

# Wavevector-dependent optical properties from wavevector-independent proper conductivity tensor

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**Abstract.** We discuss the calculation of the refractive index by means of the *ab initio* scalar dielectric function and point out its inherent limitations. To overcome these, we start from the recently proposed fundamental, microscopic wave equation in materials in terms of the frequency- and wavevector-dependent dielectric tensor, and investigate under which conditions the standard treatment can be justified. Thereby, we address the question of neglecting the wavelength dependence of microscopic response functions. Furthermore, we analyze in how far the fundamental, microscopic wave equation is equivalent to the standard wave equation used in *theoretical optics*. In particular, we clarify the relation of the “effective” dielectric tensor used there to the microscopic dielectric tensor defined in *ab initio physics*.

## 1 Introduction

More often than not, optical materials properties are accessed in *ab initio materials physics* via the standard relation between the macroscopic dielectric function  $\varepsilon(\omega)$  and the refractive index  $n(\omega)$  given by<sup>1</sup> (see Refs. [2, Eq. (8.33)], [3, Eq. (2.17)], [4, Eqs. (18.26)], [5, Eq. (6.11)], [6, p. 534] or [7, Eq. (2.203)])

$$n^2(\omega) = \varepsilon(\omega). \quad (1)$$

The macroscopic dielectric function, for its part, is defined as the limit  $\mathbf{k} \rightarrow \mathbf{0}$  of the microscopic (frequency- and wavevector-dependent) dielectric function (see Refs. [2, Eqs. (8.25)], [3, Eq. (2.18)] or [7, Eq. (2.207)]):

$$\varepsilon(\omega) = \lim_{|\mathbf{k}| \rightarrow 0} \varepsilon(\mathbf{k}, \omega). \quad (2)$$

Importantly, the dielectric function used in these equations is typically calculated from the density response function, and hence it corresponds to the longitudinal part of the dielectric tensor (see Refs. [4, Eq. (18.23)], [7, Sect. 2.6.4], [8, Vol. 1, Sect. 4]). Although the standard treatment delivers sensible results for a huge variety of materials (see Refs. [9–15] for recent examples), it ultimately proves insufficient. To make this clear, let us summarize the main conceptual problems of this standard

treatment based on the *wavevector-independent* dielectric function:

- As a matter of principle, optical properties correspond to *transverse* (although not necessarily *purely* transverse) electromagnetic waves in a material, and hence they should not be deduced from a purely *longitudinal* response function (at least not in a naïve way).
- The standard relation for the refractive index, equation (1), is only valid in the limit  $\mathbf{k} \rightarrow \mathbf{0}$ , whereas light waves definitely have a non-vanishing wavevector, i.e.,  $\mathbf{k} \neq \mathbf{0}$ .
- In particular, for anisotropic media, the refractive index should at least depend on the *direction* of the wavevector  $\mathbf{k}$ . Correspondingly, the limit (2) actually depends on the direction (cf. Ref. [4, Eq. (18.22)] or discussion below).
- Moreover, for *birefringent* or *optically active* materials, there are *polarization-dependent* refractive indices. Accordingly, the joint determination of polarization vectors and refractive indices corresponding to a given wavevector obviously requires a tensorial equation. By contrast, equation (1) yields at best one refractive index, whose polarization vector cannot – even in principle – be defined for vanishing wavevectors.

We particularly stress that the shortcomings of the standard relation between the dielectric function and the refractive index are not discovered by the authors of this article but are in fact well-known and acknowledged in the theoretical literature, where the validity of equation (1)

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<sup>1</sup> Here, as in reference [1], we suppress the subscript “r” of the relative dielectric function and simply write  $\varepsilon \equiv \varepsilon_r$ .

is typically restricted to cubic crystals (see e.g. Refs. [2, p. 32], [3, p. 294], [4, p. 432]). This does, however, not stop people from applying it to other systems as well (see Refs. [16–21]).

Befittingly, a recent, meritorious article by Sangalli et al. [22] has drawn attention to the fact that optical properties can also be calculated from the *wavevector-dependent current response tensor*. On the other hand, the above considerations make it clear that in general, the calculation of optical materials properties requires the wave- and the polarization vector to be taken into account, and this in turn requires a treatment based on the wavevector-dependent current response tensor (or equivalently, the dielectric tensor [1,23,24]). In fact, from the dielectric tensor – which naturally contains much more information than the *scalar* dielectric function – the density response function can be reconstructed by means of the Universal Response Relations [25–27], while the converse is not true (see Refs. [2,28–30]), i.e. the dielectric tensor cannot be reconstructed from the dielectric function, although this is sometimes claimed.

However, in the limit  $\mathbf{k} \rightarrow \mathbf{0}$ , such relations between different response functions cannot be evaluated due to their singular behavior (see, for example, Eq. (10) below, which relates the dielectric function to the density response function). Correspondingly, the authors of reference [22] have based their treatment on a full *ab initio* calculation of the wavevector-dependent response functions. While this is certainly the right approach in the most general case, the downside is that such wavevector-dependent calculations are in general extremely demanding, in particular if they are supposed to yield dispersion relations.

In this article, we resume this problem on a fundamental level to show that it is possible – at least in principle – to obtain wavevector-dependent optical properties without calculating wavevector-dependent response functions numerically, provided that the treatment is based on the assumption that the *proper conductivity tensor* is given in the optical limit. Fortunately, this is exactly what is common practice in numerical studies using state of the art electronic structure codes like e.g. Elk task 121 [31]. Thus, our considerations are predicated on the presupposition that the conductivity tensor has already been calculated using the appropriate *ab initio* methods, which for their part are not addressed in this article. Put differently, we are not concerned in this article with the improvement of the *ab initio* calculations, but consider the wavevector (or at least wavelength) independent conductivity tensor as an input typically provided for by some *ab initio* electronic structure code. We furthermore have to stress, in this context, that the condition of wavelength independence cannot simultaneously apply to all other response functions as well (see the discussion below).

This article is organized as follows: In Section 2, we assemble some general relations between wavevector-dependent response functions. In Section 3, we discuss the fundamental, microscopic wave equation in materials in terms of the proper conductivity tensor, and we investigate the conditions under which the treatment of optical materials properties by means of a scalar dielectric function can be justified. In particular, we define the

*effective dielectric tensor* in terms of the proper conductivity tensor, and we show that by rewriting the fundamental wave equation in terms of this effective dielectric tensor, the resulting equation agrees formally with the standard wave equation used in theoretical optics. The subsequent Section 4 is dedicated to an in-depth analysis of this wave equation and its solutions under the assumption of a wavelength-independent proper conductivity tensor. Finally, in Section 5 we reproduce some well-known facts from theoretical optics within our general formalism by assuming special forms for the effective dielectric tensor.

## 2 Response functions and wavevector-dependence

Our investigations are based on the *functional approach to electrodynamics of materials* [1,25–27,32–37], which is a paradigmatically new approach motivated by the common practice in modern *ab initio* materials science (see extensive discussion in Refs. [25–27]). In particular, the functional approach omits macroscopic averaging procedures and is hence intrinsically microscopic. Correspondingly, it replaces the traditional distinction of “free” and “bound” source terms in the Maxwell equations with the modern distinction of *external* and *internal* (or *induced*) fields as used in first principles theory. Consequently, the traditional “constitutive” equations assumed to characterize a material can be identified with *response relations* mediating between external and induced quantities, which in turn paves the way for the application of the Kubo formalism (see e.g. Refs. [28, Sect. 3], [38, Sect. 6]), so conspicuously absent from the traditional textbook literature on electrodynamics in media. On the practical side, the functional approach to electrodynamics of materials is then primarily concerned with the deduction of interrelations between different electromagnetic response functions from purely electromagnetic considerations. Entre autres, this leads to a whole number of *Universal Response Relations*, which are entirely unknown in the traditional approach, while the *ab initio* community is at least partially familiar with them.

In fact, for purely classical electrodynamic reasons it turns out that the *current response tensor*  $\chi$  already contains the complete information about all linear, electromagnetic response properties. E.g., it is related to the conductivity tensor  $\sigma$  by means of a Universal Response Relation [25, Sect. 6] reading (see Refs. [7, Eqs. (2.177) and (2.198)], [28, Eq. (3.185)], [32], and for a gauge-independent derivation see Ref. [26, Sect. 3.2.3]):

$$\overleftrightarrow{\chi}(\mathbf{x}, \mathbf{x}'; t - t') = -\partial_t \overleftrightarrow{\sigma}(\mathbf{x}, \mathbf{x}'; t - t'). \quad (3)$$

The current response tensor for its part is the spatial part of the *fundamental response tensor* [29, Sect. 7.4],

$$\chi^\mu{}_\nu(x, x') = \frac{\delta j_{\text{ind}}^\mu(x)}{\delta A_{\text{ext}}^\nu(x')}, \quad (4)$$

where  $j^\mu = (c\rho, \mathbf{j})^T$  denotes the (induced) electromagnetic four current density, and  $A^\nu = (\varphi/c, \mathbf{A})^T$  the

(external) four-potential. As is evident from equation (3), microscopic response functions constitute in general entire tensor valued integral kernels (as opposed to sheer numbers). In particular, they are typically *non-local* and *inhomogeneous*. For the latter fact, there exist two fundamentally different reasons on the theoretical level. Firstly, real probes are finitely extended in space, thereby possessing a specific geometry. Consequently, their response functions cannot be spatially homogeneous already for that reason. In theoretical materials science, however, this aspect can be discarded because here we are concerned with purely material dependent (and hence geometry independent) response functions, which would – at least conceptually – correspond to probes filling out all of space homogeneously. Concretely, this means that the response functions have to be calculated in the *thermodynamic limit*. Even so, putting aside such heuristic model systems as the homogeneous electron gas, the microscopic response function is not homogeneous due to the atomic structure of the material. Rather, in the case of *crystalline* materials it turns out that the Fourier transform of a typical electromagnetic response function, say the density response function, is of the general form

$$\chi = \chi_{\mathbf{G}\mathbf{G}'}(\mathbf{k}, \omega), \tag{5}$$

where  $\mathbf{G}$  and  $\mathbf{G}'$  are reciprocal lattice vectors while  $\mathbf{k}$  is a vector in the first Brillouin zone. Applying this to fields whose Fourier transforms are supported in the first Brillouin zone anyway, we obtain the particularly simple response relation

$$\rho_{\text{ind}}(\mathbf{k}, \omega) = \chi_{\mathbf{0}\mathbf{0}}(\mathbf{k}, \omega)\varphi_{\text{ext}}(\mathbf{k}, \omega), \tag{6}$$

whose simplistic multiplicative structure translates into

$$\rho_{\text{ind}}(\mathbf{x}, t) = c \int dt' \int d\mathbf{x}' \chi_{\mathbf{0}\mathbf{0}}(\mathbf{x} - \mathbf{x}'; t - t')\varphi_{\text{ext}}(\mathbf{x}', t') \tag{7}$$

in the space-time domain. Hence, in this case the response function appears to be strictly homogeneous (i.e., it depends exclusively on the *difference* of the spatial variables  $\mathbf{x}$  and  $\mathbf{x}'$ ). Correspondingly, we call the transition  $\mathbf{G}, \mathbf{G}' \rightarrow \mathbf{0}$  in a response relation the *homogeneous limit*. As, on the other hand, the modulus of a wavevector in the first Brillouin zone roughly fulfills the approximate inequality  $|\mathbf{k}| \lesssim \pi/a$  with a typical lattice constant  $a$ , the said homogeneous limit corresponds to the condition that the involved wavelengths are larger than at least twice the lattice constant,  $2a \lesssim \lambda$ . This is of course a version of the famous Nyquist-Shannon sampling theorem, where the external field corresponds to the signal while the crystal plays the rôle of the sampler.

In practice, i.e. in optical experiments, the involved wavelengths are even much larger than typical lattice constants and hence it is highly intuitive that at such wavelengths the material – even if crystalline on the microscopic level – appears to be homogeneous on the macroscopic level. The *homogeneous limit* can therefore be identified with the traditional *transition to macroscopic fields*, where it has to be borne in mind though that in our case the fields do not have to be averaged but are

instead simply assumed to lie in the, say, optical range (which they certainly do in the case of those experiments which are our concern here). Instead of the fields, it is therefore now the response function which is to be subjected to such a macroscopic transition, which, however, is not implemented anymore by some complicated averaging procedure (as it was in the traditional approach), but by the evaluation at small wavevectors in the sense of  $\mathbf{G}, \mathbf{G}' \rightarrow \mathbf{0}$ .

Interestingly, in the said homogeneous limit, the fundamental response tensor is now of the following general form (see e.g. Refs. [2,25,29,30]):

$$\chi_{\nu}^{\mu}(\mathbf{k}, \omega) = \begin{pmatrix} -\frac{c^2}{\omega^2} \mathbf{k}^T \overset{\leftrightarrow}{\chi}(\mathbf{k}, \omega) \mathbf{k} & \frac{c}{\omega} \mathbf{k}^T \overset{\leftrightarrow}{\chi}(\mathbf{k}, \omega) \\ -\frac{c}{\omega} \overset{\leftrightarrow}{\chi}(\mathbf{k}, \omega) \mathbf{k} & \overset{\leftrightarrow}{\chi}(\mathbf{k}, \omega) \end{pmatrix}. \tag{8}$$

In particular, the *density response function*  $\chi$  can be calculated from the current response tensor as follows [28, Eq. (3.175)]:

$$\chi(\mathbf{k}, \omega) := \frac{\delta\rho_{\text{ind}}(\mathbf{k}, \omega)}{\delta\varphi_{\text{ext}}(\mathbf{k}, \omega)} = \frac{1}{c^2} \chi_{\mathbf{0}\mathbf{0}}^0(\mathbf{k}, \omega) = -\frac{\mathbf{k}^T \overset{\leftrightarrow}{\chi}(\mathbf{k}, \omega) \mathbf{k}}{\omega^2}. \tag{9}$$

We remark that the response relations (3)–(9) hold analogously for the respective *proper* response functions, which relate the induced quantities to the total (i.e., external plus induced) quantities (see Ref. [27, Sect. 2.3]). Furthermore, the *proper* density response function is related to the dielectric function by the equation [7, Eq. (2.172)]

$$\varepsilon(\mathbf{k}, \omega) = 1 - v(\mathbf{k})\tilde{\chi}(\mathbf{k}, \omega), \tag{10}$$

where  $v(\mathbf{k}) = 1/(\varepsilon_0|\mathbf{k}|^2)$  denotes the Coulomb interaction kernel in Fourier space. We stress again that the dielectric function in this equation actually coincides with the *longitudinal* part of the dielectric tensor (see Ref. [7, Sect. 2.6.4]) given by

$$\varepsilon(\mathbf{k}, \omega) \equiv \varepsilon_{\text{L}}(\mathbf{k}, \omega) = \frac{\mathbf{k}^T \overset{\leftrightarrow}{\varepsilon}(\mathbf{k}, \omega) \mathbf{k}}{|\mathbf{k}|^2}. \tag{11}$$

With this, equation (10) can be proven directly by applying the functional chain rule (see Refs. [25] and [35, Sect. 5.1]). More directly, it can be shown as follows: The relation of the dielectric tensor to the proper current response tensor reads [35, Eq. (3.41)]

$$\overset{\leftrightarrow}{\varepsilon}(\mathbf{k}, \omega) = \overset{\leftrightarrow}{\mathbb{1}} - \overset{\leftrightarrow}{D}_0(\mathbf{k}, \omega)\tilde{\chi}(\mathbf{k}, \omega), \tag{12}$$

where

$$\overset{\leftrightarrow}{D}_0(\mathbf{k}, \omega) = \mathbb{D}_0(\mathbf{k}, \omega) \left( \overset{\leftrightarrow}{\mathbb{1}} - \frac{c^2|\mathbf{k}|^2}{\omega^2} \overset{\leftrightarrow}{P}_{\text{L}} \right), \tag{13}$$

and

$$\mathbb{D}_0(\mathbf{k}, \omega) = \frac{c^2\mu_0}{-\omega^2 + c^2|\mathbf{k}|^2}. \tag{14}$$

We then get (skipping some straightforward algebra)

$$\varepsilon_L(\mathbf{k}, \omega) = \frac{\mathbf{k}^T \overset{\leftrightarrow}{\varepsilon}(\mathbf{k}, \omega) \mathbf{k}}{|\mathbf{k}|^2} \quad (15)$$

$$= 1 - \mathcal{D}_0(\mathbf{k}, \omega) \frac{\mathbf{k}^T}{|\mathbf{k}|^2} \left( \overset{\leftrightarrow}{1} - \frac{c^2 |\mathbf{k}|^2}{\omega^2} \overset{\leftrightarrow}{P}_L \right) \overset{\leftrightarrow}{\chi}(\mathbf{k}, \omega) \mathbf{k} \quad (16)$$

$$= 1 - \frac{c^2 \mu_0}{|\mathbf{k}|^2} \left( - \frac{\mathbf{k}^T \overset{\leftrightarrow}{\chi}(\mathbf{k}, \omega) \mathbf{k}}{\omega^2} \right) \quad (17)$$

$$= 1 - \frac{\tilde{\chi}(\mathbf{k}, \omega)}{\varepsilon_0 |\mathbf{k}|^2}, \quad (18)$$

which shows the assertion, equation (10). The importance of this proof lies in the fact that the usual arguments in favor of equation (10) are either based on the isotropic limit or operate exclusively with longitudinal fields. Correspondingly, it is a priori unclear whether the general definition of the dielectric function given in equation (11) lends itself to the standard relation (10).

Importantly, as stressed already, the microscopic current response tensor – or equivalently, the microscopic conductivity tensor – already contains the complete information about all linear electromagnetic response properties (this insight can be traced back at least to Ref. [39]; the fact as such has also been stressed recently in Ref. [23]; for a systematic derivation of all linear electromagnetic response functions in terms of the conductivity tensor see also Ref. [25, Sect. 5–6]). In particular, it is possible to reconstruct from the microscopic conductivity tensor the response with respect to strictly longitudinal perturbations. However, one has to emphasize again that the corresponding response relations are formulated in terms of response functions at finite wavevectors  $|\mathbf{k}| > 0$  (see Ref. [22]). Hence, these relations can in general not be evaluated naïvely in the limit  $\mathbf{k} \rightarrow \mathbf{0}$ , but require precise knowledge about the response functions in the vicinity of the origin and the direction of its approach.

Finally, we remark again that the response relations presented above hold in this form only for homogeneous materials, while they become more involved in the general (i.e., inhomogeneous) case [25,34]. Moreover, as opposed to the (longitudinal) dielectric function such quantities as the *transverse* dielectric function are actually meaningful only in the isotropic limit (see Refs. [33, App. D.1], [35, Sect. 5.1], [36, Sect. 2.1]).

### 3 Linear wave equations in materials

#### 3.1 Fundamental, microscopic wave equation

On a fundamental level, the most general, linear electromagnetic wave equation in materials – which requires only spatial homogeneity – reads as follows (see discussions in Refs. [1,27,35,37,40]):

$$\overset{\leftrightarrow}{\varepsilon}(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k}, \omega) = 0. \quad (19)$$

Here,  $\overset{\leftrightarrow}{\varepsilon}(\mathbf{k}, \omega)$  denotes the *ab initio* dielectric tensor, which is *defined* by the linear approximation (see Refs. [7, Eq. (2.140)], [41, Eq. (5.198) and (5.203)], and [42, Eq. (E.10)]),

$$\mathbf{E}_{\text{ext}}(\mathbf{k}, \omega) \stackrel{\text{def}}{=} \overset{\leftrightarrow}{\varepsilon}(\mathbf{k}, \omega) \mathbf{E}_{\text{tot}}(\mathbf{k}, \omega), \quad (20)$$

where we will set from now on  $\mathbf{E} \equiv \mathbf{E}_{\text{tot}}$ . Although the *ab initio* derivation of the fundamental wave equation (19) is somewhat complicated [27, Sect. 4.1], its ultimate meaning is just that the external field does not penetrate the material or, put differently, that the electromagnetic waves in the material correspond to the latter's *proper oscillations*.

In the case of an isotropic medium, the fundamental wave equation decouples into

$$\varepsilon_L(\mathbf{k}, \omega) \mathbf{E}_L(\mathbf{k}, \omega) = 0, \quad (21)$$

$$\varepsilon_T(\mathbf{k}, \omega) \mathbf{E}_T(\mathbf{k}, \omega) = 0, \quad (22)$$

which are equations formulated in terms of the *longitudinal* and *transverse* dielectric function. The resulting conditions for the existence of nontrivial solutions,

$$\varepsilon_L(\mathbf{k}, \omega_{\mathbf{k}L}) = 0, \quad (23)$$

$$\varepsilon_T(\mathbf{k}, \omega_{\mathbf{k}T}) = 0, \quad (24)$$

determine the respective *dispersion relations*,  $\omega_{\mathbf{k}L}$  and  $\omega_{\mathbf{k}T}$ , of the electromagnetic proper oscillations [40, Eq. (2.34)]. In the longitudinal case described by equation (23), the resulting waves are conventionally called *plasmons* (see e.g. Refs. [28, Eq. (5.49)], [38, Eq. (14.78)], and [43, Eq. (4.92)]). The corresponding *transverse proper oscillations* determined by equation (24) – whose existence can already be deduced *per analogiam* from equation (23) – obviously describe the propagation of light in the medium. In the most general (i.e., not necessarily isotropic) case, however, both equations combine into the unified wave equation (19). It remains to discuss in how far this fundamental, microscopic wave equation translates into the standard wave equation used in theoretical optics.

#### 3.2 Wave equation used in theoretical optics

For the above purpose, we now reformulate the general wave equation (19) in terms of the microscopic *proper* conductivity tensor [27, Sect. 2.3]. As a matter of principle [27, Sect. 2.5], this quantity is related to the dielectric tensor via [40, Eqs. (2.24)–(2.25)]

$$\overset{\leftrightarrow}{\varepsilon}(\mathbf{k}, \omega) = \overset{\leftrightarrow}{1} - \overset{\leftrightarrow}{\mathbb{E}}(\mathbf{k}, \omega) \frac{\overset{\leftrightarrow}{\sigma}(\mathbf{k}, \omega)}{i\omega\varepsilon_0}, \quad (25)$$

where the *electric solution generator* [27, Sect. 2.4] is given in terms of the well-known *longitudinal* and *transverse projection operators* [35, Eqs. (2.1) and (2.2)] by the

concise formula

$$\overleftrightarrow{\mathbb{E}}(\mathbf{k}, \omega) = \overleftrightarrow{P}_L(\mathbf{k}) + \frac{\omega^2}{\omega^2 - c^2|\mathbf{k}|^2} \overleftrightarrow{P}_T(\mathbf{k}). \quad (26)$$

Equation (25) holds independently of the isotropic limit. It is, in fact, quite generally valid, i.e. regardless of the approximation used to calculate the conductivity tensor. With these preparations, the fundamental wave equation (19) can be rewritten as

$$\overleftrightarrow{\mathbb{E}}^{-1}(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k}, \omega) = \frac{1}{i\omega\varepsilon_0} \overleftrightarrow{\tilde{\sigma}}(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k}, \omega), \quad (27)$$

and further using equation (26), we can recast it into the form

$$\left( -\frac{\omega^2}{c^2} \left( \overleftrightarrow{1} - \frac{\overleftrightarrow{\tilde{\sigma}}(\mathbf{k}, \omega)}{i\omega\varepsilon_0} \right) + |\mathbf{k}|^2 \overleftrightarrow{P}_T(\mathbf{k}) \right) \mathbf{E}(\mathbf{k}, \omega) = 0. \quad (28)$$

Finally, by applying the vector identity

$$\mathbf{k} \times (\mathbf{k} \times \mathbf{E}(\mathbf{k}, \omega)) = -|\mathbf{k}|^2 \overleftrightarrow{P}_T(\mathbf{k}) \mathbf{E}(\mathbf{k}, \omega), \quad (29)$$

and by defining the *effective dielectric tensor* as

$$\overleftrightarrow{\varepsilon}_{\text{eff}}(\mathbf{k}, \omega) \stackrel{\text{def}}{=} \overleftrightarrow{1} - \frac{\overleftrightarrow{\tilde{\sigma}}(\mathbf{k}, \omega)}{i\omega\varepsilon_0}, \quad (30)$$

we can bring equation (28) into the equivalent form

$$-\frac{\omega^2}{c^2} \overleftrightarrow{\varepsilon}_{\text{eff}}(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k}, \omega) = \mathbf{k} \times (\mathbf{k} \times \mathbf{E}(\mathbf{k}, \omega)). \quad (31)$$

This equation *formally agrees* with the standard wave equation used in *theoretical optics* and solid state physics (see e.g. Refs. [44, Eq. (1)], [45, Eq. (4.11)], [46, Eq. (2.2.9)], and [47, Eq. (41.2)]). We emphasize, however, that the *effective* dielectric tensor appearing in equation (31) is actually not identical, but only approximately equal (in the limit  $|\mathbf{k}| \rightarrow \mathbf{0}$ ) to the *real* (i.e., *ab initio*) dielectric tensor defined in equation (20) (compare Eqs. (25) and (30)). Put differently, we here observe an astonishing “error cancellation”: the *phenomenological* wave equation (31) combined with the *approximate* relation between the dielectric tensor and the conductivity tensor, equation (30), is *precisely equivalent* to the *exact* wave equation (19) in the form of equation (28). This explains, in particular, why the *ab initio* treatment of the refractive index, if based on equation (28), will be *entirely correct* even though it does not explicitly start from the fundamental wave equation (19) (compare also Ref. [40], Eqs. (2.29)–(2.30), (2.33)–(2.34), and comments therein, where this insight is formulated – apparently for the first time – for the case of an isotropic dielectric function).

Next, we consider again the isotropic limit: from equations (25)–(26), it follows that equation (30) holds exactly for the longitudinal dielectric function [42, Eq. (E.11)], i.e.,

$$\varepsilon_L(\mathbf{k}, \omega) = 1 - \frac{\tilde{\sigma}_L(\mathbf{k}, \omega)}{i\omega\varepsilon_0} = \varepsilon_{\text{eff},L}(\mathbf{k}, \omega). \quad (32)$$

On the other hand, for the transverse parts we find the relation<sup>2</sup>

$$\begin{aligned} \varepsilon_T(\mathbf{k}, \omega) &= 1 - \frac{\omega^2}{\omega^2 - c^2|\mathbf{k}|^2} \frac{1}{i\omega\varepsilon_0} \tilde{\sigma}_T(\mathbf{k}, \omega) \\ &= \frac{\varepsilon_{\text{eff},T}(\mathbf{k}, \omega) \omega^2 - c^2|\mathbf{k}|^2}{\omega^2 - c^2|\mathbf{k}|^2}. \end{aligned} \quad (33)$$

Furthermore, equation (31) decouples in the isotropic limit into two separate wave equations for the longitudinal and transverse electric field components:

$$\varepsilon_{\text{eff},L}(\mathbf{k}, \omega) \mathbf{E}_L(\mathbf{k}, \omega) = 0, \quad (34)$$

$$\left( -\frac{\omega^2}{c^2} \varepsilon_{\text{eff},T}(\mathbf{k}, \omega) + |\mathbf{k}|^2 \right) \mathbf{E}_T(\mathbf{k}, \omega) = 0. \quad (35)$$

Together with equations (32)–(33), these equations are in fact equivalent to equations (21)–(22) from the previous subsection. This explains, in particular, *why in ab initio physics one usually works with two entirely different wave equations*, both formulated in terms of the dielectric function: the phenomenological wave equation (31), which is usually used for optical, i.e., transverse oscillations only, and the plasmon equation (21), which is used for longitudinal oscillations. In actual fact, however, by using the *exact* relation (25) between the dielectric tensor and the conductivity tensor, both wave equations turn out to be of the same type; and in case that longitudinal and transverse oscillations do not decouple, they have to be combined into the fundamental wave equation (19).

Finally, we also comment on the wave equation corresponding to the standard equation (1) for the refractive index, which is [7, Eq. (2.203)]

$$\left( -\frac{\omega^2}{c^2} \varepsilon_L(\omega) + |\mathbf{k}|^2 \right) \mathbf{E}_T(\mathbf{k}, \omega) = 0. \quad (36)$$

This does clearly not coincide with equation (35). Astonishingly, equation (36) constitutes instead a wave equation for a *transverse* electric field, which is, however, formulated in terms of the *longitudinal* dielectric function. Nevertheless, this equation can also be justified from the fundamental wave equation in the isotropic case, i.e., from equations (21)–(22): At optical wavelengths (i.e., for small wavevectors), it is sometimes plausible to assume that the longitudinal and transverse conductivities coincide in the sense of

$$\tilde{\sigma}_L(\mathbf{k}, \omega) = \tilde{\sigma}_T(\mathbf{k}, \omega). \quad (37)$$

With this relation, one shows directly from equation (25) that the longitudinal and transverse dielectric functions are related as follows [27, Sect. 4.4]:

$$\varepsilon_T(\mathbf{k}, \omega) = \frac{-\omega^2 \varepsilon_L(\mathbf{k}, \omega) + c^2|\mathbf{k}|^2}{-\omega^2 + c^2|\mathbf{k}|^2}, \quad (38)$$

<sup>2</sup> The identities (32) and (33) can be compared to reference [1, Eqs. (53) and (53)]; in particular, the phenomenological model defined in reference [1] has the property that  $\varepsilon_{\text{eff},L}(\mathbf{k}, \omega) = \varepsilon_{\text{eff},T}(\mathbf{k}, \omega) = \varepsilon_{\text{eff}}$  with a *constant, scalar* effective permittivity  $\varepsilon_{\text{eff}}$ .

i.e. provided the approximation (37) holds, the transverse dielectric function can be calculated from its longitudinal counterpart by this formula. Together with the fundamental wave equation (22), this implies

$$\begin{aligned} & \left( -\frac{\omega^2}{c^2} \varepsilon_L(\mathbf{k}, \omega) + |\mathbf{k}|^2 \right) \mathbf{E}_T(\mathbf{k}, \omega) \\ & = \left( -\frac{\omega^2}{c^2} + |\mathbf{k}|^2 \right) \varepsilon_T(\mathbf{k}, \omega) \mathbf{E}_T(\mathbf{k}, \omega) = 0. \end{aligned} \quad (39)$$

Finally, by neglecting the  $\mathbf{k}$ -dependence of  $\varepsilon_L$  we arrive at the assertion, equation (36). On the other hand, since the condition (37) will not always be true, and since the  $\mathbf{k}$ -dependence of  $\varepsilon_L$  cannot always be neglected, our results corroborates again our initial statement (see Sect. 1) that in the most general case, the deduction of optical materials properties should not be based on the standard relation (1).

## 4 Optical properties from conductivity tensor

### 4.1 Fundamental wave equation in optical limit

The above considerations have shown that, as a matter of principle, optical properties have to be deduced from the general wave equation encompassing the transverse subspace, i.e., from equation (19), or from its standard form given by equation (31), which can also be written as

$$\overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k}, \omega) = \frac{c^2 |\mathbf{k}|^2}{\omega^2} \overset{\leftrightarrow}{P}_T(\mathbf{k}) \mathbf{E}(\mathbf{k}, \omega). \quad (40)$$

Unfortunately, this equation requires (via the relation (30)) knowledge of the full, i.e., frequency- and wavevector-dependent conductivity tensor, which is computationally very demanding. Moreover, even if the full conductivity tensor or the corresponding effective dielectric tensor is known, the dispersion relation  $\omega = \omega_{\mathbf{k}}$  will have to be deduced from the *implicit* equation (40), where the frequency appears not only explicitly on the right-hand side but also implicitly as an argument of the effective dielectric tensor. Consequently, the refractive index, which is defined by (see Refs. [46, Eq. (1.2.5)] or [48, Eq. (11.12)])

$$n_{\mathbf{k}} = \frac{c |\mathbf{k}|}{\omega_{\mathbf{k}}}, \quad (41)$$

is also only given by an implicit equation (see Ref. [27, Sect. 4.2]).

The decisive point is now that instead of interpreting equation (41) as an implicit equation determining the frequency (or the refractive index) as a function of the wavevector, one may instead also fix the frequency and the *direction* of the wavevector,  $\hat{\mathbf{k}} := \mathbf{k}/|\mathbf{k}|$ , and regard equation (40) as an implicit equation determining the modulus of the wavevector  $|\mathbf{k}|$ . Correspondingly, one can also consider the refractive index as a function of the frequency and the *direction* of the wavevector,  $n = n(\hat{\mathbf{k}}, \omega)$ .

The modulus of the wavevector is then given in terms of this refractive index by

$$|\mathbf{k}| = \frac{\omega}{c} n(\hat{\mathbf{k}}, \omega). \quad (42)$$

Even in this case, though, the refractive index is given only implicitly by equation (40), because the modulus  $|\mathbf{k}|$  does not only appear explicitly on the right-hand side of this equation but also implicitly as an argument of the effective dielectric tensor (see also discussion in Sect. 4.4).

A pragmatic way to overcome these difficulties lies in the following assumption: although the relations between different response functions are in general *wavevector-dependent* (see Sect. 2), it is not contradictory to assume that *one particular* response function is actually *wavelength independent* (at least approximately), whereby it has to be stressed that this assumption *cannot* be upheld *simultaneously* for all response functions at once (see again Sect. 2, or the most general Universal Response Relations in Ref. [25, Sect. 6]). In particular, the wavelength independence of the conductivity tensor implies that the density response function as well as the dielectric tensor are definitely wavelength dependent (see Eqs. (9) and (25)).

Concretely, using the relation between the wavelength and the modulus of the wavevector,

$$|\mathbf{k}| = \frac{2\pi}{\lambda}, \quad (43)$$

we see that our assumption of wavelength independence concretely means that the dependence on the modulus can be dropped in the above formulary. In fact, the standard approach suggests that the assumption of wavelength independence actually applies to the *proper conductivity tensor*, or equivalently (see Eq. (3)) to the proper current response tensor. The justification for this claim is that the calculation of a genuine dielectric tensor (as opposed to a sheer dielectric function as in Eq. (10)) is, as by equation (30), in practice based on the determination of the conductivity tensor in the first place, which for its part typically comes as a completely wavevector-independent computer output. Put differently, in practical calculations one may even assume that the proper conductivity tensor is wavevector-independent altogether, i.e.  $\tilde{\sigma} = \tilde{\sigma}(\omega)$ , though conceptually this is actually overly restrictive, for which reason we do not insist on this point here. Correspondingly, for now (see Sect. 4.4 below for the more general case) we assume that at optical wavelengths, the *proper* conductivity tensor is wavelength independent in the sense that

$$\overset{\leftrightarrow}{\tilde{\sigma}}(\mathbf{k}, \omega) \equiv \overset{\leftrightarrow}{\tilde{\sigma}}(\hat{\mathbf{k}}, |\mathbf{k}|, \omega) = \overset{\leftrightarrow}{\tilde{\sigma}}(\hat{\mathbf{k}}, \omega). \quad (44)$$

In the following, we will investigate the general wave equation under this additional assumption. We remark in passing, though, that the proper conductivity tensor can of course trivially be identified with its limit  $|\mathbf{k}| \rightarrow 0$  (or equivalently  $\lambda \rightarrow \infty$ ) – which defines the so-called *optical limit* – if it is actually wavelength independent anyway.

First, equation (44) implies that the effective dielectric tensor defined by equation (30) also depends exclusively on the frequency. This, in turn, greatly simplifies the solution of the wave equation (40), which now becomes an *explicit* equation determining the modulus of the wavevector  $|\mathbf{k}|$  as a function of  $\hat{\mathbf{k}}$  and  $\omega$ . In fact, since the transverse projection operator for its part depends only on the direction (and not on the modulus) of the wavevector,

$$\overset{\leftrightarrow}{P}_T(\mathbf{k}) = \overset{\leftrightarrow}{1} - \frac{\mathbf{k}\mathbf{k}^T}{|\mathbf{k}|^2} = \overset{\leftrightarrow}{P}_T(\hat{\mathbf{k}}), \quad (45)$$

the *frequency- and direction-dependent refractive indices* are now determined by an *explicit* equation, whose directional dependence comes into play only via the transverse projection operator. To show this, we make the following ansatz for the electric field in Fourier space:

$$\begin{aligned} \mathbf{E}(\mathbf{k}, \omega) \propto & \mathbf{e}(\hat{\mathbf{k}}, \omega) \delta\left(|\mathbf{k}| - \frac{\omega}{c} n(\hat{\mathbf{k}}, \omega)\right) \\ & + \mathbf{e}^*(-\hat{\mathbf{k}}, -\omega) \delta\left(|\mathbf{k}| + \frac{\omega}{c} n(-\hat{\mathbf{k}}, -\omega)\right), \end{aligned} \quad (46)$$

where  $\mathbf{e}(\hat{\mathbf{k}}, \omega)$  is the so-called *polarization vector*, which we assume to be normalized. Note that the polarization vector may, in principle, also depend on the frequency (though *in praxi* it is usually frequency independent). The ansatz (46) fulfills the constraint condition

$$\mathbf{E}(\mathbf{k}, \omega) = \mathbf{E}^*(-\mathbf{k}, -\omega), \quad (47)$$

which guarantees that the electric field is real-valued in the space-time domain. Now, by putting equation (46) into equation (40), we see that our ansatz solves the general wave equation provided that the polarization vector fulfills the *central equation for the joint determination of refractive indices and polarization vectors*:

$$\overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) \mathbf{e}(\hat{\mathbf{k}}, \omega) = n^2(\hat{\mathbf{k}}, \omega) \overset{\leftrightarrow}{P}_T(\hat{\mathbf{k}}) \mathbf{e}(\hat{\mathbf{k}}, \omega), \quad (48)$$

as well as the analogous condition

$$\overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) \mathbf{e}^*(-\hat{\mathbf{k}}, -\omega) = n^2(-\hat{\mathbf{k}}, -\omega) \overset{\leftrightarrow}{P}_T(\hat{\mathbf{k}}) \mathbf{e}^*(-\hat{\mathbf{k}}, -\omega). \quad (49)$$

Furthermore, the reality condition on the effective dielectric tensor,

$$\overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) = (\overset{\leftrightarrow}{\varepsilon}_{\text{eff}})^*(-\hat{\mathbf{k}}, -\omega), \quad (50)$$

and the trivial fact that

$$\overset{\leftrightarrow}{P}_T(\hat{\mathbf{k}}) = (\overset{\leftrightarrow}{P}_T)^*(-\hat{\mathbf{k}}), \quad (51)$$

together imply that the condition (49) is actually equivalent to equation (48) and can therefore be discarded. On the other hand, by combining equations (48) and (49) one sees directly that

$$\mathbf{e}(\hat{\mathbf{k}}, \omega) = \mathbf{e}^*(-\hat{\mathbf{k}}, -\omega), \quad (52)$$

$$n^2(\hat{\mathbf{k}}, \omega) = n^2(-\hat{\mathbf{k}}, -\omega), \quad (53)$$

and since equation (42) should always yield a positive result for the modulus of the wavevector [49], we may even set

$$n(\hat{\mathbf{k}}, \omega) = -n(-\hat{\mathbf{k}}, -\omega). \quad (54)$$

In summary, equation (48) is the central equation for the joint determination of the frequency- and direction-dependent refractive indices and the corresponding polarization vectors. The logic of this central equation is this: the frequency and the direction of the wavevector are independent variables which, in principle, can be prescribed arbitrarily; these being given, equation (48) determines the refractive index and via equation (42) the modulus of the wavevector, as well as the possible polarization vectors of the proper oscillations with the given frequency and direction.

Finally, we remark that equation (40) is *formally analogous* to the equation  $\mathbf{D} = \varepsilon_0 n^2 \mathbf{E}_T$ , which is known from theoretical optics (see Refs. [50, Sect. 15.2, Eqs. (2) and (4)] and [51, Eq. (6.21)]), provided one plugs in the standard relation

$$\mathbf{D} \stackrel{\text{def}}{=} \varepsilon_0 \overset{\leftrightarrow}{\varepsilon}_{\text{eff}} \mathbf{E} \quad (55)$$

for the electric “displacement” field and uses equation (41). In fact, in our context equation (55) has to be interpreted as the definition of the classical field  $\mathbf{D}$ . Appropriately, the displacement field then also fulfills the standard equation

$$\nabla \cdot \mathbf{D} = \rho_{\text{ext}} \equiv \rho^{\text{“free”}}, \quad (56)$$

as follows from

$$\nabla \cdot \mathbf{D}(\omega) = \varepsilon_0 \nabla \cdot \left( \mathbf{E}(\omega) - \frac{\overset{\leftrightarrow}{\sigma}(\omega)}{i\omega \varepsilon_0} \mathbf{E}(\omega) \right) \quad (57)$$

$$= \varepsilon_0 \nabla \cdot \left( \mathbf{E}(\omega) - \frac{\mathbf{j}_{\text{ind}}(\omega)}{i\omega \varepsilon_0} \right) \quad (58)$$

$$= \rho(\omega) - \frac{\nabla \cdot \mathbf{j}_{\text{ind}}(\omega)}{i\omega} \quad (59)$$

$$= \rho_{\text{ext}}(\omega), \quad (60)$$

where we have used

$$\rho \equiv \rho_{\text{tot}} = \rho_{\text{ext}} + \rho_{\text{ind}} = \rho^{\text{“free”}} + \rho^{\text{“bound”}} \quad (61)$$

and the continuity equation in Fourier space,

$$-i\omega \rho(\mathbf{x}, \omega) + \nabla \cdot \mathbf{j}(\mathbf{x}, \omega) = 0. \quad (62)$$

By contrast, the transverse part of the displacement field,  $\nabla \times \mathbf{D}_T = \nabla \times \mathbf{D}$ , depends on the conductivity tensor under consideration. We note, however, that the thus defined displacement field would be a purely auxiliary quantity (though not a particularly helpful one) without the slightest intrinsic meaning. In particular, it would be of the most secondary character as it could only be calculated *ex post*, i.e. once the conductivity tensor is given, and can hence not serve any practical purpose. Worse, it does not even have a value on the conceptual level,

e.g. as a defining equation for the effective dielectric tensor. Rather to the contrary, the latter must be used to define the displacement field in the first place.

The present article therefore works within the paradigm of the Functional Approach to electrodynamics in media (see Refs. [25,26] for extensive discussions), which is an ab initio theory exclusively based on the microscopic Maxwell equations. Instead of introducing “macroscopic” fields, it simply distinguishes between external and induced fields as it is common practice in ab initio materials physics (see e.g. the modern textbooks [7,28,29,38]). Note, however, that one cannot – in general – apply the standard methods of theoretical optics to (40), because the effective dielectric tensor – stemming from a *retarded* response function calculated by the Kubo formula – is typically *not hermitean* (as it is assumed in theoretical optics) and hence not necessarily diagonalizable. In the following, we will study in detail the solutions of equation (48) *without* any particular assumption about the effective dielectric tensor.

## 4.2 Radiation modes and generalized plasmons

Although equation (48) is not just an eigenvalue problem for the effective dielectric tensor, it allows for a somewhat analogous mathematical treatment: nontrivial solutions exist if and only if the condition (see Refs. [45, Eq. (4.12)], [52, Eq. (77.9)] and [53, Eq. (2.14)])

$$\det \left( \overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) - n^2(\hat{\mathbf{k}}, \omega) \overset{\leftrightarrow}{P}_T(\hat{\mathbf{k}}) \right) = 0 \quad (63)$$

is fulfilled. As stressed above, for given direction  $\hat{\mathbf{k}}$  and frequency  $\omega$ , this equation determines the refractive indices  $n_\lambda(\hat{\mathbf{k}}, \omega)$ , which we label by an index  $\lambda \in \mathbb{N}$  (later, we will see that  $\lambda \in \{1, 2\}$ ). These solutions can be studied most conveniently by choosing for each direction  $\hat{\mathbf{k}}$  an orthonormal basis in  $\mathbb{R}^3$ , i.e., three (real) vectors  $\mathbf{e}_{\mathbf{k}1}$ ,  $\mathbf{e}_{\mathbf{k}2}$ ,  $\mathbf{e}_{\mathbf{k}3}$  with the property that  $\mathbf{e}_{\mathbf{k}i} \cdot \mathbf{e}_{\mathbf{k}j} = \delta_{ij}$ . We further assume that  $\mathbf{e}_{\mathbf{k}1}$  and  $\mathbf{e}_{\mathbf{k}2}$  are perpendicular to  $\hat{\mathbf{k}}$ , i.e., they lie in the transverse subspace, while  $\mathbf{e}_{\mathbf{k}3} = \hat{\mathbf{k}}$  is in the (one-dimensional) longitudinal subspace. We call this an *isotropic basis*. Equation (63) then takes the following form:

$$\det \begin{pmatrix} \varepsilon_{11} - n^2 & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22} - n^2 & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33} \end{pmatrix} = 0, \quad (64)$$

where we have defined the *optical tensor*, i.e. the matrix corresponding to the effective dielectric tensor evaluated in the isotropic basis

$$\varepsilon_{ij}(\hat{\mathbf{k}}, \omega) \stackrel{\text{def}}{=} \mathbf{e}_{\mathbf{k}i}^T \overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) \mathbf{e}_{\mathbf{k}j}. \quad (65)$$

We emphasize particularly that these components of the *effective dielectric tensor* refer to a basis in  $\mathbf{k}$ -space (and hence not to a fixed basis in real space). In particular, this implies that the matrix appearing in equation (64) does in fact depend on the direction  $\hat{\mathbf{k}}$ , *even if the original*

*effective dielectric tensor is purely frequency dependent* (as it often is). *This explains in particular why it is possible (and in fact imperative) to obtain wavevector-dependent optical properties from wavevector-independent computer outputs.*

Given a refractive index  $n_\lambda$  which solves equation (64), one further obtains the (normalized) vectors  $\mathbf{v}_\lambda = (v_\lambda^1, v_\lambda^2, v_\lambda^3)^T$  which fulfill the homogeneous equation

$$\begin{pmatrix} \varepsilon_{11} - n_\lambda^2 & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22} - n_\lambda^2 & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33} \end{pmatrix} \begin{pmatrix} v_\lambda^1 \\ v_\lambda^2 \\ v_\lambda^3 \end{pmatrix} = 0. \quad (66)$$

The corresponding (normalized) polarization vectors, which solve equation (48), can then be written as

$$\mathbf{e}_\lambda(\hat{\mathbf{k}}, \omega) = v_\lambda^1(\hat{\mathbf{k}}, \omega) \mathbf{e}_{\mathbf{k}1} + v_\lambda^2(\hat{\mathbf{k}}, \omega) \mathbf{e}_{\mathbf{k}2} + v_\lambda^3(\hat{\mathbf{k}}, \omega) \mathbf{e}_{\mathbf{k}3}, \quad (67)$$

and hence they are, in general, *not* purely transverse (and possibly complex at that).

In the remainder of this subsection, we will deduce some general properties of the solutions of equations (64) and (66). In the next subsection, we will then derive explicit expressions for the refractive indices in the most general case of an anisotropic material, and finally in Section 5 we will investigate some special cases.

The first important observation concerning equation (64) is that it leads – as a consequence of the transverse projection operator appearing on the right-hand side of equation (48) – to a polynomial equation of *second* order in  $n^2$ , and hence there are (for each direction and frequency) at most *two* (possibly complex<sup>3</sup>) refractive indices, which we denote by  $n_\lambda(\hat{\mathbf{k}}, \omega)$  with  $\lambda \in \{1, 2\}$ . This is in stark contrast to an ordinary eigenvalue problem for a  $(3 \times 3)$ -matrix, which would in general have three solutions.

However, the sole fact that there are at most two different refractive indices does not answer the question of how many *radiation modes* exist in the medium with a given frequency and direction. Hereby, we define a radiation mode as a solution  $(n^2(\hat{\mathbf{k}}, \omega), \mathbf{e}(\hat{\mathbf{k}}, \omega))$  of the central equation (48) with the following properties: (i) the refractive index is non-zero,  $n^2(\hat{\mathbf{k}}, \omega) \neq 0$ , and (ii) the polarization vector has a non-vanishing transverse part,  $\mathbf{e}_T(\hat{\mathbf{k}}, \omega) \neq 0$ . The first condition is necessary because by equation (42), a vanishing refractive index would imply that  $|\mathbf{k}| = 0$ . This means that the medium oscillates as a whole, a behavior which is also sometimes referred to as “plasmon”. The second condition excludes the purely longitudinal proper oscillations of the medium, which are usually referred to as *plasmons* (see comments below). In other words, the

<sup>3</sup> Actually, our theoretical formalism is predicated on the assumption that the refractive index is real (see Eq. (42)). Nonetheless, one might accept complex refractive indices as an outcome of the final formulae if one does not care too much about how one has arrived at these in the first place. This, however, leaves open the question of what these complex refractive indices really mean, and this problem in turn is in no way specific for the stance proposed here. We therefore feel free to ignore it for the time being.

question is now the following: for a given frequency and direction, how many linearly independent polarization vectors exist which solve equation (48) and which are not purely longitudinal? In analogy to the vacuum case, one might assume that there are at most two such modes for each direction and frequency (see Sect. 5.1). In order to prove this hypothesis, we take again recourse to theoretical optics (see e.g. Refs. [45, Sect. 4.2] and [54, p. 300]), whereby we distinguish two cases depending on the determinant of the effective dielectric tensor.

*Case 1:* The effective dielectric tensor is invertible, hence  $\det \overset{\leftrightarrow}{\varepsilon}_{\text{eff}} \neq 0$ . In this case, acting on equation (48) first with the inverse effective dielectric tensor and then with the transverse projector, we obtain

$$\begin{aligned} & \left( \overset{\leftrightarrow}{P}_{\text{T}}(\hat{\mathbf{k}}) (\overset{\leftrightarrow}{\varepsilon}_{\text{eff}})^{-1}(\hat{\mathbf{k}}, \omega) \overset{\leftrightarrow}{P}_{\text{T}}(\hat{\mathbf{k}}) \right) \mathbf{e}_{\text{T}}(\hat{\mathbf{k}}, \omega) \\ &= \frac{1}{n^2(\hat{\mathbf{k}}, \omega)} \mathbf{e}_{\text{T}}(\hat{\mathbf{k}}, \omega), \end{aligned} \quad (68)$$

where  $\mathbf{e}_{\text{T}}(\hat{\mathbf{k}}, \omega) = P_{\text{T}}(\hat{\mathbf{k}}) \mathbf{e}(\hat{\mathbf{k}}, \omega)$ . In the *transverse* subspace, this now is an eigenvalue problem, which shows that the (transverse parts of the) polarization vectors can be characterized as eigenvectors of a suitably defined  $(2 \times 2)$  matrix. The transverse part of the polarization vector being given, we can then calculate the longitudinal part by the explicit formula

$$\mathbf{e}_{\text{L}}(\hat{\mathbf{k}}, \omega) = n^2(\omega, \hat{\mathbf{k}}) (\overset{\leftrightarrow}{\varepsilon}_{\text{eff}})^{-1}(\hat{\mathbf{k}}, \omega) \mathbf{e}_{\text{T}}(\hat{\mathbf{k}}, \omega) - \mathbf{e}_{\text{T}}(\hat{\mathbf{k}}, \omega), \quad (69)$$

which follows again from the central equation (48). Consequently, there are at most two linearly independent polarization vectors which possess a transverse part and thereby qualify as radiation modes.

*Case 2:* The effective dielectric tensor is not invertible, hence  $\det \overset{\leftrightarrow}{\varepsilon}_{\text{eff}} = 0$ . In this case, one obvious solution of equation (64) is  $n^2 = 0$ , which does not qualify as a radiation mode. The other refractive index may be non-zero, and consequently, the possible polarization vectors are determined by the null space defined in equation (66) with only one possible refractive index  $n^2 \neq 0$ . This null space could in principle even be three-dimensional; however, in this case the only non-vanishing components of the effective dielectric tensor would be  $\varepsilon_{11} = \varepsilon_{22} = n^2$ , and hence there would again be two transverse and one longitudinal oscillation. Thus, even in the case of a singular effective dielectric tensor, there are at most two radiation modes.

It remains to discuss whether the kernel of the dielectric tensor has a physical meaning. For this purpose, we consider the condition

$$\overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) \mathbf{e}(\hat{\mathbf{k}}, \omega) = 0. \quad (70)$$

If this equation is supposed to give rise to a proper oscillation of the medium, then the central equation (48) has to be fulfilled as well. A comparison shows that in this case, we either have  $n^2(\hat{\mathbf{k}}, \omega) = 0$  or  $\mathbf{e}_{\text{T}}(\hat{\mathbf{k}}, \omega) = 0$ . The first possibility is not considered here. By contrast, the

second possibility states that the proper oscillation is purely longitudinal and hence corresponds to a so-called *plasmon*. Thus, equation (70) combined with the longitudinality condition constitutes the *generalized plasmon equation*, which generalizes the well-known condition (23) for isotropic media. In particular, since the effective dielectric tensor does not necessarily have a non-trivial kernel, this shows that plasmons do not necessarily have to exist in every material.

### 4.3 General formulae for refractive indices

In this final subsection, we study the refractive indices in the most general case of an anisotropic material, for which the off-diagonal components of the (effective) dielectric tensor do not vanish. In this case, equation (64) leads to the following equation, which is quadratic in  $n^2$ :

$$\begin{aligned} n^4 \varepsilon_{33} - n^2 ((\varepsilon_{11} + \varepsilon_{22}) \varepsilon_{33} - \varepsilon_{13} \varepsilon_{31} - \varepsilon_{23} \varepsilon_{32}) \\ + \det \overset{\leftrightarrow}{\varepsilon}_{\text{eff}} = 0. \end{aligned} \quad (71)$$

In theoretical optics, this is sometimes referred to as the *Fresnel equation* (in honor of the legendary Augustin Jean Fresnel (1788–1827)) (see e.g. Refs. [53, Eq. (2.14)] or [54, p. 300]). Note, however, that this equation must not be confused with the Fresnel equations used for the intensity distribution for reflection at a material interface (see Ref. [37] for a recent discussion).

For studying the solutions of equation (71), we distinguish again two cases depending on the value of  $\varepsilon_{33}$ . The latter parameter coincides, via the relation (cf. Eq. (11))

$$\begin{aligned} \varepsilon_{33}(\hat{\mathbf{k}}, \omega) \overset{\leftrightarrow}{P}_{\text{L}}(\hat{\mathbf{k}}) &= \overset{\leftrightarrow}{P}_{\text{L}}(\hat{\mathbf{k}}) \overset{\leftrightarrow}{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) \overset{\leftrightarrow}{P}_{\text{L}}(\hat{\mathbf{k}}) \\ &\equiv (\overset{\leftrightarrow}{\varepsilon}_{\text{eff}})_{\text{LL}}(\hat{\mathbf{k}}, \omega), \end{aligned} \quad (72)$$

with the longitudinal projection (see Ref. [35, Sect. 2.1]) of the effective dielectric tensor.

*Case a:* It may happen that at the given frequency and direction, we obtain  $\varepsilon_{33}(\hat{\mathbf{k}}, \omega) = 0$ . For such frequencies, equation (71) reduces to

$$n^2 (\varepsilon_{13} \varepsilon_{31} + \varepsilon_{23} \varepsilon_{32}) + \det \overset{\leftrightarrow}{\varepsilon}_{\text{eff}} = 0, \quad (73)$$

and this equation has *precisely one* solution  $n^2$  given by

$$n^2 = - \frac{\det \overset{\leftrightarrow}{\varepsilon}_{\text{eff}}}{\varepsilon_{13} \varepsilon_{31} + \varepsilon_{23} \varepsilon_{32}}. \quad (74)$$

Here, we have assumed that the denominator does not vanish (which will generally be the case for anisotropic materials).

$$(n^2)_{1/2} = \frac{1}{2} \left( \varepsilon_{11} + \varepsilon_{22} - \frac{\varepsilon_{13} \varepsilon_{31} + \varepsilon_{23} \varepsilon_{32}}{\varepsilon_{33}} \right) \pm \frac{1}{2} \sqrt{\left( \varepsilon_{11} + \varepsilon_{22} - \frac{\varepsilon_{13} \varepsilon_{31} + \varepsilon_{23} \varepsilon_{32}}{\varepsilon_{33}} \right)^2 - \frac{4 \det \overleftrightarrow{\varepsilon}_{\text{eff}}}{\varepsilon_{33}}} \quad (75)$$

Case b:  $\varepsilon_{33}(\hat{\mathbf{k}}, \omega) \neq 0$ . Now, equation (71) has generally two (possibly complex) solutions given by

See equation (75) above.

For frequencies  $\omega = \omega(\hat{\mathbf{k}})$  which satisfy

$$\det \overleftrightarrow{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) = 0, \quad (76)$$

these solutions turn into

$$n_1^2 = \varepsilon_{11} + \varepsilon_{22} - \frac{\varepsilon_{13} \varepsilon_{31} + \varepsilon_{23} \varepsilon_{32}}{\varepsilon_{33}} \quad (77)$$

and  $n_2^2 = 0$ , where the latter refractive index can be discarded. (Thus, we recover Case 2 treated in the previous subsection.)

To summarize, equations (66) and (75) solve the problem of calculating the refractive indices and polarization vectors in the most general case of a possibly non-diagonalizable effective dielectric tensor. We emphasize again that all the above formulae refer to the *effective* dielectric tensor defined by equation (30). This can be calculated from the wavevector-*independent* proper conductivity tensor, but still allows one to deduce wavevector-*dependent* optical materials properties from first principles.

#### 4.4 Wavelength dependent conductivity tensor

In the preceding subsections, we have assumed that the proper conductivity tensor is effectively wavelength independent at least in the optical range. Although this will certainly constitute an assumption which is fulfilled in almost all cases of practical relevance, we will shortly consider the possible failure of this assumption in this final subsection, if maybe only for conceptual reasons. In fact, if the proper conductivity tensor indeed perceptibly depends on the wavelength or – equivalently – on the modulus of the wavevector, so will the effective dielectric tensor, such that equation (63) has to be replaced by

$$\det \left( \overleftrightarrow{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, |\mathbf{k}|, \omega) - n^2(\hat{\mathbf{k}}, \omega) \overleftrightarrow{P}_{\text{T}}(\hat{\mathbf{k}}) \right) = 0. \quad (78)$$

At first glimpse, it now appears as if this equation was not sufficient for the determination of the optical properties anymore on the grounds that it involves yet another variable, namely the said modulus of the wavevector. Fortunately, though, it still holds true that the modulus of the wavevector is not independent of its direction and the frequency. Instead, by means of the refractive index it can be expressed in terms of these via equation (42). Plugging this back into the dielectric tensor would again turn equation (78) into a closed, though implicit equation for the

determination of the refractive indices,

$$\det \left( \overleftrightarrow{\varepsilon}_{\text{eff}} \left( \hat{\mathbf{k}}, \frac{\omega}{c} n(\hat{\mathbf{k}}, \omega), \omega \right) - n^2(\hat{\mathbf{k}}, \omega) \overleftrightarrow{P}_{\text{T}}(\hat{\mathbf{k}}) \right) = 0. \quad (79)$$

Superficially, it seems that this problem would be amenable to the methods outlined in the previous subsections. On closer inspection, however, we see that the character of the problem has changed dramatically, which might occasionally lead to qualitatively new effects. This becomes immediately clear once we perform a Taylor expansion of the dielectric tensor into powers of the inverse wavelength. To first order, we get

$$\begin{aligned} \overleftrightarrow{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, |\mathbf{k}|, \omega) &\approx \overleftrightarrow{\varepsilon}_{\text{eff}}(\hat{\mathbf{k}}, \omega) \\ &+ \frac{\omega}{c} n(\hat{\mathbf{k}}, \omega) \left( \frac{\partial}{\partial |\mathbf{k}|} \overleftrightarrow{\varepsilon}_{\text{eff}} \right)_{|\mathbf{k}|=0}(\hat{\mathbf{k}}, \omega), \end{aligned} \quad (80)$$

and this can be fed back into equation (78), where the new term completely absent in the preceding subsections would spoil the entire formulary. For the sake of simplicity, we illustrate this on the most simple example, namely on our starting point, the standard relation between the refractive index and the (longitudinal) dielectric function, equation (1), to which the above formula reduces itself under the crassest simplifying assumptions. To first order Taylor expansion, this standard formula would now have to be replaced by

$$\varepsilon(\omega) + \varepsilon'(\omega) \frac{\omega}{c} n(\omega) = n^2(\omega), \quad (81)$$

where the prime denotes the derivative w.r.t. the modulus of the wavevector. And this can be solved for the refractive indices as

$$n_{1/2}(\omega) = \frac{1}{2} \varepsilon'(\omega) \frac{\omega}{c} \pm \sqrt{\left( \frac{1}{2} \varepsilon'(\omega) \frac{\omega}{c} \right)^2 + \varepsilon(\omega)}. \quad (82)$$

The upshot of this is that for wavelength dependent dielectric functions, one may now have two essentially different refractive indices even in the completely isotropic case where the longitudinal and transverse proper conductivities coincide. Even worse – or rather even better, if one is only interested in new physics – , if the Taylor expansion was continued to arbitrarily high orders, the determining equation (78) for the refractive index would correspond to the zeros of a higher order polynomial and could therefore lead to much more essentially different refractive indices at the same frequency and direction than the usual two in the tensorial though wavelength independent case. As this, however, does not seem to apply in nature, we can safely discard the possibility that in the optical range the effective dielectric tensor would depend on the wavelength for “ordinary” materials.

Apart from the hypothetical chance that the effective dielectric tensor depends on the wavelength for some extremely specific material nonetheless, the benefit of this consideration is that we finally obtain a certain clarity about the frequently invoked optical limit  $|\mathbf{k}| \rightarrow 0$ , which is allegedly to be performed before optical properties can be accessed from *ab initio* outputs. In fact, in this respect we can say this: Either the effective dielectric tensor is independent of the wavelength anyway – in which case this limit is quite superfluous and the effective dielectric tensor can simply be calculated for infinite wavelengths at the very outset – or it is not. If it is not, however, this limit will simply be plainly wrong, no matter how unlikely this case actually is. In fact, it makes no sense to apply such a limit to the fundamental equation (40) and correspondingly in the presence of a perceptible wavelength dependence in the effective dielectric tensor we necessarily get higher order correction terms instead, which can then not be neglected (because this would precisely mean that the wavelength dependence can be neglected all the same).

## 5 Applications

For the convenience of the reader, we reproduce in this final section some standard results from the fundamental formulae (66) and (75) by introducing suitable approximations.

### 5.1 Vacuum

First, in the absence of a medium, the conductivity tensor vanishes and hence the effective dielectric tensor equals the identity matrix. Thus, in vacuo, the only solution of equation (64) is  $n^2 = 1$  corresponding to the dispersion relation  $\omega_{\mathbf{k}} = c|\mathbf{k}|$ . Furthermore, for this refractive index there are two orthogonal polarization vectors  $\mathbf{e}_{\mathbf{k}1}$  and  $\mathbf{e}_{\mathbf{k}2}$  which are both purely transverse.

### 5.2 Isotropic material

Next, for an isotropic material, we have only two independent components of the (effective) dielectric tensor<sup>4</sup>:

$$\varepsilon_{11}(\hat{\mathbf{k}}, \omega) = \varepsilon_{22}(\hat{\mathbf{k}}, \omega) = \varepsilon_{\text{eff},\text{T}}(\hat{\mathbf{k}}, \omega), \quad (83)$$

$$\varepsilon_{33}(\hat{\mathbf{k}}, \omega) = \varepsilon_{\text{eff},\text{L}}(\hat{\mathbf{k}}, \omega). \quad (84)$$

Correspondingly, the refractive indices are determined by the condition

$$\left( \varepsilon_{\text{eff},\text{T}}(\hat{\mathbf{k}}, \omega) - n^2(\hat{\mathbf{k}}, \omega) \right)^2 \varepsilon_{\text{eff},\text{L}}(\hat{\mathbf{k}}, \omega) = 0. \quad (85)$$

<sup>4</sup> In fact, for a “strictly” isotropic material, the respective dielectric functions would not even depend on the direction of the wavevector, but at best on its modulus, which is, however, excluded in the present context (see the discussion in Ref. [33, Appendix D.1]). We also note that, either way, isotropic response functions necessarily depend on the direction of the wavevector via the longitudinal and transverse projection operator. Thus, a completely wavevector-independent conductivity tensor can never be isotropic except for the case where it is proportional to the identity matrix.

We further distinguish between the following two cases. First, we consider  $\varepsilon_{\text{eff},\text{L}}(\hat{\mathbf{k}}, \omega) = 0$ . This condition determines the frequency as a function of the direction. In fact, those frequencies  $\omega = \omega(\hat{\mathbf{k}})$  for which the longitudinal dielectric function vanishes are precisely the *plasmon frequencies* in the isotropic case (see Eq. (21)). The corresponding polarization vectors are purely longitudinal. By contrast, the concept of a refractive index is not meaningful in this case because equation (85) is then fulfilled for any  $n$ . Secondly, we consider  $\varepsilon_{\text{eff},\text{L}}(\hat{\mathbf{k}}, \omega) \neq 0$ . Away from the plasmon frequencies, equation (85) has a two-fold root given in terms of the transverse dielectric function by

$$n^2(\hat{\mathbf{k}}, \omega) = \varepsilon_{\text{eff},\text{T}}(\hat{\mathbf{k}}, \omega). \quad (86)$$

We note that this equation referring to the *effective* transverse dielectric function is fully equivalent to its counterpart equation (24), which is formulated in terms of the fundamental (*ab initio*) transverse dielectric function. Furthermore, in this second case, any two transverse, mutually orthogonal vectors  $\mathbf{e}_{\mathbf{k}1}$  and  $\mathbf{e}_{\mathbf{k}2}$  can be regarded as polarization vectors, which share the same refractive index given by equation (86). Thus, for an isotropic medium we recover the well-known results described already in Section 3.1.

### 5.3 Optical activity

A more general case, which includes the isotropic limit, is defined by the absence of the longitudinal-transverse cross-couplings, i.e., by

$$\overleftrightarrow{P}_{\text{L}}(\hat{\mathbf{k}}) \overleftrightarrow{\varepsilon}_{\text{eff}}(\omega) \overleftrightarrow{P}_{\text{T}}(\hat{\mathbf{k}}) = \overleftrightarrow{P}_{\text{T}}(\hat{\mathbf{k}}) \overleftrightarrow{\varepsilon}_{\text{eff}}(\omega) \overleftrightarrow{P}_{\text{L}}(\hat{\mathbf{k}}) = 0. \quad (87)$$

In this case, the fundamental equation (64) simplifies to

$$\det \begin{pmatrix} \varepsilon_{11} - n^2 & \varepsilon_{12} & 0 \\ \varepsilon_{21} & \varepsilon_{22} - n^2 & 0 \\ 0 & 0 & \varepsilon_{33} \end{pmatrix} = 0. \quad (88)$$

Now, the generalized plasmon equation (70) together with the condition  $\mathbf{e}_{\text{T}}(\hat{\mathbf{k}}, \omega) = 0$  simply implies  $\varepsilon_{33}(\hat{\mathbf{k}}, \omega) = 0$ , and the corresponding frequencies are precisely the plasmon frequencies. Furthermore, there exist at most two *purely transverse* polarization vectors corresponding to two, in general different, refractive indices. These can be characterized as the eigenvectors and eigenvalues of the  $(2 \times 2)$  block matrix corresponding to the transverse subspace. Again, this block matrix does in general not have to be hermitean. It may, in special cases, be of the form  $\varepsilon_{11} = \varepsilon_{22}$  and  $\varepsilon_{12} = -\varepsilon_{21}$  (which, in fact, would be hermitean if only the diagonal entries were real and the off-diagonal entries were purely imaginary), such that the general formula (75) implies  $n_{\pm}^2 = \varepsilon_{11} \pm i\varepsilon_{12}$  and the eigenvectors turn out to be of the form  $\mathbf{e}_{\pm} = (\mathbf{e}_1 \pm i\mathbf{e}_2)/\sqrt{2}$  (see Ref. [51, p. 182]). In this case, the dielectric tensor induces *optical activity* in the respective direction (see e.g. Ref. [51, Eq. (6.39)]). Within the realm of solid state physics, the arch example for this type of behavior is provided by  $\alpha$ -quartz. On the other hand, for

$\varepsilon_{11} = \varepsilon_{22}$  and  $\varepsilon_{12} = \varepsilon_{21} = 0$ , we recover again the results from the previous subsection.

## 5.4 Birefringence

As a matter of principle, the formalism presented in this article encompasses birefringence as well. Experimentally, a typical example for this type of behavior is given by rutile ( $\text{TiO}_2$ ). For practical applications in the optical industry, such materials as  $\alpha$ -BBO ( $\text{Ba}(\text{BO}_2)_2$ ),  $\text{LiNbO}_3$ , or  $\text{LiTaO}_3$  are relevant examples. To demonstrate that birefringence can be described in our formalism, we consider here for the sake of simplicity the case of a *uniaxial birefringent material*, whose proper conductivity tensor is of the form

$$\overleftrightarrow{\sigma}(\omega) = \sigma_0(\omega) \overleftrightarrow{\mathbb{1}} + (\Delta\sigma)(\omega) \hat{\mathbf{a}}\hat{\mathbf{a}}^T, \quad (89)$$

where  $\hat{\mathbf{a}}$  denotes a fixed unit vector in *real space*, the so-called *optical axis*. In this case, all direction-dependent refractive indices can be calculated analytically from our formalism. For this purpose, let us define the reference indices

$$n_1^2(\omega) = 1 - \frac{\sigma_0(\omega)}{i\omega\varepsilon_0}, \quad (90)$$

$$n_2^2(\omega) = 1 - \frac{\sigma_0(\omega) + (\Delta\sigma)(\omega)}{i\omega\varepsilon_0}. \quad (91)$$

In terms of these, the *effective* dielectric tensor can be written as

$$\overleftrightarrow{\varepsilon}_{\text{eff}}(\omega) \equiv \overleftrightarrow{\mathbb{1}} - \frac{\overleftrightarrow{\sigma}(\omega)}{i\omega\varepsilon_0} = n_1^2(\omega) \overleftrightarrow{P}_T(\hat{\mathbf{a}}) + n_2^2(\omega) \overleftrightarrow{P}_L(\hat{\mathbf{a}}), \quad (92)$$

where  $P_L(\hat{\mathbf{a}})$  and  $P_T(\hat{\mathbf{a}})$  denote the longitudinal and transverse projection operators *in the direction of* the optical axis  $\hat{\mathbf{a}}$ . We first note that for  $\hat{\mathbf{k}} = \hat{\mathbf{a}}$ , i.e., if the wavevector is parallel to the optical axis, we recover the results derived in the isotropic case (see Sect. 5.2), with two purely transverse polarization vectors sharing the same refractive index  $n_1^2(\omega)$ . The situation is more complicated in the case where  $\hat{\mathbf{k}} \neq \hat{\mathbf{a}}$ . Then, we make the ansatz

$$\mathbf{e}_{\text{or}}(\hat{\mathbf{k}}) = \frac{\hat{\mathbf{k}} \times \hat{\mathbf{a}}}{|\hat{\mathbf{k}} \times \hat{\mathbf{a}}|} \quad (93)$$

for the first polarization vector, which is both orthogonal to the wavevector (i.e., purely transverse) and to the optical axis. One sees directly that this polarization vector indeed solves the central equation (48) with the refractive index  $n_{\text{or}}^2(\omega) = n_1^2(\omega)$ . Hence, the first refractive index is independent of the direction although the polarization vector is not. This solution corresponds to the so-called *ordinary ray*. In addition, one has to consider the *extraordinary ray*, for whose polarization vector we make the ansatz

$$\mathbf{e}_{\text{ex}}(\hat{\mathbf{k}}, \omega) = A(\hat{\mathbf{k}}, \omega) \hat{\mathbf{k}} + B(\hat{\mathbf{k}}, \omega) \hat{\mathbf{a}}, \quad (94)$$

such that  $\mathbf{e}_{\text{ex}}$  is perpendicular to  $\mathbf{e}_{\text{or}}$ . The two scalar functions  $A$  and  $B$  have to be determined together with the refractive index  $n_{\text{ex}}(\hat{\mathbf{k}}, \omega)$  by putting this ansatz into the central equation (48) and taking into account the normalization condition  $|\mathbf{e}_{\text{ex}}(\hat{\mathbf{k}}, \omega)| = 1$ . Defining the angle  $\alpha(\hat{\mathbf{k}})$  between  $\hat{\mathbf{k}}$  and  $\hat{\mathbf{a}}$  by

$$\cos[\alpha(\hat{\mathbf{k}})] = \hat{\mathbf{k}} \cdot \hat{\mathbf{a}}, \quad (95)$$

we thus obtain after a lengthy but straightforward calculation the refractive index of the extraordinary ray,

$$n_{\text{ex}}^2 = \frac{n_1^2 n_2^2}{n_1^2 \sin^2 \alpha + n_2^2 \cos^2 \alpha}, \quad (96)$$

as well as the coefficient functions

$$B = \left( \sin^2 \alpha + \left( 1 - \frac{n_{\text{ex}}^2}{n_1^2} \right)^2 \cos^2 \alpha \right)^{-1/2}, \quad (97)$$

and

$$A = -\frac{n_{\text{ex}}^2 \cos \alpha}{n_1^2} B. \quad (98)$$

In particular, we see that the polarization vector of the extraordinary ray is not purely transverse but encloses an angle  $\varphi_{\text{ex}}$  with the wavevector given by

$$\cos \varphi_{\text{ex}} \equiv \hat{\mathbf{k}} \cdot \mathbf{e}_{\text{ex}} = A + B \cos \alpha. \quad (99)$$

One can show that in this case the ray direction (which is – in general – defined by the Poynting vector) and the direction of the wavevector do not coincide – fact which accounts for the “extraordinary” behaviour. Finally, for  $\alpha \rightarrow 0$ , one sees that  $n_{\text{ex}}^2 \rightarrow n_1^2$  and  $(\cos \varphi_{\text{ex}}) \rightarrow 0$ , thus the extraordinary ray becomes purely transverse and shares the same refractive index with the ordinary ray, which is consistent with the results discussed in the case where  $\hat{\mathbf{k}} = \hat{\mathbf{a}}$ . All of this is in complete accordance with the standard results for birefringent materials (see e.g. Ref. [55, Sect. 4.5]).

## 6 Conclusion

We have concisely criticized the standard calculation of the refractive index from the *scalar*, wavevector-independent dielectric function. Correspondingly, we have based our treatment of the refractive index on the fundamental, microscopic wave equation in materials, equation (19), which involves in general a wavevector-dependent dielectric tensor. We have shown that this approach even unifies the equations for optical waves and plasmons. Generally, our central results are these:

- We have proven the equivalence of this fundamental, microscopic wave equation to the standard wave equation used in theoretical optics, equation (31), under the assumption that the latter refers to the *effective* dielectric tensor (30) rather than to the fundamental (*ab initio*) dielectric tensor (20).

- Thereby, we have shown that the combination of *ab initio* methods for calculating the proper conductivity tensor – which is a standard target quantity of any modern *ab initio* materials simulation code [31,56–58] (see also the discussion in Ref. [59, Sect. II]) – with the Fresnel equation from theoretical optics, equation (71), solves the problem of calculating wavevector-dependent optical properties from wavevector-independent response functions.
- Correspondingly, the central equation for the joint determination of frequency- and direction-dependent refractive indices and their respective polarization vectors is given by equation (48).
- We have clarified that the application of this entire formalism only requires the wavelength independence of the proper conductivity tensor, while a direction dependence is still possible, apart from the frequency dependence which is always assumed to be present (see Sect. 4).
- We have also investigated the consequences of a hypothetical wavelength dependence in the proper conductivity tensor. In particular, we have shown that the fundamental determining equation for both refractive indices and polarization vectors, equation (63), can then still be turned into the closed though implicit equation (79), which leads directly to correction terms as in equation (82) via a Taylor expansion in the modulus of the wavevector.
- In particular, this makes it clear that the so-called optical limit  $|\mathbf{k}| \rightarrow 0$  as performed in the dielectric tensor before calculating refractive indices is either superfluous (because the effective dielectric tensor is practically wavelength independent anyway) or wrong (because it illegitimately neglects correction terms which would be relevant). As these correction terms probably lead to qualitatively new effects, which could be detected independently of any prior *ab initio* calculation, we also obtain the not entirely trivial *empirical* conclusion that the proper conductivity tensor of typical materials does indeed not depend on the wavelength in the optical range.

Besides these results, we have also clarified some more general theoretical issues such as the following: (i) The standard formula (1) for the refractive index actually only holds under the condition of coinciding longitudinal and transverse proper conductivities (see Eq. (37)). (ii) If this condition is not fulfilled, it should be replaced by equation (40) combined with equation (41). (iii) In the standard treatment, the assumption of wavelength independence actually applies to the proper conductivity tensor (see Eq. (44)). (iv) The relation between the dielectric tensors (20) and (30) used respectively in *ab initio* physics and theoretical optics is given by equation (25).

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## Author contribution statement

All authors contributed equally to this work.

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