A local model based on the discrete dipole model has been developed in order to treat internal field effects at the surface of dielectric systems. The central part of the model is the double cell technique in which we match a freely chosen surface layer to the underlying bulk described by normal modes. We calculate the bulk and surface contributions to the anisotropic reflectance of the (110) surface of GaP obtaining results as good as the best delocalised treatments.

In order to test the validity of the double-cell technique we will calculate the internal field contribution to the surface induced optical anisotropy (SIOA) of the (110)-surface of GaP. SIOA has recently become an important tool in the investigation of semiconductor surfaces [6–14]. We will show that for a sufficiently large surface area, our results converge to those of the two-slab method [1] and we will see to what extent the bulk contributes to the anisotropy signal. As in [1] we need only three basic assumptions, or principles, to get started: the induction principle, the superposition principle and the parallel translation symmetry. All further steps are mathematical and the final solution is an exact one within the limits of the assumptions and numerical precision. The information about the physical system enters through a limited number of spatial coordinates locating the dipoles and an equal amount of polarisability tensors. As a natural result problems involving dielectric constants or boundary conditions will be avoided. Using the above mentioned principles in combination with the microscopic Hertz-potential formalism, yields for an arbitrary collection of dipole lattice planes, obeying the same parallel translation symmetry, the general description:

\[ P_i = \sum_j \left[ E_{ext,i} + \frac{1}{\varepsilon_0} \sum_j \vec{p}_j \cdot \vec{p}_i \right], \tag{1} \]

\[ \vec{p}_j = a^3 \sum_{\alpha \beta} \exp(ik\alpha \cdot \vec{r}_{\alpha \beta}) \frac{\varepsilon_\alpha \cdot \varepsilon_\beta \cdot \nabla \cdot \nabla \cdot S_j(\vec{r}, \vec{k})}{|\vec{r} - \vec{r}_{\alpha \beta}|}. \tag{2} \]

\[ S_j(\vec{r}, \vec{k}) = \sum_{n,m} \exp(ikj \cdot \vec{r}_{nm}) \frac{\varepsilon_\alpha \cdot \varepsilon_\beta \cdot \nabla \cdot \nabla \cdot S_j(\vec{r}, \vec{k})}{|\vec{r} - \vec{r}_{nm}|}. \tag{3} \]

\( \varepsilon_0 \) is a normalisation unit for the polarisability and has been defined as \( 4\pi\varepsilon_0 a^3 \). The prime in equation (3) denotes that the term with vanishing denominator has to be omitted. The coordinate system is chosen such that every lattice plane is oriented in the \( xy \)-direction and \( \hat{z} \) points in the direction of the upper half-space. The parallel translational symmetry is governed by two vectors \( \vec{s}_1 = a(1, 0, 0) \) and \( \vec{s}_2 = a(\alpha, \beta, 0) \) which span the lattice plane. We can then select an arbitrary number of lattice planes, each having a local origin at the characteristic site \( \vec{r}_j \). An arbitrary site is given by \( \vec{r}_{nm} = \vec{r}_j + ns_1 + ms_2 \) (\( n, m \) integers). We define the incident wavevector \( \vec{k} = (k_x, k_y, k_z) \) and its length \( k = |\vec{k}| \), required by the scalar planar lattice sums \( S_j(\vec{r}, \vec{k}), \vec{a} \cdot \vec{b} = a \cdot b \) the scalar improduct, \( \vec{a} \cdot \vec{b} \) the
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direct product tensor or dyad of two vectors \( \mathbf{a} \) and \( \mathbf{b} \),
and for any arbitrary vector \( \mathbf{u} = (u_z, u_r, -u_z) \) its
reflected counterpart \( \mathbf{u} \) as \( \mathbf{u} = (u_z, u_r, -u_z) \).

To the characteristic site \( r_j \) belong: the polariz-
ability tensor \( \mathbf{z}_{ij} \), the external field \( \mathbf{E}_{ext,j} \) and the charac-
teristic dipole strength \( \mathbf{p}_j \). Calculation of the interaction
tensors \( f_{ij} \) requires three different transformations of the \( S_j(r, k) \): interplanar far, interplanar near and intra-
planar. Far interplanar sums are used if \( |z_i - z_j| \) exceeds \( a \) and yield the \( f_{ij} \) according to Ewald's three-
fold integral transform [15]:

\[
\overline{f}_{ij} = \sum_{p,q} \overline{d}_{pq} \exp \left( i(k_{pq}(r_i - r_j)) \right), \quad (z_i > z_j)
\]

The validity of this

\[
d \overline{d}_{pq} = \frac{2\pi a}{\beta} \left( k^{10} - k_{pq} \right) \kappa_{pq},
\]

\( k_{pq} = \left( k_{pq}^b, \kappa_{pq} \right), \quad k_{pq} = (k^2 - K^{10})^{1/2}, \quad (4)
\]

with \( k^b_{pq} = k_i + g_{pq}^b \) and \( g_{pq}^b \) being a surface reciprocal lattice vector. If \( z_i < z_j \) one has to replace in equation

(4) \( \overline{d}_{pq} \) by \( \overline{d}_{pq} = \overline{d}_{pq}(k_{pq}) \). For the intraplanar and interplanar near (\( |z_i - z_j| \leq a \)) lattice sums one
needs a generalization of Ewald's onefold integral transform [1, 16]. The intraplanar sum governing the interaction of the characteristic dipole with its own plane will be written as \( \alpha_0^{-1} \). These integral trans-
forms have also been studied in [17, 18] and approxi-
mate expressions for \( \overline{c} \) can be found in [1, 19].

Next, two cells will be defined each obeying the same parallel translational symmetry. The surface cell
contains \( N_s \) characteristic dipoles to be chosen arbi-
trarily. This cell can take into account the influence of
surface reconstruction, relaxation and surface states.

The bulk cell, containing \( N_b \) characteristic dipoles, has

\( \mathbf{F} \) defines the number of normal modes of which only
2 lowest ones will be used. The \( q_m \)'s represent the
refracted wavevectors, the \( u_m \) the normal mode
strengths.

Values of \( q_m \) and \( u_m \) can be found through a generalization of Litzman's procedure [18, 20]. The computa-
tional effort reduces drastically if the bulk cell
can be reduced to the elementary bulk cell by means

\[
\mathbf{u}_m = e^{au_m^0} \mathbf{u}_m, \quad \mathbf{k}_m = (k_1, q_m).
\]

where \( \mathbf{u}_m \) is shorthand for \( \mathbf{u}_{m1} \). The validity of this
commensurability theorem will be discussed in [16].

The \( q_m \)'s follow from the secular determinant:

\[
det (\bar{a}(k_m)) = 0,
\]

and the normal mode strengths \( u_m \) from the secular equation:

\[
\overline{a}(k_m)u_m = 0.
\]

The explicit expression for \( \overline{a}(k_m) \) has been found to be
for the particular case of GaP(110):

\[
\overline{a}(k_m) = \alpha_0^{-1} \left[ \frac{\overline{a}_B}{\overline{a}_B} \right] \frac{a}{\alpha_3} - \sum_{v=1}^{2} e^{au_m^0} \overline{a}_{1v} + \sum_{v=1}^{2} \left( \frac{\overline{a}_{pq} e^{au_m^0}}{1 - \exp (i(q_m - \kappa_{pq})d_B)} \right) \frac{\overline{a}_{pq} e^{au_m^0}}{1 - \exp (-i(q_m + \kappa_{pq})d_B)} \right].
\]

Here \( \overline{a}_B \) represents the bulk polarizability tensor and
\( \overline{a}_{1v} \) represents the interaction tensor between the two
lattice planes of the original bulk cell. Continuing the
derivation we find two sets of equations: one set for
characteristic dipoles from the surface cell \( \mathbf{p}_j (j: 1, \ldots, N_s) \) and one for the coefficients of the 2
normal mode strengths \( v_m \). The last ones obey:

\[
- \sum_{j=1}^{N_s} e^{-ik_{ij}(\mathbf{g}_m \cdot \mathbf{d}_{ij})} d_{ij} - |\mathbf{g}_m|^2 v_m = \mathbf{g}_m E_0,
\]

\[
\mathbf{g}_m = \frac{d_{00} e^{-ik_{z}dz}}{1 - \exp (i(q_m - k_z)a)} \sum_{v=1}^{2} e^{iu_{m,v}k_{z}dz} u_m.
\]

Whereas a characteristic dipole belonging to the sur-
face cell gives rise to the equation:

\[
\sum_{j=1}^{N_s} (\overline{a}_z^{-1} \delta_{ij} - \overline{a}_0^{-1} \overline{a}_{1v}) \mathbf{p}_j - \sum_{m=1}^{M} \mathbf{g}_m v_m = E_i,
\]

\[
\mathbf{g}_m = \frac{2}{\gamma} \sum_{v=1}^{2} \sum_{p,q} \overline{d}_{pq} e^{iu_{m,v}k_{z}dz} u_m.
\]

Equations (7)-(8) yield a perfectly solvable system of
\((3N_s + 2)\) equations, which upon solution produces
all \( p_i \)'s and \( v_m \)'s. The essence of the method is to
increase \( N_s \) (by adding bulk planes) such that those 2
normal modes suffice for convergence. To obtain from
those solutions the semi-infinite reflection coefficients
is done by using:

\[ r = \left( \frac{i k^2}{2 \varepsilon_0 k_d a^2} \right) \left( \mathbf{E}_0 \cdot \mathbf{P}^d \right). \]

\[ \mathbf{P}^d = \sum_{j=1}^{N_2} e^{-i k_r p_j} + e^{i k_z d_2} \sum_{m=1}^{2} \left[ \sum_{w=1}^{2} \frac{e^{i(q_m + k)(r_m)}}{1 - \exp(i(q_m + k)a)} \right] v_m u_m. \]  

We will now specify the configuration for the particular case of the (110)-surface of GaP. The polarizable unit for bulk and surface contains one Ga atom and one P atom. We limit ourselves here to bulk-truncated GaP and we choose \( \alpha = 0, \beta = \sqrt{2} \) and \( \mathbf{d} = (a/2)(1, \beta, 1) \). The surface lattice plane generates all other planes in this case by repeatedly adding \( \mathbf{d} \). The bulk cell contains two characteristic dipoles and has height \( a \). The bulk lattice constant is \( a' = \beta a \) and has for GaP the value 5.4505 Å. The surface and bulk electronic structure of the semiconductor enter the calculation through the polarizabilities, which were derived from RPA surface and bulk dielectric constants [1, 21] using the Lorentz–Lorenz relation. Only the first three layers of the surface cell have polarizabilities different from the bulk. Surface reconstruction has been incorporated only by taking into account the rotation relaxation of 27.5° [22].

Equation (5) yields the two values for \( q_m \) which control the bulk behaviour. Since, they are essentially wavevectors, they can be compared to \( q_r \), the Fresnel wavevector. Figure 1 shows for both values of \( m \) the quantity \( |(q_m - q_r)/q_r| \). The result is apparently different from zero, but the deviations from Fresnel behaviour are very small in the optical region (of the order \( 10^{-5} \)), which proves that our normal mode decomposition is indeed capable of describing the optical response of the bulk within the discrete dipole approach and thus offers a discrete analogue of the usual continuum approaches [23, 24]. Furthermore, we also see in Fig. 1 that the two values of \( m \) produce a different curve, so even though we have isotropic bulk polarizabilities, the bulk internal fields give rise to an anisotropy. The reason for this is the following. Arguments about optically isotropic behaviour of cubic crystals rely upon simple static cubic symmetry. In reality however, this symmetry becomes distorted by the electromagnetic wave itself. As a result the orientation of the beam with respect to the bulk unit cell should play a role. From equation (5) it is immediately clear that this is indeed the case. In Fig. 2 we plot the anisotropy in the refractive index (\( \Delta n = n[\overline{1}0\overline{1}] - n[00\overline{1}] \)), as a function of the photon energy \( h\omega \). We find that for an energy of 1 eV the anisotropy is about \( 5.0 \times 10^{-6} \) or the same order of magnitude as the one experimentally observed for Si(110) in [4], proving that the phenomenon of birefringence in cubic crystals is indeed largely due to bulk internal field effects. In the discrete approach this phenomenon arises naturally; similar nonlocal descriptions are not so straightforward [5], because they need an expansion of the dielectric function \( \varepsilon(\omega, k) \) in terms of \( k \). Finally, as to the bulk behaviour we would like to comment on the mode strengths \( u_m \). The two optical normal modes \( u_m \) turn out to be polarized exactly along the (110) and (100) direction for perpendicular incidence. These modes are excited exclusively by electromagnetic waves of the same polarization, a fact which cannot be deduced from equation (5) as such.

In order to test the convergence behaviour of the double-cell method we calculated the reflectance anisotropy (RA) of the clean GaP (110)-surface as a function of the number of layers in the surface cell.
In Fig. 3. Convergence of the anisotropic reflectance of the (110)-surface of GaP. Plotted: $AR(N_s = \infty) - AR(N_s = N_s)$ for $N_s = 3, 4, 6$ and 7.

The RA is defined as $(R_{00} - R_0)/R_0$. The reflectances $R_{00}$ with respect to $R_0$ belong to polarizations of the incoming light beam along the (110) with respect to (0 0 1) direction, $R_F$ will be the classical Fresnel value. Figure 3 shows plots of the difference $(RA(\infty) - RA(N_s))$ for several values of $N_s$. The value $RA(\infty)$ corresponds to the $RA$ obtained for $N_s = 18$, whereas the $RA(N_s)$ have been obtained for values of $N_s$ varying from 3, 4, 6 through 7. It is clear that the effective range of the internal field has to be about 7 layers, most in agreement with the conclusions arrived at in [1].

Figure 4 shows the total RA for GaP(1 1 0). The results have been obtained by means of the double cell technique, using 18 dipoles in the surface cell. It appears that the bulk anisotropy effect (Fig. 2.) does not contribute substantially to the SIOA spectrum; the bulk signal is about 1% of the surface one. The same $AR$ has been calculated as well by means of the two slab method [1], but the results are virtually indistinguishable, confirming in yet another way the validity of the asymptotic continuation approach. Furthermore, we have obtained a substantial gain in processing speed compared to the two-slab method; the memory requirements are about 4 times smaller and the computing time is reduced by a factor 16.

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