Dielectric properties of multiband electron systems:

I - Tight-binding formulation

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Abstract

The screened electron-electron interaction in a multi-band electron system is calculated within the random phase approximation and in the tight-binding representation. The obtained dielectric matrix contains, beside the usual site-site correlations, also the site-bond and bond-bond correlations, and thus includes all physically relevant polarization processes. The arguments are given that the bond contributions are negligible in the long wavelength limit. We analyse the system with two non-overlapping bands in this limit, and show that the corresponding dielectric matrix reduces to a $2 \times 2$ form. The intra-band and inter-band contributions are represented by diagonal matrix elements, while the off-diagonal elements contain the mixing between them. The latter is absent in insulators but may be finite in conductors. Performing the multipole expansion of the
bare long-range interaction, we show that this mixing is directly related to the symmetry of the atomic orbitals participating in the tight-binding electronic states. In systems with forbidden atomic dipolar transitions, the intra-band and inter-band polarizations are separated. However, when the dipolar transitions are allowed, the off-diagonal elements of the dielectric matrix are of the same order as diagonal ones, due to a finite monopole-dipole interaction between the intra-band and inter-band charge fluctuations. We also calculate the macroscopic dielectric function and obtain an expression which interpolates between the well-known limits of one-band conductors and pure insulators. In particular, it is shown that the microscopic origin of the so-called self-polarization corrections is the on-site interaction which exchanges two electrons at different orbitals, combined with a finite tunneling between neighboring sites.

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1 Introduction

The present and the forthcoming work revisit the problem of the dielectric properties of multiband metals and insulators. Early calculations were carried out neglecting the discreteness of the lattice in the derivation of the screened Coulomb interaction. The result was a dielectric function built adding the intraband and interband polarizabilities. The continuous approximation in calculating the Coulomb matrix elements is however inconsistent with the multiband assumption, since the latter implies the existence of the lattice. Later works attempted to include properly the local electric field effects associated to the lattice discreteness, by formulating the problem of dielectric response in the representation of the \( k+G \) plane waves, where \( G \) are vectors of the reciprocal lattice. In this representation one comes to the dielectric matrix \( \varepsilon(q+G, q+G', \omega) \) which is of infinite order for a given \( q \). Since this latter is very difficult to handle in physical terms, it was necessary either to make additional approximations (beside the initial random phase approximation (RPA)), or to develop various numerical procedures, which in turn usually do not provide a simple physical interpretation of the results obtained.

A particular method in this direction is based on the use of the tight-binding (TB) basis in the calculation of the polarizabilities which figure in the matrix elements of \( \varepsilon(q+G, q+G', \omega) \). Providing that the atomic orbitals which represent this basis are localized enough, one gets to a good approximation a factorizable form of dielectric matrix elements, introduced earlier on through the so-called generalized shell model by Sinha et al. This factorization in principle enabled the analytical inversion...
of the dielectric matrix, and led to significant improvements in the understanding of
dielectric properties of simple TB band structures [9, 11]. The whole approach was still
burdened by the unnecessary parallel use of two basis, i.e. plane waves and TB states.

The aim of the present paper is to show that the problem of the dielectric response
is significantly simplified when the TB scheme is used from the outset, in the definition
of the dielectric matrix itself. The TB approach for the single band case was developed
before [16]. In the multiband case the elements of the TB dielectric matrix have a
transparent physical content, i.e. they are defined by the pairs of intraband and/or
interband transitions and by an index which covers pure site-site correlations as well
as the site-bond and bond-bond correlations between neighboring lattice points. By
taking into account these bond correlations we complete our earlier treatment of the
dielectric response in the TB approach [17]. Since the atomic orbitals which build the
TB Bloch functions are well localized (in contrast to usual Wannier functions), it is well
justified to keep only the bond contributions between first neighbors, so that the index
mentioned above is of the order of a coordination number for a given lattice. After
an additional truncation by keeping a finite number of presumably relevant electronic
bands, the dielectric matrix reduces to a finite, rather low order.

In order to illustrate the method, we consider here in some detail the simple case
of two non-overlapping electron bands, allowing for two types of symmetry of the TB
orbitals which form the empty higher (conducting) band. In all cases the lower band is
assumed to be partially or fully filled. We calculate all screened TB Coulomb matrix
elements within RPA, and show that in the long-wavelength limit the problem reduces to
a $2 \times 2$ dielectric matrix. The same result was obtained by an alternative analysis via the
Heisenberg equations for the electronic charge density \[17\]. Since the present approach gives in addition screened Coulomb matrix elements for all intraband and interband processes, it can be conveniently used in the calculation of the screened local fields, such as those produced by phonons in ionic multiband metals, and in particular in high $T_c$ superconductors.

Since the range of the TB atomic (molecular) orbitals is shorter than that of the two-site Coulomb interactions \[18\], the latter can be expanded into the multipole series, which is particularly well controlled in the long wavelength limit. In this limit one recognizes the decisive role of the symmetry of TB orbitals in the multiband dielectric screening. When two orbitals have the same parity, the intra-band and inter-band polarization processes are decoupled, so that the total dielectric function is the product of the corresponding intra-band and inter-band dielectric functions \[19 - 21\]. A more interesting situation occurs when the band symmetry allows for finite dipolar transitions. The finiteness of the monopole-dipole coupling in this case leads to qualitatively different results for the microscopic dielectric function (defined as the determinant of the dielectric matrix) and the TB matrix elements of screened electron-electron interaction. The effects of this coupling on the collective modes of the two-band electron liquid will be analysed in the paper II \[1\].

The multipole expansion also separates the long-range part of the bare Coulomb interaction from the on-site contributions. Because of that, we are able to trace the microscopic origin of the so-called self-polarization corrections \[3\] to the macroscopic dielectric function $\epsilon_M(q, \omega)$, which can be easily determined once the TB matrix elements of the screened Coulomb interaction are known.
The paper is organized in the following way. In Sect. 2 we derive the linear system of RPA equations for the screened Coulomb interaction and introduce the corresponding dielectric matrix. In Sects. 3 to 5 we apply these general results to the two-band model. The explicit expressions for the microscopic dielectric function and all matrix elements of the screened interaction are derived in Sect. 3. In Sect. 4 we perform the multipole expansion and discuss the role of the short-range and long-range contributions to the microscopic dielectric function. The macroscopic dielectric function and the origin of the self-polarization corrections are considered in Sect. 5. Sect. 6 contains some concluding remarks.

2 The tight-binding formulation of the dielectric matrix

The Dyson’s equation for the screened interaction in the direct space is within RPA given by

\[ V(r, r', \omega) = V(r - r') + \int dr_1 \int dr'_1 V(r - r_1) \Pi(r_1, r_1', \omega) V(r_1', r', \omega), \]

(1)

where

\[ \Pi(r, r', \omega) = -\frac{i}{\pi} \int d\omega' G^0(r, r', \omega + \omega') G^0(r', r, \omega') \]

(2)

is the bubble polarization diagram, and \( G^0(r, r', \omega) = \sum_{l} G^0_l(r, r', \omega) \) is a bare Green’s function with the contribution from the \( l \)-th band given by

\[ G^0_l(r, r', \omega) = \sum_{k} \psi_{l, k}(r) \psi_{l, k}^*(r') \times \]

\[ \times \left\{ \frac{1 - n_l(k)}{\omega - E_l(k) + i\eta} + \frac{n_l(k)}{\omega - E_l(k) - i\eta} \right\}, \]

(3)
with $\psi_{l,k}(r)$, $E_l(k)$ and $n_l$ being the corresponding Bloch function, band dispersion and occupation number respectively. Our aim is to transform the integral equation (1) into a system of algebraic equations for the TB matrix elements of the screened potential.

To this end we recall the essential property of the TB basis, namely the weak overlap of the atomic orbitals at neighboring crystal sites. Thus, it suffices to keep in various matrix elements only the contributions with pairs of orbitals centered at the same and neighboring sites, and to neglect all contributions with more distant pairs of orbitals.

In this respect it is important to compare the TB and the Wannier bases. Although the Wannier functions at different sites are exactly orthogonal, they usually have slowly converging oscillatory tails, which greatly complicates the evaluation of the one- and two-body matrix elements. We therefore continue by strictly using the TB basis. The Bloch functions are thus given by

$$\psi_{l,k}(r) = \frac{1}{\sqrt{N}} \frac{1}{[1 + S_l(k)]^{1/2}} \sum_R e^{i k R} \varphi_l(r - R).$$

(4)

Here

$$S_l(k) = \sum_{\delta \neq 0} e^{i k \delta} S_l(\delta),$$

(5)

where

$$S_l(\delta) = \int dr \varphi_l^*(r) \varphi_l(r - \delta)$$

(6)

is the direct overlap between nearest neighboring atomic orbitals $\varphi_l$, and the sum in (5) involves only first neighbors. Although the further considerations mainly do not depend on the details of the band dispersions $E_l(k)$, we remind that they include, beside $S_l(k)$, also sums of tunneling integrals due to the effective ionic potential. In these sums it again suffices to keep only nearest neighbors.
Let us now multiply eq. (6) by $e^{i q (R + \frac{1}{2} \delta_1)} \varphi_{l_1}(r - R - \delta_1) \varphi_{l_2}^*(r') \varphi_{l_1}^*(R - R) \varphi_{l_2}(r' - \delta_2)$, where $R$, $\delta_1$ and $\delta_2$ denote crystal sites. After the integration with respect to $r$ and $r'$ and summation with respect to $R$ one gets the linear system of equations

$$
\sum_{l_3} \sum_{l_3'} \left[ \delta_{l_3 l_1} \delta_{l_3 l_2} \delta_{3} \delta_{l_3} - \sum_{\delta_3'} V_{l_3 l_3 l_1'} (\delta_1, \delta_3'; q) \Pi_{l_3 l_3'} (\delta_3' - \delta_3, q, \omega) \right] \times
\times \bar{V}_{l_3 l_3 l_2} (\delta_3, \delta_2; q, \omega) = V_{l_1 l_2 l_1'} (\delta_1, \delta_2; q)
$$

for the matrix elements of the screened Coulomb interaction

$$
\bar{V}_{l_1 l_2 l_1'} (\delta_1, \delta_2; q) = \sum_R e^{i q (R + \frac{1}{2} \delta_1)} \int dr \int dr' \varphi_{l_1}^*(r - R - \delta_1) \varphi_{l_2}^*(r') \times
\times \bar{V}(r, r', \omega) \varphi_{l_1'} (r' - \delta_2).
$$

$V_{l_1 l_2 l_1'} (\delta_1, \delta_2; q)$ are the corresponding bare matrix elements, when $\bar{V}(r, r', \omega)$ in (8) is replaced by $e^2/|r - r'|$. In this TB representation of two-body interaction one again relies on the localized nature of atomic orbitals and retains only those matrix elements in which $\delta_1$ and $\delta_2$ are either zero or denote nearest neighboring sites. In other words, one distinguishes between the site-site ($\delta_1 = \delta_2 = 0$), site-bond ($\delta_1$ or $\delta_2$ equal to zero) and bond-bond ($\delta_1$ and $\delta_2$ different from zero) contributions to the given $q$-component of the Coulomb interaction. In eq. (7) we have also passed to the adiabatic screened matrix elements, with the polarization diagrams given by

$$
\Pi_{l, l'} (\delta - \delta'; q, \omega) = \frac{2}{N} \sum_k e^{-i(k + \frac{1}{2}) (\delta - \delta')} \frac{1}{[1 + S_l(k)][1 + S_{l'}(k + q)]} \times
\times \frac{n_l(k) - n_{l'}(k + q)}{\omega + E_l(k) - E_{l'}(k + q) + i\eta}.
$$

The coefficient on the left-hand sides of eq. (8),

$$
\varepsilon_{l_1 l_2 l_1' l_2'} (\delta_1, \delta_2; q, \omega) \equiv \delta_{l_1 l_1'} \delta_{l_2 l_2'} \delta_{3} \delta_{l_3} - \sum_{\delta_3'} V_{l_1 l_3 l_1'} (\delta_1, \delta_3'; q) \Pi_{l_3 l_3'} (\delta_3' - \delta_3, q, \omega)
$$

define the dielectric matrix in the TB representation. Note that the sums in eqs.(7) and (10) go over \( \delta = 0 \) and all nearest neighboring sites. The order of the matrix (10) (i.e. the number of equations in the system (2) for fixed \( l_2, l'_2 \) and \( \delta_2 \)) is equal to the number of pairs of band indices \((l_1,l'_1)\) multiplied by \( Z+1 \), where \( Z \) is the number of nearest neighbors (i.e. the coordination number for Bravais lattices). Note that the polarization diagram (9) vanishes if \( l \)-th and \( l' \)-th bands are both full or empty, so that the number of relevant pairs \((ll')\) in eqs.(8) and (10) reduces to that representing transitions between (partially) full and (partially) empty bands (including the intra-band transitions). Furthermore, the order of the matrix (10) reduces to a finite number after a physical truncation of the system (2) by which one keeps only contributions from bands which are not too far energetically from the Fermi level, and neglects e.g. those from deep core atomic states and very high empty bands.

The determinant of the matrix (10) is of the central importance for further considerations, since it carries relevant information about the microscopic dielectric properties. Indeed, it enters into the denominators of all screened matrix elements, and, furthermore, its zeros define the collective as well as the electron-hole excitations of the two-band electron gas [1]. For these reasons we call it microscopic dielectric function, 

\[
\epsilon_m(q,\omega) \equiv \text{det}[\varepsilon].
\]

We summarize this Section by comparing the present TB formulation of the dielectric matrix with the more usual one, based on the plane wave representation and indexed by an infinite number of the vectors of reciprocal lattice. The expressions (7 - 10) formally look even less convenient, since the dielectric matrix (10) is spanned by two multiplied infinities, the number of lattice sites (indices \( \delta \)) and the number of intra-
and inter-band transitions [indices \((ll')\)]. However, as argued above, both these infinities are physically reduced to finite orders, which are eventually rather low in standard TB systems. No such reduction is possible in the plane wave formulation, so that the order of the corresponding dielectric matrix remains physically very large. The reason is that the base of reciprocal lattice vectors cannot be simply truncated whenever the polarization processes take part on the scale of the unit cell.

3 Two-band model

The further analysis is concentrated on the case of two electron bands which do not overlap. To fix ideas we assume that the lower ("valence") band \((l = 0)\) is partially or completely full, while the upper ("conducting") band \((l = 1)\) is empty. The corresponding band dispersions \(E_l(k)\) will be specified in the later examples \([1]\). Furthermore, the later discussion will be mostly limited to the long-wavelength regime \((q \rightarrow 0)\). As it is shown in the Appendix A, the contributions from the Coulomb matrix elements with bond integrations \([\delta_1\text{ and/or } \delta_2\text{ finite in eq.}(8)]\) are in this limit weak and negligible in comparison to the site contributions. We therefore continue by keeping only site matrix elements in the system \([7]\), and skip \(\delta\)-indices from now on. The dielectric matrix \([10]\) then reduces to \([19, 11]\)

\[
\epsilon = \begin{bmatrix}
1 - V_{0000} \Pi_{00} & -V_{0001} \Pi_{01} & -V_{0100} \Pi_{10} & 0 \\
-V_{0010} \Pi_{00} & 1 - V_{0011} \Pi_{01} & -V_{0110} \Pi_{10} & 0 \\
-V_{1000} \Pi_{00} & -V_{1001} \Pi_{01} & 1 - V_{1100} \Pi_{10} & 0 \\
-V_{1010} \Pi_{00} & -V_{1011} \Pi_{01} & -V_{1110} \Pi_{10} & 1 \\
\end{bmatrix}.
\]
The discussion of the dielectric matrix in previous works \cite{19,11} was restricted to the cases in which the off-diagonal matrix elements were negligible. In the present work, we show that in the systems with finite interband dipolar transitions these matrix elements are essential and can by no means be treated perturbatively. On the other hand, the dielectric matrix discussed in Ref.\cite{19} allows for a Fermi surface crossing both bands due to their finite overlap. In our case of a finite gap between two bands, the matrix elements $V_{l_1l_2l_2'}(q,\omega)$ on the left-hand sides of (7) are simply multiplied by $\delta_{ll_1}\delta_{l_1l_1'}$, due to $\Pi_{11}(q,\omega) = 0$. The problem is therefore reduced to a system of three equations, as is seen from the form of the matrix (\ref{eq:matrix}).

A further simplification in $[\varepsilon]$ takes place after assuming that the products of orbitals $\phi_l^*(r)\phi_{l'}(r)$ are real for each pair $(l,l')$, as is usually true in standard cases. Then the matrix elements of the screened interaction (8) obey symmetry relations

$$V_{l_1l_2l_2'} = V_{l_2l_1l_1'2} = V_{l_1l_1l_1'2} = V_{l_1l_2l_1'l_2'},$$

(12)

which are of course also fulfilled for the corresponding bare matrix elements. Note also that

$$V_{l_1l_2l_1'l_2'}(q,\omega) = V_{l_2l_1l_1'l_1}(-q,\omega),$$

(13)

is generally valid due to $V(r,r',\omega) = V(r',r,\omega)$ and $V(r+R,r'+R,\omega) = V(r,r',\omega)$, where $R$ is any lattice vector.

Due to the first equality in (12), first three equations in the linear subsystem (\ref{eq:matrix}) reduce to two equations for $V_{0l_2l_2'}$ and e. g. $V_{0l_2l_1'2}$. The corresponding matrix is given

11
by
\[
\begin{pmatrix}
1 - V_{0000} \Pi_{00} & -V_{0001} (\Pi_{01} + \Pi_{10}) \\
-V_{1000} \Pi_{00} & 1 - V_{0011} (\Pi_{01} + \Pi_{10})
\end{pmatrix},
\]
(14)
while the microscopic dielectric function reads
\[
\varepsilon_m(q, \omega) = [1 - V_{0000}(q) \Pi_{00}(q, \omega)] \{1 - V_{0011}(q) \left[ \Pi_{01}(q, \omega) + \Pi_{10}(q, \omega) \right]\} -
-V_{0001}(q) V_{1000}(q) \Pi_{00}(q, \omega) \left[ \Pi_{01}(q, \omega) + \Pi_{10}(q, \omega) \right].
\]
(15)
Finally, among sixteen TB matrix elements of the screened interaction (8) six are
given by following relations
\[
\begin{align*}
V_{0000} &= \frac{1}{\varepsilon_m} [V_{0000} + (V_{0001} V_{0010} - V_{0000} V_{0011}) (\Pi_{01} + \Pi_{10})], \\
V_{0001} &= \frac{1}{\varepsilon_m} V_{0001}, \\
V_{0011} &= \frac{1}{\varepsilon_m} [V_{0011} + (V_{0001} V_{0010} - V_{0000} V_{0011}) \Pi_{00}], \\
V_{0101} &= \frac{1}{\varepsilon_m} [V_{0101} + (V_{0111} V_{0001} - V_{0101} V_{0011}) (\Pi_{01} + \Pi_{10})], \\
V_{0111} &= \frac{1}{\varepsilon_m} [V_{0111} - (V_{1010} V_{0001} - V_{1011} V_{0000}) \Pi_{00}], \\
V_{1111} &= V_{1111} + \frac{1}{\varepsilon_m} \{V_{1010} \Pi_{00} [V_{0101} + (V_{0001} V_{0111} - V_{0101} V_{0011}) (\Pi_{01} + \Pi_{10})] \\
&+ V_{1110} (\Pi_{01} + \Pi_{10}) [V_{0111} + (V_{0010} V_{0101} - V_{0111} V_{0000}) \Pi_{00}]\}.
\end{align*}
\]
(16-21)
while the remaining ten matrix elements follow from the relations (12) and (13).

The dimension of the matrix (14) coincides with the number of intra-band and inter-band transitions for the present two-band model, in accordance with the general conclusion about the dimension of the dielectric matrix in the tight-binding approach from Ref.[17] and Sect.2. Evidently, the result (14) simply reduces to the one-dimensional matrices in the particular cases of the two-band insulator (when $\Pi_{00} = 0$), and of the one-band conductor (when $\Pi_{01} = \Pi_{10} = 0$).
4 Multipole expansion

For further considerations it is appropriate to write the bare Coulomb matrix elements as sums of local and long-range (two-site) parts,

\[ V_{t_1t_2' t_1' t_2'}(\mathbf{q}) = U_{t_1t_2' t_1' t_2'}(\mathbf{q}) + W_{t_1t_2' t_1' t_2'}(\mathbf{q}), \]  

(22)

with

\[ U_{t_1t_2' t_1' t_2'} = \int d\mathbf{r} \int d\mathbf{r}' \varphi^*_{l_1}(\mathbf{r}) \varphi^*_{l_2}(\mathbf{r}') \frac{e^2}{|\mathbf{r} - \mathbf{r}'|} \varphi_{l_1'}(\mathbf{r}) \varphi_{l_2'}(\mathbf{r}') \]  

(23)

and

\[ W_{t_1t_2' t_1' t_2'}(\mathbf{q}) = \sum_{\mathbf{R} \neq 0} e^{i\mathbf{q}\mathbf{R}} \int d\mathbf{r} \int d\mathbf{r}' \varphi^*_{l_1}(\mathbf{r} - \mathbf{R}) \varphi^*_{l_2}(\mathbf{r}') \frac{e^2}{|\mathbf{r} - \mathbf{r}'|} \varphi_{l_1'}(\mathbf{r} - \mathbf{R}) \varphi_{l_2'}(\mathbf{r}'). \]  

(24)

Integrations in the sums (24) are localized within the ranges of TB-orbitals. It was already noted that they are small not only with respect to the range of interaction, but also in comparison with the distances between neighboring crystal sites. It is therefore natural to perform the multipole expansions of the long-range parts. This is particularly useful in the long wavelength limit in which the hierarchy of the leading multipole terms is well defined. In this Section we calculate dominant contributions to the long-range bare matrix elements (24), and indicate for which matrix elements (22) the on-site contributions (23) are relevant for the dielectric response in the long wavelength limit.

Let us start from the expansion of the bare Coulomb interaction up to the second order in the atomic coordinates [22],

\[ \frac{1}{|\rho + \mathbf{R} - \rho'|} = \frac{1}{R} - \frac{\mathbf{R}(\rho - \rho')}{R^3} + \frac{\rho \rho' - 3(\rho \cdot \mathbf{R}_0)(\rho' \cdot \mathbf{R}_0)}{R^3} - \frac{1}{2} \sum_{i,j} (\rho_i \rho_j + \rho'_i \rho'_j) \frac{R^2 \delta_{i,j} - 3 X_i X_j}{R^5} + ..., \]  

(25)
where \( R_0 \equiv R/R \), and \( \rho \) and \( \rho' \) are local electron positions at the \( R \)-th site and at the origin respectively. \( X_i \) and \( \rho_i \) are i-th Cartesian components of the vectors \( R \) and \( \rho \) respectively. In the next step we chose the parity of the TB orbitals by imposing

\[
\varphi_0(-r) = \varphi_0(r),
\]

(26)

and

\[
\varphi_1(-r) = \pm \varphi_1(r).
\]

(27)

Since the two possibilities defined by the upper [model A] and lower [model B] sign in eq.(27) lead to qualitatively different properties of the dielectric response, we shall treat both in parallel. Furthermore, we choose the cubic lattice, the simplest one for the calculation of lattice sums in the matrix elements of the terms in the expansion (25). To fix ideas, we also specify in the further considerations that \( \varphi_0 \) is an s - orbital, while \( \varphi_1 \) is an d (upper sign) or p (lower sign) orbital, i.e. that the models A and B are characterized by \((s,d)\) and \((s,p)\) bands respectively. With these choices and with the assumption that there is no orbital degeneracy, the crystal symmetry is strictly speaking lower than cubic. Still this is not a serious inconsistency. On the one side, one may suppose that the Bravais lattice is built from some elongated molecules with e.g. \( \sigma \) and \( \pi \) electronic orbitals and that the differences in the lattice constants along three orthogonal crystal directions are still small and have negligible effects on the lattice sums. Alternatively, the present analysis can be straightforwardly completed by including cubic orbital degeneracy into electronic spectrum. The dielectric matrix for the cubic crystal with e.g. one (partially) full s and three empty p bands will be derived in the Appendix B.
Let us now determine the leading terms in the expansion (25) of the matrix elements (22), paying particular attention to those present in the dielectric matrix (14).

$V_{0000}(q)$ belongs to the set of matrix elements $V_{l_1,l_2,l_1',l_2'}(q)$ for scatterings in which electrons do not change bands. The leading long range contribution $W_{l_1,l_2,l_1',l_2'}(q)$ comes from the monopole-monopole term which is same for all $l_1$ and $l_2$, no matter whether one ($l_1 = l_2$) or two ($l_1 \neq l_2$) bands are involved. Since this term diverges in the limit $q \to 0$, the on-site contribution $U_{l_1,l_2,l_1',l_2'}$ to the total bare interaction (22) can be neglected. Keeping only the most divergent part of the monopole-monopole term, one gets for both models A and B

$$V_{l_1,l_2,l_1',l_2'}(q) = \frac{4\pi e^2}{a^3 q^2},$$

where $a^3$ is the volume of the primitive cell.

The bare Coulomb matrix elements in which one or both electrons change bands will depend on the choice of TB orbitals $\varphi_0$ and $\varphi_1$. The decisive difference comes from the fact that the matrix element for the dipole transition

$$\mu_{01} = \mu_{10}^* \equiv e \int d\rho \varphi_0^*(\rho) \rho \varphi_1(\rho)$$

vanishes in the model A and is finite in the model B. We therefore continue by considering each model separately.

Let us start with the model B. The leading long range contribution to the matrix elements in which one electron changes the band comes from the monopole-dipole term in the expansion (25), and reads

$$W_{l_1l_2l_1'l_2'}(q) = \sum_{R \neq 0} e^{iqR} \frac{\mu_{l_1l_1'} \cdot R_0}{R^2}. \quad (30)$$
\( (l \neq l') \). The divergent sum in this expression depends on the angle between \( \mathbf{q} \) and \( \mathbf{\mu}_{ll'} \), where the direction of \( \mathbf{\mu}_{ll'} \) is determined by \( \mathbf{r} \) - dependences in \( \varphi_0 \) and \( \varphi_1 \) \((23)\). With \( \varphi_1 \) chosen to be the \( p \) - orbital elongated along the \( x \) - axis, \( \mathbf{\mu}_{01} \) is also directed along this axis. Noting in addition that in the model B the on-site term \( U_{ll'} \) vanishes due to symmetry reasons, one gets

\[
V_{ll'}(\mathbf{q}) = -V_{l'll'}(\mathbf{q}) = \frac{4\pi i e \mu q_x}{a^3} q^2
\]

where \( \mu \equiv |\mathbf{\mu}_{01}| \). The leading long range contribution to matrix elements with both electrons changing bands is the dipole-dipole one, which after performing the dipolar summations \((23)\) for the cubic crystal lattice in the limit \( \mathbf{q} \rightarrow 0 \), reads

\[
V_{ll'}(\mathbf{q}) = \frac{4\pi \mu^2}{3a^3} \left( \frac{3q_x^2}{q^2} - 1 \right) + U_{ll'}
\]

with \( l \neq l' \). Eqs.(28), (31) and (32) exhaust all matrix elements for the model B. We conclude that for \( \mathbf{q} \rightarrow 0 \) the on-site and the long range contributions enter on the equal footing only into the matrix elements (32), while in the matrix elements (28) and (31) the former are negligible with respect to the latter.

In the model A the leading long range contributions to the matrix elements \( V_{ll'}(\mathbf{q}) \) and \( V_{ll'}(\mathbf{q}) \) come from monopole-quadrupole and quadrupole-quadrupole terms in the expansion \((23)\), respectively. If the matrix elements of the quadrupolar transition between the orbitals \( \varphi_l \) and \( \varphi_{l'} \) are finite, these contributions behave in the limit \( \mathbf{q} \rightarrow 0 \) as \( q^0 \) and \( q^2 \), respectively. We do not need their detailed forms, since the further conclusions will be based solely on the fact the corresponding total matrix elements \( V_{ll'}(\mathbf{q}) \) and \( V_{ll'}(\mathbf{q}) \) are regular in the long wavelength limit (and \( V_{ll'} \simeq U_{ll'} \) in addition).

The most important difference between the \( \mathbf{q} \rightarrow 0 \) limits of models A and B appears
in processes in which one electron changes its band. While in the model B the corresponding matrix element \( (31) \) comes from the long range part and diverges as \( q^{-1} \), the analogous matrix element in the model A is the sum of the long range part and the on-site contribution and behaves as \( q^0 \).

It remains to determine the asymptotic behavior of the polarization diagrams which figure in the dielectric matrix \( (14) \). We limit the present discussion to the regime \( (q \to 0, \omega \neq 0) \) in which the real parts of the polarization diagrams are given by the well-known expressions

\[
\text{Re} \, \Pi_{00}(q, \omega) = \frac{n_e}{m^*} \frac{q^2}{\omega^2}, \quad (33)
\]

and

\[
\text{Re}[\Pi_{01}(q, \omega) + \Pi_{10}(q, \omega)] = 2n_e \frac{E_g}{\omega^2 - E_g^2}. \quad (34)
\]

\( m^* \) is the effective mass and \( n_e \) is the number of electrons per site in the lower band, while \( E_g \equiv E_1 - E_0 \) is the energy difference between the band centers. Here we neglect the bandwidths in the expression \( (34) \) assuming that they are much smaller than \( E_g \). Note that the expressions \( (33) \) and \( (34) \) do not depend on the symmetry of TB orbitals \( \varphi_0 \) and \( \varphi_1 \). Furthermore, we do not introduce the corresponding imaginary parts, relegating the discussion of the effects of Landau damping to the paper II. We only note that in the above \( (q, \omega) \)-regime \( \text{Im} \, \Pi_{00} = 0 \) while \( \text{Im}[\Pi_{01} + \Pi_{10}] \) may be finite.

We are now ready to compare the microscopic dielectric functions for models A and B. Let us start with the simpler case of model A, already considered in Ref.\((19)\) and explored in Refs.\((20, 21)\). As it is seen from \( (33) \) and \( (34) \), the term in the expression \( (13) \) which comes from the off-diagonal elements in the matrix \( (14) \) vanishes in the long
wavelength limit. In other words, the intra-band and inter-band polarization processes, given by the diagonal elements in this matrix, become decoupled, and it is legitimate to distinguish between the intra-band ($\epsilon_{\text{intra}}$) and inter-band ($\epsilon_{\text{inter}}$) dielectric functions [19], the total dielectric function being the product of the two. Due to this factorization the collective modes, defined by the real zeros of $\epsilon_m$ in the $\omega$-plane, are also either of intraband or of interband origin. They will be analysed in detail in the paper II.

Furthermore, the expressions (16-21) for the screened Coulomb interaction reduce to

$$\nabla_{lll'}/V_{lll'} = 1/\epsilon_{\text{intra}}$$  \hspace{1cm} (35)

(with either $l = l'$ or $l \neq l'$), and

$$\nabla_{lll'}/V_{lll'} = \nabla_{lll'}/V_{lll'} = 1/\epsilon_{\text{inter}}$$  \hspace{1cm} (36)

(with $l \neq l'$). Thus all purely intraband scatterings are screened by $\epsilon_{\text{intra}}$, while the scatterings with at least one interband electron transition are screened by $\epsilon_{\text{inter}}$.

The above decoupling does not hold in the model B. The product of two off-diagonal matrix elements in (14) now remains finite in the long wavelength limit, and introduces a coupling between the intraband and interband contributions into the microscopic dielectric function (13). The present analysis shows that this coupling comes from the divergent monopole-dipole contribution (31) to the matrix element $V_{lll'}(q)$. Its most important consequence, the hybridization of the intraband and interband collective modes in conductors, will be considered in the paper II.
5 Macroscopic dielectric function

The aim of this section is to establish the relation between the macroscopic dielectric function $\varepsilon_M(q, \omega)$ and our microscopic dielectric function $\varepsilon_m(q, \omega)$. The former is defined in a standard way as the ratio between the Fourier components of the spatial averages of the bare (external) and screened (total) Coulomb potential. To get macroscopic averages, it is enough to take the unit cell as the averaging volume [6]. Let us denote the averaged bare and screened interactions between the unit cells at sites $R$ and $R'$ by $V_{av}(R, R')$ and $V_{av}(R, R', \omega)$ respectively. Since at long wavelengths details in the space dependence of the true (non-averaged) interactions on the scale of unit cell are irrelevant, the true and averaged interactions are in this limit indistinguishable, and one gets the connection

$$V_{av}(q + \mathbf{G}, q + \mathbf{G}', \omega) \approx \delta_{\mathbf{G},0} \delta_{\mathbf{G}',0} V(q, q, \omega)$$

(37)

between the corresponding Fourier transforms. Here $\mathbf{G}$ and $\mathbf{G}'$ denote the vectors of the reciprocal lattice. The same relation holds for the averaged bare interaction $V_{av}(q)$.

The macroscopic dielectric function is now defined by

$$\frac{1}{\varepsilon_M(q, \omega)} = \frac{V_{av}(q, q, \omega)}{V_{av}(q)} = \varepsilon^{-1}(q, q, \omega).$$

(38)

The ratio $V_{av}(q, q, \omega)/V_{av}(q)$ can be also interpreted as the $q$-component of the total charge density, redistributed due to the Coulomb screening, averaged across the unit cell and divided by the corresponding $q$-component of the probe charge [7]. The last equality in eq.(38) follows from the standard definition of the dielectric matrix in the plane wave representation, $V(q, G, G', \omega) = \varepsilon^{-1}(q + G', q + G, \omega)V(q, G, G)$, and the
relation (37). Our aim is however to express \( \epsilon_M(q, \omega) \) in terms of the TB bare and screened Coulomb matrix elements (22, 8). The TB matrix elements (8) can be expressed as double sums over the Fourier transforms \( V(q + G, q + G', \omega) \) through the relation

\[
\sum_{G, G'} I_{l_1 l_2} \left( q + G, q + G' \right) V(q + G, q + G', \omega) = \sum_{G, G'} I_{l_1 l_2} \left( q + G, q + G' \right) V(q + G, q + G', \omega)
\]

(39)

with

\[
I_{l_1 l_2}(q) \equiv \int d\mathbf{r} \varphi_{l_1}^*(\mathbf{r}) e^{i\mathbf{q}\cdot\mathbf{r}} \varphi_{l_2}(\mathbf{r}).
\]

(40)

The corresponding relation for the averaged matrix elements follows after inserting the relation (37) into (39). Expanding the matrix elements (40) in powers of \( q \) we get in the limit \( q \to 0 \)

\[
\frac{\nabla_{av,l_1 l_2 l_1' l_2'}(q, \omega)}{\nabla_{av}(q, \omega)} \approx \frac{\delta_{l_1, l_1'} + \frac{iq \mu_{l_1 l_1'}}{e} + O(q^2)}{\delta_{l_2, l_2'} + \frac{iq \mu_{l_2 l_2'}}{e} + O(q^2)} \nabla_{av}(q, \omega)
\]

(41)

with \( \mu_{l_1 l_1'} \) being the dipolar matrix element (29). Thus, whenever \( l_1 \neq l_1' \) and/or \( l_2 \neq l_2' \), the corresponding ratios \( \frac{\nabla_{av,l_1 l_2 l_1' l_2'}(q, \omega)}{\nabla_{av}(q, \omega)} \) tend to zero as \( q \to 0 \). In other words, since the averaged interactions do not vary within a unit cell, the contributions from the dipolar and higher transitions to the corresponding TB matrix elements \( \nabla_{av,l_1 l_2 l_1' l_2'}(q, \omega) \) vanish. For \( l_1 = l_1' \) and \( l_2 = l_2' \), i.e. for matrix elements with a finite monopole-monopole contribution, we have

\[
\nabla_{av,l_1 l_2 l_1 l_2}(q, \omega) \approx \nabla_{l_1 l_2 l_1 l_2}(q, \omega) \approx \nabla_{av}(q, \omega)
\]

(42)

for any pair of orbital indices \( l_1 \) and \( l_2 \). The first equality in (42) is based on the same argument as the relation (37). Since the relations (41) and (42) are also valid for the bare interaction \( V(\mathbf{r} - \mathbf{r}') \), the macroscopic dielectric function reads

\[
\epsilon_M(q, \omega) = \frac{V_{l_1 l_2 l_1 l_2}(q)}{\nabla_{l_1 l_2 l_1 l_2}(q, \omega)}.
\]

(43)
This result is generally valid for any number of bands. It also does not depend on the method of calculation of the linear dielectric response, which brings in a particular relationship between the screened and bare Coulomb interaction. The independence of the right-hand side of (43) on the indices $l_1$ and $l_2$ may serve as a check of the consistency of a given approximation in the macroscopic limit. It is easy to see that this independence is realized for the RPA result (16-21).

Although $\epsilon_M(q, \omega)$ is the ratio of the bare and screened monopole-monopole interactions, this of course does not mean that other higher order terms in the multipole expansion do not contribute to the macroscopic response. This becomes clear already for the simple two-band models from Sects. 3, 4. For the model A the result (33) reads

$$
\epsilon_M(q, \omega) = \epsilon_{\text{intra}}(q, \omega) = 1 + 4\pi\alpha_c(q, \omega),
$$

(44)

with

$$
\alpha_c(q, \omega) \equiv -\frac{e^2}{4\pi q^2}\Pi_{00}(q, \omega)
$$

(45)

representing the intraband polarizability. Thus, the macroscopic dielectric function is entirely determined by the intraband processes, and reduces to unity when the lower band is fully occupied. In other words, the interband modes are not directly optically active, and can possibly be observed only by Raman scattering.

The macroscopic dielectric response for the model B follows after inserting any of the expressions (16,19,21) into (43), and taking into account the long wavelength asymptotic expressions (28,31,32). One gets

$$
\frac{\epsilon_m}{\epsilon_M} = 1 + \left[\frac{4\pi \mu^2}{3a^3} - U_{0011}\right] (\Pi_{01} + \Pi_{10}).
$$

(46)
Generally, the right-hand side in this expression depends on the direction of \( q \) due to the anisotropy of the polarization diagram \((\Pi_{01} + \Pi_{10})\). However, in the particular limit given by eq.(34) this is not the case, so that the orientational dependence of \( \varepsilon_M(q,\omega) \) is determined solely by that of the microscopic function \( \varepsilon_m(q,\omega) \). Let us consider here only two particular directions, \( q \perp \mu \) and \( q \parallel \mu \).

For \( q = q_\perp \perp \mu \) the bare monopole-dipole matrix element (31) vanishes, so that \( \varepsilon_m = \varepsilon_{\text{intra}} \varepsilon_{\text{inter}} \) with \( \varepsilon_{\text{inter}} \) equal to the right-hand side od (46). Hence one gets

\[
\varepsilon_M(q_\perp,\omega) = 1 + 4\pi \alpha_c(q_\perp,\omega),
\] (47)

as should be expected, since for \( q \perp \mu \) the interband longitudinal polarization processes are not possible.

For \( q \parallel \mu \) the (intraband) monopole - (interband) dipole coupling is finite and, in addition, even for an insulator there is no simplification in (46) as for \( q \perp \mu \). The macroscopic dielectric function can be then written in the form

\[
\varepsilon_M(q_\parallel,\omega) = 1 + 4\pi \alpha_c + \frac{4\pi \alpha_I}{1 - \frac{4\pi}{3} \alpha_I},
\] (48)

Here

\[
\alpha_I(q_\parallel,\omega) \equiv \frac{\alpha_I(q_\parallel,\omega)}{1 + U_{0011}a^3\alpha_I(q_\parallel,\omega)/\mu^2},
\] (49)

with \( \alpha_I(q_\parallel,\omega) \) being the dipolar interband polarizability

\[
\alpha_I(q_\parallel,\omega) \equiv -\frac{\mu^2}{a^3} \left[ \Pi_{01}(q_\parallel,\omega) + \Pi_{10}(q_\parallel,\omega) \right].
\] (50)

The three quantities which enter into the expressions (49, 50) represent the intraband \( (\alpha_c) \), interband \( (\alpha_I) \) and intra-atomic \( [U_{0011}(\Pi_{01} + \Pi_{10})] \) contributions to the macroscopic dielectric response. Consequently, this expression covers, and interpolates be-
tween, limiting cases of one-band conductor, two (multi)-band insulator, and atomic (zero bandwidth) insulator. Before going into particular limits, we remind that Adler parametrized any deviation of the expression (48) from the usual form containing only the dipolar interband polarizability (50) by introducing a self-polarization correction, defined as \( \alpha_I(q||,\omega) - \bar{\alpha_I}(q||,\omega) \equiv C \). When expressed in terms of TB quantities, this parameter reads

\[
C = \frac{\mu^2}{a^3} \frac{U_{0011}(\Pi_{01} + \Pi_{10})^2}{1 - U_{0011}(\Pi_{01} + \Pi_{10})}.
\]

As it will be shown below, the TB results (49) and (50) provide a direct physical insight into this correction for atomic insulators as well as for band insulators and conductors.

Let us now consider some characteristic limits. The usual Lindhard (i.e. Sellmeyer) expression for a metal follows after neglecting interband polarization (\( \alpha_I = 0 \)), while the opposite limit for an atomic insulator is obtained after putting \( \alpha_c = 0 \) and neglecting all bandwidths [11]. The latter result is the well known Lorentz-Lorenz (i.e. Clausius-Mossotti) expression. In this limit the polarizability \( \bar{\alpha_I} \) which enters into eq.(48) is given by

\[
\bar{\alpha_I}(q,\omega) = -\frac{2n_e}{a^3} \frac{E_{g,eff}\mu^2_{eff}}{\omega^2 - E_{g,eff}^2},
\]

i.e. it does not differ in form from the expression (50). Here \( \mu^2_{eff} = \mu^2 E_g/E_{g,eff} \) and \( E_{g,eff} = E_g^2 + 2nE_gU_{0011} \) are the renormalized values of the dipole matrix element and the level spacing, i.e. instead of the initial (e.g. atomic Hartree-Fock) parameters one obtains those which include the on-site RPA screening. In other words, the Adler’s self-polarization correction in this case simply reduces to a renormalization of the parameters in the dipolar (intra-atomic) polarizability. This reflects the fact that the RPA scheme
proposed in the present work treats both nonlocal and on-site screenings at the same level of approximation. In this respect, we warn that in the limit the RPA is usually not a reliable scheme at the atomic (or molecular) level [25], and that it is obviously invalid when the on-site interactions are large in comparison with $E_g$.

In the cases of insulators and conductors with finite bandwidths the effective dipolar polarizability (49) does not have the simple form (52) and, moreover, cannot be reduced to the form of an effective interband polarization diagram, cf. (50). Instead, one has a nontrivial $\omega$-dependence in the denominator of the expression (49). The self-polarization correction is no more of entirely local (i.e. intra-atomic) origin, but appears to be a combined effect of the on-site Coulomb interaction $U_{0011}$ in which two electrons exchange their orbital states, and of the finite inter-site tunneling (i.e. of the finite bandwidths). The role of $U_{0011}$ in the macroscopic dielectric function was already noticed in Ref. [11]. We note that some later results for $\epsilon_M$ [7, 26], although based on the RPA method, cannot be expressed in the form (49, 50) i.e. (51). The reason might be traced in the additional approximations which, unlike the present RPA scheme, do not include the local on-site screening on the same footing with the long range one.

Furthermore, it is appropriate to point out that $\overline{\alpha_T}$ appears in the denominator of eq.(48) due to the finiteness of $G \neq 0$ terms (local fields) in the Fourier transform of the dipole-dipole interaction (32). The $G \neq 0$ terms are comparable to the $G = 0$ contribution and should not be neglected, as already stressed in the Introduction. Their omission transforms the formula (48) into an expression which is additive in the polarizabilities $\overline{\alpha}$.
6 Conclusion

The most important step in the present work is the use of the complete set of TB states in the representation of the bare and screened Coulomb interactions. In this basis, the symmetry properties of the Coulomb matrix elements follow directly from the symmetry of molecular orbitals participating in the band states. As a consequence, the initial system of linear equations for the screened matrix elements decomposes into smaller subsets with a common dielectric matrix. E.g., in the two-band case sixteen linear equations were reduced to two equations. This agrees with our general conclusion \cite{17} that for a multi-band system with only two-center Coulomb interactions taken into account, the TB dielectric matrix within RPA has a dimension equal to the number of non-vanishing intra-band and inter-band polarization diagrams \cite{9}.

The present approach is particularly convenient in the long wavelength limit, in which the multipole expansion is feasible. Again, the symmetry of molecular orbitals is of central importance, since it determines the degree of mixing between intra-band and inter-band polarization processes in multiband conductors. In particular, whenever the interband processes are dipolar the off-diagonal elements in the TB dielectric matrix are finite and cannot be treated perturbatively. The inclusion of the long-range monopole-dipole interaction induced by these processes leads to the correct expression for the macroscopic dielectric function, and, as will be shown in paper II \cite{1}, essentially influences the spectrum of collective modes for a multiband electron liquid (see also Ref. \cite{17}). In this respect it is important to recognize that the results for $\epsilon_m(q, \omega)$ \cite{11} and
\( \epsilon_M(q, \omega) \) reduce to the additive form

\[
\epsilon(q, \omega) - 1 = 4\pi \alpha_c + 4\pi \sum_I \alpha_I,
\]

only in the high frequency limit, i.e. for frequencies much higher than all interband energy differences \( E_{gI} \). [The \( I \)-summation in eq.(53) goes over all relevant interband polarizabilities (50)]. It is interesting to note that in this limit we apparently recover the well-known expression for the high frequency plasma edge expressed through the standard plasmon sum rule, involving the free electron mass.

In conclusion, we point out that the present approach might have numerous applications, particularly in systems with dipolar degrees of freedom which are sensitive to the variations of the valence band properties induced by pressure, changes in band filling, etc. We expect that the present method will prove very useful in such investigations.

A Appendix A

In this Appendix we consider the influence of the site-bond (\( \delta_1 \) or \( \delta_2 \) different from zero) and bond-bond (\( \delta_1 \) and \( \delta_2 \) different from zero) Coulomb interaction (8) on the dielectric response in the long-wavelength limit (\( q \rightarrow 0, \omega \) finite). We take the simplest examples of one-band and two-band systems with the one-dimensional lattice \([\delta_i \text{ in eqs.(7 - 10)}\) equal to 0 and \( \pm 1 \)]. The extensions to more complex band structures and lattices are straightforward.

The system (7) simplifies after assuming, like in Sects.3 and 4, that the orbitals \( \phi_l(\mathbf{r}) \) are even or odd, and that the products \( \phi_{l_i}^*(\mathbf{r} - \delta) \phi_{l_j}(\mathbf{r}) \) are real. This leads to additional symmetry relations between bare Coulomb matrix elements (8), which in the case of
two-band system \((l_i = 0, 1)\) read

\[
V_{l_i l_1 l_2 l_2'}(\delta_1, \delta_2) = (-1)^{l_i} V_{l_i l_1 l_1', l_2 l_2'}(\delta_2, \delta_1) = (-1)^{l_i} V_{l_2 l_1 l_2, l_1 l_1'}(-\delta_2, -\delta_1) = \\
V_{l_1 l_1 l_2 l_2'}(\delta_1, -\delta_2) = V_{l_2 l_1 l_1', l_2 l_1'}(-\delta_1, \delta_2) = V_{l_1 l_1 l_2, l_1 l_1'}(-\delta_1, -\delta_2).
\] (A.1)

Due to these symmetry relations, the number of equations in (7) reduces to \(N_e/2\) or \((N_e + 1)/2\) for even or odd \(N_e\) respectively, where \(N_e\) is the product of \(Z + 1\) and the number of the non-zero polarization diagrams among \(\Pi_{00}, \Pi_{01}\) and \(\Pi_{10}\).

In particular, taking into account these relations, we obtain the microscopic dielectric function for one-band metal in the form of a \(2 \times 2\) determinant

\[
\varepsilon_m = \left| 1 - \sum_\delta V(0, \delta) \Pi_0(\delta) & - \sum_\delta V(1, \delta) \Pi_0(\delta) \\
- \sum_\delta V(0, \delta) [\Pi_0(\delta - 1) + \Pi_0(\delta + 1)] & 1 - \sum_\delta V(1, \delta) [\Pi_0(\delta - 1) + \Pi_0(\delta + 1)] \right|
\] (A.2)

with \(V(\delta_1, \delta_2) \equiv V_{0000}(\delta_1, \delta_2, q)\) and \(\Pi_0(\delta) \equiv \Pi_{00}(\delta, q, \omega)\). In the limit \(q \to 0\) the leading monopole-monopole terms in the bare matrix elements appearing in (A.2) are simply related by

\[
V(1, 1, q) = SV(1, 0, q) = S^2 V(0, 0, q) = S^2 \frac{4\pi e^2}{a^3 q^2},
\] (A.3)

where \(S\) is the direct overlap (3). After inserting these relations and the expressions (8) for the polarizability diagrams \(\Pi_0(0), \Pi_0(\pm 1)\) and \(\Pi_0(\pm 2)\) into the expression (A.2), it reduces to

\[
\varepsilon_m = 1 - V(0, 0, q) \Pi_0(0) \left[ 1 + O(q^2) \right].
\] (A.4)

Thus, we conclude that in the limit \(q \to 0, \omega\) finite, the microscopic dielectric function for a one-band metal does not depend on the overlap factor \(S\) which enters into the
site-bond and bond-bond interactions through the expressions (A.3). In other words, it is completely determined by the site-site correlations.

In the case of the two-band insulator the most important simplification comes from the fact that the only non-vanishing polarization diagrams are the inter-band ones with the zero phase factor $[\delta - \delta' = 0$ in eq. (A.3)]. The microscopic dielectric function then reduces to

$$
\varepsilon_m = 1 - V_1(1,1)\Pi_1 - V_1(0,1)\Pi_1 - V_1(-1,1)\Pi_1
\begin{vmatrix}
-1 - V_1^*(0,1)\Pi_1 & 1 - V_1(0,0)\Pi_1 & -V_1(0,1)\Pi_1 \\
-V_1^*(-1,1)\Pi_1 & -V_1^*(0,1)\Pi_1 & 1 - V_1(1,1)\Pi_1 \\
\end{vmatrix}
\quad (A.5)
$$

with $V_1(\delta, \delta') \equiv V_{1001}(\delta, \delta'; \mathbf{q})$. Since only one polarization factor $[\Pi_{01}(0, \mathbf{q}, \omega) + \Pi_{10}(0, \mathbf{q}, \omega) \equiv \Pi_1]$ figures in this determinant, the criterion of weakness of site-bond and bond-bond correlations with respect to the site-site ones obviously involves only the corresponding matrix elements of bare Coulomb interaction, i.e. $V_1(0,1), V_1(1,1)$ and $V_1(-1,1)$ vs $V_1(0,0)$. More precisely, the three- and four-center correlations may be considered as weak perturbations provided the matrix element of bond dipolar transition

$$
\mu(\delta) = \int d\mathbf{r} \varphi_0^*(\mathbf{r}) \mathbf{r} \varphi_1(\mathbf{r} - \delta)
\quad (A.6)
$$

and local site-bond and bond-bond matrix elements are much smaller than the respective site parameter (28) and $U_{0011}$ [see eq. (32)].

For a two-band metal $\varepsilon_m$ is given by a $5\times5$ determinant, not written here for the sake of space. Although this determinant contains intraband and interband polarization diagrams (3) with various phase factors, the analysis of the long wavelength limit leads to the same conclusion, namely that the criterion for the weakness of site-bond and bond-bond correlations in the dielectric function involves only the corresponding bare
Coulomb matrix elements.

B Appendix B

In Sections 4 and 5 we have assumed within the model B that there is one orbital with $p_x$ symmetry at each site of a cubic Bravais lattice. In order to complete the discussion, we take here the simplest generalization by which the electron bands have the cubic symmetry. We assume that there are one lower s-orbital and three degenerate upper $p$-orbitals with the symmetry

$$\varphi_x(-x, y, z) = -\varphi_x(x, y, z) \quad (B.1)$$

and equivalently for $x \rightarrow y \rightarrow z$.

Following the procedure from Sec.3 we get for this four-band model a linear Dyson system (7) which contains 256 equations and is decomposed into 16 subsystems of 16 equations. After assuming, like in Sec.3, that all products $\varphi_i^*(r)\varphi_i'(r)$ are real, the problem reduces to a system of four equations

$$\begin{align*}
V_{000x} &= V_{000x} + V_{0000}V_{000x} \Pi_0 + V_{000x}V_{00xx} (\Pi_{0x} + \Pi_{x0}), \\
V_{00xx} &= V_{00xx} + V_{x000}V_{00xx} \Pi_0 + V_{00xx}V_{00xx} (\Pi_{0x} + \Pi_{x0}), \\
V_{00yx} &= V_{00yy}V_{00yx} (\Pi_{0y} + \Pi_{y0}), \\
V_{00zx} &= V_{00zz}V_{00zx} (\Pi_{0z} + \Pi_{z0}).
\end{align*} \quad (B.2-B.5)$$

Here subscripts $x, y$ and $z$ stand for $p_x, p_y$ and $p_z$ orbitals respectively, and the wave vector $\mathbf{q}$ is oriented along the crystal axis $x$. Due to the isotropy of the cubic crystal this special choice does not harm the generality of the system (B.2-B.3) (see also Ref.[17]).
The corresponding dielectric matrix is given by

\[
\begin{bmatrix}
1 - V_{0000} \Pi_{00} & -V_{000z} \Pi_{0z} & 0 & 0 \\
-V_{2000} \Pi_{00} & 1 - V_{00zx} \Pi_{0x} & 0 & 0 \\
0 & 0 & 1 - V_{00yy} \Pi_{0y} & 0 \\
0 & 0 & 0 & 1 - V_{00zz} \Pi_{0z} \\
\end{bmatrix},
\]

(B.6)

and all TB matrix elements \( V_{l_1 l_2 l'_1 l'_2} \) with \( l_i = 0, x, y, z \) follow straightforwardly, but we do not write them in order to save space.

The dielectric properties of the isotropic crystal in the direction parallel and perpendicular to the wave vector are described by the upper left 2 \( \times \) 2 block, and by the third and fourth diagonal element in the matrix (B.6), respectively. Note that the parallel and perpendicular responses with respect to any direction of the wave vector in the cubic crystal are identical to those along the respective preferred dipolar axis \( x \), and the perpendicular axes \( y \) and \( z \) within the model B. Thus, all conclusions derived within the model B for the preferred direction \( x \) are valid also for any direction in the isotropic cubic crystal. In particular the expression \((B8)\) for \( \epsilon_M(q, \omega) \) is now valid for any orientation of \( q \).
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