Dielectric properties of multiband electron systems:

II - Collective modes

P. Županović

Department of Physics, Faculty of Science and Art, University of Split, Teslina 10, 21000 Split, Croatia

A. Bjeliš and S. Barišić

Department of Physics, Faculty of Science, University of Zagreb, P.O.B. 162, 10001 Zagreb, Croatia

Abstract

Starting from the tight-binding dielectric matrix in the random phase approximation we examine the collective modes and electron-hole excitations in a two-band electronic system. For long wavelengths ($q \to 0$), for which most of the analysis is carried out, the properties of the collective modes are closely related to the symmetry of the atomic orbitals involved in the tight-binding states. In insulators there are only inter-band charge oscillations. If atomic dipolar transitions are allowed, the corresponding collective modes reduce in the asymptotic limit of vanishing bandwidths to Frenkel excitons for an atomic insulator with weak on-site interactions. The finite bandwidths renormalize the dispersion of these modes and introduce a continuum of incoherent inter-band electron-hole excitations. The possible Landau damping of collective modes due to the presence of this continuum is discussed in detail.
In conductors the intra-band charge fluctuations give rise to plasmons. If the atomic dipolar transitions are forbidden, the coupling of inter-band collective modes and plasmons tends to zero as $q \to 0$. On the contrary, in dipolar conductors this coupling is strong and nonperturbative, due to the long range monopole-dipole interaction between intra-band and inter-band charge fluctuations. The resulting collective modes are hybrids of intra-band plasmons and inter-band dipolar oscillations. It is shown that the frequency of the lower hybridized longitudinal mode is proportional to the frequency of the transverse dipolar mode when the latter is small. The dielectric instability in a multi-band conductor is therefore characterized by the simultaneous softening of a transverse and a longitudinal mode, which is an important, directly measurable consequence of the present theory.

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**Key words:** collective modes, Landau damping, dielectric instability
1 Introduction

The present paper completes the analysis of the dielectric screening for a two band system, derived in the preceding paper I ([1]). Using the tight-binding (TB) scheme, we showed that the dielectric matrix $\varepsilon$ reduces to a simple $2 \times 2$ form. The determinant of this matrix, $\varepsilon_m$, has the significance of the microscopic dielectric function. In particular, its zeros and poles in the $\omega$-plane define all collective and incoherent (electron-hole) excitations, respectively. Here we examine these excitations for two-band insulators and metals, emphasizing in particular two aspects; the Landau damping of the collective modes caused by the inter-band electron-hole continua, and the hybridization of the intra-band and inter-band collective modes in the dipolar conductors.

The RPA dielectric matrix for the TB system with two bands is given by

$$
\begin{bmatrix}
1 - V_{0000}(q)\Pi_{00}(q, \omega) & -V_{0001}(q)[\Pi_{01}(q, \omega) + \Pi_{10}(q, \omega)] \\
-V_{1000}(q)\Pi_{00}(q, \omega) & 1 - V_{0011}(q)[\Pi_{01}(q, \omega) + \Pi_{10}(q, \omega)]
\end{bmatrix}.
$$

(1)

The matrix elements of the bare Coulomb interaction $V_{lll'1'l'}$ and the bubble polarization diagrams $\Pi_{ij}$ are introduced in paper I [see eqs.(I.8) and (I.9)]. The two diagonal elements in the matrix (1) come from the intraband (00,00) and interband (01,01) polarization processes, while the product of the off-diagonal elements (00,01) and (10,00) introduces a finite mixing between them. Obviously the latter vanishes for insulators, since then $\Pi_{00} = 0$. For conductors, the intraband-interband mixing depends in an essential way on the bare matrix element $V_{0001}$. More precisely, the product of off-diagonal elements in eq.(1) is finite in the long wavelength limit $q \to 0$ if the long range part of $V_{0001}$ diverges as $q^{-1}$ [1]. For Bravais lattices, such divergence takes place when the
matrix element for the dipolar transition between two atomic TB orbitals, $\mu_{01}$, is finite, and $V_{0001}$ is dominated by the monopole-dipole term

$$V_{0001}(q) = \frac{4\pi ie\mu q_x}{a^3 q^2}.$$  

(2)

If $\mu_{01}$ is zero, $V_{0001}(q \to 0)$ is the sum of the on-site contribution and the long range monopole-quadrupole terms which both behave as $q^0$, and thus do not compensate the dependence $\Pi_{00}(q \to 0) \sim q^2$ in the off-diagonal matrix element $(10,00)$.

Having in mind the above remarks we divide the analysis of the collective and electron-hole excitations in the range $(q \to 0, \omega \neq 0)$ into two parts. First, we shall consider in Sect.2 cases for which the condition

$$\lim_{q \to 0} V_{0001}^2 \Pi_{00} [\Pi_{01} + \Pi_{10}] = 0,$$

(3)

is fulfilled for any direction of $q$, so that the microscopic dielectric function has a factorized form

$$\epsilon_m = \epsilon_{intra} \epsilon_{inter},$$

(4)

where $\epsilon_{intra}$ and $\epsilon_{inter}$ are respectively the diagonal elements $(00,00)$ and $(01,01)$ in eq.(1). As we have seen above, the condition (3) covers two interesting cases, namely those of two band insulators and of two-band conductors for which the intra-atomic selection rules forbid the interband dipolar transitions. In Sect.3, we examine the conditions under which the collective modes from the Sect.2 become damped due to the crossing of their dispersion lines with the borders of the inter-band electron-hole continuum. (Some illustrative examples are considered in the Appendix). In Sect.4, we discuss the effects of the finite interband-intraband mixing (3) on the collective modes. Sect.5 contains concluding remarks.
2 Pure intraband and interband collective modes

The factor $\epsilon_{\text{intra}}$ in eq.(4) is the usual one-band RPA dielectric function [2, 3]. The corresponding collective excitations are intraband plasmons, the collective excitations of the valence band electron gas. Their frequencies are finite if the band is metallic, i.e. partially filled.

On the other hand, the spectrum of collective interband excitations is defined by zeros of the interband dielectric function $\epsilon_{\text{inter}}$. These excitations are dispersive, i.e. undamped, provided that the imaginary part of the interband polarization diagram

$$\Pi_{01}(q, \omega) + \Pi_{10}(q, \omega) = \frac{2}{N} \sum_k \left\{ \frac{n_0(k)}{\omega + E_0(k) - E_1(k + q) + i\eta} \right. \\
- \frac{n_0(k + q)}{\omega + E_1(k) - E_0(k + q) + i\eta} \left. \right\}$$

vanishes. In other words, $\omega(q)$, the zeros of $\epsilon_{\text{inter}}(q, \omega)$, have to satisfy the condition

$$\omega(q) \pm [E_0(k) - E_1(k + q)] \neq 0$$

for any $k$, and to be the roots of the equation

$$\frac{4}{N} V_{0011}(q) \sum_k n_0(k) \frac{E_1(k + q) - E_0(k)}{\omega^2 - [E_1(k + q) - E_0(k)]^2} = 1.$$ \hspace{1cm} (7)

If the condition (8) does not hold, the corresponding collective excitation is damped, i.e. it decays into the quasi continuum of incoherent interband electron-hole transitions [3].

Let us start from the hypothetical crystal with zero bandwidths for which $E_1(k) = E_1$, $E_0(k) = E_0$. The dispersion of collective interband modes which follows from eq.(7) is then given by

$$\omega^2(q) = E_g [E_g + 2n_e V_{0011}(q)],$$ \hspace{1cm} (8)
where $E_g = E_1 - E_0$ is the difference of the band centers, and $n_e$ is the number of valence band electrons per site.

For small values of $q$ we distinguish two qualitatively different types of dispersion emerging from eq.(8). If $\mu_{01} = 0$ the leading term in $V_{0011}(q)$ is the intra-atomic interaction $U_{0011}$, while the lowest order possible dependence on $q$ is quadratic and comes from the quadrupole-quadrupole term in the multiple expansion of the inter-atomic interaction $W_{0011}(q)$. This means that in the limit $q \to 0$ the frequency $\omega(q = 0)$ does not depend on the orientation $q/|q|$, but has a single value which is entirely determined by the intra-atomic parameters, and thus represents a transition between two atomic orbitals.

If $\mu_{01} \neq 0$ the dipole-dipole term in the multipole expansion of $W_{0011}(q)$ is finite at $q = 0$, and varies with the orientation of $q$ [see eq.(I.32) for the particular case of cubic lattice]. As a consequence, the frequency $\omega(q \to 0)$ varies continuously with the change of relative orientation between $q$ and the fixed direction of $\mu_{01}$. In other words, the expression (7) represents a single dipolar excitation band, in accordance with the reduced crystal symmetry introduced by the choice of TB orbitals [model B of paper I]. The limiting upper and lower edges which correspond to the pure longitudinal ($q \parallel \mu_{01}$) and transverse ($q \perp \mu_{01}$) dipolar modes are given by

$$\omega_L^2(q \to 0) \equiv \omega_L^2 = E_g(E_g + 2n_eU_{0011} + \frac{16\pi n_e\mu^2}{3a^3}), \tag{9}$$

$$\omega_T^2(q \to 0) \equiv \omega_T^2 = E_g(E_g + 2n_eU_{0011} - \frac{8\pi n_e\mu^2}{3a^3}). \tag{10}$$

respectively. As $|q|$ increases both $\omega_L(q)$ and $\omega_T(q)$ approach the value of $\sqrt{E_g(E_g + 2n_eU_{0011})}$, i.e. the width of the dipolar excitation band decreases (Fig.1).
Let us remind that the result \[(9)\] for the longitudinal dipolar mode is valid provided only that the valence band is full.

It is worthwhile to note that eqs.\((9,10)\), written in terms of the effective polarization \(\mu_{\text{eff}}^2 = \mu^2 E_g / E_{g,\text{eff}}\) and the gap \(E_{g,\text{eff}}^2 = E_g^2 + 2n E_g U_{0011}\) [see eq.\((I.52)\)], are just the standard expressions for the Frenkel excitons. Indeed, after the slight generalization to the band structure which has a cubic symmetry \[\Pi\], these expressions reproduce completely the longitudinal and the transverse excitonic collective branches for a cubic crystal \[\mathbb{I}\]. Moreover, they include two distinct types of dipolar terms treated separately in Ref.\[\mathbb{I}\], i.e. those representing the simultaneous excitation and deexcitation of two atomic electron-hole pairs and the simultaneous excitations of two electron-hole pairs at different crystal sites. It is therefore useful to compare the present derivation of the collective interband modes via RPA (which is strictly limited to the weak coupling regime), with derivations starting from atomic electron-hole pairs \[\mathbb{I}\].

The collective excitonic state with the wave vector \(q\) is given here by a linear superposition in which all interband Bloch electron-hole pairs \(a_1^\dagger(k+q) a_0(k)\) enter with the same weights and phase factors. When expressed in terms of local atomic orbitals, this superposition has the form

\[
\sum_\mathbf{R} e^{i\mathbf{qR}} a_1^\dagger(\mathbf{R}) a_0(\mathbf{R}),
\]  

(11)

where \(a_0^\dagger(\mathbf{R})\) creates the electron in the orbital state 0(1) at the crystal site \(\mathbf{R}\). The expression \(\mathbb{I}\) is just the standard description of Frenkel exciton as the running wave of the atomic electron-hole pairs \[\mathbb{I}\].

Actually, the zero band-width case \(\mathbb{I}\) presents a common asymptotic limit of two
incompatible physical regimes. On the one side, one may have the on-site electron-hole repulsion \( U_{0101} \) which is much larger than the band-widths \( t_{0,1} \). This binds the electron and the hole into the pair at a given site. The corrections to eqs. (9,10) and (11) are then of the order of \( t_{0,1}^2/U_{0101} \) [5]. Alternatively, in our weak coupling regime, \( U_{0101} \) is small with respect to the band-widths, which are in turn assumed to be small with respect to the band separation \( E_g \). The corresponding corrections to eqs. (9,10) and (11) are now of the order of \( t_{0,1}^2/E_g \).

From the terminological side, the name of Frenkel exciton is nowadays usually attached to the regime \( t_{0,1}/U_{0101} < 1 \) [3]. Since we are considering here primarily the limit \( t_{0,1}/E_g < 1 \) at \( U_{0101} \) small, we shall use for the collective mode (9,10) the term ”dipolar excitation” all throughout the present paper. We note that the above interpretation does not agree with that established in numerous early works (e.g. [7, 8, 9]) on collective excitations in insulators, and adopted by Giaquinta et al [10] whose results for a two-band insulator coincide with our expressions (9,10). In Ref. [10] the longitudinal mode (9) is identified as the renormalized ”total” plasmon (i.e. \( \omega_p^2 = E_g^2 + \frac{2}{3} \omega_{pl,t}^2 \)), due to the simple relation between the frequency of the ”total” plasmon and the dipolar matrix element, \( \omega_{pl,t}^2 = 8\pi \mu^2 n_e E_g/a^3 \), to which the f-sum rule reduces for an insulating two-band system. This coincidence however does not hold for multiband systems, in which the ”total” plasmon contains all interband contributions, while the number of longitudinal collective modes increases with the order of the TB dielectric matrix [4]. We emphasize that \( \omega_{pl,t} \) does not represent the frequency of a collective excitation, but appears as a parameter which characterizes the asymptotic limit \( \omega \to \infty \) for \( \epsilon_m \). On the other hand, the longitudinal exciton from Refs. [7-10] is associated with the exchange and correlation
(beyond RPA) contributions to the screening, and is expected to be inside or below the interband electron-hole continuum $[10]$. These are precisely the properties of Wannier excitons which, as argued in Sect. 5, have to be distinguished from the present dipolar collective modes.

### 3 Damping of interband collective modes

In this Section we discuss the interband Landau damping of collective modes, following only the more complex case of dipolar excitations $[9, 10]$. In the limit of zero bandwidths the inter-band electron-hole excitations coincide with the inter-atomic transitions between two orbitals, and are represented by the horizontal line $\omega = E_g$ in Fig. 1. For finite bandwidths this line is replaced by a region of finite width $[8]$, with the boundaries $E_{m(M)}(q)$ given by

$$E_{m(M)}(q) = \min_{k}(\max_{k} [E_1(k + q) - E_0(k)])$$

(12)

The illustration of the interband electron-hole continuum for particular TB band dispersions is shown in the Appendix. The collective modes are damped in the region $E_m(q) < \omega < E_M(q)$. The finite bandwidths which enter into eq.(12) however also alter the dispersion curves of the collective modes through eq.(7). In particular, the longitudinal and transverse edges of the band of collective modes for a given $|q|$ cease to exist when (and if) the renormalised energies $\omega_L(q)$ and $\omega_T(q)$ cross the lines $E_M(q)$ and $E_m(q)$ respectively. Note that the interband electron-hole continuum always covers a part of the "interior" of the band of collective excitations from Fig.1, i.e. there is a finite range of orientations of the wave vector $q$ for which the collective dipolar modes
are damped. This is a property of the particular band model B of paper I.

The question which arises now is, whether and under which conditions the dispersion curves of the pure longitudinal and/or transverse collective modes enter into the interband electron-hole continuum. The further discussion is mostly limited to the experimentally interesting range \( q \to 0 \). Note that Figs. 1 and 3 suggest that modes from this range are usually the last which become damped as the bandwidths increase.

At first, it follows directly from eq. (7) that the crossing of lines \( \omega_L(q) \) and \( E_M(q) \) is impossible if the sum on the left-hand side diverges after replacing \( \omega(q) \) by \( E_M(q) \). The same is true for the lines \( \omega_T(q) \) and \( E_m(q) \), with the replacement \( \omega(q) \to E_m(q) \). If this condition holds for all values of \( q \) (with \( q \parallel \mu_0 \) and \( q \perp \mu_0 \) respectively), the whole line \( \omega_L(T)(q) \) will remain above (below) the boundary \( E_M(m)(q) \) of the interband electron-hole continuum. Obviously, such argument can be also straightforwardly extended to collective modes within the band of Fig. 1.

The integral in eq. (7) diverges if the locus of zeros of the equation

\[
E_{M(m)}(q) - E_1(k + q) + E_0(k) = 0
\]  

(13)

in the \( d \)-dimensional \( k \)-space has the dimension \( d - 1 \). For lower dimensions of the locus the \( k \)-integration is regular, so that the crossing may occur for some finite value of \( |q| \). As the width of the interband electron-hole continuum increases the crossing point usually moves towards smaller values of \( |q| \), and eventually reaches the point \( q = 0 \).

The properties of electron-hole locus in the \( k \)-space are directly linked to the details in the band dispersions \( E_{0(1)}(k) \), in particular to their dimension. E.g., for strictly one-dimensional bands the crossing between the dipolar collective modes and the electron-
hole lines is in principle not possible for any value of the wave vector. Analogously, the plasmon dispersion curve $\omega_{pl}(q)$ does not enter into the intraband electron-hole continuum $[11, 12]$. For crystals with two- and three-dimensional electron bands the locus from eq.(13) is usually a point, so that the crossing between the collective modes and the electron-hole lines is not forbidden.

4 Hybridization of intraband and interband collective modes

In the two-band conductors with finite interband dipolar transitions the condition (3) is not fulfilled, and the off-diagonal intraband-interband elements of the matrix (1) enter into the equation $\epsilon_m = 0$. In order to determine analytically collective modes which follow from this equation, let us consider the limit $q \rightarrow 0$ and assume, like in Sec.2, that the widths of the valence and conducting band are small in comparison with $E_g$ and $|E_g^2 - \Omega^2|^{1/2}$, where $\Omega$ stands for the frequencies of collective modes, yet to be determined. We also use the effective mass form of the valence ($l = 0$) band dispersion. The intraband and interband polarization diagrams are then given by eqs.(I.33) and (I.34) respectively, and the equation $\text{Re} \epsilon_m(q, \omega) = 0$ reduces to

$$
\Omega^4 - \left[ E_g^2 + \frac{n_e q^2}{m^*} V_{0000}(q) + 2 n_e E_g V_{0011}(q) \right] \Omega^2 + \\
+ E_g \left[ V_{0000}(q)V_{0011}(q) + V_{0001}(q) \right] \frac{2n_e q^2}{m^*} + \frac{n_e E_g^2}{m^*} V_{0000}(q) q^2 = 0. \quad (14)
$$

where $m^*$ is the effective band mass and $n_e$ is, as before, the number of valence band electrons per site. Here it is taken into account that, in accordance with the previous assumption $t_i \ll E_g$, the regions of intraband and interband electron-hole continua do
not overlap, i.e. that
\[
\text{Im}[\Pi_{00}(q, \omega)] \cdot \text{Im}[\Pi_{01}(q, \omega) + \Pi_{10}(q, \omega)] = 0. \tag{15}
\]

For wave vectors perpendicular to \( \mu \) \((q_x = 0)\) the matrix element \( V_{0001} \) vanishes, so that the longitudinal intraband plasmons are not coupled to the transverse dipolar modes (10). However, the dipolar modes which propagate in other directions, do couple with plasmons. This coupling is strongest in the longitudinal direction \( q = q_x \to 0, q_y = q_z = 0 \) which we consider further on. It is important to note that in this limit the square of the monopole-dipole interaction (2) and the product \( V_{0000}(q) V_{0011}(q) \) [see eqs. (I.28), (I.31) and (I.32)] have the same structure and the numerical factors of the same order, so that the monopole-dipole interaction cannot be treated perturbatively.

After inserting the bare Coulomb matrix elements, the solutions of eq.(14) read
\[
\Omega^2_\pm = \frac{1}{2} \left\{ \omega_L^2 + \omega_{pl}^2 \pm \left[ (\omega_{pl}^2 + \omega_L^2)^2 - 4 \omega_T^2 \omega_{pl}^2 \right]^{1/2} \right\} \tag{16}
\]
Here \( \omega_{pl} = \sqrt{4\pi n e^2/(m^* a^3)} \) is the frequency of the bare intraband plasmon. Let us discuss the result (16) by distinguishing between the two opposite limits, \( \omega_{pl}^2 \gg \omega_L^2 \) and \( \omega_{pl}^2 \ll \omega_L^2 \). For \( \omega_{pl}^2 \gg \omega_L^2 \) the renormalized frequencies reduce to
\[
\Omega_+^2 = \tilde{\omega}_{pl}^2 \approx \omega_{pl}^2 \left[ 1 + \frac{\omega_T^2}{\omega_{pl}^2} \right] = \omega_{pl}^2 + \frac{4\pi n e \alpha}{a^3} \left[ 1 + \frac{\omega_T^2}{\omega_{pl}^2} \right] E_g^2 \tag{17}
\]
and
\[
\Omega_-^2 = \tilde{\omega}_L^2 \approx \omega_L^2 \left[ 1 - \frac{\omega_T^2}{\omega_{pl}^2} \right], \tag{18}
\]
where \( \alpha = 2\mu^2/E_g \) is the molecular polarizability. It follows from eq.(17) that the monopole-dipole interaction increases the effective monopole-monopole interaction, causing an increase of the plasmon frequency \( \tilde{\omega}_{pl} \). The factor \( 4\pi n e \alpha/a^3[1+\omega_T^2/\omega_{pl}^2] \) is usually
of the order of unity in metals. The renormalized plasmon frequency \( (17) \) can thus be identified as the so-called interband plasmon frequency \( (13, 14) \), usually taken as equal to \( \sqrt{\omega_{pl}^2 + E_g^2} \). On the other hand, the renormalized longitudinal dipolar mode \( (18) \) is shifted below the transverse one. Furthermore, the band of dipolar modes becomes extremely narrow, i.e. \( (\omega_T^2 - \omega_L^2)/(\omega_L^2 - \omega_T^2) \approx \omega_T^2/\omega_{pl}^2 \ll 1 \). Since \( \omega_T \leq \min(E_m, E_M) \), this band, if not damped, lies below the interband electron-hole continuum.

In the opposite limit \( \omega_L^2 \gg \omega_{pl}^2 \) the frequencies \( \Omega_{\pm} \) reduce to

\[
\Omega_{\pm}^2 = \tilde{\omega}_L^2 = \omega_L^2 \left[ 1 + \frac{\omega_T^2}{\omega_{pl}^2} \frac{\omega_{pl}^2}{\omega_L^2} \right] (\omega_L^2 - \omega_T^2)
\]

and

\[
\Omega_-^2 = \tilde{\omega}_{pl}^2 = \omega_{pl}^2 \left[ 1 - \frac{\omega_{pl}^2}{\omega_L^2} \right].
\]

Within this limit one may distinguish between the two opposite cases, related to the ratio \( E_g^2/(\omega_L^2 - \omega_T^2) \). For \( E_g^2/(\omega_L^2 - \omega_T^2) \gg 1 \) the renormalized plasmon frequency \( (20) \) is close to its bare value \( \omega_{pl} \), due to \( \omega_L/\omega_T \approx 1 \). The opposite limit represents the regime in which \( \omega_T \) may be critically softened, or even become unstable \( (13) \). As is seen from eq.(20), \( \tilde{\omega}_{pl}^2 \) is proportional to, and smaller than \( \omega_T^2 \). Note that in both cases the shift of the pure longitudinal dipolar mode, given by eq.(19), is small and positive \( (\tilde{\omega}_L^2 > \omega_L^2) \).

The most important common outcome of the above discussion is the proportionality of the frequencies of the lower longitudinal mode \( (\Omega_-) \) and the transverse dipolar mode \( (\omega_T) \). Moreover, it follows from eq.(16) that in the regime of critical softening the instabilities \( \omega_T \to 0 \) and \( \Omega_- \to 0 \) proceed simultaneously, irrespectively of the relation between \( \omega_{pl} \) and \( \omega_L \), with the ratio

\[
(\omega_T/\Omega_-)^2 \simeq 1 + (\omega_L/\omega_{pl})^2 > 1.
\]

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Thus, after taking properly into account the coupling of the longitudinal dipolar mode and intraband plasmon, one obtains a common critical behavior for the lower longitudinal mode and the transverse mode, although the latter is not coupled to the plasmon. This, to some extent unexpected, result brings a new insight into the problem of the instability of dipolar collective modes. We note that it is not restricted to the present two band structure [4].

The finite monopole-dipole interaction also modifies the conclusions of Sect.3, concerning the damping of the renormalized collective longitudinal modes due to the electron-hole transitions. Let us keep the assumption that the intraband and interband electron-hole continua do not overlap, as expressed by eq.(15). The imaginary part of the microscopic dielectric function $\epsilon_m$ reads

$$\text{Im } \epsilon_m = -\text{Im } \Pi_{00} V_{0000} [1 - V_{0011} \text{Re} (\Pi_{01} + \Pi_{10})] - \text{Im} (\Pi_{01} + \Pi_{10}) V_{0011} \times$$

$$\times (1 - V_{0000} \text{Re } \Pi_{00}) + V_{0011}^2 [\text{Re } \Pi_{00} \text{Im} (\Pi_{01} + \Pi_{10}) + \text{Im } \Pi_{00} \text{Re} (\Pi_{01} + \Pi_{10})]. \quad (22)$$

Obviously, without the monopole-dipole interaction the intraband plasmons are not damped inside the $(q, \omega)$ region covered by the interband electron-hole continuum. The analogous conclusion holds for the band of dipolar modes, even if it has a finite overlap with the intraband valence electron-hole continuum.

The inclusion of $V_{0001}(q)$ does not change the boundaries of the electron-hole continua. However, it brings into eq.(22) a new term, which is finite at all crossings of the electron-hole continua and the dispersion curves of longitudinal collective modes. Consequently, the longitudinal collective modes become damped by entering into both intraband and interband electron-hole continua. In particular, this means that the propagation of the
renormalized long wavelength plasmon [eqs. (17) or (20)] is damped within the interband electron-hole continua. On the other hand, collective modes with finite frequencies at \( q \to 0 \) are not damped by the intraband electron-hole continuum, since the latter has an upper boundary, which starts at the origin of the \((q, \omega)\) space. For finite values of \(|q|\) this argument does not hold, and one has to establish criteria for the absence of damping of longitudinal collective excitations along the lines discussed in Sect.3. Note that the pure transverse modes exist within the intraband electron-hole continuum, since they do not couple to this continuum.

The frequencies (16) are the zeros of the macroscopic dielectric function [see eq. (I.48)] which is in the absence of the electron-hole damping given by

\[
\epsilon_M(q_\parallel, \omega) = \frac{(\omega^2 - \Omega_-^2)(\omega^2 - \Omega_+^2)}{\omega^2(\omega^2 - \omega_T^2)}.
\] (23)

This result includes the well-known special cases of the one band conductor and the two-band insulator. In the former case \( \Omega_- = \omega_{pl} \) and \( \Omega_+ \approx \omega_T \to \infty \), so that \( \epsilon_M \) reduces to the RPA expression for the plasmon edge. In the latter case one recovers, after \( \omega_{pl} = \Omega_- = 0 \) and \( \Omega_+ = \omega_L \), the Lorentz-Lorenz expression \( \epsilon_M(q_\parallel, \omega) = (\omega^2 - \omega_L^2)/(\omega^2 - \omega_T^2) \), i.e. the Lyddane-Sachs-Teller relation \( \epsilon_M(\omega = 0)/\epsilon_M(\omega = \infty) = \omega_L^2/\omega_T^2 \).

Eq. (23) and Fig. 2 also clearly show how the simultaneous instability of the transverse dipolar mode and the lower longitudinal mode in a conductor is manifested in the macroscopic dielectric properties. This instability is preceded by the shifts of both, the zero \( (\omega = \Omega_-) \) and the pole \( (\omega = \omega_T) \) of \( \epsilon_M(q_\parallel, \omega) \), towards the origin. Note that for the insulator, the zero is at the origin all the time \( [\omega = \Omega_- = 0] \), compensating the metallic \( \omega^{-2} \) divergence in the expression (23). The instability of the transverse dipolar mode is
then manifested solely as a shift of the pole at $\omega = \omega_T$ towards the origin.

Eq. (23) and Fig. 2 apply also, with a somewhat modified content, to the isotropic crystal with three degenerate $p$-bands [see Appendix B in paper I]. The expression (23) is then valid for any orientation of $\mathbf{q}$, with the frequencies $\omega_T$ and $\Omega_\pm$ being the true branches of collective modes, in contrast to the above anisotropic case in which they represent the edges of a continuous band as it is specified in Sect. 2.

5 Conclusion

The present analysis, although based on the simplest nontrivial example of a multiband system, leads to some results of broad significance. First, one may distinguish between purely intra-band and inter-band collective modes only in the long wavelength limit. Even then, this is justified only in the particular cases of an insulator and a conductor with forbidden dipolar transitions. Furthermore, in the zero bandwidth limit of the former case the dispersion and the coherent electron-hole distribution of dipolar interband modes are just those of Frenkel excitons. We stress again that our identification is at variance with that from earlier literature [7-10], and that Frenkel excitons are not associated only with strong coupling systems [1], but are also an asymptotic limit within the RPA approach appropriate for the weak coupling regime, provided that the conditions specified in Sect. 2 are fulfilled.

In the case of dipolar conductors there is a finite "off-diagonal" coupling between the intra-band and inter-band polarization processes. It is explicitly shown that in the long wavelength limit this coupling originates from the diverging long-ranged monopole-
dipole interaction which cannot be treated perturbatively. As a consequence, the collective modes are hybridized from the intra-band (plasmon) and inter-band (dipolar) oscillations. The most interesting property of this hybridization is the proportionality of the frequencies of the lower longitudinal mode and the (non-hybridized) transverse inter-band mode when both are small. In other words, the corresponding dielectric instability in conductors proceeds by the simultaneous softening of two collective modes. The search for systems in which phenomenon of this kind takes place would be very desirable.

It should be pointed out that these results, as well as those regarding the Landau damping of the collective modes due to finite regions of incoherent electron-hole continua in the \((k, \omega)\) plane, follow from the TB microscopic dielectric function \(\Pi\) which includes properly the most relevant ingredients of the interactions and symmetries of a multiband system, and remains transparent to an analytical approach. They could not be obtained even approximately from the dielectric function with the intra-band and inter-band polarizabilities entering additively, which is the usual form given in textbooks \[3\].

The present work deals with the simple (i.e. electron-hole) version of the RPA, formulated in paper I. It can be however extended to the generalized RPA which also includes exchange (ladder) contributions \[15, 16\] to the polarization diagram. Postponing the full account of this calculation within the present TB approach for a separate paper, let us mention here that the spectrum of excitations then acquires a qualitatively new feature, namely the bottom of the interband electron-hole continuum (or continua for more than two bands) exhibits a discrete structure. The localised levels, defined by poles of the imaginary part of the dielectric function, represent the bound states of interband
electron-hole excitations, i.e. the Wannier excitons. On the other hand, the dipolar modes from Sects. 2 and 4 are coherent superpositions of electron-hole pairs, specified by eq. (1). Hence those excitations and Wannier excitons are two qualitatively different types of interband excitations. The former are collective bosonic dipolar fluctuations, while the latter are the electron-hole states which are localized in space and may propagate with a finite velocity. There is no a priori reason, at least in the weak coupling regime, against the coexistence of those excitations, as far as they do not overlap in the \((q, \omega)\) plane. In the narrow region of overlap the calculation of \(\varepsilon_m(q, \omega)\) would have to be extended to diagrams beyond the exchange RPA. In that respect the discussion in Sect. 3 may be applied only to the continuous part above the discrete Wannier structure in the region of interband electron-hole excitations. Finally, additional complications occur in low dimensional systems.

### Appendix

In order to illustrate the discussion of Sect. 3 on the crossing between the interband collective modes and the boundaries of interband electron-hole continuum, we consider here two cubic TB bands with bandwidths \(t_0\) and \(t_1\) and assume that the lower (valence) band is either almost empty (A.I) or full (A.II).

#### A.I Nearly empty valence band

In this case the band dispersions can be approximated by the effective mass form \(E_l(k) = E_l - (6 - k^2a^2)t_l\) with \(l = 0, 1\) and \(k = |k|\). The corresponding electron-hole continuum
is sketched in Fig.3. The expression (7) then reduces in the limit \( q \to 0 \) to
\[
\sqrt{E_m - \omega} \arctan \frac{\sqrt{E}}{\sqrt{E_m - \omega}} + \sqrt{E_m + \omega} \arctan \frac{\sqrt{E}}{\sqrt{E_m + \omega}} = 2\sqrt{E} + \frac{E^{3/2}}{3n_e V_{0011}},
\]
with \( V_{0011} \equiv V_{0011}(q \to 0) \), \( E_m \equiv E_g - 6(t_1 - t_0) \), \( \mathcal{E} \equiv (t_1 - t_0)k_F^2a^2 \) and \( k_F = (3\pi^2n_eN/L^3)^{1/3} \). Eq.(A.1) implicitly determines the renormalization of the dipolar collective modes due to finite widths of electronic bands. In Fig.4 we show the dependence of the limiting frequencies \( \omega_T \) and \( \omega_L \) and of the boundaries of the electron-hole continuum \( E_m \) and \( E_M \) on the difference of band-widths \( \mathcal{E} \). Evidently, for any finite \( \mathcal{E} \) a part of the excitation spectrum will be forbidden. In order to organize the discussion, it is instructive to distinguish four ranges.

(i) \( \mathcal{E} > 0, \ 0 < \omega < E_m \). The left-hand side of eq.(A.1) is now bounded between 0 and \( 2\sqrt{E} \). The pure transverse dipolar modes exist for \( \mathcal{E} < \mathcal{E}_{crT} \) where \( \mathcal{E}_{crT} \) is the solution of the equation
\[
\sqrt{2E_m} \arctan \frac{\sqrt{\mathcal{E}_{crT}}}{\sqrt{2E_m}} = 2\sqrt{\mathcal{E}_{crT}} + \frac{\mathcal{E}_{crT}^{3/2}}{3n_e[U_{0011} - 4\pi\mu^2/(3a^3)]}.
\]
At \( \mathcal{E} = \mathcal{E}_{crT} \) the frequency of the pure transverse mode becomes equal to the lower boundary of the electron-hole continuum. Putting \( \mathcal{E}_{crT} \ll E_m \) in eq.(A.2), in agreement with the assumption that the valence band is nearly empty, one gets \( \mathcal{E}_{crT} \approx 4\pi n_e \mu^2/a^3 - 3n_eU_{0011} \). \( \mathcal{E}_{crT} \) is well defined as far as \( \mathcal{E}_{crT}/(k_F^2a^2) < [E_g - 2n_eU_{0011} - 8n_e\pi\mu^2/(3a^3)]/6 \), since only then the pure transverse mode disappears at a finite frequency. Otherwise one would have an instability of the pure transverse mode \( (\omega_T \to 0) \) at the value of \( \mathcal{E}_{crT}/(k_F^2a^2) \) equal to the right-hand side of this inequality. In considering the Frenkel excitons \([5, 6, 17]\) it is usually assumed that the above condition is fulfilled. The excitonic instability has been however invoked recently in the context of excitonic mechanism for
the superconductivity in high \( T_c \) systems \[3\].

(ii) \( \mathcal{E} > 0, \omega > E_M = E_m + \mathcal{E} \). The left-hand side of eq.\((A.1)\) is now given by
\[
\sqrt{\omega - E_m} \arctan(\sqrt{\mathcal{E}}/\sqrt{\omega - E_m}) + \sqrt{E_m + \omega} \arctan(\sqrt{\mathcal{E}}/\sqrt{E_m + \omega}).
\]
This function tends to \( 2\sqrt{\mathcal{E}} \) for \( \omega \to \infty \), and to infinity for \( \omega \) approaching the upper boundary of electron-hole continuum \( E_M \). The pure longitudinal mode remains outside the electron-hole continuum, and therefore does not disappear in the whole region \( \mathcal{E} > 0 \). This result is the consequence of the fact that the Fermi surface coincides with the surface of the poles in the integral of eq.(7).

(iii) \( \mathcal{E} < 0, 0 < \omega < E_M \). Now \( E_m \) and \( E_M \) are the upper and lower boundaries of the electron-hole continuum respectively. The function at the left-hand side of eq.\((A.1)\) is given by
\[
\sqrt{E_m - \omega} \arctan(\sqrt{|\mathcal{E}|}/\sqrt{E_m - \omega}) + \sqrt{E_m + \omega} \arctan(\sqrt{|\mathcal{E}|}/\sqrt{E_m + \omega}).
\]
The arguments analogous to those in (ii) lead to the conclusion that the pure transverse mode exists in the whole range \( \mathcal{E} < 0 \), approaching asymptotically the lower boundary of the electron-hole continuum, as shown in Fig.4.

(iv) \( \mathcal{E} < 0, \omega > E_m \). The function at the left-hand side of eq.\((A.1)\) is now given by
\[
\sqrt{\omega - E_m} \arctan(\sqrt{|\mathcal{E}|}/\sqrt{\omega - E_m}) + \sqrt{E_m + \omega} \arctan(\sqrt{|\mathcal{E}|}/\sqrt{E_m + \omega}).
\]
The frequency of the pure longitudinal mode \( \omega_L \) touches the upper boundary of electron-hole continuum at the frequency \( \omega = E_m \), i.e. for \( \mathcal{E}_{crL} \) determined as the zero of the equation
\[
\sqrt{2E_m \arctan(\sqrt{|\mathcal{E}_{crL}|}/\sqrt{2E_m})} = 2\sqrt{|\mathcal{E}_{crL}|} - \frac{|\mathcal{E}_{crL}|^{3/2}}{3n_e[U_{0011} + 8\pi\mu^2/(3a^3)]}. \tag{A.3}
\]
Again, taking into account that \( E_m \gg |\mathcal{E}_{crL}| \), one gets \(|\mathcal{E}_{crL}| \approx 8\pi n_e \mu^2/a^3 + 3n_e U_{0011} \).

Note that \(|\mathcal{E}_{crL}| \geq 2\mathcal{E}_{crT} + 9n_e U_{0011} \), i.e. on the absolute scale of the bandwidth broadening, the transverse part of the band of collective modes disappears before the
longitudinal one, provided that $U_{0011} > -\mathcal{E}_{crT}/(9n_e)$.

A.II Full (and nearly full) valence band

When the lower band is full eq.(7) for $q = 0$ reduces to

$$
\frac{4}{N} \left( \frac{L}{2\pi} \right)^3 V_{0011}(q) E_g \int_{-\pi/a}^{\pi/a} \prod_{i=1}^3 dk_i \times
$$

$$
\times \frac{\omega^2 - E_g^2 + 4\mathcal{E}^2(\sum_{i=1}^3 \cos k_i a)^2}{(\omega^2 - E_g^2 - 4\mathcal{E}^2(\sum_{i=1}^3 \cos k_i a)^2)^2 - 16 E_g^2 \mathcal{E}^2(\sum_{i=1}^3 \cos k_i a)^2} = 1. \quad (A.4)
$$

The energies of the interband electron-hole transitions with $q = 0$ lie between $E_m \equiv E_g - 6\mathcal{E}$ and $E_M = E_g + 6\mathcal{E}$, where now $\mathcal{E} = t_1 - t_0$. From eq.(A.4) it follows that for $E_g \gg 6|\mathcal{E}|$ the pure transverse mode is damped for $|\mathcal{E}| > |\mathcal{E}_{crT}| = 3I[4\pi \mu^2/(3a^3) - U_{0011}]/(8\pi^3)$, where

$$
I = \int_{-\pi/a}^{\pi/a} \prod_{i=1}^3 dx_i \frac{1}{9 - (\sum_{i=1}^3 \cos x_i)^2}. \quad (A.5)
$$

The numerical calculation gives $I = 41.784$. Due to the reduction of the Fermi surface to the corners of the first Brillouin zone the pure transverse mode can disappear for both $\mathcal{E} > 0$ and $\mathcal{E} < 0$. Note also that this mode becomes unstable ($\omega_T = 0$) for $|\mathcal{E}| = E_g \sqrt{E_g/[8\pi \mu^2/(3a^3) - 2U_{0011}]} - 2$.

The analogous analysis performed for pure longitudinal mode shows that it becomes damped for both signs of the difference $\mathcal{E}$ provided that $|\mathcal{E}| > |\mathcal{E}_{crL}| = 3I[8\pi \mu^2/(3a^3) + U_{0011}]/(8\pi^3)$. Note that $|\mathcal{E}_{crL}| \approx 2|\mathcal{E}_{crT}| + 9I/(8\pi^3)U_{0011}$, i.e. the pure transverse mode disappears before the pure longitudinal one as the difference increases, provided that $U_{0011} > -\mathcal{E}_{crT}8\pi^3/(9I)$. The frequencies of the pure longitudinal and transverse modes as well as the lines $E_m$ and $E_M$ are depicted in the Fig.5.
For the nearly full valence band \( k_F = \sqrt{3} \pi/a - k_{h_F}, \ k_{h_F} \ll \sqrt{3} \pi/a \) the results for \( \mathcal{E}_{crT} \) and \( \mathcal{E}_{crL} \) are modified in a following way. If \( \mathcal{E} > 0 \) (here again \( \mathcal{E} = t_1 - t_0 \)), the pure transverse mode with \( \mathbf{q} = 0 \) reaches the value \( E_m \) for \( \mathcal{E}_{crT} = 3I[4\pi\mu^2/(3a^3) - U_{0011}][1 - 2\pi^3n_h/(9I)]/(8\pi^3) \), where \( n_h = k_{h_F}^3L^3/(3\pi^2N) \). Due to the finite Fermi surface which is the locus of electron-hole poles in eq (7), the frequency of the longitudinal mode remains above the value \( E_M(\mathbf{q} = 0) \).

For \( \mathcal{E} < 0 \) one has the opposite situation in which the pure longitudinal mode disappears for \( |\mathcal{E}_{crL}| = 3I[8\pi\mu^2/(3a^3) + U_{0011}][1 - 2\pi^3n_h/(9I)]/(8\pi^3) \), while the pure transverse mode remains below the electron-hole continuum.
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Figure captions

Fig.1. The band of collective dipolar excitations for $16\pi n_e\mu^2/(3a^2E_g) = 0.82$ and $U_{0011} = 0$.

Fig.2. The schematic frequency dependence of the macroscopic dielectric function for the (a) two band metal, (b) one band metal and (c) insulator.

Fig.3. The region of interband electron-hole excitations for the band dispersion specified in front of eq.(A.1).

Fig.4. The frequencies of collective dipolar excitations for $q=0$ and the electron-hole continuum vs bandwidth difference $\mathcal{E} \equiv (t_1 - t_0)k_F^2a^2$ for a nearly empty valence band and $U_{0011} = 0$. Note that $|\mathcal{E}_{crL}| \neq 2|\mathcal{E}_{crT}|$ since the condition $E_m \gg \mathcal{E}$ is not fulfilled.

Fig.5. The frequencies of dipolar excitations and the electron-hole continuum vs bandwidth difference $\mathcal{E} \equiv t_1 - t_0$ for a full valence band and $U_{0011} = 0$. Note that $|\mathcal{E}_{crL}| \neq 2|\mathcal{E}_{crT}|$ since the condition $E_g \gg \mathcal{E}$ is not fulfilled.
Fig. 1
Fig. 2

\[ \varepsilon_M(\omega) \]

(a) 

(b) 

\[ \omega_{pl} \]

(c) 

\[ \omega_T, \omega_L \]

\[ \Omega_-, \Omega_+ \]
Fig. 3
Fig. 4
Fig. 5

![Graph showing fields and eigenfrequencies](image-url)