

## Linear Optical Response in Silicon and Germanium Including Self-Energy Effects

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We calculate the optical response of silicon,  $\epsilon_1(\omega)$ , below the optical-absorption threshold and the static dielectric constant of germanium. The time-dependent local-density approximation (LDA) is modified by the addition of a self-energy term taken to be a "scissors operator,"  $P_{c\mathbf{k}}\Delta_{\mathbf{k}}$ . This form leads to a Ward-identity replacement  $\mathbf{p} \rightarrow (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}}^{\text{LDA}})^{-1}(\epsilon_{n\mathbf{k}} - H_{\mathbf{k}})\mathbf{p}$ . For silicon, we obtain 11.3 for  $\epsilon_1(\omega=0)$ , compared to 11.7 for experiment, 13.1 in our LDA calculation, and 8.4 in a naive self-energy-corrected theory (i.e., simply modifying the eigenvalues without modifying the momentum operator). For germanium, the corresponding values are 16.5, 15.8, 21.3, and 10.4.

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The most serious error in local-density-approximation (LDA) calculations of the band structure of semiconductors is the discrepancies between the Kohn-Sham eigenvalues<sup>1</sup> and the optical-absorption energies. The problem was resolved in the mid 1980's, first by the suggestion that the exchange-correlation functional was discontinuous for energies on two sides of the exchange-correlation gap,<sup>2</sup> and then by a new generation of *GW* calculations,<sup>3-5</sup> which have allowed agreement with experimental optical-absorption energies at the level of 0.1 eV.

Recently, high-quality LDA calculations of silicon<sup>6,7</sup> and related crystals<sup>7</sup> have become available. They indicate that the LDA overestimates the static dielectric constant of silicon by about 10% and some 25% in germanium.<sup>8</sup> This overestimate is related to the underestimate of the band gaps in the LDA, which is more severe in germanium than silicon. However, as noted by Hybertsen and Louie,<sup>7</sup> simply replacing the LDA eigenvalues by their *GW* or empirical counterparts leads to far too small a dielectric constant. A calculation by the empirical pseudopotential method<sup>9</sup>—which has the appropriate band gap built in—leads to the value of 9.0 for the static dielectric constant, compared to 11.7 for experiment.<sup>10</sup>

Since the *GW* theory depends upon a knowledge of the dielectric response, which in turn depends on the quasiparticle energies, one would hope that an improved knowledge of the quasiparticle energies could be used to improve the *GW* calculation itself. Up to the present, the practice has been to use the LDA or RPA dielectric response.

In this paper, we show how to properly include the many-body effect of a quasiparticle energy shift in the optical response and do so for silicon and germanium at the level of a scissors operator<sup>5,11</sup>  $\Delta_{\mathbf{k}}P_{c\mathbf{k}}$ , i.e., a  $\mathbf{k}$ -dependent energy shift in the conduction bands which does not change their wave functions. (Here,  $P_{c\mathbf{k}}$  is the projection operator onto all conduction bands at wave vector  $\mathbf{k}$ .) Since the quasiparticle wave functions calculated in the *GW* approximation<sup>3-12</sup> are in excellent agreement

(wave-function overlaps exceeding 0.999) with the LDA wave functions,<sup>4</sup> this is a promising *Ansatz*. The magnitude of the shift can be found semiempirically,<sup>13</sup> from a quasiparticle local-density calculation<sup>14</sup> or other simple models,<sup>15,16</sup> or from *GW* calculations.<sup>3-5</sup>

Self-consistent frequency-dependent response theory<sup>17</sup> including local-field corrections<sup>18</sup> and exchange-correlation effects<sup>19,20</sup> is known as the time-dependent local-density approximation (TDLDA). In this formulation, a number density is induced according to

$$\delta n(\mathbf{r};\omega) = \int d\mathbf{r}' \chi_0(\mathbf{r},\mathbf{r}';\omega) \phi^{\text{scf}}(\mathbf{r}';\omega), \quad (1)$$

where the self-consistent field  $\phi^{\text{scf}}(\mathbf{r}';\omega)$  is related to the external field  $\phi^{\text{ext}}(\mathbf{r};\omega)$  by

$$\phi^{\text{scf}}(\mathbf{r};\omega) = \phi^{\text{ext}}(\mathbf{r};\omega) + \phi^C(\mathbf{r};\omega) + \phi^{\text{xc}}(\mathbf{r};\omega). \quad (2)$$

Here,  $\phi^C(\mathbf{r};\omega)$  is the induced Coulomb potential

$$\phi^C(\mathbf{r};\omega) = e^2 \int d\mathbf{r}' \frac{\delta n(\mathbf{r}';\omega)}{|\mathbf{r} - \mathbf{r}'|}, \quad (3)$$

and  $\phi^{\text{xc}}(\mathbf{r};\omega)$  is the induced exchange-correlation potential

$$\phi^{\text{xc}}(\mathbf{r};\omega) = \frac{\delta V_{\text{xc}}(\mathbf{r})}{\delta n(\mathbf{r})} \Big|_{n(\mathbf{r})=n_0(\mathbf{r})} \delta n(\mathbf{r};\omega) \quad (4)$$

linearized about its ground-state value. Frequency-dependent corrections to this formula have been considered, but are believed to be small.<sup>21</sup> The choice of Eq. (3) is characteristic of the Coulomb gauge.<sup>22</sup>

The independent-particle susceptibility is given by

$$\chi_0(\mathbf{r},\mathbf{r}';\omega) = \sum_{ij} (f_i - f_j) \frac{\varphi_i^*(\mathbf{r})\varphi_j(\mathbf{r})\varphi_j^*(\mathbf{r}')\varphi_i(\mathbf{r}')}{\epsilon_i + \hbar\omega - \epsilon_j + i\eta}, \quad (5)$$

where the sum is taken over all single-particle states, the  $f_i$  are occupation factors, and  $\eta$  is a positive infinitesimal. We take each  $f_i$  to be 0 or 1. Fundamentally the energies and wave functions in Eq. (5) refer to the associated many-body states.<sup>19</sup> In TDLDA, these quantities are replaced by their LDA values; a modified

choice is made in this work.

Consider the case of a crystalline solid. Let  $\{\cdot\cdot\cdot\}$  denote the quantities which are periodic in the primitive direct lattice vectors of the crystal; for example, the electron density is in the form  $\delta n(\mathbf{r};\omega) = \Omega_0^{-1} e^{i\mathbf{q}\cdot\mathbf{r}} \{\delta n(\mathbf{r};\omega)\}$  with the periodic part of the electron density given by

$$\{\delta n(\mathbf{r};\omega)\} = \bar{\Omega}_0 \int_{\text{BZ}} d\mathbf{k} \sum_{n\pm}^{\text{occ}} \langle \{n\mathbf{k}\} | \mathbf{r} \rangle \langle \mathbf{r} | (\epsilon_{n\mathbf{k}} \pm \hbar\omega - H_{\mathbf{k}+\mathbf{q}})^{-1} \{\phi^{\text{scf}}(\omega)\} | \{n\mathbf{k}\} \rangle, \quad (6)$$

where  $H_{\mathbf{k}}$  is the “ $k$ -dot- $p$ ” Hamiltonian,<sup>23</sup> given in the LDA by

$$H_{\mathbf{k}}^{\text{LDA}} = \frac{1}{2} (\mathbf{p} + \mathbf{k})^2 + V_{\mathbf{k}} \quad (7)$$

in (Hartree) atomic units. In general,  $V_{\mathbf{k}}$  is nonlocal;<sup>6,7</sup> in the present work a nonlocal ionic pseudopotential is used.<sup>24-26</sup> The unit-cell volume is  $\Omega_0$ , and  $\bar{\Omega}_0 = \Omega_0 / (2\pi)^3$ . An expanded exposition will be given elsewhere.<sup>27</sup> In the presence of a self-energy term, the  $k$ -dot- $p$  Hamiltonian takes the form

$$H_{\mathbf{k}} = \frac{1}{2} (\mathbf{p} + \mathbf{k})^2 + V_{\mathbf{k}} + \Sigma_{\mathbf{k}}, \quad (8)$$

where  $\Sigma_{\mathbf{k}}$  is the self-energy operator with crystal momentum  $\mathbf{k}$ . We will be interested in small- $\mathbf{q}$  expansions; in

this limit longitudinal response theory (presented here) is equivalent to transverse, or optical, response.<sup>28</sup> Define

$$H_1 = \mathbf{q} \cdot (\mathbf{p} + \mathbf{k} + \nabla_{\mathbf{k}} V_{\mathbf{k}} + \nabla_{\mathbf{k}} \Sigma_{\mathbf{k}}), \quad (9)$$

and

$$H_2 = \frac{1}{2} q^2 + \mathbf{q} \cdot \nabla_{\mathbf{k}} [\mathbf{q} \cdot \nabla_{\mathbf{k}} (V_{\mathbf{k}} + \Sigma_{\mathbf{k}})]. \quad (10)$$

The modification of the momentum operator given by Eq. (9) is a Ward identity.<sup>29</sup>

The electron density and self-consistent fields of Eq. (6) can be broken into long-wave ( $\mathbf{G}=0$ ) and short-wave or local-field ( $\mathbf{G}\neq 0$ ) parts. The long-wave charge induced by a long-wave potential  $\phi_0 e^{i\mathbf{q}\cdot\mathbf{r}}$  is given from Eq. (6) by

$$\Omega_0^{-1} \int_0 d\mathbf{r} \{\delta n(\mathbf{r};\omega)\} = \phi_0 (\hbar\omega)^{-2} \bar{\Omega}_0 \int_{\text{BZ}} d\mathbf{k} \sum_{n\pm}^{\text{occ}} [\langle \{n\mathbf{k}\} | H_1 (\epsilon_{n\mathbf{k}} \pm \hbar\omega - H_{\mathbf{k}})^{-1} H_1 | \{n\mathbf{k}\} \rangle + \langle \{n\mathbf{k}\} | H_2 | \{n\mathbf{k}\} \rangle]; \quad (11)$$

related formulas describe the local-field corrections.<sup>27</sup> By perturbation theory,

$$\langle \{n\mathbf{k}\} | H_1 (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}})^{-1} H_1 | \{n\mathbf{k}\} \rangle + \langle \{n\mathbf{k}\} | H_2 | \{n\mathbf{k}\} \rangle = \frac{1}{2} \mathbf{q} \cdot \nabla_{\mathbf{k}} (\mathbf{q} \cdot \nabla_{\mathbf{k}} \epsilon_{n\mathbf{k}}). \quad (12)$$

Because of the periodicity of the eigenvalue, the integral of this expression over the Brillouin zone vanishes; this proves the  $f$ -sum rule for crystals<sup>30</sup> in the present context. Combining the integral of Eq. (12) with Eq. (11) leads to

$$\Omega_0^{-1} \int_0 d\mathbf{r} \{\delta n(\mathbf{r};\omega)\} = \phi_0 (\hbar\omega)^{-2} \bar{\Omega}_0 \int_{\text{BZ}} d\mathbf{k} \sum_{n\pm}^{\text{occ}} [\langle \{n\mathbf{k}\} | H_1 (\epsilon_{n\mathbf{k}} \pm \hbar\omega - H_{\mathbf{k}})^{-1} H_1 | \{n\mathbf{k}\} \rangle - \langle \{n\mathbf{k}\} | H_1 (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}})^{-1} H_1 | \{n\mathbf{k}\} \rangle], \quad (13)$$

from which the static limit<sup>6,7</sup>

$$\Omega_0^{-1} \int_0 d\mathbf{r} \{\delta n(\mathbf{r};0)\} = 2\phi_0 \bar{\Omega}_0 \int_{\text{BZ}} d\mathbf{k} \sum_n^{\text{occ}} \langle \{n\mathbf{k}\} | H_1 (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}})^{-3} H_1 | \{n\mathbf{k}\} \rangle \quad (14)$$

may be derived by a Taylor expansion in powers of  $\omega$ . Neither Eq. (13) nor the formulas for the local-field corrections depend explicitly on  $H_2$ .

We now invoke the scissors Ansatz  $\Sigma_{\mathbf{k}} = \Delta_{\mathbf{k}} P_{c\mathbf{k}}$ . This implies<sup>27</sup>

$$H_1 | \{n\mathbf{k}\} \rangle = (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}}^{\text{LDA}})^{-1} (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}}) H_1^{\text{LDA}} | \{n\mathbf{k}\} \rangle, \quad (15)$$

with  $H_1^{\text{LDA}} = \mathbf{q} \cdot (\mathbf{p} + \mathbf{k} + \nabla_{\mathbf{k}} V_{\mathbf{k}})$ . The self-energy  $\Sigma_{\mathbf{k}}$  has  $\mathbf{k}$  dependence through the projection operator  $P_{c\mathbf{k}}$  even if  $\Delta_{\mathbf{k}}$  is a constant. In the static limit, the self-energy-corrected static dielectric constant (without local-field correction) is given by

$$\Omega_0^{-1} \int_0 d\mathbf{r} \{\delta n(\mathbf{r};0)\} = 2\phi_0 \bar{\Omega}_0 \int_{\text{BZ}} d\mathbf{k} \sum_n^{\text{occ}} \langle \{n\mathbf{k}\} | H_1^{\text{LDA}} (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}}^{\text{LDA}})^{-1} (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}})^{-1} (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}}^{\text{LDA}})^{-1} H_1^{\text{LDA}} | \{n\mathbf{k}\} \rangle. \quad (16)$$

Thus, in contrast to a naive attempt to incorporate self-energy effects by the substitution  $(\epsilon_{n\mathbf{k}} - H_{\mathbf{k}}^{\text{LDA}})^{-3} \rightarrow (\epsilon_{n\mathbf{k}} - H_{\mathbf{k}})^{-3}$  while retaining  $H_1^{\text{LDA}}$  without modification in the LDA version of Eq. (14), we have shown that the net correction is to only one of the three energy denominators.

We have extended the plane-wave pseudopotential code of Allan and Teter<sup>25</sup> to include linear response. We solve the modified Sternheimer equation,<sup>31,32</sup> using iterative techniques; the solution is equivalent to a sum over all “virtual intermediate states.”<sup>27</sup> We use a separable, norm-conserving pseudopotential.<sup>24,33</sup> A 12-Ry energy cutoff was selected for this work, which is somewhat lower than the 14 Ry used in similar studies.<sup>6,7</sup> For germanium, we chose a 20-Ry cutoff,

TABLE I. Calculated values of the static dielectric constant for silicon. Sixty special points were chosen in all cases. The third line is from Eq. (14) with LDA operators; fourth line, from Eq. (14) with  $H_1 = H_1^{\text{LDA}}$  and  $H_{\mathbf{k}} = H_{\mathbf{k}}^{\text{LDA}} + \Delta_{\mathbf{k}}$ ; fifth line, from Eq. (16).  $\epsilon_{00}$  represents the calculation without local-field corrections (Ref. 17);  $\epsilon_M^{\text{RPA}}$  excludes  $\phi^{\text{sc}}$  of Eq. (4);  $\epsilon_M$  includes LDA values, self-energy-corrected calculations, and experiment. The ideal value for the  $f$  sum is unity. The  $f$ -sum integral is identical for the LDA and the present model.

	Silicon				Germanium		
	$\epsilon_{00}$	$\epsilon_M^{\text{RPA}}$	$\epsilon_M$	$f$ sum	$\epsilon_{00}$	$\epsilon_M$	$f$ sum
BR LDA <sup>a</sup>	13.4	12.0	12.7				
HL LDA <sup>b</sup>	13.6	12.2	13.0		21.9	20.7	
Present LDA	13.8	12.4	13.1	1.013	22.0	21.3	0.993
"Naive," $\Delta_{\mathbf{k}} = 0.7$ eV	8.8		8.4	0.887	10.8	10.4	0.857
Present, $\Delta_{\mathbf{k}} = 0.7$ eV	11.8		11.3	1.013	17.0	16.5	0.993
Expt. <sup>c</sup>			11.7			15.8	

<sup>a</sup>Baroni and Resta, Ref. 6.

<sup>b</sup>Hybertsen and Louie, Ref. 7.

<sup>c</sup>Reference 10.

which is the same as in Ref. 7. We slightly exceed these studies' published values in the calculation of  $\epsilon_{00}$ ,  $\epsilon_M^{\text{LDA}}$ , and  $\epsilon_M^{\text{RPA}}$  for both ten and sixty special points;<sup>34</sup> our results are 0.3–0.4 units in  $\epsilon$  above the work of Baroni and Resta<sup>6</sup> and 0.1–0.6 units above the work of Hybertsen and Louie.<sup>7</sup> (The symbol  $\epsilon_M$  denotes the macroscopic dielectric constant, which may be compared to experiment.) Integration of all terms other than  $\frac{1}{2}q^2$  appearing in the  $f$ -sum rule Eq. (12) yields values of 1.031 (1.045) with ten special points and 1.013 (0.993) with sixty special points for silicon (germanium), relative to a unit normalization. It is numerically critical to use Eq. (13) rather than Eq. (11), to avoid a huge error in  $\epsilon$ ,  $4\pi("f$  sum" - 1)  $(\hbar\omega/1H)^{-2} \geq 16$  for  $\hbar\omega \leq 2.7$  eV.

We chose  $\Delta_{\mathbf{k}} = 0.7$  eV, independent of  $\mathbf{k}$ , based on a suggestion by Godby, Schlüter, and Sham; this introduces errors at the level of 0.1 eV for silicon and 0.2 eV for germanium.<sup>5</sup> The static calculations are summarized in Table I. As can be seen, the self-energy correction improves agreement with the experimental values.

In Fig. 1, we present the dielectric response for silicon in the frequency range 0–2.7 eV. Local-field effects impose a significant, if featureless, correction; more significant corrections are to be expected in the presence of absorption resonances.<sup>35</sup> The self-energy-corrected theory results in a curve which starts 4% below the experiment and rises somewhat too gently; by contrast the TDLDA results start some 12% above the experimental value and rise rather too steeply. The error reduction is about a factor of 3. A reduction in the scissors constant from the value chosen 0.7 to about 0.55 eV would considerably improve agreement with experiment; increasing the energy cutoff will affect the eigenvalue spectrum at the 0.1-eV level.<sup>5</sup> A host of other effects, such as error in the Brillouin-zone integration, core polarizabilities, phonon corrections,  $GW$  vs LDA wave-function difference,  $\mathbf{k}$  dependence of the scissors operator, corrections

to the linearized exchange-correlation assumption of Eq. (4), transverse versus longitudinal response, and deviations of the pseudopotential phase shifts from the LDA phase shifts, are each expected to enter at the level of 0.1%–2%. Also significant is our neglect of the renormalization constant and the diffuse part of the spectral response; in our approximation, oscillator strength properly belonging to the background is placed in the quasi-particle peaks.

We have shown how to include the effect of a self-energy operator in self-consistent-field response theory. We have given a new, explicit formula to compute the effect of the self-energy on the response in the important case that the self-energy operator has a scissors form.

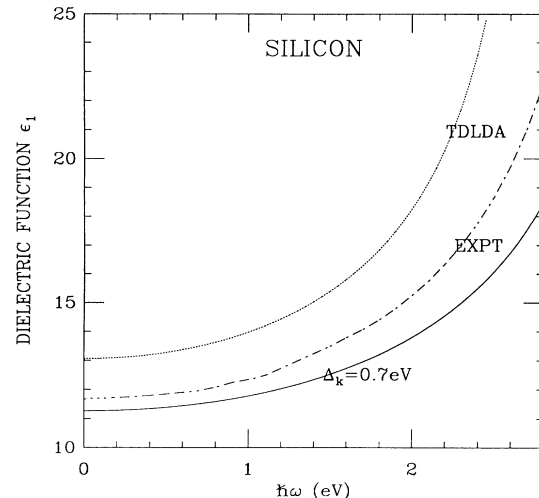


FIG. 1. The longitudinal dielectric response for silicon  $\epsilon_1(\omega)$  below the direct gap. The dotted line is the TDLDA with the local-field correction. The solid line is the present self-energy-corrected theory for  $\Delta_{\mathbf{k}} = 0.7$  eV with the local-field correction. The chain-dotted line is from experiment (Ref. 10).

We have performed a well-converged self-energy- and local-field-corrected response calculation for silicon, as well as a comparison TDLDA calculation (itself the first well-converged calculation for any crystalline solid). The correction proposed here considerably improves the agreement with experiment.

The static dielectric constant is usually regarded as a ground-state property, and therefore amenable to explanation by density-functional theory. The LDA is thought to be a reasonably good implementation thereof.<sup>5</sup> By contrast, in our formulation, we invoke excited-state properties, properties regarded as not being a part of the density-functional theory,<sup>2</sup> in an essential way to understand the dielectric constant.

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