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SiC: linear and nonlinear optical properties of four polytypes

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We calculated ε_∞ and second-order nonlinear coefficients $d = \chi^{(2)}/2$ for four SiC polytypes as well as ε_∞ for diamond within the LDA and using the LDA wave functions with a self-energy correction in the form of a "scissors operators" $\Delta P_{c\vec{k}}$ to adjust the band gaps. For all the wide band gap materials we have studied so far, we find that the Δ needed to reproduce the optical response properties is less than the Δ deduced by GW calculations or needed to reproduce the experimentally observed band gaps. This is in contrast to the small to medium gap semiconductors we studied previously. For nH SiC with $n \geq 4$, our predictions for d are close to the only experimental data. Our calculations show that both d and the local-field corrections to d for nH SiC form a trend as $n \to \infty$. The d_{113}/d_{333} ratios we predicted for different SiC polytypes differ from those of bond-charge-model predictions.

I. INTRODUCTION

SiC is the only IV-IV compound known to form stable long-range ordered structures.[1] Over 200 polytypes have been observed among which the 6H polytype is predominant. One form of polytype, 3C, is cubic, exhibiting the zincblende structure (space group T_d^2-F$\bar{4}$3m). Another form, 2H, has the wurtzite structure (space group C_{6v}^4-P6_3mc). All other polytypes have either a hexagonal unit cell (with $c/a > 2$) or a rhombohedral unit cell (space group C_{3v}^5-R3m).[2] In all polytypes, every atom is tetrahedrally surrounded by four atoms of the other species. The crystal structure may be regarded as a close packing of atoms of any one kind, say Si, with the other (C) atoms occupying half of the tetrahedral voids. The structure for 3C (zincblende) corresponds to the hexagonal close packing with a repeat unit of ABC. For 2H (wurtzite) the repeat unit is AB; for 4H it is ABCB; and for 6H it is ABCACB.

In all SiC polytypes, the chemical bonds between the Si and C atoms are the same. So it is an interesting problem to find how the packing order of the atomic layers alone can affect different physical properties, such as the dielectric constants ε_∞ and the second-order nonlinear susceptibilities $d = \chi^{(2)}/2$ which are studied in this paper. The experimental dielectric constants[1] show small changes from one polytype to another, which is readily captured by our calculations. The bond-charge models are, in principle, particularly powerful on a crystal with a single type of bond. The assumption that all the bonds contribute in an additive fashion constrains the ratios of the components of d. Our calculations are not consistent with this picture. Some of our calculated d values for different SiC polytypes do agree with the only data available on SiC that was measured from an unknown polytype.[3] In particular, our 6H and ∞H SiC values of d are within the experimental uncertainty region, and our 4H values are plausible.

In our calculations, we consider both the Kohn-Sham local density approximation[4] (LDA) as well as the LDA with a self-energy correction in the form of a "scissors-operator".[5]

The new Hamiltonian takes on the form

$$H_{\vec{k}} = H_{\vec{k}}^{LDA} + \Delta P_{c\vec{k}}, \tag{1}$$

in which $P_{c\vec{k}}$ is the projection operator on to the conduction bands and Δ is the constant energy shift. Surprisingly, the shift of the conduction bands leads modifies the velocity operator in the matrix elements as $\vec{p}+\vec{k} \rightarrow \vec{p}+\vec{k}+\nabla_{\vec{k}}(\Delta P_{c\vec{k}})$. The issue of gauge invariance of the Hamiltonian of Eq (1) has been considered recently.[6]

The motivation for the "scissors-operator" approximation comes from GW calculations.[7,8] A simple (read: moderately accurate and subject to corrections) way to represent the results of these calculations is by assuming the eigenvalues are subject to a rigid shift, but the quasiparticle wave functions are unchanged. It is a common approximation within the GW community to assume the LDA wave functions are adequate representations of the quasiparticle states.[7,9,10] Our point of view is that we are building the best one-particle Green's function that we can short of undertaking a full GW calculation. The fact that we rely on wave-function methods is an incidental feature of the calculation; the equivalence of a formulation in terms of wave functions and in terms of independent particle Green's functions is well known.[11] Following the practice within GW theory,[10] all calculations are performed at the experimental lattice constant.

The need for a shift of the LDA spectrum for the interpretation of optical response was noted over a decade ago by Wang and Klein.[12] The approximation of Eq. (1) has been used to calculate the dielectric constants of Si,[13,14,15] Ge,[13,15] AlP,[5] AlAs,[5] GaN,[15] GaP,[5] GaAs,[5,15] Se,[16] α-quartz[16,17] and stishovite.[18] Volume or pressure dependence has been calculated for Si,[13,14,15] Ge,[13,15] AlP,[5] AlAs,[5] GaP,[5] and GaAs.[5,15] Frequency-dependence of the dielectric constant has been considered within the approximation for Si,[13] Ge,[13] GaP,[5] GaAs,[5] Se,[16] and α-quartz.[16] All photoelastic tensor components have been presented for Si.[14] Second-harmonic susceptibilities have been given for AlP,[5] AlAs,[5] GaP,[5] GaAs,[5] Se,[16] and α-quartz[16] including frequency-dependence for GaP[5] and GaAs.[5] In the vast majority of cases, the results have been very encouraging: the use of a shift in the LDA eigenvalues suggested by GW calculations typically leads to a calculated value within experimental error bars or a few per cent of the experimental value. The results have been recently reviewed by one of us.[19] Another form of the scissors operator has been used for the calculation of second-harmonic susceptibilities by Huang and Ching.[20]

We provide an extensive list of the various successes of the "scissors" approximation to suggest that a systematic failure of this approximation is interesting. In contrast to the small to medium gap semiconductors, we found that for wide gap semiconductors we studied in this paper — diamond and 2H, 4H, 6H, and 3C SiC — the scissors operators needed to reproduce the experimental dielectric constants are less than the Δ needed to match the band gaps. This is also true for the only other wide gap material we studied, namely urea.[21]

The use of Eq. (1) for linear optical response calculations has been criticized recently by Dal Corso, Baroni, and Resta.[22] These authors argue that the well-known underestimate of the Local Density Approximation of the optical band gaps of solids is irrelevant because the Kohn-Sham eigenvalues should not be associated with optical transition energies and indeed are unphysical quantities. Instead, they suggest that to improve upon LDA's ability to predict the static dielectric constants (ε_∞) of solids one should work within the density-functional theory but with an improved approximation to the exchange-correlation energy (and more specifically, its second functional derivative with respect to the electron density). These authors propose a gradient-corrected density-functional theory for the static dielectric constant, and calculate it for the case of silicon. Some improvement over the LDA value compared to experiment is demonstrated.[23]

Dal Corso *et al.* suggest that to go beyond density-functional theory requires the calculation of a two-particle Green's function. Here, our point of view differs. Our belief is that such a calculation would be desirable, but is too large for current computers. Eq. (1) is an attempt to approximate the single-particle Green's function and to calculate linear and second-harmonic response within that formalism.

The linear response calculations were preparatory to the main goal of our present study, the calculation of the second-order nonlinear susceptibilities d for different SiC polytypes. In view of the linear response data, we decided our best guess for the second-harmonic susceptibility would be given by taking the self-energy correction Δ to be a parameter to be adjusted to the fit ε_∞. In a previous study of an insulator, α-quartz, the use of Δ to match the static dielectric constant resulted in an excellent account of d_{111}.[16] We urge and welcome experimental measurements of $\chi^{(2)}$ for the SiC polytypes. Nevertheless, we present three sets of results throughout the paper: LDA, band-gap-matching Δ_{gap}, and ε_∞-matching Δ_ε. The qualitative features of the calculation are not affected by the self-energy shift, only the calculated numerical values.

II. RESULTS FOR OPTICAL RESPONSE

Before studying SiC, we first calculated ε_∞ of diamond.[24] Our LDA calculations for diamond — with and without local-field corrections — agreed with a previous calculation,[25] suggesting an overall reliability of our pseudopotential and other aspects of the calculation. However, we were surprised to find that adjusting the gap from the LDA value to the experimental one, using the scissors-operator approach (Δ=1.8 eV), failed to reproduce the experimental ε_∞ as shown in Table I. This is in contrast to the work for a large class of small to medium gap semiconductors listed in the introduction. In diamond, we need increase the energy gap to only 84% of the experimental gap from the 77% given by the LDA. To deduce this, we did a linear interpolation using the computed ε_∞ at Δ=0 and 0.9 eV. As a check on the linearity, we found that ε_∞ obtained with Δ=0.9 eV differs by less than 1% from the linear interpolation with Δ=0 and 1.8 eV. The value Δ=1.8 eV for diamond may be obtained by averaging Δ_k over the points M, X, and L from either of two GW calculations.[7,8] Another recent GW calculation[26] leads to a somewhat larger average Δ value (2.2 eV), which would only make the problem more extreme.

In wide-gap materials, for which until this paper we had only the example of urea,[21] there seem to be a "gap problem", that the effective gap to reproduce ε_∞ is less than the experimentally observed gap. (Although we have also studied α-quartz, the band gap is not well defined in this case.[27]) In the previous paragraph, we found this "gap problem" in diamond and we will see it for the SiC polytypes too. It may be generic in wide band gap materials.

As a first step to calculate the optical response of different polytypes of SiC, we studied the Δ dependence of ε_∞ and d in the case of 2H SiC; we found that they are almost linear in Δ in the range studied, $0 \leq \Delta \leq 1.5$ eV. As shown in Fig. 1, by linear interpolation using the computed values at Δ=0 and Δ=1.5 eV, we introduced an error of less than 1% for ε_∞ and less than 5% for d as compared to the computed values at Δ=0.9 eV. In the remainder of the paper, we will assume ε_∞ and d to be linear in Δ in the range $0 \leq \Delta < 1.5$ eV not only in 2H SiC but also in the other three polytypes of SiC: 3C, 4H, and 6H.

Table II shows that the same Δ=0.6 eV reproduces the experimental ε_o and ε_e within 2% for all four SiC polytypes studied. The fact that only a single Δ is needed to reproduce the experimental values of 2H SiC for both ε_o and ε_e can also be seen in Fig. 1(a). In this figure, the two values of Δ corresponding to the experimental ε_o and ε_e are essentially the same and are close to 0.6 eV. The situation is almost the same in the other three polytypes 3C, 4H, and 6H SiC. Although for 6H SiC the best choice of Δ is closer to 0.7 eV, we kept Δ=0.6 eV (same as the other three polytypes) for simplicity. Hence we chose 0.6 eV as Δ_ε, the self-energy correction to match ε_∞ for all four SiC polytypes we studied.

These results for the "gap problem" of SiC are in agreement with the work by Lambrecht *et al.*[28] They did experiments and LDA calculations on 3C and 4H SiC and found that the

Δ needed to match the indirect band gaps are a few hundred meV greater the Δ to match the reflectivities. A recent study[26] of 3C SiC (zincblende) band structure by GW and LDA calculations (using Gaussian orbitals) indicates that Δ should be 1.26 eV, almost identical with our choice of Δ_{gap} for this polytype. In that same paper,[26] the calculated ε_∞ for 3C SiC within the LDA is reported to be 6.7, compared to 6.95 for the present work.

To study the trends for nH SiC polytypes, we used a special structural relation between 3C and ∞H SiC to obtain the physical properties (e.g., ε_∞ and d) of ∞H SiC from those of 3C SiC. This structural relation between 3C and ∞H SiC is easy to understand in the Hägg notation[2] which describes relations between successive Si-C bilayers. A passage from bilayers A$\rightarrow$B$\rightarrow$C$\rightarrow$A involves a translation of (1/3, 2/3) in the basal plane, whereas the passage from layers A$\rightarrow$C$\rightarrow$B$\rightarrow$A involves a translation of ($-1/3$, $-2/3$). Hägg denoted the former by "+" and the latter by "$-$". In the Hägg notation, 2H SiC (AB$\cdots$) is represented by $+-\cdots$, 4H SiC (ABCB$\cdots$) by $++--\cdots$, and 6H SiC (ABCACB$\cdots$) by $+++---\cdots$. The 3C SiC structure (ABC$\cdots$) is now represented by $+++\cdots$, or alternatively $---\cdots$ if we rotate the 3C SiC crystal 180° along the z-axis (perpendicular to the Si-C planes). So if we cut the bulk 3C SiC crystal in half along the x-y plane and rotate the upper half by 180° along the z-axis, we will get the structure of $+++\cdots---\cdots$, which is equivalent to the structure of ∞H SiC. Since the boundary does not affect the bulk properties of the crystals, physical properties of ∞H SiC are directly related to those of 3C SiC. In particular, the relations we used are

$$\begin{aligned} \varepsilon_o^{(\infty H)} &= \varepsilon_e^{(\infty H)} = \varepsilon^{(3C)}, \\ d_{333}^{(\infty H)} &= -2d_{113}^{(\infty H)} = 2d_{123}^{(3C)}/\sqrt{3}, \end{aligned} \tag{2}$$

as can be derived by transformation between cubic and hexagonal coordinates.[29]

As can be seen in Table II, the computed ε_∞ for nH SiC with the same scissors operators Δ=0.6 eV capture the same trend as the experimental facts, namely ε_o, ε_e, and $\varepsilon_e - \varepsilon_o$ approach those of ∞H SiC as n increases. In particular, ε_o remains more or less constant while ε_e and $\varepsilon_e - \varepsilon_o$ decrease with increasing n in nH SiC. The trend is also true for the LDA calculations. We note here that E_{cut} for 6H SiC is 1 hartree smaller than others (because of memory limits on the Cray YMP) and this might have caused the computed values of 6H SiC to lie somewhat out of the trend. We also observed that using the band-gap-matching Δ, which means different Δ for different polytypes, does not produce the correct trend.

Table III shows that the second-order nonlinear coefficients d_{113} and d_{333} calculated for nH SiC also follow the trend that with Δ=0.6 eV they approach those of ∞H SiC as $n \to \infty$. (Although the point group for crystals with C_{6v} symmetries including 2H, 4H, and 6H SiC permits three independent tensor components, namely d_{113}, d_{311}, and d_{333}, Kleinman symmetry,[30] which is exact at zero frequency, implies $d_{113} = d_{311}$.) The d we predicted for all four SiC polytypes have the same order of magnitude as the experimental data on an unknown SiC polytype.[3] Furthermore, with Δ=0.6 eV, the calculated d of 6H SiC are close to the experimental values within uncertainty region. Table III also shows that d_{333} of 2H SiC is unexpectedly small as compared with the other three polytypes. The trend for d in nH SiC can be observed in Fig. 2 in which we choose $1/n$ to be the x-axis. For both d_{113} and d_{333}, the variation is sublinear with $1/n$. The common end point value is somewhat below a linear fit to the small n calculations.

Bond-charge-model estimates, shown in Table III, of the second-order nonlinear coefficients for the SiC polytypes are model dependent.[3,31] The bond-charge model of Levine[31] gives essentially double the predictions as the two-band-model[32] on d which are three times greater than the experimental values. The second bond-charge model[3] used different core radii corrections for the ions as compared to the first model listed in Table III and gives the correct order of magnitude predictions for d. Because Si and C ions have the same ionic charge, the bond-charge model predictions for d are very sensitive to the choice of bond-charge and the ion radii as pointed out by Singh *et al.*[3] All bond-charge models share tensoral addition of the single-bond second-order polarizability β, but differ as to how that β

is defined. Hence the ratios of the macroscopic second-order susceptibilities are independent of the particular bond-charge model chosen. This ratio is d_{113}/d_{333}=$-1/2$ for all nH SiC.[29] This is different from our calculations most manifestly in 2H SiC for which we predicted $d_{113}/d_{333} \approx -1.5$. In each bond-charge model, the d for *all* nH SiC are the same, which is again in contrast to our predictions.

Fig. 3 shows that the local-field corrections for both d_{113} and d_{333} for nH SiC smoothly approach those for ∞H SiC as n increases. As n increases the splitting in the local-field decreases monotonically to the degenerate $n = \infty$ case. The local-field corrections are at least one order of magnitude smaller than the main contribution. Our calculations show that the local-field corrections to ε_o and ε_e are also small. For the four polytypes considered, they all lie between -3.5% and -4%.

We close with a few remarks concerning the integration scheme and the convergence of the calculation. We choose integration points by dividing the two-dimensional hexagonal Brillouin zone of the SiC polytypes into equilateral triangles, with vertices at Γ and other high symmetry points. We choose our integration points to be the centers of these equilateral triangles. For the calculations presented here, in 2H SiC two layers of points with 10 integration points per plane in the irreducible Brillouin zone (IBZ) are used with the integration scheme made as identical as possible for the other nH polytypes. For 3C SiC, the standard 28 special points in the are chosen.[33]

The sum rules[5] associated with ε_∞ and d for all four SiC polytypes, as summarized in Table IV, are all satisfied within 2%. This indicates that the energy cutoff for the plane waves and the number of integration points are probably adequate for calculating ε_∞ and d. The following remarks apply to the 2H SiC calculation. Varying the number of integration points in the plane from 6 to 10, with two layers, led to a variation in the two components of ε_∞ and d by no more than 0.4% and 1% respectively. Variation of the number of planes from 2 to 3 (with 6 integration points per plane) led to corresponding variations of no more than 1% and 3% respectively. Based on past experience with the sum rules,[5] these results and the results in Table IV imply that the d calculations are converged to 5%, and that the ε_∞ values are converged to within 2%.

III. CONCLUSIONS

We calculated dielectric constants ε_∞ and second-order nonlinear coefficients $d = \chi^{(2)}/2$ for four polytypes of SiC in addition to ε_∞ for diamond. We used the LDA as well as the LDA with a self-energy correction in the form of a "scissors operator"[5] $\Delta P_{c\vec{k}}$ which is applied to the conduction bands to adjust the band gap. The LDA overestimates the experimental values of ε_∞ by 3% in diamond and 6-8% in the SiC polytypes. In all the wide band gap materials we have studied so far (urea,[21] diamond, and four polytypes of SiC) we observed the curious fact that the Δ needed to reproduce the dielectric constants is less than that needed to reproduce the band gap. We found in particular that with Δ=0.6 eV for all four SiC polytypes, we could reproduce the experimental ε_∞ within 2%. We further found that with the same Δ=0.6 eV, the calculated d_{113} and d_{333} for nH SiC with $n \geq 4$ are all close to the values of the only experimental data available which were measured from an unknown SiC polytype. In the calculations of d, we found that d_{333} of 2H SiC (wurtzite) is unexpectedly small as compared to the other three polytypes. The results are not consistent with a bond-charge model in which the hyperpolarizability is a tensoral sum of bond hyperpolarizabilities. This is a surprise for this σ-bonded system. The local-field corrections to ε_∞ of diamond and ε_∞ and d of the four SiC polytypes are all at least one order of magnitude smaller than the main contributions. For nH SiC, both the values and the local-field corrections to d form a trend among the polytypes. The qualitative features of the calculation appear in the LDA as well as the calculation with a self-energy correction.

The main role of this correction is to reduce the numerical values by about 6% for ε_∞ and 20% for $\chi^{(2)}$.

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TABLE I. Effects of self-energy corrections and of E_{cut} on the dielectric constant of diamond. In contrast to the GW prediction[7,8] of $\Delta = 1.8$ eV, we found that $\Delta \approx 0.5$ eV should reproduce the experimental dielectric constant by linearly interpolating between the results at Δ=0 (i.e., the LDA) and 0.9 eV. The E_{cut} we used is smaller than used in Ref. 25 because we used a softer pseudopotential.[24] However, the corresponding ε_∞ are essentially the same. We did not carry out a convergence study on the number of $\vec{k}$ points for integration relying instead on the study in Ref. 25. The calculations done in Ref. 26 and Ref. 34 use Gaussian orbitals and orthogonalized linear combination of atomic orbitals (OLCAO), respectively.

Self-energy correction Δ (eV)	E_{cut} (hartree)	No. of $\vec{k}$ points	ε_{00} (long wave)	ε_∞ (tot)
0	17	10	6.03	5.86
0	20	10	6.02	5.85
0.9	17	10	5.66	5.51
0.9	20	10	5.65	5.50
1.8	17	10	5.34	5.20
0 (Ref. 25)	25	10	6.06	
0 (Ref. 25)	25	28	6.06	5.90
0 (Ref. 26)	Gaussian	10		5.5
0 (Ref. 34)	OLCAO	505		4.34
Exp. (Ref. 1)				5.67

TABLE II. Computed ordinary and extraordinary dielectric constants for 4 polytypes of SiC compared with experiment. The self-energy correction Δ_ε to reproduce (within 2% for all polytypes) the dielectric constant (also used in Table III to predict d) is roughly half the value needed to reproduce the indirect band gap Δ_{gap}. Results with $\Delta_\varepsilon = 0.6$ eV are obtained by linearly interpolating between LDA and Δ_{gap} values. Tables III and IV have the same E_{cut}, Δ_{gap}, and number of $\vec{k}$ points as this table. (The slight variation in E_{cut} is due to memory limitations on the Cray YMP.)

	Method	2H	4H	6H	∞H (3C)
Δ_{gap} (eV)		1.17	1.07	1.04	1.27
No. $\vec{k}$ points		20	10	10	28
E_{cut} (eV)		17	17	16	17
ε_o	With Δ_{gap}	6.13	6.22	6.27	6.09
	LDA	6.91	6.96	7.00	6.95
	$\Delta_\varepsilon = 0.6$ eV	6.51	6.55	6.58	6.54
	Exp. (Ref. 1)	6.51	6.56	6.52	6.52
ε_e	With Δ_{gap}	6.45	6.41	6.46	6.09
	LDA	7.32	7.20	7.24	6.95
	$\Delta_\varepsilon = 0.6$ eV	6.87	6.76	6.79	6.54
	Exp. (Ref. 1)	6.84	6.78	6.68	6.52
$\varepsilon_e - \varepsilon_o$	With Δ_{gap}	0.32	0.19	0.19	0
	LDA	0.41	0.24	0.24	0
	$\Delta_\varepsilon = 0.6$ eV	0.36	0.21	0.21	0
	Exp. (Ref. 1)	0.33	0.22	0.16	0

TABLE III. Computed static second-order nonlinear coefficients $d = \chi^{(2)}/2$ (pm/V) for 4 polytypes of SiC. Kleinman symmetry gives $d_{113} = d_{311}$.[30] Values for ∞H SiC are obtained from 3C SiC results by $d_{333}^{(\infty H)} = -2d_{113}^{(\infty H)} = 2d_{123}^{(3C)}/\sqrt{3}$.[29] The values of Δ_{gap} are given in Table II. The experimental data is from an unknown SiC polytype,[3] the static limits of which are obtained by using a Miller's rule,[35] namely, $d_{ijk}(0)/[\chi_{ii}(0)\chi_{jj}(0)\chi_{kk}(0)] = d_{ijk}(\omega)/[\chi_{ii}(2\omega)\chi_{jj}(\omega)\chi_{kk}(\omega)]$. The bond-charge models and the two-band model predict the same d for all SiC polytypes. The two bond-charge models listed differ in the treatment in the core radii corrections of the ions.

	With Δ_{gap}		LDA		Interpolation Δ_ε =0.6 eV	
	d_{113}	d_{333}	d_{113}	d_{333}	d_{113}	d_{333}
2H SiC	4.4	−2.9	6.6	−4.3	5.5	−3.6
4H SiC	5.0	−7.8	7.4	−11.6	6.1	−9.5
6H SiC	5.2	−9.3	7.5	−13.8	6.2	−11.2
∞H SiC	4.6	−9.2	7.1	−14.1	5.9	−11.8
Exp. ($\lambda = \infty$)					6.9±0.8	−11.6±1.3
Exp. ($\lambda = 1.06\mu$m) (Ref. 3)					7.9±0.9	−13.5±1.5
Bond-charge model 1 (Ref. 31)					42	−83
Bond-charge model 2 (Ref. 3)					4	−8
Two-band model (Ref. 32)					20	−40

TABLE IV. The sum rules for ε_∞ and d are satisfied within 2% for four SiC polytypes and diamond. In other calculations,[5] this has indicated that the energy cutoff and the number of integrations points are probably adequate for the evaluation of ε_∞ and d. $1-\Sigma_f$ and Σ_ρ are sum rules associated with ε_∞ and d, respectively. The diamond study used E_{cut}=17 hartree and 10 special $\vec{k}$ points.

Material	$1-\Sigma_f$		Σ_ρ^{LDA}	
	o	e	113	333
2H SiC	0.0019	−0.0002	−0.0180	−0.0165
4H SiC	0.0044	0.0055	−0.0069	−0.0020
6H SiC	0.0048	0.0048	−0.0048	−0.0037
3C SiC	0.0016		0.0092	
diamond	−0.0184			

FIG 1. Dielectric constants ε_o and ε_e and second-order nonlinear coefficients d_{113} and d_{333} for 2H SiC versus the self-energy correction Δ. Interpolating between the computed values at $\Delta = 0$ eV and $\Delta = 1.5$ eV yields an error, compared to the computed value, of less than 1% for ε and less than 5% for d. The experimental values for ε_o and ε_e correspond to $\Delta \approx$0.6 eV, which is then used to predict d. Calculations are done with E_{cut}=17 hartree and 12 special $\vec{k}$ points in the irreducible Brillouin zone for integration.

FIG 2. The trend of d_{113} and d_{333} in nH SiC for different Δ. We plotted $-d_{333}/2$ to combine the two d components in one figure. In ∞H SiC, the two d components are related by Eq. (2).

FIG 3. Local-field corrections to second-order nonlinear coefficients d_{113} and d_{333} of nH SiC with self-energy correction $\Delta = 0.6$ eV. We observe two facts: (1) All local-fields corrections are within 10% of the total contribution. (2) The local-field form a trend among the polytypes. In the case of ∞H SiC, $d_{113} = -d_{333}/2$ by symmetry. So the local-field corrections are identical. With LDA and band-gap-matching Δ, the local-field corrections are very similar to the present case.

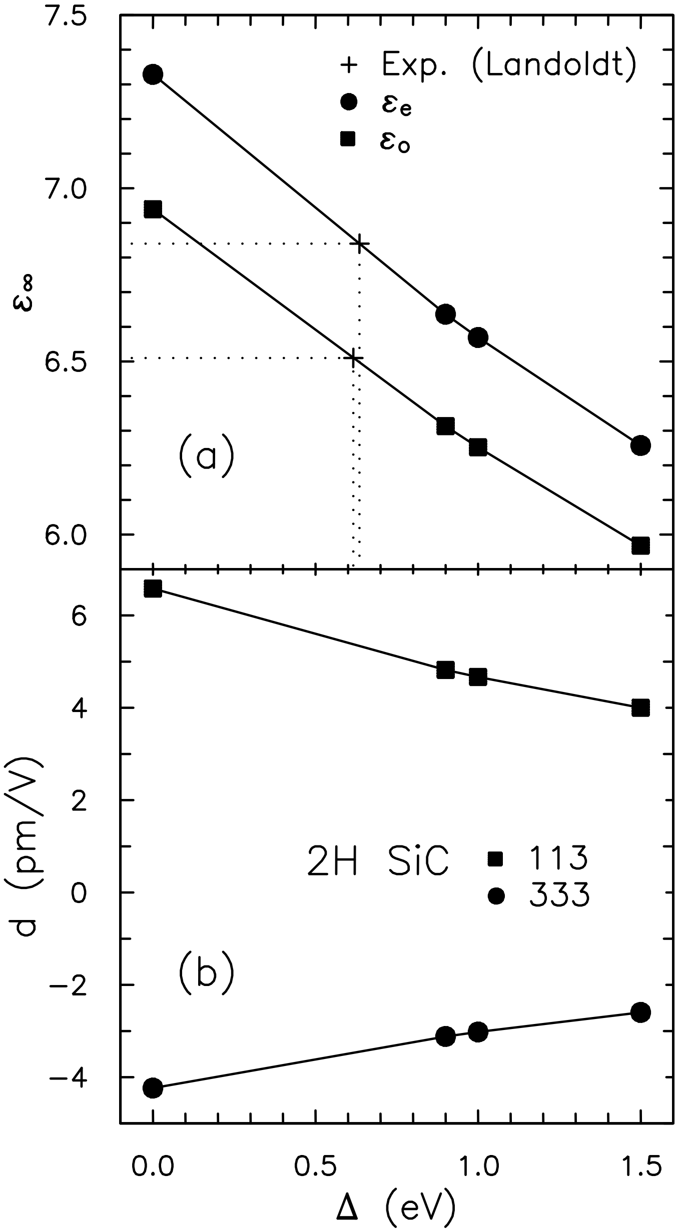

Fig. 1

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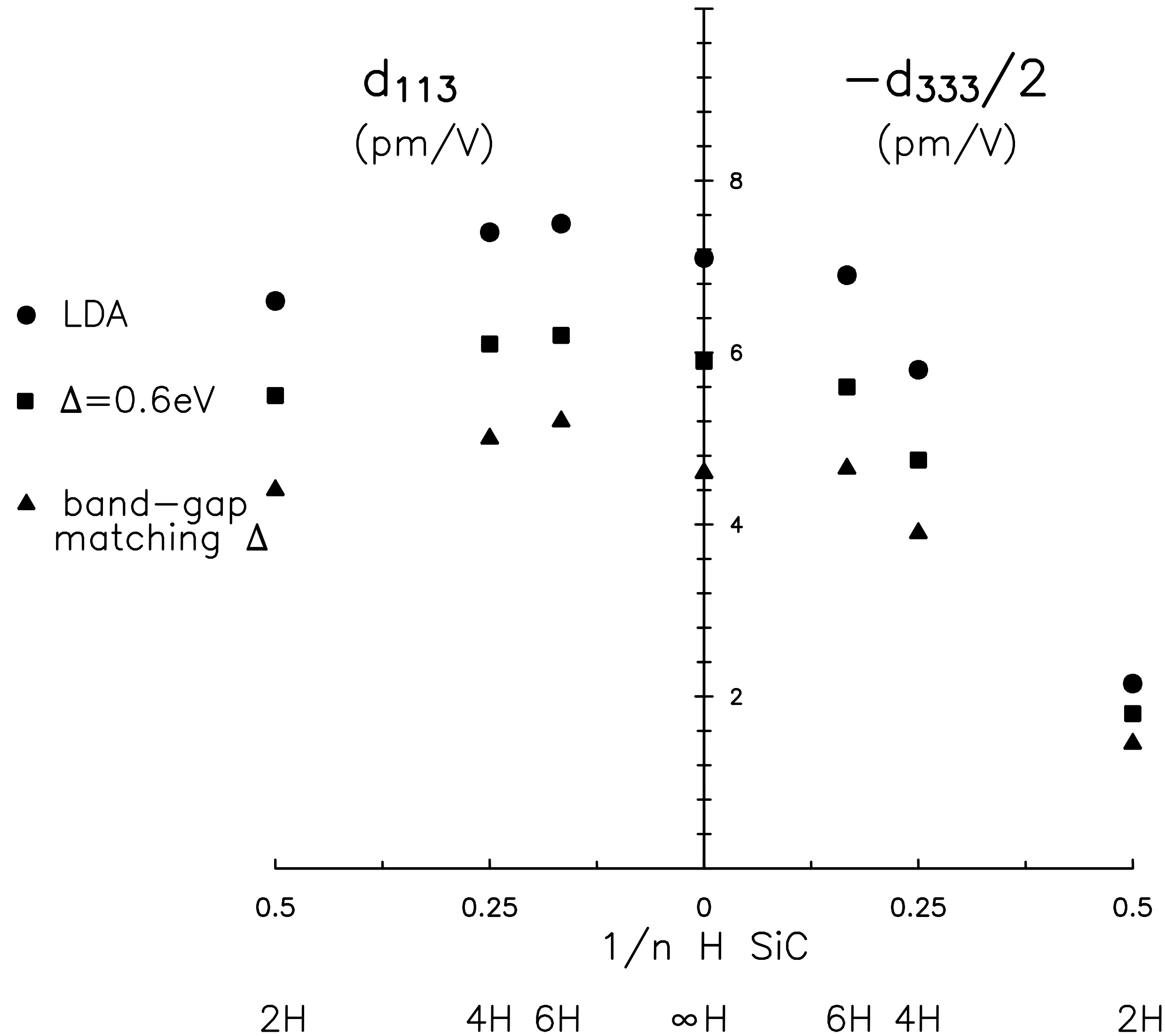

Fig. 2

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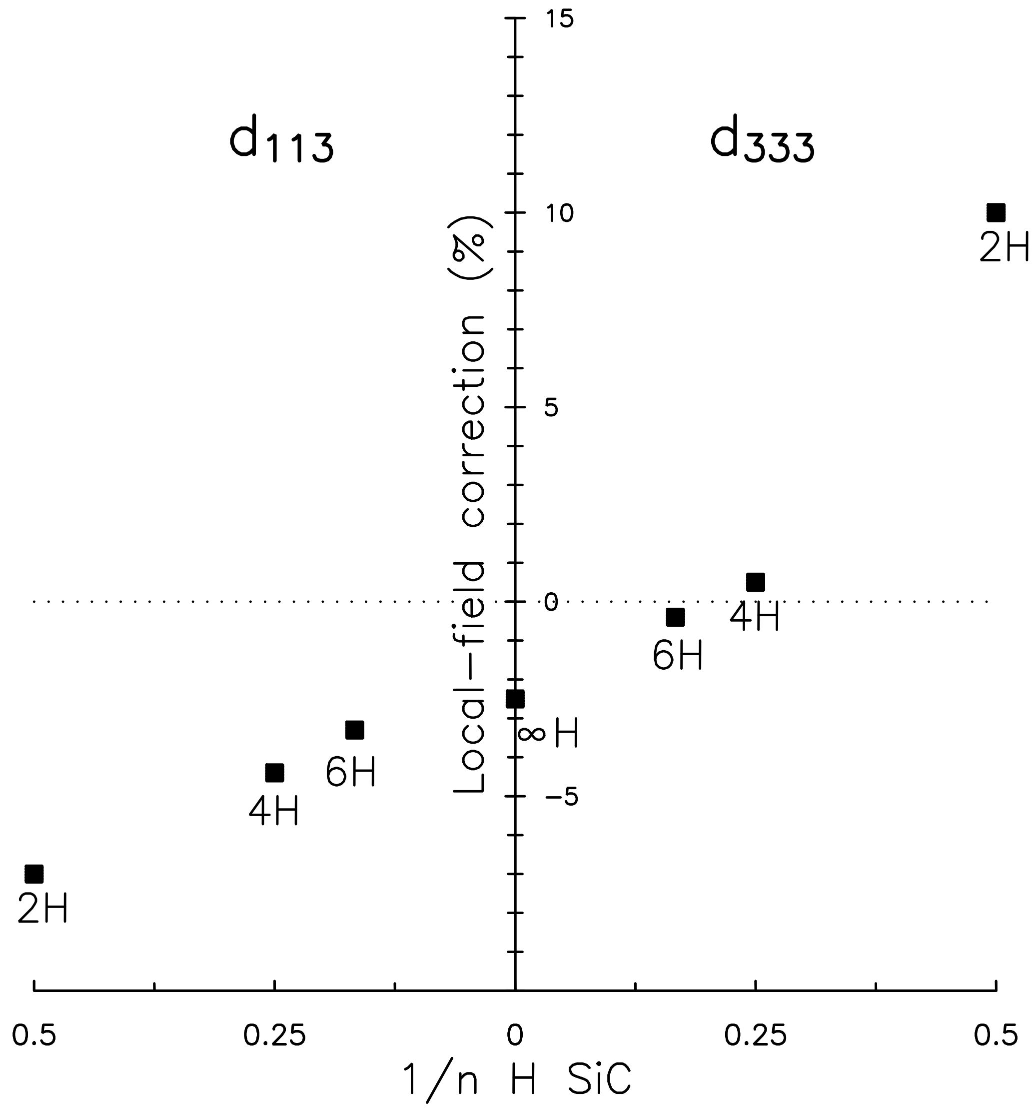

Fig. 3

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