# Quantum optics of plasmon polaritons and velocity sum rules

O. Al-Dossary,* M. Babiker, and N. Enfati
*Department of Physics, University of Essex, Colchester CO4 3SQ, United Kingdom*
(Received 17 November 1995)

We demonstrate that the reciprocal-space Hamiltonian method for quantizing the electromagnetic fields in a dispersive dielectric medium leads to inconsistencies when applied to the cases of plasmon polaritons and coupled phonon-plasmon polaritons. In particular, we show that the use of the standard expressions for the dielectric functions in these two cases leads to the violation of the Huttner-Barnett velocity-ratio sum rule and, hence, to the breakdown of the equal-time canonical field commutation relations. We show that correct behavior is recoverable after the dielectric functions are appropriately regularized. Regularization effectively leads in each case to the introduction of an additional transverse low-frequency polariton branch. This is sufficient to rectify the theory and there is no need to invoke the presence of the longitudinal modes. The theory is illustrated with reference to the coupled phonon-plasmon polaritons of Si-doped GaAs. [S1050-2947(96)04409-5]

PACS number(s): 42.50.−p, 03.70.+k

## I. INTRODUCTION

Over the last decade or so considerable research effort has been aimed at extending quantum optics theory to the realm of continuous dielectric media [1–18]. Recent interest has been fueled by advances in the detection of quantum optical processes in media [19]. An essential first step in the study of quantum processes in media is the development of an unambiguous field quantization program that can be implemented straightforwardly, preferably in a manner analogous to the case of nonrelativistic quantum electrodynamics in vacuum. The issue of quantization has been complicated by the nonlocal nature of most dielectric media in both space and time, as reflected in the dependence of the dielectric function $\epsilon(\omega,\mathbf{k})$ on frequency and wave vector.

The problem is considered to have been solved for a certain class of linear media, namely, those characterized by a single frequency-dependent dielectric function $\epsilon(\omega)$. The procedure used follows the guidelines of classical electrodynamics with the fields being expressed as linear combinations of monochromatic components and with Maxwell's equations determining the spatial dependence. Quantization is achieved by replacing the amplitudes with operators that are made to obey boson commutation relations. The recent work by Huttner and Barnett [16] provided justification from a canonical perspective that for such media quantization can proceed in this standard fashion. One of the main requirements of the general theory is that the momentum which is canonically conjugate to **A**, the transverse vector potential of light in the medium, is $-\mathbf{D}$ as is the case in earlier work on microscopic molecular quantum electrodynamics [20]. Huttner and Barnett, however, based their work on model media; they did not proceed to discuss application to a specific dielectric medium nor did they consider the details of coupled modes.

The formal procedure of quantizing the three-dimensional polariton fields in a real medium characterized by a given $\epsilon(\omega)$ consists of writing down the Hamiltonian for each mode and constructing the quantum-mechanical operator of the vector potential that conforms with Maxwell's equations, together with the boson commutation relations between the mode annihilation and creation operators [21]. This enables the equal-time canonical commutation relations between the total field components to be evaluated. One of the characteristic features of the transverse electromagnetic fields quantized in this manner is their dependence on the group and phase velocities $v_g^j$ and $v_p^j$ of the polariton branches $j$. There are two sum rules involving the velocities, both of which have been derived by Huttner and Barnett [16]. The first is the sum of products of the velocities in the form $\Sigma_j v_g^j v_p^j/\overline{c^2}=1$, where $\overline{c^2}=(\epsilon_0\epsilon_\infty\mu_0)^{-1}$ and the second is the sum of the velocity ratios $\Sigma_j v_g^j/v_p^j=1$. As we explain shortly, the validity of the velocity-ratio sum rule is crucial for the identification of the electric displacement field **D** as the minus of the momentum canonically conjugate to the transverse vector potential **A**. Because of its importance in the quantization program, we shall refer to the velocity-ratio sum rule as the Huttner-Barnett sum rule.

In this paper we examine the implications of applying the reciprocal-space quantization methods to two important examples of polariton fields in real media, namely, plasmon polaritons and coupled phonon-plasmon polaritons. Plasmon polaritons are well characterized modes of light in bulk metals and in suitably doped semiconductors. If the semiconductor is a polar material the plasmons can couple strongly with transverse-optical phonons leading to coupled phonon-plasmon modes. These modes have been studied extensively by both theory and experiment and are now considered to be well understood. However, as we show below, the quantum optics theory of such polaritons is beset by undesirable features that need to be rectified before the theory can be used to study associated quantum optical processes.

In Sec. II we give a brief outline of the general formalism involving the quantization of polaritons with a given dielectric function $\epsilon(\omega)$. In Sec. III we discuss plasmon polaritons using analytical methods to exhibit the breakdown of the

*Permanent address: Physics Department, King Saudi University, Riyadh 11451, Saudi Arabia.

 

Huttner-Barnett velocity sum rule and the conflicting identification of canonical field variables for this particular case. In Sec. IV we analyze the case of coupled modes and, using typical parameters for doped GaAs, we display the behavior of the phase and group velocities for the various polariton branches and the associated sum rules. We thus demonstrate both analytically and numerically the breakdown of the quantization scheme for this case, too. In Sec. V we show how the discrepancies that are inherent in the plasmon and coupled phonon-plasmon dielectric functions can be remedied by regularization. Section VI contains our comments and final conclusions.

## II. QUANTIZED POLARITONS

For simplicity, we confine our attention to transverse fields within dielectric media characterized by real dielectric functions. The assumption of the existence of a dielectric function is central to the reciprocal-space Hamiltonian method of field quantization in continuous media. A homogeneous nonmagnetic medium characterized by a single frequency-dependent dielectric function $\epsilon(\omega)$ has a discrete polariton Hamiltonian which, in the radiation gauge, can be written in terms of a transverse vector potential in the following form:

$$H_{j\lambda}(\mathbf{k_n}) = \frac{1}{2}\,\epsilon_0 \int_{L^3} dV \left[ \left( \frac{\partial(\omega\epsilon)}{\partial\omega} \right)_j \dot{\mathbf{A}}^2_{j\lambda}(\mathbf{r},t,\mathbf{k_n}) + c^2\{\boldsymbol{\nabla}\times\mathbf{A}_{j\lambda}(\mathbf{r},t,\mathbf{k_n})\}^2 \right]. \tag{1}$$

This is the familiar general polariton field Hamiltonian [22], written here in terms of the vector potential. It is well known that this Hamiltonian embodies both the electromagnetic field energy and the mechanical energy of the medium. We have verified by explicit calculations that for polar optical phonons, plasmons, and coupled modes the Hamiltonian can be derived from a Lagrangian starting point [23] or from the requirement of conservation of energy flow [24]. In these cases the results can be cast in the general form given in Eq. (1) once the dielectric function has been identified.

The system of fields is assumed to be confined in a cubic cavity of volume $L^3$ subject to periodic boundary conditions. The fields appearing in Eq. (1) are associated with a well-defined mode labeled by $j$, $\lambda$, and $\mathbf{k_n}$ where $\mathbf{n}\equiv(n_x,n_y,n_z)$. The complete fields arise by the superposition of such independent modes. The modes of frequency $\omega_j(k_\mathbf{n})$, wavevector $\mathbf{k_n}$, polarization $\lambda=1,2$, in general, belong to one of several possible branches $j$ satisfying the polariton dispersion relation

$$k^2 = \frac{\omega^2\epsilon}{c^2}. \tag{2}$$

The notation is such that the presence of the label $j$ indicates that an expression should be evaluated at frequency $\omega_j$. The quantization is done mode by mode and begins by writing for the vector potential associated with the discrete polariton

$$\mathbf{A}_{j\lambda}(\mathbf{r},t,\mathbf{k_n}) = \widetilde{\mathbf{A}}_{j\lambda}(\mathbf{r},t,\mathbf{k_n}) + \widetilde{\mathbf{A}}^\dagger_{j\lambda}(\mathbf{r},t,\mathbf{k_n}). \tag{3}$$

Superposition gives rise to the complete transverse vector potential in the form

$$\mathbf{A}(\mathbf{r},t) = \sum_{j,\lambda}\sum_n \{\widetilde{\mathbf{A}}_{j\lambda}(\mathbf{r},t,\mathbf{k_n}) + \widetilde{\mathbf{A}}^\dagger_{j\lambda}(\mathbf{r},t,\mathbf{k_n})\}. \tag{4}$$

In the three-dimensional bulk, in the limit $L\to\infty$, the explicit form of $\widetilde{\mathbf{A}}_{j\lambda}$ can easily be specified in terms of transverse plane waves with $\mathbf{k}$ a continuous variable. The remaining requirement is that the polariton Hamiltonian Eq. (1) reduces to the canonical form. The complete transverse vector potential operator can then be written in the form

$$\mathbf{A}(\mathbf{r},t) = \sum_{j,\lambda} \int d^3\mathbf{k} \left\{ \frac{\hbar}{2(2\pi)^3\omega_j\epsilon_0\epsilon_j} \left( \frac{v_g^j}{v_p^j} \right) \right\}^{1/2} \times\{\hat{\mathbf{e}}^j(\mathbf{k},\lambda)a^j(\mathbf{k},\lambda)e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_j(k)t} + \text{H.c.}\}, \tag{5}$$

where H.c. denotes Hermitian conjugate and $\hat{\mathbf{e}}^{\,j}(\mathbf{k},\lambda)$ are unit polarization vectors satisfying [25,26]

$$\sum_{\lambda=1,2} \hat{e}^j_\alpha(\mathbf{k},\lambda)\hat{e}^{j'}_\beta(\mathbf{k},\lambda) = \delta_{jj'}\left[ \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} \right]. \tag{6}$$

For each allowed polariton branch $j$ the phase and group velocities are $v^{\,j}_p=(\omega/k)_j$ and $v^{\,j}_g=(\partial\omega/\partial k)_j$. Their ratio satisfies the identity [27]

$$\frac{v_p^j}{v_g^j} \equiv 1 + \left( \frac{\omega}{2\epsilon}\,\frac{\partial\epsilon}{\partial\omega} \right)_j. \tag{7}$$

There are two other important identities involving the group and phase velocities in sums over contributions arising from the allowable polariton branches. The first is the velocity-product sum in the form

$$\sum_j v_p^j v_g^j = \sum_j \left( \frac{\omega}{k}\,\frac{\partial\omega}{\partial k} \right)_j = \frac{1}{2k}\,\frac{\partial}{\partial k} \sum_j \omega_j^2 \tag{8}$$

and the second is the velocity-ratio sum in the form

$$\sum_j \left( \frac{v_g^j}{v_p^j} \right) = \sum_j \left( \frac{k}{\omega}\,\frac{\partial\omega}{\partial k} \right)_j = \frac{1}{2}\,k\,\frac{\partial}{\partial k}[\ln\,\Pi_j(\omega_j^2)], \tag{9}$$

where $\Pi_j(\,)$ stands for the product of $(\,)$. The above two identities become sum rules when the right-hand sides are evaluated.

The operators $a^j(\mathbf{k},\lambda)$ in Eq. (5) are boson operators satisfying the commutation relations

$$[a^j(\mathbf{k},\lambda), a^{j'^\dagger}(\mathbf{k}',\lambda')] = \delta_{jj'}\delta_{\lambda\lambda'}\delta(\mathbf{k}-\mathbf{k}') \tag{10}$$

and the complete polariton Hamiltonian has the canonical form

$$H = \sum_{j,\lambda} \int d^3\mathbf{k}\; H_{j\lambda}(\mathbf{k}) = \tfrac{1}{2}\sum_{j\lambda} \int d^3\mathbf{k}\;\hbar\omega_j(k) \times[a^{j\dagger}(\mathbf{k},\lambda)a^j(\mathbf{k},\lambda) + a^j(\mathbf{k},\lambda)a^{j\dagger}(\mathbf{k},\lambda)]. \tag{11}$$

The complete electric field operator $\mathbf{E}$ and the complete electric displacement field operator $\mathbf{D}$ corresponding to Eq. (5) are obtainable in the forms

$$\mathbf{E}(\mathbf{r},t)=i\sum_{j,\lambda}\int d^3\mathbf{k}\left\{\frac{\hbar\omega_j}{2(2\pi)^3\epsilon_0\epsilon_j}\left(\frac{v_g^j}{v_p^j}\right)\right\}^{1/2}\times\{\hat{\mathbf{e}}^j(\mathbf{k},\lambda)a^j(\mathbf{k},\lambda)e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_j(k)t}-\text{H.c.}\} \quad (12)$$

and

$$\mathbf{D}(\mathbf{r},t)=i\sum_{j,\lambda}\int d^3\mathbf{k}\left\{\frac{\hbar\omega_j\epsilon_0\epsilon_j}{2(2\pi)^3}\left(\frac{v_g^j}{v_p^j}\right)\right\}^{1/2}\times\{\hat{\mathbf{e}}^j(\mathbf{k},\lambda)a^j(\mathbf{k},\lambda)e^{i\mathbf{k}\cdot\mathbf{r}-i\omega_j(k)t}-\text{H.c.}\}. \quad (13)$$

Equations (5) and (13) allow the explicit evaluation of the equal-time commutator between Cartesian components $\alpha$ and $\beta$ of these fields. With the use of Eqs. (10) and (6), we obtain, after some algebra,

$$[A_\alpha(\mathbf{r},t),-D_\beta(\mathbf{r}',t)]=i\hbar\mathbf{I}_{\alpha\beta}(\mathbf{r},\mathbf{r}'), \quad (14)$$

where

$$I_{\alpha\beta}(\mathbf{r},\mathbf{r}')=\frac{1}{(2\pi)^3}\sum_j\int d^3\mathbf{k}\left(\frac{v_g^j}{v_p^j}\right)\left[\delta_{\alpha\beta}-\frac{k_\alpha k_\beta}{k^2}\right]e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}. \quad (15)$$

The above theory is general in the sense that it applies to any dielectric function $\epsilon(\omega)$. Clearly both $v_g^j$ and $v_p^j$ are, in general, $k$ dependent and we cannot without further information proceed to evaluate the $k$ integral in Eq. (15); the sum $\Sigma_j v_g^j/v_p^j$ must be obtained first.

In the most trivial case of a frequency-independent real $\epsilon$, we deduce at once from Eq. (2) that there is only one branch $j=1$ and, from Eq. (7), that the ratio $v_g^j/v_p^j$ is unity for all $k$. The integral in Eq. (15) can be evaluated and written in a closed form and Eq. (14) then becomes

$$[A_\alpha(\mathbf{r},t),-D_\beta(\mathbf{r}',t)]=i\hbar\mathbf{I}_{\alpha\beta}(\mathbf{r},\mathbf{r}')=i\hbar\,\delta^\perp_{\alpha\beta}(\mathbf{r}-\mathbf{r}'), \quad (16)$$

where $\delta^\perp_{\alpha\beta}(\mathbf{r}-\mathbf{r}')$ is the transverse Dirac $\delta$ function [25,26].

Thus we have shown that the general quantization program applied to this simple case yields the expected result of microscopic theory, namely, that $-\mathbf{D}$ is the momentum canonically conjugate to $\mathbf{A}$. For the desired result Eq. (16) to be true, in general the Huttner-Barnett sum rule

$$\sum_j\frac{v_g^j}{v_p^j}=1 \quad (17)$$

must be valid in general.

## III. PLASMON POLARITONS

Plasmon polaritons have the well-known dielectric function

$$\epsilon(\omega)=\epsilon_\infty\left(1-\frac{\omega_p^2}{\omega^2}\right), \quad (18)$$

where $\epsilon_\infty$ is the high-frequency dielectric constant of the material and $\omega_p$ is the plasma frequency. This dielectric function has an excellent record in the study of the transverse response of an electron gas in metals and in doped semiconductors in both the homogeneous and inhomogeneous cases [28–31].

The quantization procedure outlined in Sec. II can now be applied to the plasmon polariton case by simply substituting from Eq. (18). We see immediately that we have a single polariton branch and the evaluation of the velocity ratio in Eq. (7) gives at once

$$\frac{v_g}{v_p}=\epsilon(\omega)/\epsilon_\infty, \quad (19)$$

which conflicts with the Huttner-Barnett sum rule. On substituting in Eq. (15) we immediately conclude that the desired commutation relations do not hold and it is not valid to interpret $-\mathbf{D}$ as the momentum canonically conjugate to $\mathbf{A}$. On the other hand, we can easily show that the commutation relation between components of $\mathbf{A}$ and those of $-\epsilon_0\mathbf{E}$ for the plasmon polariton case [defined by Eqs. (18) and (19)] does reduce exactly to the canonical form. From Eqs. (5) and (12) we obtain, in a manner analogous to that leading to Eq. (14),

$$[A_\alpha(\mathbf{r},t),-\epsilon_0E_\beta(\mathbf{r}',t)]=i\hbar\,\delta^\perp_{\alpha\beta}(\mathbf{r}-\mathbf{r}'). \quad (20)$$

This immediately suggests that it is $-\epsilon_0\mathbf{E}$ and not $-\mathbf{D}$ that is the momentum canonically conjugate to $\mathbf{A}$. Paradoxically, the velocity-product sum rule

$$\sum_j\frac{v_g^jv_p^j}{\overline{c^2}}=1 \quad (21)$$

is valid for this case. This can be checked explicitly for the one-plasmon polariton branch using Eqs. (2) and (18) and with $\overline{c^2}=1/(\mu_0\epsilon_0\epsilon_\infty)$.

The conclusion based on Eq. (20) incorrectly leads to doubts about the validity of the quantization program and of the Huttner-Barnett sum rule. As we show later, both the quantization program and the Huttner-Barnett sum rule are valid, but it is the model dielectric function commonly used for the plasmons that is the cause of the inconsistencies.

## IV. COUPLED PHONON-PLASMON POLARITONS

In doped polar semiconductor materials the coupled phonon-plasmon polaritons are characterized by the dielectric function

$$\epsilon(\omega)=\epsilon_\infty\left[\frac{(\omega_L^2-\omega^2)}{(\omega_T^2-\omega^2)}-\frac{\omega_p^2}{\omega^2}\right], \quad (22)$$

where $\omega_L$ and $\omega_T$ are the longitudinal- and transverse-optical frequencies of the material and $\omega_p$ is the plasma frequency due to conduction electrons. This dielectric function also has an excellent record in the theory and experiment of coupled phonon-

plasmons [28–31]. The dispersion relation for the polaritons is given by Eq. (2) with Eq. (22). We obtain

$$\omega^4-\omega^2[k^2\overline{c^2}+\omega_L^2+\omega_p^2]+\omega_T^2[k^2\overline{c^2}+\omega_p^2]=0. \quad (23)$$

There are two branches $j=(+)$ and $j=(-)$ and the dispersion curves have the forms shown in Fig. 1. The validity of the

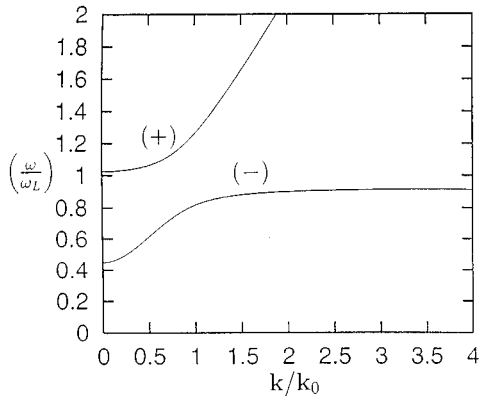


FIG. 1. Dispersion curves $\omega$ versus $k$ for phonon-plasmon polaritons in doped bulk GaAs. The modes satisfy the dispersion relation Eq. (2) with $\epsilon$ given by Eq. (22). The frequency $\omega$ is in units of $\omega_L$—the longitudinal-optical frequency of GaAs ($\hbar\omega_L \approx 36$ meV). The doping is such that $\omega_p = 0.5\omega_L$. The wave vector is in units of $k_0 = (\omega_L/\overline{c})$ where $\overline{c^2} = c^2/\epsilon_\infty$ with $\epsilon_\infty = 10.89$ the high-frequency dielectric constant of GaAs. The branches are labeled by $j = (+)$ and $j = (-)$.

Huttner-Barnett velocity-product sum rule can be shown at once using Eq. (23). We have, using Eq. (8),

$$\sum_{j=\pm} \frac{v_p^j v_g^j}{\overline{c^2}} = \frac{1}{2k\overline{c^2}} \frac{\partial}{\partial k} [\omega_+^2 + \omega_-^2] = 1. \tag{24}$$

On the other hand, the velocity-ratio sum rule is obtained using Eq. (9) as follows:

$$\sum_{j=\pm} \frac{v_g^j}{v_p^j} = \frac{1}{2} k \frac{\partial}{\partial k} \ln\{\omega_+^2 \omega_-^2\} = \frac{k^2\overline{c^2}}{k^2\overline{c^2} + \omega_p^2}. \tag{25}$$

The manifest $k$ dependence of the result of summing the velocity ratios indicates that the corresponding Huttner-Barnett sum rule does not hold in this case. Clearly, as $k \to \infty$ the right-hand side approaches unity and the sum rule applies in this large $k$ limit. However, strong deviations from unity occur at small $k$. To clarify this point further we need to examine the dependence on $k$ of the velocities $v_p^j$ and $v_g^j$ for each branch $j$. These are obtained in the form

$$v_p^j = \frac{c}{\sqrt{(\epsilon)_j}} \tag{26}$$

and

$$v_g^j = v_p^j \left\{ \frac{\omega^2(\omega^2-\omega_L^2)(\omega^2-\omega_T^2) - \omega_p^2(\omega^2-\omega_T^2)^2}{\omega^2(\omega^2-\omega_L^2)(\omega^2-\omega_T^2) + \omega^4(\omega_L^2-\omega_T^2)} \right\}_j . \tag{27}$$

Consider a real system in which the coupled modes are known to have been characterized by both theory and experiment, namely, Si-doped GaAs. Using typical parameters for this system it is possible to illustrate the variations of the phase and group velocities with wave vector for each polariton branch $j$ and then examine the variations of their product and ratio with $k$. In this manner we can examine the validity of the sum rules numerically.

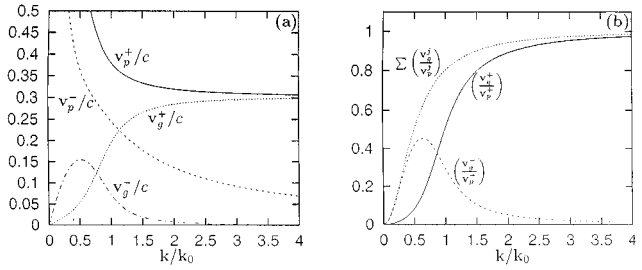


FIG. 2. (a) Variations with wave vector of the phase and group velocities $v_g^j$ and $v_p^j$ (in units of the velocity of light $c$) of the $\pm$ branches of Fig. 1. (b) Variations with wave vector of the ratio $v_g^j/v_p^j$ for $j = (\pm)$ branches and the sum of this ratio over the two branches.

We choose a typical doping density $n$ such that $\omega_p = 0.5\omega_L$. The dispersion curves for this case are displayed in Fig. 1 and they have the typical polariton resonance discussed by Huttner and Barnett [16]. The modes, however, carry both plasmon and optical phonon characters, depending on the value of $k$. In Fig. 2(a) we display the variations of the velocities $v_g$ and $v_p$ for each of the two branches and in Fig. 2(b) we display the corresponding variations of their ratio $v_g^j/v_p^j$, together with the sum of this quantity over the two branches. As we inferred from the analytical results, Fig. 2(b) confirms that the sum $\Sigma v_g^j/v_p^j$ is not equal to unity for all values of $k$. In fact the sum deviates drastically from the value unity across a range of wave vectors and shows a tendency to attain the asymptotic value of unity at large $k$. The discrepancy of the ratio sum is a demonstration of an apparent breakdown of the Huttner-Barnett sum rule and through it follows the breakdown of the quantization scheme for the phonon-plasmon polaritons.

The consequences of the result in Eq. (25) for the canonical commutation relations can be seen at once by direct substitution in Eqs. (14) and (15). We have

$$[A_\alpha(\mathbf{r},t), -D_\beta(\mathbf{r}',t)] = i\hbar\,\delta_{\alpha\beta}^{\perp}(\mathbf{r}-\mathbf{r}') - \frac{i\hbar}{(2\pi)^3} \times \int d^3\mathbf{k} \left( \frac{\omega_p^2}{k^2\overline{c^2} + \omega_p^2} \right) \times \left[ \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} \right] e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}. \tag{28}$$

This differs from the expected result in Eq. (16) by the additional integral on the right-hand side. The integral may be evaluated by choosing Cartesian components $\alpha$ and $\beta$, but for brevity, the results will not be presented here. The mere presence of this additional integral in Eq. (28) serves to further highlight the breakdown of the quantization scheme. As we show in Sec. V this breakdown is not a consequence of the invalidity of the Huttner-Barnett sum rule, but is symptomatic of the model embodied in the functional form of the dielectric function $\epsilon(\omega)$ Eq. (22).

## V. REGULARIZATION

The results for the pure plasmon polariton given in Sec. III, specifically Eqs. (19) and (20), conflict with those ex-

pected from the reciprocal-space quantization scheme in that they lead to the assertion that $-\epsilon_0\mathbf{E}$ is the momentum canonically conjugate to the vector potential, not $-\mathbf{D}$. Moreover, we have seen in Sec. IV that the coupled phonon plasmons deviate strongly from the expected behavior over a range of wave vectors and have shown explicitly the consequences of this on the canonical commutation relations. The theory of polaritons involves only transverse fields for which $\epsilon(\omega)\neq 0$. It could be argued that the longitudinal modes satisfying $\epsilon(\omega)=0$, which we have not considered, may have a role in the breakdown of the quantization scheme of plasmon polaritons and phonon-plasmon polaritons. However, it is easy to show that in the infinite bulk the longitudinal modes are quantizable separately [32] and are completely independent of the transverse polaritons.

The clue to the resolution of the problem stems from the fact that the quantization scheme works well for the pure optical phonon modes [21], obtainable from the dielectric function in Eq. (22) by setting $\omega_p=0$. Furthermore, we have seen in Sec. II that the use of the pure plasmon dielectric function Eq. (18) [obtainable from Eq. (22) by setting both $\omega_L$ and $\omega_T$ to zero] has undesirable consequences. The discrepancy must then be attributed to the plasmon part of the dielectric function and it is the low wave-vector limit of the dispersion curve that appears to be the source of the problem. This suggests a modification of the dielectric function Eq. (18) to read

$$\epsilon(\omega)=\epsilon_\infty\left(1+\frac{\omega_p^2}{\omega_0^2-\omega^2}\right), \tag{29}$$

where $\omega_0 \ll \omega_p$ is a small frequency whose sole purpose is to regularize the quantization program. With the use of the regularized plasmon dielectric function Eq. (29) the dispersion relation Eq. (2) yields for this case

$$\omega^4-\omega^2(\omega_p^2+\omega_0^2+k^2\overline{c^2})+k^2\overline{c^2}\omega_0^2=0. \tag{30}$$

The solution of this equation leads to two branches labeled $(\pm)$ with the new branch labeled $(-)$ in the low-frequency region. The validity of the Huttner-Barnett velocity-ratio sum rule can be checked analytically for the two branches $(\pm)$ satisfying Eq. (30). We obtain

$$\sum_{j=\pm}\left(\frac{v_g^j}{v_p^j}\right)=\frac{1}{2}\,k\,\frac{\partial}{\partial k}\ln(\omega_+^2\omega_-^2)=\frac{1}{2}\,k\,\frac{\partial}{\partial k}\ln(k^2\omega_0^2\overline{c^2})=1. \tag{31}$$

This proves that the Huttner-Barnett velocity-ratio sum rule is valid for the regularized plasmon polaritons and that this conclusion holds irrespective of the value of $\omega_0$, although the physical situation demands that $\omega_0\to 0$. It is also easy to check that as $k\to 0$ the frequency of the new branch $\omega_-\to 0$. In this limit the group- and phase-velocities both vanish ($v_g^-\to 0$ and $v_p^-\to 0$), but in such a manner that their ratio $v_g^-/v_p^-$ approaches unity. Clearly for small but finite $k$, the discrepancy in the contribution of the $(+)$ branch is exactly compensated by that from the new branch. The validity of the Huttner-Barnett sum rule for the regularized plasmon polaritons immediately leads us to correctly identify the momentum canonically conjugate to the vector potential as $-\mathbf{D}$. This (correct) identification should be contrasted with the (incorrect) identification made in Sec. II arising from the conventional form of the dielectric function.

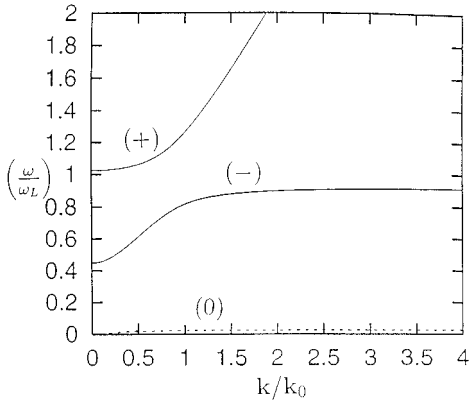


FIG. 3. Dispersion curves $\omega$ versus $k$ for regularized phonon-plasmon polaritons in doped bulk GaAs. The modes satisfy the dispersion relation Eq. (2) with $\epsilon=\epsilon_R$ as given by Eq. (32) and with $\omega_0=0.1\omega_p$. All other parameters are the same as those in Fig. 1. There are three branches here: two are analogues of the $(\pm)$ branches in Fig. 1 and the third is a low-frequency branch labeled (0) arising from the regularization procedure.

Consider next the case of coupled modes. By analogy we modify Eq. (22) to read

$$\epsilon_R(\omega)=\epsilon_\infty\left[\frac{(\omega_L^2-\omega^2)}{(\omega_T^2-\omega^2)}+\frac{\omega_p^2}{(\omega_0^2-\omega^2)}\right]. \tag{32}$$

With the use of the regularized dielectric function Eq. (32) the dispersion relation acquires a new polariton branch labeled (0) in the low-frequency region. This is shown in Fig. 3 for the same parameters as in Fig. 1 except that $\omega_0=0.1\omega_p$. There are corresponding velocity variations as shown in Fig. 4(a) with the ratio variations shown in Fig. 4(b). The sum of velocity ratios is seen in Fig. 4(b) to give unity for all values of $k$. This result can fortunately be verified analytically. It is easy to check that the dispersion relation is cubic in $\omega^2$ and has three distinct solutions $\omega(+)$, $\omega(-)$, and $\omega(0)$ for any given $k$ and that the product of the three solutions is given by the $\omega$-independent term. The analogue of Eq. (31) is

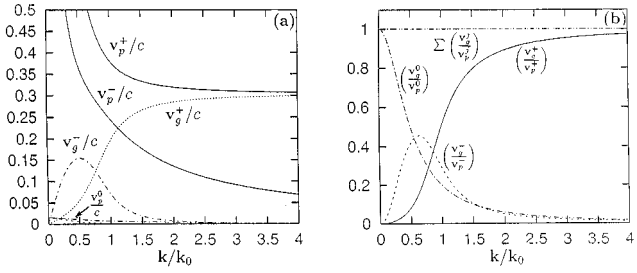


FIG. 4. (a) Variations of the phase and group velocities of the three polariton branches (+), (−), (0) with wave vector. The curves corresponding to the new polariton branch are in the low-velocity region and for convenience we have chosen to label only the upper curve $v_p^0/c$. Both curves have the same value at $k=0$, but the $v_g^0/c$ curve lies below the $v_p^0/c$ curve. (b) Variations with wave vector of the ratio $v_g^j/v_p^j$ for the three branches $j=(+)$, $(-)$, (0) and the sum of the ratio over the three branches.

$$\sum_{j=\pm,0}\left(\frac{v_g^j}{v_p^j}\right)=\frac{1}{2}\,k\,\frac{\partial}{\partial k}\ln\{\omega(+)^2\omega(-)^2\omega(0)^2\}$$

$$=\frac{1}{2}\,k\,\frac{\partial}{\partial k}\ln\{k^2\omega_{\mathrm{T}}^2\omega_0^2\overline{c^2}\}=1. \tag{33}$$

We conclude that the Huttner-Barnett velocity-ratio sum rule is valid for this case, too. As a result, the regularized polariton theory for coupled phonon-plasmon modes is amenable to quantization in reciprocal space with conclusions in line with those demanded by the general polariton theory described in Sec. II.

## VI. COMMENTS AND CONCLUSIONS

This paper has examined the validity of the reciprocal-space quantization program for two important cases of electromagnetic fields in media, namely, plasmon polaritons and coupled phonon-plasmon polaritons. The functional forms of these dielectric functions have been widely applied in the study of solid-state plasmas in metals, semimetals, and in doped semiconductors, both in light scattering and in electric transport. We have shown that the use of these dielectric functions in the context of the reciprocal-space quantization program is not justified. For the pure plasma case the results are misleading in that the momentum canonically conjugate to the vector potential is proportional to the electric field vector, rather than the displacement vector. This is a peculiar feature of the theory, leading as it does to a definite analytical result, emphasizing a wrong identification of the canonical field variables. In the coupled phonon-plasmon case discussed in Sec. IV we have obtained an analytical result for the sum of the velocity ratios and explored the effect of this on the commutation relations. Numerical results have clearly exhibited the breakdown of the corresponding Huttner-Barnett sum rule for the practical situation of coupled phonon plasmons in Si-doped GaAs. We have also shown how the regularized forms of the dielectric function stemming from Eq. (32) are free from the problems inherent in the conventional forms.

The manifestations of the inconsistencies of the dielectric functions were established by direct consideration of the commutation relations between field variables. Since quantum processes rely on these, their validity is important. The evaluation of the commutation relations has led us to the investigation of the validity of the Huttner-Barnett sum rule involving the ratio of the group and phase velocities of the polariton. It is the small-$k$ behavior of this ratio for the polariton branches that is responsible for the discrepancies. It follows that any effects involving this region of $k$ will, in principle, be prone to mishandling using the conventional theory, while the corresponding effects based on the regularized theory should be free from such difficulties. The regularization introduces a parameter $\omega_0$ whose precise value is unimportant provided it is much smaller than the plasma frequency. Clearly, a finite but small $\omega_0$ helps to soften the singularity at $\omega=0$ through the involvement of a low-frequency polariton branch.

Throughout this paper we have assumed that we are dealing with homogeneous unbounded media. There has been further effort recently to extend quantum optics theory in dielectric media to encompass the inhomogeneous case [33] and the theory could accommodate the case of media that show both dispersion and loss, i.e., those characterized by complex dielectric functions. In the inhomogeneous case electromagnetic and matter fields are made to obey boundary conditions at the interfaces between media characterized by different dielectric functions. One of the important features of the inhomogeneous case is the appearance of interface modes, which can contribute significantly to the quantum processes involving polaritons [32,34]. Furthermore, the presence of interfaces can lead to hybrid modes that possess linear combinations of transverse and longitudinal fields. It can, therefore, be suggested that the reciprocal space quantization techniques should be applied to interface problems to assess how a regularized theory would modify the details of the interaction of quantum systems in the vicinity of interfaces. These matters are currently under investigation and the results will be reported in due course.

## ACKNOWLEDGMENTS

The authors would like to thank Professor Rodney Loudon FRS and Dr. Steven Barnett FRSE for useful discussions.

[1] M. Hillery and L. D. Mlodinow, Phys. Rev. A **30**, 1860 (1984).
[2] M. D. Levenson, R. M. Shelby, A. Aspect, M. Reid, and D. F. Walls, Phys. Rev. A **32**, 1550 (1985).
[3] R. M. Shelby, M. Levenson, S. H. Perlmutter, R. G. deVoe, and D. F. Walls, Phys. Rev. Lett. **57**, 691 (1986).
[4] M. D. Levenson, R. M. Shelby, R. M. Reid, and D. F. Walls, Phys. Rev. Lett. **57**, 2473 (1986).
[5] B. Yurke, P. Grangier, R. E. Slusher, and M. J. Potasek, Phys. Rev. A **35**, 3586 (1987).
[6] M. J. Potasek and B. Yurke, Phys. Rev. A **35**, 3974 (1987).
[7] R. E. Slusher, P. Grangier, A. LaPorta, B. Yurke, and M. J. Potasek, Phys. Rev. Lett. **59**, 2566 (1987).
[8] C. M. Caves and D. D. Crouch, J. Opt. Soc. Am. B **4**, 1535 (1987).
[9] I. Abram, Phys. Rev. A **35**, 4661 (1987).
[10] M. Shirasaki and H. A. Haus, J. Opt. Soc. Am. B **7**, 30 (1990).
[11] K. J. Blow, R. Loudon, S. J. D. Phoenix, and T. J. Shepherd, Phys. Rev. A **42**, 4102 (1990).
[12] F. S. Johnson, Am. J. Phys. **58**, 1044 (1990).
[13] B. Huttner, S. Serulnik, and Y. Ben-Aryeh, Phys. Rev. A **42**, 5594 (1990).
[14] P. D. Drummond, Phys. Rev. A **42**, 6845 (1990).
[15] R. J. Glauber and M. Lewenstein, Phys. Rev. A **43**, 467 (1991).
[16] B. Huttner and S. M. Barnett, Phys. Rev. A **46**, 4306 (1992).
[17] P. W. Milonni, J. Mod. Opt. **42**, 1991 (1995).
[18] K. Koshino and A. Shimizu (unpublished).
[19] A. M. Steinberg, P. G. Kwiat, and R. Y. Chiao, Phys. Rev. Lett. **20**, 2421 (1992).

[20] M. Babiker and R. Loudon, Proc. R. Soc. London Ser. A **385**, 439 (1983).
[21] N. C. Constantinou, O. Al-Dossary, and M. Babiker, J. Phys. Condens. Matter **5**, 5581 (1993).
[22] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, 2nd ed. (Pergamon, Oxford, 1984).
[23] C. Kittel, *Quantum Theory of Solids*, 2nd ed. (Wiley, New York, 1987), Chap. 3.
[24] M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Clarendon, Oxford 1968).
[25] E. A. Power, *Introductory Quantum Electrodynamics* (Longmans, London, 1964).
[26] R. Loudon, *Quantum Theory of Light*, 2nd ed. (Clarendon, Oxford, 1983).
[27] D. L. Mills and E. Burstein, Rep. Prog. Phys. **37**, 817 (1974).
[28] P. M. Platzman and P. A. Wolff, *Waves and Interactions in Solid State Plasmas* (Academic, New York, 1973), p. 87.
[29] M. G. Cottam and D. R. Tilley, *Introduction to Surface and Superlattice Excitations* (Cambridge University Press, Cambridge, 1989).
[30] A. Mooradian and G. B. Wright, Phys. Rev. Lett. **16**, 999 (1966).
[31] E. L. Albuquerque and M. G. Cottam, Phys. Rep. **2**, 67 (1993).
[32] M. Babiker, Semicond. Sci. Technol. **7**, B52 (1992).
[33] For a review see the introduction to the paper by R. Matloob, R. Loudon, S. M. Barnett, and J. Jeffers, Phys. Rev. A **52**, 4823 (1995).
[34] M. Babiker, N. C. Constantinou, and B. K. Ridley, Phys. Rev. B **48**, 2236 (1993).