PHYSICAL REVIEW B VOLUME 44, NUMBER 24 15 DECEMBER 1991-II

Plasmon excitations in crystals

Behnam Farid and G. E. Engel
Department of Physics, Cavendish Laboratory, University of Cambridge, Madingley Road,
Cambridge CB3 0HE, United Kingdom

R. Daling and W. van Haeringen
Department of Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB, Eindhoven, The Netherlands

(Received 9 July 1991)

Using an analogy between the one-particle Green function G on the one hand and the dynamically screened electron-electron Coulomb interaction function W on the other for a many-electron system, we obtain a biorthonormal type of representation for the Fourier transform with respect to time of the function W. Based on the assumption that this function can be approximated in terms of simple poles in the complex energy plane, the connection between these poles and the energies of (damped) collective excitations, which are to be identified with plasmon excitations, is made clear. The biorthonormal-type representation of the screened interaction function W offers alternative computational prospects for the calculation of the electron self-energy in crystals.

I. INTRODUCTION

This is the first of two papers dealing with the validity and usefulness of the plasmon picture in describing the dielectric properties of crystalline materials. The present paper provides the theoretical background of such a discussion by drawing analogies between the equations governing collective charge-density oscillations and those describing quasiparticle excitations. In the second paper,[1] hereafter referred to as paper II, we shall discuss in more detail the analytic structure of the dielectric function and its inverse and propose a practical scheme allowing a highly accurate description of the latter in terms of plasmonlike excitations and its consequences for the definition of plasmon energies and lifetimes.

According to common usage, elementary excitations of a many-electron system may be classified into two categories: particlelike and collective excitations.[2–4] The related quantum-mechanical quanta of the former excitations are generally referred to as quasiparticles, which may be quasielectrons, quasiholes, polarons, etc., while those of the latter are designated plasmons, phonons, magnons, etc., depending on the phenomenon they account for. For instance, a quasielectron is an electronlike entity which consists of an electron surrounded by a polarization cloud. A plasmon is associated with an intrinsic charge oscillation in the system.

Not only quasiparticle band structures in crystals, but also the knowledge of plasmon band structures, offers valuable information about the physical properties of the system. Plasmon energies can be easily measured by, e.g., fast-electron experiments, as it is known that fast electrons are slowed down by plasmon excitations.[5,6]

The theory of plasmon excitations and their energy dispersion has mainly been studied for the uniform electron gas. The results can be applied to simple metals (see, however, Ref. 7). Investigations concerning the effect of ion potentials on the plasmon band structures are also reported.[8,9] In this paper, we will concentrate on the latter aspect.

We consider a crystal with immobile ions in which, moreover, the electrons interact via the spin-independent Coulomb interaction function only. In such a system, we expect to have quasielectrons, quasiholes, and plasmons. Plasmon excitations are directly related to the long range of the Coulomb interaction, for by the latter property a local disturbance of the electron charge density may give rise to an organized self-sustained oscillation of a macroscopic number of electrons in the system. As the long range of the bare Coulomb interaction is responsible for the well-known $1/|\mathbf{q}|^2$ singularity of the interaction, we expect plasmons to be of particular significance in the limit of small $|\mathbf{q}|$, i.e., in the limit of long wavelengths. Moreover, due to the possibility of exciting electron-hole pairs, plasmons cannot be long lived in the range of very high energies. On the other hand, in the case of a uniform electron gas, self-sustained oscillations cannot be excited at too low energies either, for due to the strong screening effects at low energies, the Coulomb interaction is effectively of short range. In crystals such as semiconductors, the long range of the interaction will only be partially screened, so that the minimum energy of plasmons may be lower (in paper II we shall expound on the actual meaning of the plasmon energy in the context of inhomogeneous systems).

As much of our discussions in this and the following paper revolve around the analogy between collective excitations and quasiparticles, a short review of the properties of quasiparticles seems in place.

Quasiparticles are considered as well-defined entities as long as their lifetimes are long enough to be observed in an experimental situation. This is in fact the case for low-lying excitations of the electronic system. For a uniform gas the lifetimes of quasiparticles decrease inversely proportional to the square of the difference between the quasiparticle wave vector and the Fermi wave vector.[10] As a consequence, the fast quasiparticles are very short lived. In semiconductors, it can be made plausible that the quasiparticle lifetimes are extremely large for energies within the energy range of $\frac{3}{2}$ times the gap energy around the midgap energy value.[11] As the quasiparticles in the higher bands are loosely bound and therefore behave as particles in a uniform system, their lifetimes are expected to be short.

Quasiparticle energies show up as peaks in the imaginary part of the one-particle Green function G. A convenient representation of this one-particle Green function, the so-called biorthonormal representation, is in terms of the eigenvalues and eigenfunctions of a Schrödinger-like wave equation in which an energy-dependent nonlocal operator, the self-energy operator, plays the role of a potential.[12,13] The energy dependence and non-Hermiticity of the latter potential give rise to a broadening of the quasiparticle peaks in the imaginary part of the Green function, which is generally associated with a damping of the quasiparticle excitations.

Similarly, plasmons show up as peaks in the imaginary part of the screened interaction function W, which is closely related to the inverse of the dielectric function. In a spatially uniform system, plasmons are known to coincide with the (high-energy) zeros of the dielectric function of the system;[14] these zeros occur at real energies, giving rise to δ-function peaks in the imaginary part of W. In a crystalline material, the existence of self-sustained oscillations in the absence of an external field leads to the requirement that the determinant of the dielectric matrix in the plane-wave representation should vanish at plasmon energies.[8] However, as will be explained in more detail in Sec. II, this requirement is fulfilled nowhere on the physical Riemann sheet,[15] and one has to look for possible zeros of the analytical continuation of the dielectric function ϵ through the branch cut. Again, the energy dependence and non-Hermiticity of the dielectric function give rise to a broadening of the plasmon peaks. In Sec. II we derive a biorthonormal representation for W in complete formal analogy to the above-mentioned representation for the Green function. By considering our biorthonormal-type representation for the *static* screened interaction function, we recover the concept of dielectric band structure introduced earlier by Baldereschi and Tosatti.[16,17] Making approximations similar to those used in deriving the quasiparticle representation of the one-electron Green function from its exact biorthonormal representation, we will then assume that it is possible to describe the energy dependence of W in terms of simple poles, an assumption which will be justified numerically in paper II. The connection of these poles with charge-density oscillations will be explained.

Section III is devoted to the discussion of an application of this theory in the calculation of the electron self-energy operator in the GW scheme.[18] The thus-obtained result will be used in paper II to calculate a highly accurate self-energy operator for a model semiconductor. We end with conclusions.

II. THE THEORY

Most of the following is based on the observation that there is an analogy between the equations governing the behavior of the one-particle Green function on the one hand, and the dynamically screened interaction function on the other. To be specific, there is a Dyson-type integral equation for the screened interaction function W similar to the one for the one-particle Green-function G in which the role of the unperturbed Green function G^0 is played by the bare Coulomb interaction v and the role of the self-energy operator (function) Σ by the polarization function P. Put otherwise, in the case of the one-particle Green function we have, symbolically, $G=G^0+G^0\Sigma G$, whereas in case of the screened interaction function we have $W=v+vPW$. Interestingly, like the unperturbed Green function G^0, which satisfies a differential equation with the Dirac δ function as the source term, the bare Coulomb interaction satisfies an equation, Poisson's equation, with almost the same structure. This analogy suggests that, just as for the one-particle Green function G which can be given in terms of a biorthonormal representation,[12,13] there also exists a biorthonormal-type representation for the dynamically screened interaction function W. More importantly, and this is what we really are aiming at, this similarity suggests that if some particular approximation would work for the biorthonormal representation of G, the same might apply to W. As known, the usual quasiparticle approximation for G is considered to be a sufficiently accurate one, at least as far as the behavior of G at low excitation energies is concerned. The question arises whether a similar type of approximation does exist for W, which would then offer substantial computational advantages for applications in real materials.

We start by giving first the basic relations concerning the bare and screened Coulomb interaction functions $v(1,2)$ and $W(1,2)$, where $j=1,2$ stands for a space-time point $(\mathbf{r}_j,t_j)$. Poisson's equation reads

$$-\frac{\epsilon_0}{e^2}\nabla_1^2 v(1,2)=\delta(1,2)\ , \tag{1}$$

while the relation between W and v can be expressed as

$$W(1,2)=v(1,2)+\int d(3)d(4)v(1,3)P(3,4)W(4,2)\ . \tag{2}$$

This relation can alternatively be expressed in terms of the inverse dielectric function ϵ^{-1}:

$$W(1,2)=\int d(3)\epsilon^{-1}(1,3)v(3,2)\ . \tag{3}$$

The solution of (1) can be written

$$v(1,2)=\frac{e^2}{4\pi\epsilon_0}\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}\delta(t_1-t_2)\ , \tag{4}$$

while the function P is related to the dielectric function ϵ by means of

$$\epsilon(1,2)=\delta(1,2)-\int d(3)v(1,3)P(3,2)\ . \tag{5}$$

The functions ϵ and ϵ^{-1} are related via

$$\int d(3)\epsilon(1,3)\epsilon^{-1}(3,2)=\int d(3)\epsilon^{-1}(1,3)\epsilon(3,2)=\delta(1,2)\ . \tag{6}$$

The similarity of Eq. (2) with the Dyson equation relating perturbed and unperturbed Green functions and of Eq. (1) with the Schrödinger-type equation defining G^0 is apparent [see Eqs. (10) and (9) of Ref. 13].

Let us now concentrate on the first relation given in Eq. (6). In view of Eqs. (3) and (5) it will be clear that Eq. (6) is nothing but the integral equation which the function W fulfills. Starting from Eq. (6) is therefore equivalent to starting from Eq. (2) in which the formal solution of the integral equation for W has been written down. By Fourier transforming Eq. (6) with respect to time according to Eq. (15) of Ref. 13 we obtain [cf. Eq. (16) of Ref. 13]:

$$\int d^3r_3\,\epsilon(\mathbf{r}_1,\mathbf{r}_3;\varepsilon)\epsilon^{-1}(\mathbf{r}_3,\mathbf{r}_2;\varepsilon)=\delta(\mathbf{r}_1-\mathbf{r}_2)\ . \tag{7}$$

It is not difficult to verify that the function $\epsilon^{-1}(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)$, completely similar to the Green function $G(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)$, may be expressed in the form of a biorthonormal representation:

$$\epsilon^{-1}(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)=\sum_n \frac{\xi_n(\mathbf{r}_1;\varepsilon)\zeta_n^*(\mathbf{r}_2;\varepsilon)}{D_n(\varepsilon)}\ , \tag{8}$$

in which the functions ξ_n and ζ_n satisfy the equation [cf. Eqs. (18) and (19) of Ref. 13]:

$$[D_n(\varepsilon)-\mathcal{H}(\varepsilon)]\xi_n(\mathbf{r};\varepsilon)=0\ , \tag{9a}$$

$$[D_n^*(\varepsilon)-\mathcal{H}^\dagger(\varepsilon)]\zeta_n(\mathbf{r};\varepsilon)=0\ , \tag{9b}$$

in which the operator $\mathcal{H}(\varepsilon)$ has been defined by

$$\mathcal{H}(\varepsilon)f(\mathbf{r})=\int d^3r'\epsilon(\mathbf{r},\mathbf{r}';\varepsilon)f(\mathbf{r}')\ , \tag{10}$$

where the function $f(\mathbf{r})$ is an arbitrary function. The adjoint operator $\mathcal{H}^\dagger(\varepsilon)$ has been defined through the relation $\langle \mathcal{H}(\varepsilon)f,g\rangle=\langle f,\mathcal{H}^\dagger(\varepsilon)g\rangle$, where $\langle\ ,\ \rangle$ stands for the inner product, defined according to the expression $\langle f,g\rangle=\int d^3r\,f^*(\mathbf{r})g(\mathbf{r})$. As shown in Ref. 13, the functions ξ_n and ζ_n can be constructed to be biorthonormal, in the sense that

$$\langle \xi_m,\zeta_n\rangle=\delta_{m,n}\ . \tag{11}$$

Moreover, these functions are supposed to satisfy the closure relation [cf. Eq. (27) of Ref. 13]:

$$\sum_n \xi_n(\mathbf{r}_1;\varepsilon)\zeta_n^*(\mathbf{r}_2;\varepsilon)=\delta(\mathbf{r}_1-\mathbf{r}_2)\ . \tag{12}$$

In view of the fact that $\epsilon(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)$ has the dimension m^{-3}, we observe from Eq. (10) that $\mathcal{H}(\varepsilon)$ and therefore also $D_n(\varepsilon)$ are dimensionless. The dimension of the product $\xi_n(\mathbf{r}_1;\varepsilon)\zeta_n^*(\mathbf{r}_2;\varepsilon)$ in Eq. (8) is m^{-3}, which allows us to postpone the choice of dimension for the functions ξ_n and ζ_n themselves, as long as their product has dimension m^{-3}. It follows easily from Eq. (10) that

$$\mathcal{H}^\dagger(\varepsilon)f(\mathbf{r})=\int d^3r'\epsilon^*(\mathbf{r}',\mathbf{r};\varepsilon)f(\mathbf{r}')\ , \tag{13}$$

again for arbitrary functions $f(\mathbf{r})$.

Substituting Eq. (8) in the right-hand side of Eq. (3), we obtain

$$W(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)=\sum_n \frac{\xi_n(\mathbf{r}_1;\varepsilon)\zeta_n'^*(\mathbf{r}_2;\varepsilon)}{D_n(\varepsilon)}\ , \tag{14}$$

in which

$$\zeta_n'(\mathbf{r};\varepsilon)=\int d^3r'v(\mathbf{r}-\mathbf{r}')\zeta_n(\mathbf{r}';\varepsilon)\ . \tag{15}$$

From Eqs. (12) and (15) it follows that

$$\sum_n \xi_n(\mathbf{r}_1;\varepsilon)\zeta_n'^*(\mathbf{r}_2;\varepsilon)=v(\mathbf{r}_1-\mathbf{r}_2)\ . \tag{16}$$

By using Eqs. (5), (9b), (13), and (15) one can readily show that $\zeta_n'(\mathbf{r};\varepsilon)$ satisfies the equation

$$\int d^3r'\left[\delta(\mathbf{r}'-\mathbf{r})-\int d^3r''P^*(\mathbf{r}',\mathbf{r}'';\varepsilon)v(\mathbf{r}''-\mathbf{r})\right]\zeta_n'(\mathbf{r}';\varepsilon)=D_n^*(\varepsilon)\zeta_n'(\mathbf{r};\varepsilon)\ . \tag{17}$$

In the system we are dealing with (i.e., no spins and magnetic fields), $P(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)$ is equal to $P(\mathbf{r}_2;\mathbf{r}_1;\varepsilon)$, so that we have in fact

$$[D_n^*(\varepsilon)-\mathcal{H}^*(\varepsilon)]\zeta_n'(\mathbf{r};\varepsilon)=0\ . \tag{18}$$

Therefore the functions ξ_n and $\zeta_n'^*$ actually satisfy the same equation.

In a crystal, where $\epsilon(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)=\epsilon(\mathbf{r}_1+\mathbf{R},\mathbf{r}_2+\mathbf{R};\varepsilon)$, in which $\mathbf{R}$ stands for a lattice vector, the functions $\xi_n(\mathbf{r};\varepsilon)$ and $\zeta_n'(\mathbf{r};\varepsilon)$ can be chosen to be Bloch functions and may be denoted as $\xi_{l,\mathbf{k}}(\mathbf{r};\varepsilon)$ and $\zeta_{l,\mathbf{k}}'(\mathbf{r},\varepsilon)$; l is a band index and $\mathbf{k}$ is a vector in the first Brillouin zone (1BZ). As the functions $\zeta_{l,\mathbf{k}}'^*(\mathbf{r};\varepsilon)$ have to be linear combinations of the $\xi_{l,\mathbf{k}}(\mathbf{r};\varepsilon)$ functions, and as both $\langle \xi_{l,\mathbf{k}'}^*,\xi_{l,\mathbf{k}}\rangle$ and $\langle \zeta_{l,\mathbf{k}'}',\xi_{l,\mathbf{k}}\rangle$ are equal to zero for any $\mathbf{k}'\neq-\mathbf{k}$ (due to the Bloch property), it follows directly from Eq. (11) that $\zeta_{l,\mathbf{k}}'^*(\mathbf{r};\varepsilon)$ has to be identified, apart from a possible numerical factor, with $\xi_{l,-\mathbf{k}}(\mathbf{r};\varepsilon)$ (compare the similar discussion in Ref. 13). By choosing the factor equal to unity, i.e., by writing $\zeta_{l,\mathbf{k}}'^*(\mathbf{r};\varepsilon)=\xi_{l,-\mathbf{k}}(\mathbf{r};\varepsilon)$, we simultaneously fix the dimension of the involved eigenfunctions ζ' and ξ to $(J\,m^{-3})^{1/2}$. Denoting $D_n(\varepsilon)$ as $D_{l,\mathbf{k}}(\varepsilon)$, we may now write Eq. (14) as

$$W(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)=\sum_{l,\mathbf{k}} \frac{\xi_{l,\mathbf{k}}(\mathbf{r}_1;\varepsilon)\xi_{l,-\mathbf{k}}(\mathbf{r}_2;\varepsilon)}{D_{l,\mathbf{k}}(\varepsilon)}\ . \tag{19}$$

As $\xi_{l,\mathbf{k}}$ and $\xi_{l,-\mathbf{k}}$ are both eigenfunctions to the same eigenvalue, we find that $D_{l,\mathbf{k}}(\varepsilon)$ and $D_{l,-\mathbf{k}}(\varepsilon)$ are in fact identical. The static values $D_{l,\mathbf{k}}(\varepsilon=0)$ correspond to the eigenvalues defining the concept of dielectric band structure introduced by Baldereschi and Tosatti.[16,17] Note that $\xi_{l,-\mathbf{k}}(\mathbf{r};\varepsilon)$ is only equal to $\xi_{l,\mathbf{k}}^*(\mathbf{r};\varepsilon)$ if $P(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)$ is real valued. By expanding the functions ξ in Eq. (19) in plane waves $\exp[i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}]$, where $\mathbf{G}$ denotes a reciprocal-lattice vector, according to the relation

$$\xi_{l,\mathbf{k}}(\mathbf{r};\varepsilon)=\frac{1}{\sqrt{\Omega}}\sum_{\mathbf{G}}\Xi_{l,\mathbf{k}}(\mathbf{G};\varepsilon)e^{i(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}}\ , \tag{20}$$

with Ω the volume of the crystal, and Fourier transforming $W(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)$ according to Eq. (37) of Ref. 13, we obtain the plane-wave matrix elements of $W(\mathbf{r}_1,\mathbf{r}_2;\varepsilon)$ as follows ($\mathbf{K}$ and $\mathbf{K}'$ are reciprocal-lattice vectors):

$$W_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)=\sum_{l}\frac{\Xi_{l,\mathbf{k}}(\mathbf{K};\varepsilon)\Xi_{l,-\mathbf{k}}(-\mathbf{K}';\varepsilon)}{D_{l,\mathbf{k}}(\varepsilon)}\ , \tag{21}$$

where the $\Xi_{l,\mathbf{k}}$ functions have the dimension $J^{1/2}$. From Eqs. (9), (10), and (20) it follows directly that the plane-wave coefficients $\Xi_{l,\mathbf{k}}(\mathbf{K};\varepsilon)$ satisfy the following system of linear equations:

$$\sum_{\mathbf{K}'}\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)\Xi_{l,\mathbf{k}}(\mathbf{K}';\varepsilon)=D_{l,\mathbf{k}}(\varepsilon)\Xi_{l,\mathbf{k}}(\mathbf{K},\varepsilon)\ . \tag{22}$$

If the system under consideration is symmetric with respect to the origin, it can be shown that $\Xi_{l,\mathbf{k}}(\mathbf{K};\varepsilon)$ and $\Xi_{l,-\mathbf{k}}(-\mathbf{K};\varepsilon)$ are in fact identical.

The closure relation in Eq. (16) transforms to

$$\sum_{l}\Xi_{l,\mathbf{k}}(\mathbf{K};\varepsilon)\Xi_{l,-\mathbf{k}}(-\mathbf{K}';\varepsilon)=v_{\mathbf{K},\mathbf{K}'}(\mathbf{k})\ , \tag{23}$$

whereas the normalization is fixed by the Fourier transform of Eq. (12) with ζ_n replaced by using Eq. (15);

$$\sum_{\mathbf{K}}\frac{1}{v_{\mathbf{K},\mathbf{K}}(\mathbf{k})}\Xi_{l,\mathbf{k}}(\mathbf{K};\varepsilon)\Xi_{l',-\mathbf{k}}(-\mathbf{K};\varepsilon)=\delta_{l,l'}\ , \tag{24}$$

where

$$v_{\mathbf{K},\mathbf{K}'}(\mathbf{k})=\frac{e^2}{\epsilon_0}\frac{1}{|\mathbf{k}+\mathbf{K}|^2}\delta_{\mathbf{K},\mathbf{K}'} \tag{25}$$

is the Fourier transform of the bare Coulomb interaction function.

Let us now consider $W_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)$ for large values of $|\varepsilon|$. The asymptotic behavior is known to be

$$W_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)\to v_{\mathbf{K},\mathbf{K}'}(\mathbf{k})+O(\varepsilon^{-2})\quad\text{as } |\varepsilon|\to\infty\ . \tag{26}$$

Similarly, the matrix elements of the dielectric matrix satisfy

$$\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)\to\delta_{\mathbf{K},\mathbf{K}'}+O(\varepsilon^{-2})\quad\text{as } |\varepsilon|\to\infty\ . \tag{27}$$

As a consequence of this, it follows from (22) that

$$D_{l,\mathbf{k}}(\varepsilon)\to 1\ ,\quad\text{as } |\varepsilon|\to\infty\ . \tag{28}$$

Therefore, to our aim it is rather advantageous to introduce a function $\mathcal{D}_{l,\mathbf{k}}(\varepsilon)$ via the relation

$$\frac{1}{D_{l,\mathbf{k}}(\varepsilon)}=1+\frac{1}{\mathcal{D}_{l,\mathbf{k}}(\varepsilon)}\ , \tag{29}$$

in which, owing to Eq. (28), $1/\mathcal{D}_{l,\mathbf{k}}(\varepsilon)\to 0$, as $|\varepsilon|\to\infty$. Due to the fact that the plane-wave coefficients $\Xi_{l,\mathbf{k}}(\mathbf{K};\varepsilon)$ approach constant values for large $|\varepsilon|$, and owing to Eq. (26), we must have $1/\mathcal{D}_{l,\mathbf{k}}(\varepsilon)=O(\varepsilon^{-2})$ as $|\varepsilon|\to\infty$. Introducing Eq. (29) in Eq. (21), while making use of Eq. (23), we obtain

$$W_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)=v_{\mathbf{K},\mathbf{K}'}(\mathbf{k})+\widetilde{W}_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)\ . \tag{30}$$

The function

$$\widetilde{W}_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)\equiv\sum_{l}\frac{\Xi_{l,\mathbf{k}}(\mathbf{K};\varepsilon)\Xi_{l,-\mathbf{k}}(-\mathbf{K}';\varepsilon)}{\mathcal{D}_{l,\mathbf{k}}(\varepsilon)} \tag{31}$$

is in fact the screening part of the electron-electron interaction.

As mentioned above, the biorthonormal representation in Eqs. (19) and (30) is formally equivalent to the representation of the one-particle Green function in terms of eigenfunctions and eigenvalues of an energy-dependent Hamiltonian.[12,13] Therefore it seems natural to make an approximation for W similar to the quasiparticle approximation of the Green function. In such an approximation, the eigenvalues $\mathcal{D}_{l,\mathbf{k}}(\varepsilon)$ are assumed to have M_l *simple* zeros $e_l^m(\mathbf{k})$ for which it holds that

$$\mathcal{D}_{l,\mathbf{k}}(e_l^m(\mathbf{k}))=0\Longleftrightarrow D_{l,\mathbf{k}}(e_l^m(\mathbf{k}))=0\ . \tag{32}$$

Equation (31) is then written as a sum over residues:

$$\widetilde{W}_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)\simeq\sum_{l}\sum_{m=1}^{M_l}f_{l,\mathbf{k}}^{m}\frac{\Xi_{l,\mathbf{k}}^{m}(\mathbf{K})\Xi_{l,-\mathbf{k}}^{m}(-\mathbf{K}')}{\varepsilon-e_l^m(\mathbf{k})}\ , \tag{33}$$

in which the function $\Xi_{l,\mathbf{k}}^{m}(\mathbf{K})$ stands for $\Xi_{l,\mathbf{k}}(\mathbf{K};e_l^m(\mathbf{k}))$ and in which

$$f_{l,\mathbf{k}}^{m}\equiv\left[\frac{\partial\mathcal{D}_{l,\mathbf{k}}(\varepsilon)}{\partial\varepsilon}\right]^{-1}_{\varepsilon=e_l^m(\mathbf{k})}=\left[\frac{\partial D_{l,\mathbf{k}}(\varepsilon)}{\partial\varepsilon}\right]^{-1}_{\varepsilon=e_l^m(\mathbf{k})}\ . \tag{34}$$

Note that $e_l^m(\mathbf{k})=e_l^m(-\mathbf{k})$. Obviously, at $\varepsilon=e_l^m(\mathbf{k})$ the solution of Eq. (22) with vanishing right-hand side [cf. Eq. (32)] is nontrivial if and only if

$$\det[\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};e_l^m(\mathbf{k}))]=0\ , \tag{35}$$

which is just the dispersion relation for plasmon energies[8] [see also Eq. (41) and the text following it]. Therefore we have shown that the poles $e_l^m(\mathbf{k})$ of our biorthonormal type of representation can be identified with plasmon excitation energies.[19] Note that only those solutions of Eqs. (32) and (35) are physically acceptable for which it holds that either $\mathrm{Re}[e_l^m(\mathbf{k})]>0$ and $\mathrm{Im}[e_l^m(\mathbf{k})]<0$ *or* $\mathrm{Re}[e_l^m(\mathbf{k})]<0$ and $\mathrm{Im}[e_l^m(\mathbf{k})]>0$. We remark here that, for practical purposes, it is advantageous to look for the poles of $\mathrm{Tr}[\epsilon^{-1}_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)]$ rather than for the solutions of Eq. (35) as we have

$$\sum_{\mathbf{K}}\epsilon^{-1}_{\mathbf{K},\mathbf{K}}(\mathbf{k};\varepsilon)=\sum_{l}\frac{1}{D_{l,\mathbf{k}}(\varepsilon)}\ . \tag{36}$$

In this connection, we have to realize that the function $\det[\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)]$, for semiconductors, is real valued for real energy values ε within a finite interval around $\varepsilon=0$. Furthermore, the function is analytic everywhere in the complex ε plane, except for branch cuts on the real axis. Consequently, the reflection principle of Schwarz[20] implies that the function takes complex conjugate values at complex conjugate energies. Therefore the zeros of the above function, if any, should be symmetrically located with respect to the real axis. This would imply poles of W in all four quadrants of the complex energy plane, which contradicts the above assertion about the location of poles of W, unless they lie precisely on the real axis.

The latter possibility, however, is in conflict with the observation that, for three-dimensional systems, the function W has to be continuous[21] and therefore bounded. The solution to this seemingly contradictory situation is that we have to search for the possible zeros of the *analytic continuation*[22] of $\det[\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)]$ across the branch cut on the real axis, i.e., on the nonphysical Riemann sheets.[15]

In paper II we shall derive a formalism which circumvents the practical difficulties in solving Eqs. (32), (34), and (35) directly, which arise from the necessity to analytically continue $\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)$ into the nonphysical Riemann sheet. This is achieved by making an inspired guess for the pole positions e_l^m and using the weights $f_{l,\mathbf{k}}^m$ as fitting parameters. If one is merely interested in a good numerical approximation for W, it is even possible to choose pole positions independent of the band index l, i.e.,

$$\widetilde{W}_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)\simeq\sum_m \left[\sum_l f_{l,\mathbf{k}}^m \Xi_{l,\mathbf{k}}^m(\mathbf{K})\Xi_{l,-\mathbf{k}}^m(-\mathbf{K}')\right] \times\left[\frac{1}{\varepsilon-e_m(\mathbf{k})}-\frac{1}{\varepsilon+e_m(\mathbf{k})}\right] , \quad (37)$$

where, concerning the minus sign between the two terms inside the last set of large parentheses, use has been made of the fact that the dielectric matrix $\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)$ is an *even* function of ε.

If, however, we are interested in explaining the main features of the plasmon spectrum in terms of as few collective excitations as possible, direct solution of Eqs. (32), (34), and (35) yields a physically more appealing picture, in particular in view of calculating plasmon band structures. As this requires knowledge of the analytic continuation of W across the branch cut along the real energy axis, the answers one obtains for the excitation energies and lifetimes become the more ambiguous the further away from the real axis the corresponding pole lies, and it becomes more and more difficult to distinguish between individual excitations. It should be noted that whatever choice of parameters in Eq. (33) has been made, Eqs. (32), (34), and (35) will always be satisfied exactly if we replace the exact functions $\epsilon_{\mathbf{K},\mathbf{K}'}$ and $\mathcal{D}_{l,\mathbf{k}}$ by those which would be obtained from the approximation to $\widetilde{W}$ in Eq. (33). The technique of analytic continuation has been used successfully to determine plasmon bands along the Δ and Λ directions in silicon.[23]

In order to find the connection between the expanding functions $\xi_{l,\mathbf{k}}(\mathbf{r};\varepsilon)$ and the charge-density functions taking part in plasmon oscillations, we remind the reader that the *total* potential $\delta\chi(\mathbf{r};\varepsilon)$ due to an *external* potential $\delta\phi(\mathbf{r};\varepsilon)$ is equal to[24]

$$\delta\chi(\mathbf{r};\varepsilon)=\delta\phi(\mathbf{r};\varepsilon)+\int d^3r' v(\mathbf{r}-\mathbf{r}')\delta\rho(\mathbf{r}';\varepsilon) , \quad (38)$$

in which $\delta\rho(\mathbf{r}';\varepsilon)$ stands for the charge-density deviation from the charge-density distribution in the ground state. Moreover, $\delta\phi(\mathbf{r};\varepsilon)$ is related to $\delta\chi(\mathbf{r};\varepsilon)$ through the relation[25]

$$\delta\phi(\mathbf{r};\varepsilon)=\int d^3r' \epsilon(\mathbf{r},\mathbf{r}';\varepsilon)\delta\chi(\mathbf{r}';\varepsilon) . \quad (39)$$

In the absence of the external potential, making use of Eq. (38), this expression reduces to

$$\int d^3r_1 d^3r_2 \epsilon(\mathbf{r},\mathbf{r}_1;\varepsilon)v(\mathbf{r}_1-\mathbf{r}_2)\delta\rho(\mathbf{r}_2;\varepsilon)=0 , \quad (40)$$

or, in the plane-wave representation,

$$\sum_{\mathbf{K}'}\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)v_{\mathbf{K}',\mathbf{K}'}(\mathbf{k})\delta\rho(\mathbf{k}+\mathbf{K}';\varepsilon)=0 . \quad (41)$$

It is just Eq. (41) from which the dispersion relation for plasmon band structures follows; due to the fact that $\det[v_{\mathbf{K},\mathbf{K}'}(\mathbf{k})]\neq 0$, Eq. (41) can have nontrivial solutions if and only if $\det[\epsilon_{\mathbf{K},\mathbf{K}'}(\mathbf{k};\varepsilon)]=0$. It immediately follows that the eigenfunctions $\Xi_{l,\mathbf{k}}^m$ can be identified, apart from a multiplicative constant α, with the amplitudes of charge-density fluctuations:

$$\delta\rho(\mathbf{k}+\mathbf{K};e_l^m(\mathbf{k}))=\alpha\frac{\epsilon_0}{e^2}|\mathbf{k}+\mathbf{K}|^2\Xi_{l,\mathbf{k}}^m(\mathbf{K}) . \quad (42)$$

III. PROSPECTS FOR CALCULATIONS OF THE ELECTRON SELF-ENERGY IN THE *GW* SCHEME

Most of the calculations of the electron self-energy operator Σ in crystalline materials thus far have employed the *GW* approximation, in which only contributions to lowest order in the screening potential W are accounted for.[26–28] If we replace the Green function G by its unperturbed version G^0, the plane-wave matrix elements of Σ can be written as[11]

$$\Sigma_{\mathbf{G},\mathbf{G}'}(\mathbf{k};\varepsilon)=\frac{i}{2\pi\hbar^2\Omega}\sum_{\mathbf{k}'}\sum_{\mathbf{K},\mathbf{K}'}F_{\mathbf{K},\mathbf{K}'}(\varepsilon;\mathbf{G},\mathbf{G}',\mathbf{k},\mathbf{k}') , \quad (43)$$

where

$$F_{\mathbf{K},\mathbf{K}'}(\varepsilon;\mathbf{G},\mathbf{G}',\mathbf{k},\mathbf{k}')=\hbar\sum_n d_{n,\mathbf{k}-\mathbf{k}'}(\mathbf{K})d_{n,\mathbf{k}-\mathbf{k}'}^*(\mathbf{K}')\int_{-\infty}^{\infty}d\varepsilon'\frac{W_{\mathbf{G}-\mathbf{K},\mathbf{G}'-\mathbf{K}'}(\mathbf{k}';\varepsilon')e^{-i\varepsilon'\eta_0/\hbar}}{\varepsilon-\varepsilon'-\varepsilon_n(\mathbf{k}-\mathbf{k}')-i\eta_1\mathrm{sgn}[\mu-\varepsilon_n(\mathbf{k}-\mathbf{k}')]} . \quad (44)$$

In Eq. (44) the $d_{n,\mathbf{k}}(\mathbf{K})$ coefficients are plane-wave components of the Bloch eigenfunctions of the unperturbed system with band index n and wave vector $\mathbf{k}$; $\varepsilon_n(\mathbf{k})$ is the related (real) energy; μ separates occupied from unoccupied bands; and η_0,η_1 are infinitesimally small positive numbers. Making use of Eqs. (30) and (31), we obtain from Eq. (44)

$$F_{\mathbf{K},\mathbf{K}'}(\varepsilon;\mathbf{G},\mathbf{G}',\mathbf{k},\mathbf{k}')=2\pi i\hbar\sum_{n}d_{n,\mathbf{k}-\mathbf{k}'}(\mathbf{K})d^{*}_{n,\mathbf{k}-\mathbf{k}'}(\mathbf{K}')\times[\Theta(\mu-\varepsilon_{n}(\mathbf{k}-\mathbf{k}'))v_{\mathbf{G}-\mathbf{K},\mathbf{G}'-\mathbf{K}'}(\mathbf{k}')+H_{\mathbf{G}-\mathbf{K},\mathbf{G}'-\mathbf{K}'}(\mathbf{k}';\varepsilon,\varepsilon_{n}(\mathbf{k}-\mathbf{k}'),\mu)]\ , \tag{45}$$

in which

$$H_{\mathbf{G}-\mathbf{K},\mathbf{G}'-\mathbf{K}'}(\mathbf{k}';\varepsilon,\varepsilon_{n}(\mathbf{k}-\mathbf{k}'),\mu)\equiv\frac{1}{2\pi i}\int_{-\infty}^{\infty}d\varepsilon'\frac{\widetilde{W}_{\mathbf{G}-\mathbf{K},\mathbf{G}'-\mathbf{K}'}(\mathbf{k}';\varepsilon')}{\varepsilon-\varepsilon'-\varepsilon_{n}(\mathbf{k}-\mathbf{k}')-i\eta_{1}\mathrm{sgn}[\mu-\varepsilon_{n}(\mathbf{k}-\mathbf{k}')]}\ . \tag{46}$$

In arriving at Eq. (45), use has been made of the residue theorem to obtain the term containing the Θ function. By substituting Eq. (37) in Eq. (46) and again using the residue theorem, we obtain

$$H_{\mathbf{G}-\mathbf{K},\mathbf{G}'-\mathbf{K}'}(\mathbf{k}';\varepsilon,\varepsilon_{n}(\mathbf{k}-\mathbf{k}'),\mu)=-\sum_{l,m}f^{m}_{l,\mathbf{k}'}\frac{\Xi^{m}_{l,\mathbf{k}'}(\mathbf{G}-\mathbf{K})\Xi^{m}_{l,-\mathbf{k}'}(-\mathbf{G}'+\mathbf{K}')}{\varepsilon-\varepsilon_{n}(\mathbf{k}-\mathbf{k}')+e_{m}(\mathbf{k}')\mathrm{sgn}[\mu-\varepsilon_{n}(\mathbf{k}-\mathbf{k}')]}\ . \tag{47}$$

The practical advantage of Eq. (45) along with Eq. (47) above Eq. (44) is obvious: there is no energy integration left to be carried out numerically. The price to be paid is an additional summation over the plasmon band index l and over the pole positions m. In paper II, an efficient method for obtaining the coefficients $f^{m}_{l,\mathbf{k}'}$ will be proposed, which makes evaluation of Σ^{GW} using Eqs. (43), (45), and (47) comparable in terms of numerical cost with schemes employing crude approximations for W, like the plasmon-pole model,[26] in which each matrix element of W is represented in terms of two simple poles.

IV. CONCLUSIONS

We have proposed a general representation for the dynamically screened interaction function W of a many-electron system. To this end, we have made use of the fact that both the one-particle Green function G and the screened interaction function W obey Dyson-type integral equations whose formal solutions can be expressed in terms of eigenfunctions and eigenvalues of the related operators, the Hamiltonian, and the dielectric function, respectively. It should be mentioned that the latter two operators are energy dependent and non-Hermitian in most of the energy range of interest. Analogously to the way quasiparticle energies are found to correspond to the poles of the Green function, plasmons have been shown to be related to the poles of W. Moreover, the corresponding eigenvectors of the dielectric function are shown to be related to the amplitudes of the charge-density oscillations. We have discussed the merits of using our representation in self-energy calculations within the GW approximation scheme. In paper II we further develop the theory presented here and give the numerical results of our calculations on a model semiconductor.

ACKNOWLEDGMENTS

B.F. and W.v.H. acknowledge useful discussions with D.J. Wielaard. One of us (G.E.E.) would like to thank Gateway Corporation and Churchill College, Cambridge, for financial support. This work was supported by the Science and Engineering Research Council (United Kingdom).

---

[1] G. E. Engel, B. Farid, C. M. M. Nex, and N. H. March, following paper, Phys. Rev. B **44**, 13 349 (1991).

[2] D. Pines and P. Nozières, *The Theory of Quantum Liquids*, Vol. I of Normal Fermi Liquids (Benjamin, New York, 1966), pp. 111–118.

[3] R. Mattuck, *A Guide to Feynman Diagrams in the Many-Body Problem* (McGraw-Hill, New York, 1967), pp. 12–15.

[4] N. H. March and M. Parrinello, *Collective Effects in Solids and Liquids* (Hilger, Bristol, 1982).

[5] L. Marton, Rev. Mod. Phys. **28**, 172 (1956).

[6] K. Sturm, Adv. Phys. **31**, 1 (1982).

[7] E.-N. Foo and J. J. Hopfield, Phys. Rev. **173**, 635 (1968).

[8] W. M. Saslow and G. F. Reiter, Phys. Rev. B **7**, 2995 (1973).

[9] N. H. March and M. P. Tosi, Proc. R. Soc. London, Ser. A **330**, 373 (1972); Philos. Mag. **28**, 91 (1973).

[10] A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Dover, New York, 1963), pp. 15–18.

[11] B. Farid, R. Daling, D. Lenstra, and W. van Haeringen, Phys. Rev. B **38**, 7530 (1988).

[12] A. J. Layzer, Phys. Rev. **129**, 897 (1963).

[13] W. van Haeringen, B. Farid, and D. Lenstra, Phys. Scr. **T19**, 282 (1987).

[14] D. Pines and P. Nozières, *The Theory of Quantum Liquids* (Ref. 2), pp. 210–215.

[15] An n-valued function of a complex variable can be considered to consist of n single-valued functions, each called a *branch* of the original function. Alternatively, and equivalently, one can extend the domain of the above n-valued function into n so-called Riemann sheets, thereby making the many-valued function single valued over the larger domain of n Riemann sheets. The dielectric function we are considering in this paper, for instance, is a many-valued function of energy. By simply *replacing* the real energy variable in the usual defining expression for this function by a complex energy, one obtains what we in this paper refer to as the dielectric function on the physical Riemann sheet; other branches of this function correspond to other, nonphysical, Riemann sheets.

[16] A. Baldereschi and E. Tosatti, Solid State Commun. **29**, 131 (1979).

[17] R. Car, E. Tosatti, S. Baroni, and S. Leelaprute, Phys. Rev. B **24**, 985 (1981).

[18] L. Hedin, Phys. Rev. **139**, A796 (1965).

[19] B.F. is indebted to D. J. Wielaard for pointing out this result.

[20] E. C. Titchmarch, *The Theory of Functions* (Oxford University Press, London, 1985), pp. 155–157.

[21] L. van Hove, Phys. Rev. **89**, 1189 (1953).

[22] K. Knopp, *The Theory of Functions* (Dover, New York, 1945), Part I, pp. 92–111; *ibid.* (Dover, New York, 1947), Part II, pp. 93–118.

[23] R. Daling, W. van Haeringen, and B. Farid, Phys. Rev. B **44**, 2952 (1991).

[24] L. Hedin and S. Lundqvist, in *Solid State Physics: Advances in Research and Applications,* edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1969), Vol. 23, p. 46.

[25] L. Hedin and S. Lundqvist, in *Solid State Physics: Advances in Research and Applications* (Ref. 24), p. 14.

[26] M. S. Hybertsen and S. G. Louie, Phys. Rev. Lett. **55**, 1418 (1985); Phys. Rev. B **34**, 5390 (1986).

[27] R. W. Godby, M. Schlüter, and L. J. Sham, Phys. Rev. Lett. **56**, 2415 (1986); Phys. Rev. B **35**, 4170 (1987); **37**, 10 159 (1988).

[28] W. von der Linden and P. Horsch, Phys. Rev. B **37**, 8351 (1988).