$$

(the $-\omega_0^2/\omega^2$ coefficient is inserted for convenience), and our attention will be focused on the function $\mathcal{V}(\omega) = \mathcal{V}'(\omega) + i\mathcal{V}''(\omega)$. Thus for $k \to 0$

$$\left.\frac{ds}{d\gamma}\right|_{\gamma=0} = \frac{2}{\omega_0}\,\frac{\alpha_0(\vec{k} = 0, \omega = \omega_0)}{\dfrac{d}{d\omega}\alpha_0(\vec{k} = 0, \omega = \omega_0)}\,\mathcal{V}(\omega_0) = -\mathcal{V}(\omega_0)\,. \tag{9}$$

Since $\alpha_0(\omega_0) < 0$, $(d/d\omega)\alpha_0(\omega_0) > 0$, it is the signature of $\mathcal{V}(\omega_0)$ which is to be determined.

The high-frequency expansion of $\alpha'(\vec{k}, \omega)$ can be derived from a similar expansion of $\hat{\alpha}(\vec{k}, \omega)$, the external polarizability,

$$\hat{\alpha}(\vec{k},\omega)=\alpha(\vec{k},\omega)/\epsilon(\vec{k},\omega)\,. \tag{10}$$

As will be discussed below, $\alpha''(\omega\to\infty)\to 0$ very quickly; thus, it is sufficient to consider

$$\hat{\alpha}'(\vec{k},\omega)\simeq\alpha'(\vec{k},\omega)/[1+\alpha'(\vec{k},\omega)] \tag{11}$$

or

$$\alpha'(\vec{k},\omega)\simeq\hat{\alpha}'(\vec{k},\omega)/[1-\hat{\alpha}'(\vec{k},\omega)]\,. \tag{12}$$

The expansion of $\hat{\alpha}'(\omega)$ in powers of $\omega^{-2}[\hat{\alpha}'(\omega)$, being an even function, cannot contain odd powers of $\omega]$, is related to the frequency moments of $\hat{\alpha}''(\omega)$ through the Kramers-Kronig relations

$$\hat{\alpha}'(\omega)=\int\frac{d\mu}{\pi}\frac{\hat{\alpha}''(\mu)}{\mu-\omega}=-\sum_{n=1}^{\infty}\int\frac{d\mu}{\pi}\hat{\alpha}''(\mu)\mu^{2n-1}\frac{1}{\omega^{2n}}\,. \tag{13}$$

The latter then can be calculated by evoking the fluctuation-dissipation theorem which links the frequency moments of $\hat{\alpha}''(\omega)$ to the time derivatives of the equal-time two-point correlations in the system. We note, however, that the above derivation rests on two considerations: (i) that $\hat{\alpha}(\omega)$ is a plus function, and (ii) that $\hat{\alpha}''(\omega)$ converges quickly enough to allow for the μ/ω expansion in the integrand. Although on physical grounds one is confident that both of these conditions are satisfied for the exact $\hat{\alpha}(\omega)$, it is to be carefully examined whether they are not violated in a particular approximation. If we expand $\hat{\alpha}$ in γ and restrict our attention to the $O(\gamma^0)$ and $O(\gamma^1)$ terms, evidently

$$\epsilon'=\frac{1}{[1-(\hat{\alpha}'_0+\Delta\hat{\alpha}'_0)-\gamma\hat{\alpha}'_1]}\,. \tag{14}$$

$\Delta\hat{\alpha}_0$ is an $O(\gamma^0)$ contribution of non-RPA origin: the existence of such terms in the high-frequency expansion of $\hat{\alpha}$ has been pointed out by ITTP and will also be discussed in the following.

The zeros of $\epsilon(\omega)$ come from the infinities of the denominator in Eq. (12). If $\hat{\alpha}'(\omega)$ is expanded in ω^{-2}, no finite number of terms can lead to such an infinity. The infinite series representing $\hat{\alpha}'_0(\omega)$ can be easily resummed into the RPA $\hat{\alpha}'_0(\omega)=\alpha'_0(\omega)/[1+\alpha'_0(\omega)]$, expressed in terms of the known RPA polarizability $\alpha'_0(\omega)$. No such resummation, however, is known for $\hat{\alpha}'_1(\omega)$ or $\Delta\hat{\alpha}'_0(\omega)$. Moreover, if one restricts oneself to a finite number of terms in the latter, a glance at Eq. (14) makes it evident that no shift in the plasmon dispersion can result.

This observation should be sufficient to demonstrate the fallacy of any derivation that proposes to calculate the shift of the plasmon dispersion from the analysis of a finite-order ω^{-2} expansion of $\hat{\alpha}'(\omega)$. It will, however, be instructive to see the origin of the purported negativeness of $ds/d\gamma|_{\gamma=0}$ in the derivation of ITTP. This is quite simple. Consider the expansion of $\hat{\alpha}'$, in powers of γ and, in addition, of $x=\omega_0^2/\omega^2$ and $y=k^2/\kappa^2$:

$$\hat{\alpha}(\vec{k},\omega)=\sum_{n,m}(\hat{a}_{nm}\gamma^n y^m x+\hat{b}_{nm}\gamma^n y^m x^2+\hat{c}_{nm}\gamma^n y^m x^3+\cdots)\,. \tag{15}$$

We know (e.g., from ITTP) that

$$\begin{aligned}&\hat{a}_{00}=-1\,, && \hat{a}_{nm}=0(m\neq 0,n\neq 0)\,,\\ &\hat{b}_{00}=-1\,, && \hat{b}_{01}=3\,, \qquad \hat{b}_{0m}=0(m\neq 0,1)\,,\\ &\hat{b}_{10}=0\,, && \hat{b}_{11}\neq 0\,,\\ &\hat{c}_{00}=+1\,, && \hat{c}_{01}\neq 0\,,\\ &\hat{c}_{10}=0\,, && \hat{c}_{11}=0\,,\end{aligned} \tag{16}$$

Expansion of (14) to order γ now yields

$$\begin{aligned}&\epsilon'=\bar{\epsilon}_0+\gamma\bar{\epsilon}_0^2\hat{\alpha}_1\,,\\ &\bar{\epsilon}_0=1/[1-(\hat{\alpha}_0+\Delta\hat{\alpha}_0)]\,.\end{aligned} \tag{17}$$

Substituting this into (7) and taking into account that, as previously pointed out, momentum conservation requires both $\hat{\alpha}_1$ and $\Delta\hat{\alpha}_0$ to be at least of $O(k^2)$ [cf. (14)], we find that the order k^2 contribution of $\delta\omega(k)$ is

$$\delta\omega(k)\simeq-\gamma\hat{\alpha}_1(\vec{k},\omega=\omega_0)\frac{\epsilon_0^2(\vec{k}=0,\omega=\omega_0)}{\frac{d}{d\omega}\epsilon_0(\vec{k}=0,\omega=\omega_0)}\,. \tag{18}$$

Since $\epsilon_0(\vec{k}=0,\omega=\omega_0)=0$, this is in accordance with our previous observation that, to any finite order, $\delta\omega(k)=0$. However, if we now formally apply an x-expansion, *before* setting $x=1$, we can write

$$\begin{aligned}\delta\omega(k)&=\tfrac{1}{2}\gamma y x^{3/2}(\hat{b}_{11}x^2+\hat{c}_{11}x^3+\cdots)(1-x)^2\\ &\simeq\tfrac{1}{2}\gamma y x^{3/2}\hat{b}_{11}x^2+(\hat{c}_{11}-2\hat{b}_{11})x^3+\cdots\,.\end{aligned} \tag{19}$$

Depending now on the order x at which one elects to terminate the series, one finds, successively,

$$\begin{aligned}&\delta\omega(k)=\tfrac{1}{2}\gamma y\hat{b}_{11}\,,\quad \delta\omega(k)=\tfrac{1}{2}\gamma y(\hat{c}_{11}-\hat{b}_{11})\,,\\ &\delta\omega(k)=\tfrac{1}{2}\gamma y(\hat{d}_{11}-\hat{c}_{11})\,.\end{aligned} \tag{20}$$

In particular, in view of (14), termination at x^2 yields

$$\left.\frac{ds}{d\gamma}\right|_{\gamma=0}=-\frac{4}{15}\,, \tag{21}$$

while termination at x^3 converts this into

$$\left.\frac{ds}{d\gamma}\right|_{\gamma=0}=+\frac{4}{15}\,. \tag{22}$$

Equation (21) is the earlier result of Ichimaru and Tange,[6] and Eq. (22) is the "corrected" result of ITTP. It is obvious that by suitably choosing the power of x at which the series is terminated, practically any desired value of $\delta\omega(k)$ can be generated: none of them can claim superiority over any of the others. As already pointed out by Baus,[4] the arbitrary termination of the high-frequency expansion for the purpose of the analysis of dispersion phenomena in the vicinity of $x=1$ is completely unjustified.

We now turn to examining the consequences of the Kramers-Kronig (KK) relations. First we note that the real and imaginary parts of $\epsilon(\vec{k},\omega)$ have rather different analytic structures as functions of γ: while ϵ' is analytic in γ (at least for ω's in the vicinity of ω_0) i.e., $\lim_{\gamma\to 0}\epsilon' \sim \gamma$, ϵ'', in addition to having a γ-proportional contribution, contains a so-called "dominant" term which renders $\lim_{\gamma\to 0}\epsilon'' \sim \gamma\log\gamma^{-1}$. In view of the linear relationship between ϵ' and ϵ'', as required by the KK relations, the Hilbert transform of the dominant term must vanish. The second observation we make is that, to assure the requisite plus-function character of $\epsilon(\omega)$ in the $k\to 0$ limit in the expression $\lim_{k\to 0}\alpha_1(\vec{k},\omega) = (\omega_0^4/\omega^4)(k^2/\kappa^2)\mathcal{V}(\omega)$, the $1/\omega^4$ is to be properly interpreted as $(1/\omega)\lim_{o\to 0} 1/(\omega+io)^3$. ITTP, on the other hand, implied the assumed dominance of the Hilbert transform of $\alpha''_{1,\mathrm{dom}}$ in determining α'_1 and claimed that

$$\alpha'_{1,\mathrm{dom}}(\omega_0) = I + J$$

with

$$I = \int_{-\infty}^{-\omega_1} \frac{d\mu}{\pi}\,\frac{\alpha''_{1,\mathrm{dom}}(\mu)}{\mu-\omega_0} + \int_{\omega_1}^{\infty} \frac{d\mu}{\pi}\,\frac{\alpha''_{1,\mathrm{dom}}(\mu)}{\mu-\omega_0}\,,$$

$$J = \int_{-\omega_1}^{\omega_1} \frac{d\mu}{\pi}\,\frac{\alpha''_1(\mu)}{\mu-\omega_0}\,. \quad \text{(ITTP)} \tag{23}$$

In the literature[3,4,7] there is a general agreement concerning the dominant part of $\mathcal{V}''(\omega)$,

$$\mathcal{V}''_{\mathrm{dom}}(\omega) = (8/15\sqrt{\pi})(\log\gamma^{-1})(\omega_0/\omega)\,. \tag{24}$$

In choosing ω_1 in Eq. (23) to be sufficiently small, ITTP correctly estimate that I is negative. However, they erroneously argue that J is also negative, leading to $\alpha'_1(\omega_0)$ negative and $ds/d\gamma$ positive. Their line of reasoning rests on two premises: (a) that $\alpha''(\mu) \equiv \alpha''_0(\mu) + \gamma\alpha''_1(\mu) > 0$ for $\mu > 0$ and, (b) that for small k, $\alpha''_1 \sim k^2$, $\alpha''_0 \sim \exp(-\mu^2/k^2 v_{\mathrm{th}}^2)$, and thus α''_1 dominates. The fallacy of this argument lies in ignoring the fact that "small k" actually means $kv_{\mathrm{th}}/\mu \ll 1$ in this context, and in calculating the Hilbert transform for any small, but fixed k, one always runs into (positive) μ domains where $kv_{\mathrm{th}}/\mu > 1$, and where α''_1 is not dominant. Thus, for μ sufficiently small in the interval $(0,\omega_1)$, α'' is positive, as it should be, even though α''_1 does not have to be: that α''_1 is indeed negative and therefore J positive is demonstrated, however, below.

The precise reason for which ITTP's evaluation of Eq. (23) is incorrect can be understood by letting $\omega_1 \to 0$; with this choice ITTP would have $I \to -\infty$ and $J \to 0$, whereas the properly interpreted singular denominator $(1/\mu)^3 = P(1/\mu)^3 - (i\pi/2)\ddot{\delta}(\mu)$ yields, instead of (23),

$$\alpha'_{1,\mathrm{dom}}(\omega_0) = \omega_0^3\,\frac{k^2}{\kappa^2}\int \frac{d\mu}{\pi}\,\frac{1}{(\mu-\omega_0)}\,\mathrm{Im}\,\frac{\mathcal{V}_{\mathrm{dom}}(\mu)}{\mu^3}$$
$$= K + L\,, \tag{25}$$

with

$$K = \omega_0^3\,\frac{k^2}{\kappa^2}\int \frac{d\mu}{\pi}\,\frac{1}{(\mu-\omega_0)}\,\frac{\mathcal{V}''_{\mathrm{dom}}(\mu)}{\mu^3}$$
$$= \frac{k^2}{\kappa^2}\left[\mathcal{V}'_{\mathrm{dom}}(\omega_0) - \mathcal{V}'_{\mathrm{dom}}(0) - \tfrac{1}{2}\omega_0^2\ddot{\mathcal{V}}'_{\mathrm{dom}}(0)\right],$$

$$L = -\frac{\omega_0^3}{2}\,\frac{k^2}{\kappa^2}\pi\int \frac{d\mu}{\pi}\,\frac{1}{(\mu-\omega_0)}\,\ddot{\delta}(\mu)\mathcal{V}'_{\mathrm{dom}}(\mu)$$
$$= \frac{k^2}{\kappa^2}\left[\mathcal{V}'_{\mathrm{dom}}(0) + \tfrac{1}{2}\omega_0^2\ddot{\mathcal{V}}'_{\mathrm{dom}}(0)\right].$$

The equivalent of ITTP's I is now K [due to $P(1/\mu)^3$], which is exactly cancelled by L [due to $\ddot{\delta}(\mu)$] giving $\alpha'_{1,\mathrm{dom}}(\omega_0) = 0$, consistent with our first observation that the Hilbert transform of the dominant term must vanish. At the same time,

$$\alpha'_1(\omega_0) = \frac{k^2}{\kappa^2}\int \frac{d\mu}{\pi}\,\frac{1}{(\mu-\omega_0)}\left(\frac{\omega_0}{\mu}\right)^3 \mathcal{V}''_{\text{non-dom}}(\mu)\,. \tag{26}$$

In the above line of reasoning, we have tacitly assumed that the ω^{-5} behavior of $\alpha''_1(\omega)$, known to be exact for frequencies above the collision frequency, can be used to represent α''_1 down to $\omega\to 0$. This is almost certainly not so, but this point is not crucial to our argument. Our model only illustrates that the plus-function character has to be preserved correctly to any order of the approximation. Any other reasonable perturbative model (e.g., a "hydrodynamic" one[4] with $\alpha''_1 \sim \omega^{-3}$ for $\omega\to 0$) gives basically the same result: a large positive contribution to the integral for $\omega\to 0$, coming from a large negative value of α''_1.[8]

We conclude that neither of the proposed proofs of the positiveness of $(ds/d\gamma)$ can be regarded as being correct and that at the present time there is no general information available to determine the sign of $(ds/d\gamma)$. Nevertheless, a concrete calculation based on Coste's[3] formulation can be performed, which provides both $\mathcal{V}'(\omega)$ and $\mathcal{V}''(\omega)$. They can easily be shown[2] to satisfy the combined Eqs. (25) and (26), with $\alpha'_1(\omega_0) > 0$ leading to $(ds/d\gamma) < 0$.

We have already remarked that the satisfaction of Eq. (13) requires a sufficiently rapid vanishing of $\hat{\alpha}''(\omega) \simeq \alpha''(\omega)$ for $\omega\to\infty$. The calculated $\mathcal{V}''(\omega)$, however, does not satisfy this criterion. This can already be seen from Eq. (24): $\lim_{\omega\to\infty}\hat{\alpha}''_{1,\mathrm{dom}}(\omega) = C_1/\omega^5$. At the same time, $\hat{\alpha}''_{1,\mathrm{non\text{-}dom}}(\omega)$ can be shown to have the behavior $\lim_{\omega\to\infty}\hat{\alpha}''_{1,\mathrm{non\text{-}dom}}(\omega) = (C_2 \log|\omega| + C_3)/\omega^5$.

This unphysical behavior of $\alpha''_1(\omega)$ for $\omega\to\infty$ is also reflected in the pathological high-frequency properties of $\alpha'_1(\omega)$. Indeed, while the calculated value of $\alpha'_1(\omega)$ satisfies the $1/\omega^4$ moment sum rule, it already fails to satisfy the $1/\omega^6$ sum rule. Moreover, in the high-frequency expansion of $\alpha'_1(\omega)$ an anomalous $1/|\omega|^5$ term appears. The origin of all these problems can be traced back to the well-known nonuniformity of the γ expansion: it is expected to fail both for $\omega < O(\gamma)$ and $\omega > O(1/\gamma)$. Thus the actual high-frequency behavior of $\alpha(\omega)$ cannot be predicted on the basis of the γ expansion. On physical grounds, however, one expects the collisional damping to vanish exponentially for frequencies higher than the inverse duration time of a typical collision, ω_0/γ. Conjecturing that this effect can be well represented by cutting off the integral in Eq. (26) at $\mu = \omega_0/\gamma$, all the anomalies cure themselves in a straightforward manner. At the same time, however, the introduction of the γ-dependent cutoff in the Hilbert transform eliminates the simple power-by-power correspondence between $\alpha'(\omega)$ and $\alpha''(\omega)$ in their respective γ expansions. In general, a term of order $\gamma^{n''}$ in $\alpha''(\omega)$ contributes to a term to order $\gamma^{n'}$ ($n' < n''$) in $\alpha'(\omega)$. In particular, $\alpha''_1(\omega)$ contributes to $\alpha'_0(\omega)$, thus providing a zero-order term of non-RPA origin. ITTP discovered the existence of such contributions by analyzing the structure of equal time two-point correlations; the qualitative analysis outlined above indicates how terms of this type emerge in the perturbation expansion of the dielectric function. Once the convergence of all the moment integrals is assured, there is no $1/|\omega|^5$ term generated in $\alpha'(\omega)$; moreover, the possible contribution of the higher order $\gamma^2\alpha''_2(\omega)$ to $\gamma\alpha'_1(\omega)$ opens the way to the emergence of a correct coefficient for the $1/\omega^6$ term.

ACKNOWLEDGMENTS

This work was supported in part by AFOSR Grant No. 76-2960; the work of one of us (G.K.) was partially supported by NATO Grant No. 1211.

---

[1] J. P. Hansen, E. L. Pollock, and I. R. McDonald, Phys. Rev. Lett. 32, 277 (1974); J. P. Hansen, I. R. McDonald, and E. L. Pollock, Phys. Rev. A 11, 1025 (1975).

[2] K. I. Golden and G. Kalman, Phys. Rev. A 19, 2112 (1979); P. Carini, G. Kalman, and K. I. Golden, Bull. Am. Phys. Soc. 23, 820 (1978).

[3] J. Coste, Nucl. Fusion 5, 284 293 (1965).

[4] M. Baus, Phys. Rev. A 15, 744 (1977).

[5] S. Ichimaru, H. Totsuji, T. Tange, and D. Pines, Prog. Theor. Phys. 54, 1077 (1975).

[6] S. Ichimaru and T. Tange, Phys. Rev. Lett. 32, 102 (1974).

[7] Y. H. Ichikawa, Prog. Theor. Phys. 24, 1083 (1960); C. R. Willis, Phys. Fluids 5, 219 (1962); D. DuBois, V. Gilinsky, and M. Kivelson, Phys. Rev. Lett. 8, 419 (1962).

[8] One could also correctly observe that *all* perturbation expansions in γ break down below the collision frequency ($\sim\gamma$). Should one use the unexpanded $\alpha''(\mu)$ for $-\omega_1 < \mu < \omega_1$, the splitting of $\alpha''(\mu)$ into "dominant" and "nondominant" parts becomes, of course, meaningless in this domain, and one expects that the resulting J, in addition to cancelling I, provides a contribution to the nondominant $\alpha'(\omega_0)$.