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Symmetry peculiarities of dielectric tensor in incommensurately modulated crystal phases

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Abstract. The dielectric permittivity tensor for incommensurately modulated phases in insulating crystals is considered. The main attention is paid to the spatial dispersion giving rise to optical gyration effects and the influence of the modulation phase on those effects. General properties of the dielectric tensor are analysed in relation to the lattice and incommensurate superlattice periodicity requirements, the Onsager principle and the condition of absence of the radiation losses in the optical medium. The structure of the microscopic components of the tensor and a macroscopic averaging procedure for the plane-wave modulation region are discussed. The possibility of existence of an anti-Hermitian part in the dielectric tensor of lossless incommensurate crystals originating from the modulation-induced spatial dispersion is revealed. The performed analysis shows a necessity for introduction into the constitutive equation of the term including spatial derivatives of the optical activity tensor.

1. Introduction

In the last decade optics of the insulating crystals possessing intermediate phases with incommensurately modulated superstructure has been extensively explored. A particular example for those materials is the crystals of A_2BX_4 family [1], of which crystal optical properties have been a subject of a permanent interest of researchers [2]. Within the above field, we should at least mention the problem of the optical activity effect observed in incommensurate (IC) phases of some A_2BX_4 crystals, which has been widely disputed in the literature (see, e.g. [3–7] and references therein). Since the effect is in no simple way compatible with the inversion symmetry of the average structure of the IC phase, it still needs a detailed experimental and theoretical investigation.

From the viewpoint of electrodynamics, the response of the nonmagnetic IC crystals on the electromagnetic wave at optical frequency is determined by the dielectric permittivity tensor. General properties of this tensor were discussed in a number of studies (e.g. [3, 8–12]). Golovko and Levanyuk [8] and Fousek and Kroupa [9] described the dielectric function as a property deviating locally from that determined by the average symmetry. Instead of concept of the local symmetry, Meekes and Janner [3] worked in terms of the superspace groups which represented a full description of the IC phases. It was shown that, despite the orthorhombic basic structure, the superspace symmetry did not forbid the existence of real off-diagonal components of the dielectric tensor, as well as the gyration tensor. Stasyuk and Shvaika [11] developed further the approach [3] and, in particular, discussed some properties of the microscopic dielectric tensor. Finally, Dijkstra *et al* [12] gave a general characterization of the dielectric properties of the IC phases at optical frequencies, including the transition from a microscopic to macroscopic

level. It was revealed that the permittivity tensor should be regarded as periodic along the IC modulation direction, with the mesoscopic spatial period.

The aim of the present work is to consider the main features of the dielectric tensor for the IC crystals and analyse the points related to the symmetry of that part of the tensor originating from a non-locality of the optical response, i.e. the spatial dispersion effect. We are mainly interested in considering a plane-wave modulation region of the IC phases which is the simplest to deal with analytically. The mentioned subject has been in brief regarded in our preliminary study [13].

2. Basic constitute equation and microscopic components of the dielectric tensor

Let us consider a general case of a linear optical medium. From the standpoint of phenomenological electrodynamics, the response of the medium on the field of electromagnetic wave is not local in time and space, i.e. it is determined by the effects of both time (or frequency) and spatial dispersions. The corresponding constitutive equation is as follows (summation over repeated indices is henceforth understood) [14]:

$$D_i(t, \mathbf{r}) = \iint \varepsilon_{ij}(t, t', \mathbf{r}, \mathbf{r}') E_j(t', \mathbf{r}') dt' d\mathbf{r}' \quad (1)$$

where \mathbf{E} and \mathbf{D} are the electric field and displacement vectors of the electromagnetic wave, ε the kernel of the integral operator defining the optical response and the indices refer to Cartesian coordinates. Crystalline media are homogeneous in time ($\varepsilon_{ij}(t, t', \mathbf{r}, \mathbf{r}') = \varepsilon_{ij}(\tau, \mathbf{r}, \mathbf{r}')$, where $\tau = t - t'$), but spatially inhomogeneous on the microscopic level due to a lattice periodicity. However, a model of a continuous medium can be arrived at in the classical optics after appropriate macroscopic averaging [14], resulting in $\varepsilon_{ij}(\tau, \mathbf{r}, \mathbf{r}') = \varepsilon_{ij}(\tau, \mathbf{R})$, where $\mathbf{R} = \mathbf{r} - \mathbf{r}'$.

When the IC modulation is present, there exists an extra inhomogeneity whose period is not commensurate with respect to the underlying lattice. It can be therefore shown that the IC crystal should be regarded as inhomogeneous already on the macroscopic scale. One may treat this situation in terms of additional dependence of the kernel $\varepsilon_{ij}(\tau, \mathbf{R}, \mathbf{r})$ on the coordinate \mathbf{r} [15]. Another equivalent approach suggested by Shvaika *et al* [10] and Stasyuk and Shvaika [11] (see also Janner and Janssen [16]) is to describe the inhomogeneity imposed by a periodic modulation via a formal dependence of the optical parameters on the phase φ of the modulation wave. Then the equation (1) may be modified to

$$D_i(t, \mathbf{r}, \varphi) = \iiint \varepsilon_{ij}(\tau, \mathbf{r}, \mathbf{r}', \varphi, \varphi') E_j(t', \mathbf{r}', \varphi') dt' d\mathbf{r}' d\varphi' \quad (2)$$

where a possible non-locality in φ is taken into account. Making a standard Fourier transformation of (2), one has

$$D_i(\omega, \mathbf{r}, \varphi) = \iint \varepsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}', \varphi, \varphi') E_j(\omega, \mathbf{r}', \varphi') d\mathbf{r}' d\varphi' \quad (3)$$

with

$$\varepsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}', \varphi, \varphi') = (2\pi)^{-1} \int \varepsilon_{ij}(\tau, \mathbf{r}, \mathbf{r}', \varphi, \varphi') e^{i\omega\tau} d\tau. \quad (4)$$

In what follows we shall disregard the influence of frequency dependence of the permittivity tensor on the optical properties of the IC crystals, paying instead closest attention to the spatial dispersion effects.

The tensor $\varepsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}', \varphi, \varphi')$ should be invariant with respect to the transformations included in the corresponding superspace symmetry group of the IC phase (see, e.g. [16]). In particular, for the translation subgroup of the latter one has

$$\varepsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}', \varphi, \varphi') = \varepsilon_{ij}(\omega, \mathbf{r} + \mathbf{n}, \mathbf{r}' + \mathbf{n}, \varphi + \mathbf{q}_{IC}\mathbf{n}, \varphi' + \mathbf{q}_{IC}\mathbf{n}) \quad (5)$$

where \mathbf{q}_{IC} is the IC modulation wave vector, $\mathbf{n} = l_i \mathbf{a}_i$ the translation vectors of the underlying lattice of the parent phase, \mathbf{a}_i the three basic lattice vectors and l_i integers.

The dielectric function that answers the demand (5) may be written in the form (see also Agranovich and Ginzburg [14])

$$\varepsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}', \varphi, \varphi') = \sum_{n_i, m} f_{ij}^{n_i, m}(\omega, \mathbf{R}, \Delta\varphi) e^{im\varphi'} e^{-i\mathbf{h}\cdot\mathbf{r}'} \quad (6)$$

where $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, $\Delta\varphi = \varphi - \varphi'$ and the summation is performed over all the Fourier vectors \mathbf{h} occurring in the crystal structure when the IC modulation is taken into account [3]:

$$\mathbf{h} = n_i \mathbf{a}_i^* + m \mathbf{q}_{IC}. \quad (7)$$

In formula (7) \mathbf{a}_i^* denote the basic reciprocal lattice vectors, and n_i and m are integers. Note that the IC modulation in the A_2BX_4 group crystals is one-dimensional. According to crystallographic orientation adopted in the work [3], it occurs along the \mathbf{a}_3 axis, so that the wave vector \mathbf{q}_{IC} may be represented as follows:

$$\mathbf{q}_{IC} = \gamma \mathbf{a}_3^* \quad \gamma = r/s + \delta \quad (8)$$

where the integer numbers r and s characterize the crystal structure of the commensurate lock-in phase (the corresponding wave vector $\mathbf{q}_C = (r/s)\mathbf{a}_3^*$), and $\delta \ll 1$ is a small irrational incommensurability parameter which depends on temperature [1].

Passing from the real (coordinate) space to the Fourier one, one obtains the transform of the equation (3),

$$D_i(\omega, \mathbf{k}, \varphi) = \iint \varepsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}', \varphi, \varphi') E_j(\omega, \mathbf{k}', \varphi') d\mathbf{k}' d\varphi' \quad (9)$$

with \mathbf{k} denoting the wave vector of light, and

$$\varepsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}', \varphi, \varphi') = (2\pi)^{-6} \iint \varepsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}', \varphi, \varphi') e^{i(\mathbf{k}'\mathbf{r}' - \mathbf{k}\mathbf{r})} d\mathbf{r} d\mathbf{r}'. \quad (10)$$

Substituting formula (6) into (10) yields

$$\varepsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}', \varphi, \varphi') = \sum_{n_i, m} \varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) \delta(\mathbf{k}' - \mathbf{k} - \mathbf{h}) e^{im\varphi'} \quad (11)$$

where $\delta(\mathbf{k})$ is the Dirac delta function, and

$$\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) = (2\pi)^{-3} \int f_{ij}^{n_i, m}(\omega, \mathbf{R}, \Delta\varphi) e^{-i\mathbf{k}\cdot\mathbf{R}} d\mathbf{R} \quad (12)$$

the microscopic Fourier components of the dielectric permittivity tensor. They are understood to have the periodicities $\lambda_h = 2\pi/|\mathbf{h}|$ related to the corresponding reciprocal lattice vectors \mathbf{h} . In agreement with the comment presented above, formulae (11) and (12) testify that the inhomogeneity of the overall tensor $\varepsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}', \varphi, \varphi')$ is owing to both the lattice and the IC superlattice periodicities.

The result (11) differs from the corresponding relation [14] for the crystals without the IC superstructure by the additional exponential factor. In the limiting case of a commensurate, purely lattice inhomogeneity ($\mathbf{q}_{IC} = \mathbf{q}_C$, $\delta = 0$ and $\mathbf{h} = n_{0i} \mathbf{a}_i^*$ —see formulae (7), (8)) the

phase of the modulation is the same at each atom. Then the latter factor becomes unity owing to the obvious equality $\exp(i\mathbf{a}_i^* \cdot \mathbf{a}_i) = 1$, and we have the well known relation [14]

$$\varepsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}') = \sum_{n_{0i}} \varepsilon_{ij}^{n_{0i}}(\omega, \mathbf{k}) \delta(\mathbf{k}' - \mathbf{k} - n_{0i} \mathbf{a}_i^*) \quad (13)$$

where the summation is now performed over the reciprocal lattice vectors \mathbf{h}_0 of the non-modulated crystal.

Let us now ascertain the properties of the Fourier components $\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi)$ associated with the symmetry principle for the kinetic coefficients and other general considerations [14]. The attempt looks somewhat artificial, because the symmetry properties of the macroscopic dielectric tensor are only of importance. Consideration of symmetry relations for the microscopic $\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi)$ components may be, however, justified by a need of comparison with the results known for the macroscopic tensor and clarification of their origin.

The Onsager symmetry principle for the case of a nonmagnetic IC crystal and the absence of external low-frequency (or static) magnetic field postulates that the kernel of the integral operator in (2) or (3) should satisfy (cf [14, 17]):

$$\varepsilon_{ij}(\tau, \mathbf{r}, \mathbf{r}', \varphi, \varphi') = \varepsilon_{ji}(\tau, \mathbf{r}', \mathbf{r}, \varphi', \varphi). \quad (14)$$

On the basis of formulae (6), (12) this leads to

$$\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) e^{im\varphi'} = \varepsilon_{ji}^{n_i, m}(\omega, -\mathbf{k} - \mathbf{h}, -\Delta\varphi) e^{im\varphi}. \quad (15)$$

A real physical field \mathbf{E} should induce a real electric displacement \mathbf{D} . A necessary and sufficient condition for that is a real value of the kernel $\varepsilon_{ij}(\tau, \mathbf{r}, \mathbf{r}', \varphi, \varphi')$ [14]. When using formulae (4), (6) and (12), one can prove that

$$\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) = [\varepsilon_{ij}^{-n_i, -m}(-\omega^*, -\mathbf{k}^*, \Delta\varphi)]^* \quad (16)$$

where * denotes complex conjugation. Considering a lossless optical medium (i.e. real values of ω and \mathbf{k}), we have

$$\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) = [\varepsilon_{ij}^{-n_i, -m}(-\omega, -\mathbf{k}, \Delta\varphi)]^*. \quad (17)$$

Then taking into account that $\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi)$ is an even function of frequency (see, e.g. formula (4)) and combining (15) and (17) give a final relation

$$\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) e^{im\varphi'} = [\varepsilon_{ji}^{-n_i, -m}(\omega, \mathbf{k} + \mathbf{h}, -\Delta\varphi) e^{-im\varphi}]^*. \quad (18)$$

Formula (18) shows that the microscopic components of the dielectric tensor of transparent incommensurately modulated crystals are not Hermitian, in contrast to the well known situation with the macroscopic dielectric tensor [17]. As can be easily seen, the same result equally refers to the microscopic dielectric tensor of the non-modulated crystals with the lattice periodicity alone. It should be therefore reasonable to attribute the above result to inhomogeneity of crystals on the microscopic scale (the presence of non-zero microscopic indices n_i and m in (18)). For the case of a classical commensurate crystal optics it is known [14] that a proper procedure of macroscopic averaging allows one to express all the microscopic Fourier components $\mathbf{E}(\mathbf{k} + \mathbf{h}_0)$ of electric field in terms of the macroscopic field $\mathbf{E}(\mathbf{k})$ and thus reduce the dielectric tensor to the $\varepsilon_{ij}^0(\mathbf{k})$ component concerned with the reciprocal lattice vector $\mathbf{h}_0 = \mathbf{0}$ alone. According to (18), this would provide a Hermitian character of the macroscopic permittivity tensor.

3. Macroscopic dielectric tensor

3.1. Introduction of macroscopic dielectric tensor

Our next step is to discuss the most important points of the corresponding macroscopic averaging in the case of crystals with the IC phases (see also [12, 14]). We start from substituting formula (11) into (9), which becomes

$$D_i(\omega, \mathbf{k}, \varphi) = \sum_{n_i, m} \int \varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) E_j(\omega, \mathbf{k} + \mathbf{h}, \varphi') e^{im\varphi'} d\varphi'. \quad (19)$$

In ordinary (commensurate) crystals all the vectors \mathbf{h}_0 are much longer than the typical wave vectors \mathbf{k} of the visible light ($\mathbf{k} + \mathbf{h}_0 \approx \mathbf{h}_0$), so that in the sum (19) the term with $\varepsilon_{ij}^{0,0}(\omega, \mathbf{k}, \Delta\varphi)$ labelled with zero microscopic indices, which characterizes a homogeneous optical material, will finally remain. For the IC crystals accounting for this term only implies adopting a lowest-order ‘space-averaged structure’ approximation (see, e.g. Dijkstra *et al* [12] and Kushnir [18]). The latter turns out to be too rough, since long enough superlattice periodicities can exist in the IC structure. That is why it is necessary to take into account the Fourier components $\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi)$ associated with relatively long-wavelength reciprocal lattice vectors $\mathbf{h} \neq \mathbf{0}$ [3, 12, 18]. In the plane-wave modulation region, the most important of these vectors is that determined by the small deviation of the soft-mode wave vector \mathbf{q}_{IC} from the corresponding vector \mathbf{q}_C of the lock-in phase, for it affects notably different physical properties of the IC phases (see [18–20]). It is introduced through the relations

$$\mathbf{q} = s(\mathbf{q}_{IC} - \mathbf{q}_C) = s\delta\mathbf{a}_3^*. \quad (20)$$

A comparison of (8) with (20) enables us to identify \mathbf{q} with the reciprocal lattice vectors $\pm\mathbf{h}$, of which the indices are equal to $n_1, n_2 = 0, n_3 = \mp r$ and $m = \pm s$. The modulation wavelength λ_q related to \mathbf{q} ($\lambda_q = a_3/(s\delta)$) exceeds by far the lattice parameters a_i though is still smaller than the light wavelength λ ($\lambda_q/\lambda \approx 10^{-2}$ to 10^{-1} —see e.g. [3, 12, 18]).

Keeping those points in mind, we distinguish the contributions of the vectors $\mathbf{h} = \mathbf{0}, \mathbf{q}$ among all the other microscopic dielectric components and rewrite (19) as

$$D_i(\omega, \mathbf{k}, \varphi) = \varepsilon_{ij}^{0,0}(\omega, \mathbf{k}) E_j(\omega, \mathbf{k}) + \varepsilon_{ij}^{\pm s}(\omega, \mathbf{k}, \varphi) E_j(\omega, \mathbf{k} \pm \mathbf{q}, \varphi) + \sum_{n_1, n_2; n_3 \neq \mp r; m \neq \pm s; n_i, m \neq 0} \varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi) E_j(\omega, \mathbf{k} + \mathbf{h}, \varphi') e^{im\varphi'} d\varphi' \quad (21)$$

where

$$\begin{aligned} \varepsilon_{ij}^{0,0}(\omega, \mathbf{k}) &= \int \varepsilon_{ij}^{0,0}(\omega, \mathbf{k}, \Delta\varphi) d\varphi' \\ \varepsilon_{ij}^{\pm s}(\omega, \mathbf{k}, \varphi) &= \int \varepsilon_{ij}^{\pm s}(\omega, \mathbf{k}, \Delta\varphi) e^{\pm is\varphi'} d\varphi'. \end{aligned} \quad (22)$$

Note that the term $\varepsilon_{ij}^{\pm s}(\omega, \mathbf{k}, \Delta\varphi)$ in (22) does not contain any microscopic indices n_i associated with the underlying lattice periodicities (see formula (20)). The same is also true of the component $\varepsilon_{ij}^{\pm s}(\omega, \mathbf{k}, \varphi)$ in formula (21). The non-locality dependent on the phase of the modulation wave, which very much complicates the constitutive equation (19), has been removed from (21) in the following way. We have expanded the field $\mathbf{E}(\omega, \mathbf{k} \pm \mathbf{q}, \varphi')$ in the r.h.s. of (19) in a Taylor series around the point φ and restricted ourselves to the zero-order approximation (i.e. $\mathbf{E}(\varphi') \approx \mathbf{E}(\varphi)$). One can substantiate that the accuracy of the approximation remains fairly high, since we have neglected the terms of the order of a_i/λ , while the component $\varepsilon_{ij}^{\pm s}(\omega, \mathbf{k}, \Delta\varphi)$ itself includes already small modulation-dependent terms of the same order of magnitude.

Following the procedures suggested by Agranovich and Ginzburg [14] (see also Dijkstra *et al* [12]), one can arrive at a macroscopic material equation. Namely, having neglected the transverse parts $\mathbf{E}_\perp(\omega, \mathbf{k} + \mathbf{h}, \varphi)$ of the short-wavelength microscopic fields $\mathbf{E}(\omega, \mathbf{k} + \mathbf{h}, \varphi)$ (the corresponding accuracy being of the order of $(a_i/\lambda)^2$, since $\mathbf{k} + \mathbf{h} \approx \mathbf{h}$ for the reciprocal lattice vectors with the indices $n_i \neq 0$; $n_3 \neq \mp r$; $m \neq 0 \pm s$) and expressed the longitudinal parts $\mathbf{E}_\parallel(\omega, \mathbf{k} + \mathbf{h}, \varphi)$ in terms of the fields $\mathbf{E}(\omega, \mathbf{k})$ and $\mathbf{E}(\omega, \mathbf{k} \pm \mathbf{q}, \varphi)$, we reduce formula (21) to the form

$$D_i(\omega, \mathbf{k}, \varphi) = \varepsilon_{ij}^0(\omega, \mathbf{k})E_j(\omega, \mathbf{k}) + \varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi)E_j(\omega, \mathbf{k} \pm \mathbf{q}, \varphi) \quad (23)$$

where $\varepsilon_{ij}^0(\omega, \mathbf{k})$ and $\varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi)$ include now the appropriate contributions from the last sum in (21), beside of the initial contributions $\varepsilon_{ij}^{0,0}(\omega, \mathbf{k})$ and $\varepsilon_{ij}^{\pm s}(\omega, \mathbf{k}, \varphi)$. The relation (23) represents the constitutive equation for the IC crystals in the Fourier space obtained by means of the macroscopic averaging. Now the analysis of some important consequences of this equation is straightforward.

3.2. Characterization of mesoscopic approximation

It is worth mentioning that the result similar to (23) has been obtained by Dijkstra *et al* [12]. Equation (23) corresponds to a semi-macroscopic (or ‘mesoscopic’, in terms of [12]) approximation in the description of optical properties of the IC phases, which takes into account both the ‘homogeneous’ macroscopic field $\mathbf{E}(\omega, \mathbf{k})$ and the mesoscopic field $\mathbf{E}(\omega, \mathbf{k} \pm \mathbf{q}, \varphi)$. The latter is characterized by spatial periods larger than the underlying lattice parameters but smaller than the light wavelength. That is why the mesoscopic approach is more refined when compared with the usual ‘macroscopic’ one, which should have retained only the first term in the r.h.s. of formula (23) governed by the point symmetry of spatially averaged structure of the IC crystal (cf also the conclusion by Pick [5]).

When substantiating equation (23), we have confined ourselves to considering a single mesoscopic modulation wavelength λ_q and a single dominant wave vector \mathbf{q} which corresponds to the reciprocal lattice vector $\mathbf{h} = (0; 0; \mp r; \pm s)$. This, by the way, means that we disregard in fact the soliton region of the IC phases where higher modulation harmonics become important. We have also neglected the contributions of the vectors \mathbf{h} with very high indices n_i and m . The latter procedure appears to be evident in the case of a classic crystal optics. Then increasing the indices n_{0i} is accompanied with decreasing a structural importance of the reciprocal lattice vectors, as is typically observed in morphological and x-ray-diffraction investigations, and the same is believed of the contribution of high-index Fourier components $\varepsilon_{ij}^{n_{0i}}(\omega, \mathbf{k})$ into the optical properties (see [3, 14]). Justification of this assumption for the IC crystals does not seem to be so obvious, despite the point of view in [3] and [6]. Indeed, from the entire set of the reciprocal lattice vectors it is possible to select a specific sequence of $\mathbf{h}_l = (0; 0; n_3; m)$, for which $\lim_{n_3, m \rightarrow \infty} (n_3/m) = -\gamma$ (see the explanations below formula (12)). Unlike the situation with the classical crystals, the length of those vectors decreases with increasing microscopic indices, and the corresponding modulation wavelength λ_{h_l} can be comparable with (or even larger than) the wavelength of the visible light [3]. This is why the importance of the ‘ultralong-wavelength’ vector \mathbf{h}_l represents a disputable problem.

In our opinion, the morphological importance of the reciprocal vectors depends upon their length rather than the indices themselves, and this should hold true for the IC crystals, too (see the results by Kremers *et al* [21] and Kremers [22]). Similar regularities are believed to take place also for the x-ray-diffraction reflections labelled with the vectors \mathbf{h} , including the satellite reflections ($m \neq 0$). So, it is known that ‘temperature quenching’ of the x-ray-diffraction intensity becomes more pronounced for the reflections with longer \mathbf{h} [23]. Although theoretical

studies of the importance of the IC-satellite crystal faces [21] showed a tendency for more stability of the faces with lower indices, the faces referring to the vector \mathbf{h}_l were not dealt with directly. Regarding the appropriate experimental results, both the morphological and the x-ray-diffraction characteristics associated with the ultralong-wavelength reciprocal lattice vectors \mathbf{h}_l still remain to be studied in much more detail. In any case, we have adopted the common point of view concerning a negligible contribution of \mathbf{h}_l to the optical response of the IC crystals. The additional reason for this is the analysis of the quantum-mechanical expression for the $\varepsilon_{ij}^{n_i, m}(\omega, \mathbf{k}, \Delta\varphi)$ components derived in a way similar to that used in [14]. These results are reported elsewhere [24].

3.3. Symmetry properties

Symmetry properties of the tensors $\varepsilon_{ij}^0(\omega, \mathbf{k})$ and $\varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi)$ imposed by Onsager's principle and the condition of transparency of the medium, may be clarified on the basis of (15), (18) and (21), (22) etc. For the homogeneous components we have the well known relations [14, 17]

$$\varepsilon_{ij}^0(\omega, \mathbf{k}) = \varepsilon_{ji}^0(\omega, -\mathbf{k}) \quad (24)$$

$$\varepsilon_{ij}^0(\omega, \mathbf{k}) = [\varepsilon_{ji}^0(\omega, \mathbf{k})]^* \quad (25)$$

while for the mesoscopic components

$$\varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi) = \varepsilon_{ji}^{\pm q}(\omega, -\mathbf{k} \mp \mathbf{q}, -\mathbf{k}, \varphi) \quad (26)$$

and

$$\varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi) = [\varepsilon_{ji}^{\mp q}(\omega, \mathbf{k} \pm \mathbf{q}, \mathbf{k}, \varphi)]^*. \quad (27)$$

Formula (27) testifies that, even in the case of a lossless optical medium, the small modulated contributions $\varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi)$ related to spatial dispersion in the IC crystal are not in general Hermitian. Naturally, the reason for the effect is again the inhomogeneity of the crystal on the mesoscopic scale originating from the IC character of structural modulation. Thus, the effect cannot be neglected unless we adopt the average structure approximation.

In our opinion, the just mentioned result is not really unusual. We should recall in this relation another fact known in electromagnetic theory of transparent non-modulated gyrotropic crystals, a weak non-orthogonality of the normal light waves [14]. Mathematically speaking, this means that the tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ is not self-conjugated (Hermitian), because the eigenvectors of a self-conjugated operator referred to different eigenvalues (in other words, refractive indices) must be mutually orthogonal (see, e.g. [25]). Restricting our attention to the first-order spatial dispersion (or the optical activity effect) only, we can write the dielectric permittivity in the Fourier space as [14]

$$\varepsilon_{ij}(\omega, \mathbf{k}) = \varepsilon_{ij}(\omega) + i\gamma_{ijl}(\omega)k_l \quad (28)$$

where the purely real tensors $\varepsilon_{ij}(\omega)$ and $\gamma_{ijl}(\omega)$ are, respectively, symmetric and anti-symmetric in the indices i and j . The tensor (28) appears to be Hermitian on inspecting its general form with formula (25). However, closer examination shows that, in the case of the spatial dispersion, $\varepsilon_{ij}(\omega, \mathbf{k})$ depends on the parameters of the normal waves (namely, their wave vectors $\mathbf{k}^{(1)} \neq \mathbf{k}^{(2)}$ which differ slightly due to different refractive indices). We have, e.g. $\varepsilon_{x,y}(\omega, \mathbf{k}) = i\gamma_{xyz}(\omega)k_z^{(1)}$ and $\varepsilon_{yx}(\omega, \mathbf{k}) = -i\gamma_{xyz}(\omega)k_z^{(2)}$ for the \mathbf{k} -dependent off-diagonal component, in contradiction with the formula (25). Thus, the effect of non-orthogonality of the normal waves arises from the fact that the dielectric permittivity of the optical medium with spatial dispersion, when written in a real coordinate space, is not simply a material constant but a differential operator which depends on the normal wave characteristics. In addition, the

dielectric permittivity of the IC crystals is not rigorously Hermitian owing to a mesoscopic inhomogeneity of the modulated medium.

3.4. Analysis of spatial dispersion phenomena

In order to understand better the consequences of the constitutive equation (23), it is convenient to expand $\varepsilon_{ij}^0(\omega, \mathbf{k})$ and $\varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi)$ in a series of \mathbf{k} (or, equivalently, in a series of $\mathbf{k} \pm \mathbf{q}$), maintaining only the linear term:

$$\varepsilon_{ij}^{\pm q}(\omega, \mathbf{k}, \mathbf{k} \pm \mathbf{q}, \varphi) = \varepsilon_{ij}^{\pm q}(\omega, \varphi) + i\gamma_{ijl}^{\pm q}(\omega, \varphi)k_l. \quad (29)$$

On averaging the influence of the IC modulation, the point group which describes symmetry of the IC phases in the A_2BX_4 crystals includes the inversion centre, so that the spatially homogeneous, linear in \mathbf{k} , terms in the dielectric tensor are forbidden ($\varepsilon_{ij}^0(\omega, \mathbf{k}) = \varepsilon_{ij}^0(\omega)$). At the same time, the inhomogeneous perturbations $\varepsilon_{ij}^{\pm q}(\omega, \varphi)$ and $\gamma_{ijl}^{\pm q}(\omega, \varphi)$ of the dielectric tensor imposed by the modulation are symmetry allowed [3, 12]. Therefore formula (23) modifies to

$$D_i(\omega, \mathbf{k}, \varphi) = \varepsilon_{ij}^0(\omega)E_j(\omega, \mathbf{k}) + [\varepsilon_{ij}^{\pm q}(\omega, \varphi) + i\gamma_{ijl}^{\pm q}(\omega, \varphi)k_l]E_j(\omega, \mathbf{k} \pm \mathbf{q}, \varphi). \quad (30)$$

Now let us pass with (30) to the coordinate space. In the following procedure, the most important fact is that the originals of the $\varepsilon_{ij}^{\pm q}(\omega, \varphi)$ and $\gamma_{ijl}^{\pm q}(\omega, \varphi)$ tensors should be spatially dependent. Therefore we deal in (30) with a product of two functions of the coordinate \mathbf{r} . For the arbitrary originals $f(t)$ and $g(t)$ and their Fourier transforms $F(x)$ and $G(y)$ one can write the relations (see, e.g. [26])

$$\begin{aligned} \iint F(x)G(y-x)e^{iyt} dx dy &= f(t)g(t) \\ \iint F(x)iyG(y-x)e^{iyt} dx dy &= \frac{d}{dt}[f(t)g(t)]. \end{aligned} \quad (31)$$

Making use of formulae (30), (31), with a formal correspondence $t \rightarrow \mathbf{r}$, $x \rightarrow \mp \mathbf{q}$, $y \rightarrow \mathbf{k}$, $F \rightarrow \varepsilon$, γ , and $G \rightarrow \mathbf{E}$, \mathbf{D} leads to the equation

$$D_i(\omega, \mathbf{r}, \varphi) = [\varepsilon_{ij}^0(\omega) + \varepsilon_{ij}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})]E_j(\omega, \mathbf{r}, \varphi) + \nabla_l[\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})E_j(\omega, \mathbf{r}, \varphi)] \quad (32)$$

where

$$\begin{aligned} E_i(\omega, \mathbf{r}, \varphi) &= \int E_i(\omega, \mathbf{k} \pm \mathbf{q}, \varphi) e^{i(\mathbf{k} \pm \mathbf{q}) \cdot \mathbf{r}} d(\mathbf{k} \pm \mathbf{q}) \\ \varepsilon_{ij}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r}) &= \int \varepsilon_{ij}^{\pm q}(\omega, \varphi) e^{\mp i\mathbf{q} \cdot \mathbf{r}} d(\pm \mathbf{q}) \end{aligned} \quad (33)$$

etc. For the reason of conciseness we do not write out the definitions for the originals of all the other material tensors and field vectors in (32). The equation (32) may be further specified after defining the exact spatial dependence of $\varepsilon_{ij}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})$ and $\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})$. For instance, when solving a practical problem of light propagation in the plane-wave modulation region of the A_2BX_4 group crystals, one can put $\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r}) = \gamma_{ijl}(\omega) \sin(\varphi_0 + \mathbf{q} \cdot \mathbf{r})$, where φ_0 has the meaning of the initial phase value (see [3, 12, 18]).

Note that spatial derivatives in (32) refer to both $\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})$ and $E_j(\omega, \mathbf{r}, \varphi)$. Thus, a consistent consideration of spatial dispersion in incommensurately modulated crystals leads to the conclusion that variation of the dielectric tensor in space should be accounted for directly in the constitutive equation. It should be mentioned that the result similar to (32) has already been obtained by Agranovich and Yudson [27], while considering the problems

of phenomenological electrodynamics of the gyrotropic media with sharp boundaries. In our case, however, the inhomogeneity is an intrinsic one, being concerned with long periodicities available in the structure of the IC crystals, rather than the sharp surface boundaries and the corresponding transition layer where the material tensors depend on spatial coordinates (see [27]).

Finally, let us elucidate more the meaning of the symmetry properties for the dielectric tensor derived in the previous section. Considering a simple plane electromagnetic wave field of the form $\mathbf{E}(\omega, \mathbf{r}, \varphi) = \mathbf{E}_0(\omega, \varphi + \mathbf{q} \cdot \mathbf{r}) \exp i\mathbf{k} \cdot \mathbf{r}$ propagating in the periodic modulated medium (see [18]), and the same for the electric displacement, one has instead of formula (32)

$$D_{0i}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r}) = \{\varepsilon_{ij}^0(\omega) + \varepsilon_{ij}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r}) + i\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})k_l + \nabla_l[\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})]\} \\ \times E_{0j}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r}) + \gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})\nabla_l[E_{0j}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})] \quad (34)$$

where the derivatives are meant to refer only to the expressions in square brackets. As pointed out above, the optical activity tensor $\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})$ is real and antisymmetric in its first two indices. Eventually, this can be strictly proved after analysing the equations (32) or (34) with the aid of the Onsager principle and the condition of absence of the radiation losses in the medium. Therefore the following relation holds true for the contribution to the dielectric permittivity in (34) proportional to \mathbf{k} :

$$\Delta\varepsilon_{ij}^I = i\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})k_l = (\Delta\varepsilon_{ji}^I)^*. \quad (35)$$

As seen from (35), the term $\Delta\varepsilon_{ij}^I$ is imaginary and antisymmetric in its first two indices and so Hermitian if we disregard the remarks quoted while discussing equation (28). All the other contributions occurring in (34) are also Hermitian, except for the term related to spatial derivatives of the gyration tensor. It satisfies the relation

$$\Delta\varepsilon_{ij}^{II} = \nabla_l[\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})] = -(\Delta\varepsilon_{ji}^{II})^* \quad (36)$$

being both real and antisymmetric in the i and j indices and thus showing anti-Hermitian behaviour. This clarifies, at last, the content of a potential possibility for existence of a non-Hermitian part in the dielectric function of an incommensurately modulated optical medium predicted by formula (27).

It is worthwhile to stress that the term in the constitutive equation containing $\nabla_l[\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})]$ has been disregarded in all the previous studies on the subject. At the same time, it cannot be excluded that the latter term plays a crucial role in the explanation of the macroscopic optical activity effect observed in a number of crystals of the A_2BX_4 group [3, 4, 7]. Indeed, in the plane-wave region of the IC phases the dielectric permittivity tensor will include a contribution $q_l\gamma_{ijl}(\omega) \cos(\varphi_0 + \mathbf{q} \cdot \mathbf{r})$ proportional to the mesoscopic wave vector \mathbf{q} (see subsection 3.4 and formula (34)). The amplitude of this contribution is at least order of magnitude larger than that of the term $ik_l\gamma_{ijl}(\omega) \sin(\varphi_0 + \mathbf{q} \cdot \mathbf{r})$ commonly accounted for within the classical theory of optical activity (see [14, 17]). We recall in this respect that the available theoretical models for the optical activity in the A_2BX_4 family crystals just have serious difficulties when explaining the magnitude of the effect observed in the experiments [3, 5, 6, 18]. Now one can see that neglecting the gradient term $\nabla_l[\gamma_{ijl}(\omega, \varphi + \mathbf{q} \cdot \mathbf{r})]$ for the IC phases is in no case admissible, even as a kind of a rough approximation. In fact, the presence of spatial derivatives of the gyration tensor in the material equation reveals an additional mechanism for the spatial dispersion effect in a periodic, mesoscopically modulated medium. This mechanism has to be evaluated by the parameter a_i/λ_q rather than the parameter a_i/λ , as in ordinary cases (see, e.g. [14, 17]). Thus the analysis performed in the present work proves once more a nontrivial origin of the optical activity in incommensurately modulated materials, as has been pointed out in [18, 28–30].

4. Conclusion

In this work we have shown that the IC modulation causes several extra peculiarities of the dielectric permittivity function which determines the optical response of the crystal to an electromagnetic wave. The material tensors of the incommensurately modulated medium, including the tensor that describes the spatial dispersion effect, should be regarded as depending on the phase of the modulation wave. The procedure of the macroscopic averaging substantiated by us for the case of the IC crystals demonstrates a need in accounting for the inhomogeneity of the medium on a mesoscopic scale. This semi-macroscopic inhomogeneity results in the appearance of a non-Hermitian part in the dielectric tensor even in transparent, lossless medium. The latter effect is demonstrated to originate from the term proportional to spatial derivative of the modulation-induced gyration tensor.

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