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Pressure-dependent dynamical and dielectric properties of cubic SiC

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Abstract. The plane-wave pseudopotential approach to the density-functional theory (DFT) in the local-density approximation (LDA) is used for an *ab initio* calculation of pressure-dependent structural, lattice-dynamical, and dielectric properties of cubic silicon carbide (3C SiC). Whereas the influence of hydrostatic pressure on the structural and the underlying bonding properties is treated within the DFT-LDA, the linear-response theory is applied to describe the pressure dependence of the zone-centre phonon frequencies, the dielectric constant, and the Born effective charge of 3C SiC. Furthermore, the results for 3C SiC are compared with the corresponding ones for silicon and diamond.

1. Introduction

Silicon carbide has recently attracted increasing interest due to its outstanding mechanical, chemical, thermal, and electronic properties [1, 2]. The potential of SiC as a microelectronic device material for high-power and high-temperature applications is related to its large bandgap and strong bonding behaviour, which result in its resistance to high temperatures and radiation damage. SiC can be n- and p-type doped and possesses a higher thermal conductivity than silicon [3]. In spite of such unique properties, electronic device applications based on SiC have been delayed [4]. Moreover, SiC is the prototype material showing polytypism. With more than 200 polytypes, SiC exhibits the fascinating phenomenon of long-range structural ordering, which despite enduring efforts is still in debate.

In the past, high-pressure studies have been carried out mainly on the zinc-blende phase of silicon carbide (3C SiC). In particular, the Raman modes have been studied up to 0.4 Mbar [5–8]. Very recently, x-ray diffraction experiments have been made on cubic 3C SiC under ultrahigh pressure up to 1.05 Mbar [9]. The zinc-blende structure undergoes a phase transition to a rock-salt-type structure at about 1.0 Mbar, accompanied by a remarkable volume reduction of about 20%. This pressure-induced phase transition of 3C SiC was predicted theoretically by Chang and Cohen *et al* [10] and matches well with the large ionicity of SiC [11].

Theoretical studies of the pressure-dependent properties of cubic silicon carbide have been performed on the basis of empirical [6] and *ab initio* pseudopotential methods [10–13]. First-principles studies of the ground-state properties of hexagonal polytypes of SiC including the influence of the internal parameters have also been performed [14–18].

The purpose of the present work is to give a detailed description of the behaviour of lattice-dynamical and dielectric properties of 3C SiC under hydrostatic pressure using density-functional theory (DFT) [19] and the linear-response approach to the DFT [20, 21]. In section 2 we give a brief description of the method employed and some computational details. Then, in section 3 we investigate the pressure dependence of the lattice-dynamical and dielectric properties of the cubic phase of SiC. In particular, we calculate the optical phonon frequencies, the Born effective charges, and the high-frequency dielectric constant of 3C SiC as functions of an external hydrostatic pressure. In order to point out the different roles played by the constituent atoms we compare the pressure-dependent properties of 3C SiC with those of diamond and silicon crystals. Finally, our conclusions are drawn in section 4.

2. Method

The present *ab initio* calculations are performed within the framework of DFT using the local-density approximation (LDA) [22]. The exchange–correlation contribution to the total electron–electron interaction is described by the electron gas data of Ceperley and Alder [23] parametrized by Perdew and Zunger [24]. In order to describe the lattice-dynamical and dielectric properties within the harmonic approximation we employ the density-functional perturbation theory (DFPT) [20, 21, 25]. DFPT is a powerful and accurate tool for determining derivatives of the ground-state electronic energy and has been successfully applied to the calculations of elastic, dielectric, and piezoelectric constants [26–28], and various lattice-dynamical properties [20, 21, 29–34].

Table 1. The calculated static equilibrium lattice constant a (au), and bulk modulus B_0 (Mbar), and its pressure derivative B'_0 , of diamond, 3C SiC, and silicon.

		a	B_0	B'_0
C	Theory	6.68	4.64	3.5
	Experiment	6.74 ^a	4.42 ^a	4.0 ^b
3C SiC	Theory	8.21	2.24	3.8
	Experiment	8.24 ^a	2.27 ^c	3.6 ^c
Si	Theory	10.21	0.99	5.8
	Experiment	10.26 ^a	0.99 ^a	4.1 ^d

^a[60].

^b[61].

^c[7].

^d[62].

In the explicit calculations only the valence electrons, which contribute dominantly to the chemical bonds, are taken into account. The effect of the core electrons is incorporated into pseudopotentials. Soft norm-conserving pseudopotentials are generated for carbon and silicon using the method proposed by Troullier and Martins [35].

The pseudopotential scheme is combined with the plane-wave expansion of the single-particle wave functions of the valence electrons. The number of plane waves at equilibrium volume is restricted to a kinetic energy cut-off of 36, 50, and 16 Ryd for 3C SiC, C, and Si, respectively. The corresponding numbers of approximately 500, 460, and 280 plane waves ensure well-converged results for the physical quantities considered. To guarantee consistent results for the different physical properties at varying crystal volumes, the number of plane waves per atom has been fixed. The integrals over the irreducible wedge of the Brillouin

zone (BZ) are replaced by summations over sets of 28 special points [36]. The pressure and volume dependence of the total energy and bulk modulus are determined using the Vinet [37] equations of state. The converged results for the static equilibrium parameters are shown in table 1 and compared with the corresponding experimental values; the agreement obtained is very good.

3. Dynamical and dielectric properties

3.1. Optical phonon frequencies at the zone centre

The pressure dependence of the Raman lines can supply useful information about vibrational properties, structural phase transitions, and the ionic or covalent character of a crystal. Furthermore, the direct comparison of the predicted pressure dependence of such frequencies with the experimental findings is an additional tool for testing the applicability and reliability of lattice-dynamical approaches.

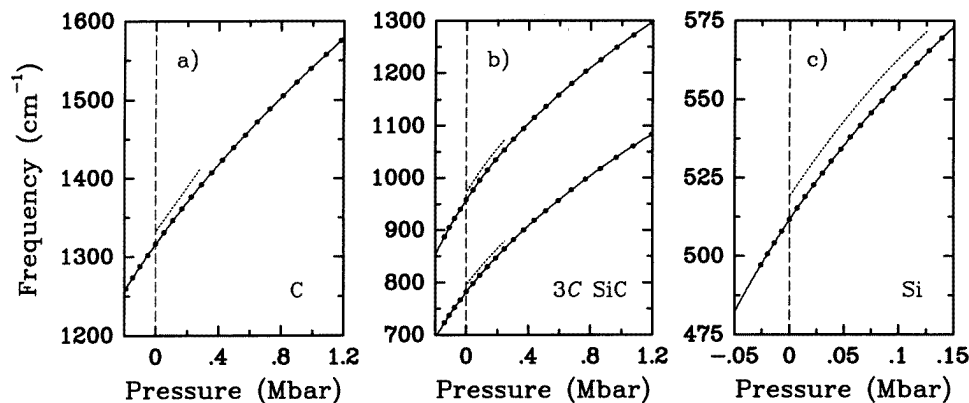


Figure 1. The pressure dependence of the optical frequencies of diamond (a), 3C SiC (b), and silicon (c) versus hydrostatic pressure. For comparison, the experimental first-order Raman data from [38] (C), [6] (3C SiC), and [39] (Si) are plotted as dotted lines.

The calculated transverse optical (TO) and longitudinal optical (LO) phonon frequencies of 3C SiC are plotted in figure 1 as functions of hydrostatic pressure. For comparison, the results for diamond and silicon are also shown in this figure. The pressure dependences of the optical phonon frequencies of diamond, 3C SiC, and silicon derived from theory are in very good agreement with those obtained from experimental first-order Raman spectra [38, 6, 39]. The only difference between the available experimental data and our calculations is a small almost pressure-independent shift. The absolute values of the calculated frequencies are about 2% too small. However, for 3C SiC this discrepancy is smaller by a factor of 2 than that of previous *ab initio* calculations [13]. The pressure dependence of the calculated phonon frequencies can be described by a fourth-order polynomial:

$$\omega(p) = \sum_{n=0}^4 \alpha_n p^n. \quad (1)$$

The corresponding expansion coefficients for diamond, 3C SiC, and silicon are given in table 2. The values of the calculated coefficients up to second order for diamond, 3C SiC,

Table 2. Calculated and experimental polynomial expansion coefficients of various pressure-dependent vibrational quantities of diamond, the 3C phase of SiC, and silicon. The Born effective charge is given in units of the elementary charge, frequencies in cm^{-1} , and the pressure in Mbar.

		α_0	α_1	α_2	α_3	α_4
C						
$\omega_{\text{TO}}(\Gamma)$	Theory	1316.7	280.0	-81.0	34.8	-8.7
	Experiment ^a	1332.5	287 ± 10	—	—	—
ε_∞	Theory	5.69	-0.32	0.20	-0.11	0.03
	Experiment ^b	5.70	—	—	—	—
3C SiC						
$\omega_{\text{TO}}(\Gamma)$	Theory	779.2	389.9	-231.0	162.0	-54.4
	Experiment ^c	796.2 ± 0.3	388 ± 8	-220 ± 40	—	—
$\omega_{\text{LO}}(\Gamma)$	Theory	952.8	458.7	-306.8	226.1	-77.5
	Experiment ^c	972.7 ± 0.3	475 ± 9	-250 ± 40	—	—
ε_∞	Theory	7.02	-1.33	1.85	-1.38	0.40
	Experiment ^b	6.52	—	—	—	—
ε_0	Theory	10.50	-2.40	2.79	-2.18	0.70
	Experiment ^b	9.73	—	—	—	—
Z^{B}	Theory	2.72	0.33	-0.35	0.28	-0.09
	Experiment ^c	2.70	—	—	—	—
Si						
$\omega_{\text{TO}}(\Gamma)$	Theory	511.5	521.3	-1092.1	3247.4	-5879.6
	Experiment ^d	519.5 ± 0.8	520 ± 30	-700 ± 200	—	—
ε_∞	Theory	13.08	-4.80	37.5	-149.1	289.5
	Experiment	12.10^{b}	$-4.8 \pm 1.2^{\text{e}}, -3.4 \pm 0.5^{\text{f}}$	—	—	—

^a[38].

^b[60]

^c[6].

^d[39].

^e[63].

^f[64].

and silicon agree well with the experimental data from [38], [6], and [39], respectively. The pressure dependence of the optical phonon frequencies is rather nonlinear, in particular for 3C SiC and silicon. At very high pressure there is a weaker pressure dependence of the frequency curves of 3C SiC but no indication for a saturation as observed experimentally in the case of 6H SiC [40].

The mode Grüneisen parameters $\gamma_j(\mathbf{k}) = -\partial \ln \omega_j(\mathbf{k}) / \partial \ln V$ for the TO(Γ) and LO(Γ) phonons are derived from the polynomial expansion of the phonon frequencies in equation (1) using the Vinet EOS and theoretical data for the bulk modulus B_0 and its pressure derivative B'_0 . The resulting values are $\gamma_{\text{TO}}(\Gamma) = 0.99$ (C), 1.10 (3C SiC), and 0.98 (Si) and $\gamma_{\text{LO}}(\Gamma) = 1.06$ (3C SiC). The calculated mode Grüneisen parameters of diamond and silicon agree well with the corresponding experimental data: 0.97 (C) and 0.98 (Si) from [41] and [7], respectively. The mode Grüneisen parameters of 3C SiC are consistent with other theoretical results [12, 13], but smaller than the previously reported experimental values of $\gamma_{\text{TO}}(\Gamma) = 1.56 \pm 0.01$ and $\gamma_{\text{LO}}(\Gamma) = 1.55 \pm 0.01$ [6]. However, the results reported in [6] were obtained using values of B_0 and B'_0 obtained from the averages of the corresponding data for diamond and silicon. These averaged values of B_0 and B'_0 differ remarkably from experimental and theoretical data [7–9, 12–14, 29, 42].

Therefore, if we rescale the experimental mode Grüneisen parameters using our theoretical values for B_0 and B'_0 (see table 1), we find values of $\gamma_{\text{TO}}(\Gamma) = 1.11$ and $\gamma_{\text{LO}}(\Gamma) = 1.10$ in reasonable agreement with our data. Furthermore, more recent experimental values of the mode Grüneisen parameters of 3C SiC, $\gamma_{\text{TO}}(\Gamma) = 1.10$ and $\gamma_{\text{LO}}(\Gamma) = 1.09$, are in good agreement with our theoretical results [7].

3.2. High-frequency dielectric constants

In the case of cubic diatomic crystals the long-range Coulomb interaction causes the splitting of the threefold-degenerate optic modes at $\mathbf{q} = 0$ by raising the frequency of the LO mode above that of the TO mode. The LO(Γ)–TO(Γ) splitting, which is visible in figure 1(b), can directly be related to the transverse (Born) effective charge Z^{B} and high-frequency dielectric constant ε_{∞} , according to the relation [43]

$$\omega_{\text{LO}}^2(\Gamma) - \omega_{\text{TO}}^2(\Gamma) = \frac{4\pi e^2 (Z^{\text{B}})^2}{V\mu\varepsilon_{\infty}} \quad (2)$$

where μ is the reduced atomic mass and V denotes the volume of the unit cell.

The phonon frequencies, the high-frequency dielectric constant ε_{∞} , and the Born effective charge Z^{B} depend on the pressure. However, a reliable pressure dependence of these quantities has not yet been derived theoretically. Moreover, reliable experimental studies on the pressure dependence of ε_{∞} and Z^{B} for a wide pressure region are not available. In the literature, various approximate relations for the volume dependence of the dielectric constant for covalent materials can be found, e.g. [6, 40, 44]

$$\varepsilon_{\infty}(V) = \varepsilon_{\infty}(V_0) \left(\frac{V}{V_0} \right)^{\kappa} \quad (3)$$

$$\varepsilon_{\infty}(V) - 1 = [\varepsilon_{\infty}(V_0) - 1] \left(\frac{V}{V_0} \right)^{\nu} \quad (4)$$

with $V_0 = V(p = 0)$. Together with an appropriate equation of state these relations can give the pressure dependence of ε_{∞} . However, the application of relations (3) and (4) is strongly limited to volumes rather close to the equilibrium one. Olego *et al* [6] applied the relation (3) to 3C SiC with an exponent $\kappa = 0.6$. This value has also been used for 6H SiC [40] and for AlN, BN, and BP [45]. The relation (4) has been suggested by Harrison [44] with an exponent $\nu \approx 0.333$ and is based upon the argument that the susceptibility $(\varepsilon_{\infty} - 1)/4\pi$ varies linearly with the bond length for an elemental semiconductor. For a wide class of semiconductors besides the elemental crystals C, Si, and Ge, the exponent ν should be somewhat larger than 0.333 (cf. references given in [44]).

The high-frequency dielectric tensor ε_{∞} relates the macroscopic (screened) electric field \mathbf{E} to the applied external homogenous field $\mathbf{E}_0 = \varepsilon_{\infty} \mathbf{E}$ and can be determined from the derivative of the electronic polarization \mathbf{P}^{el} with respect to the macroscopic electric field at fixed ionic positions:

$$(\varepsilon_{\infty})_{\alpha\beta} = \delta_{\alpha\beta} + 4\pi \left. \frac{\partial P_{\alpha}^{\text{el}}}{\partial E_{\beta}} \right|_{\{u\}=0}. \quad (5)$$

For crystals of cubic symmetry the dielectric tensor has a diagonal form with equal elements. The calculated high-frequency dielectric constants of 3C SiC and of the elemental semiconductors diamond and silicon are shown in figure 2. For comparison, we show in figure 2 the pressure dependence of the dielectric constants described by the relation (3) using the suggested exponent $\kappa = 0.6$ and also using a best least-squares fit of κ to the

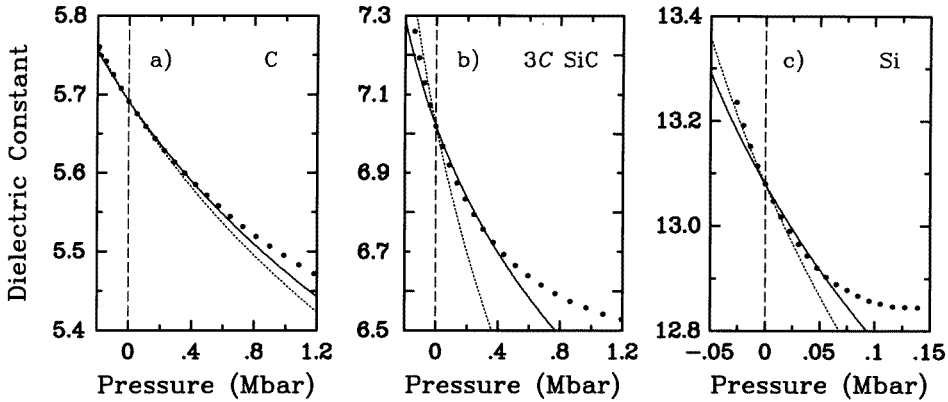


Figure 2. The pressure dependence of the high-frequency dielectric constant of diamond (a), 3C SiC (b), and silicon (c). The data actually calculated are shown as filled circles. The solid lines correspond to the best least-squares fit obtained using equation (3) with $\kappa = 0.238$ for diamond, $\kappa = 0.342$ for 3C SiC, and $\kappa = 0.268$ for silicon, whereas the dotted lines represent relation (3) with the fixed value $\kappa = 0.6$.

calculated data. As can be seen from this figure the proposed relation is only valid around the equilibrium volume, using exponents from least-squares fits to the calculated data. The relation (4) gives a similarly insufficient description of the pressure dependence of ϵ_∞ . The so-obtained exponents κ and ν of the materials studied are not universal and differ considerably from the values $\kappa = 0.6$ and $\nu = 0.333$ suggested by Olego *et al* [6] and Harrison [44], respectively. The calculated high-frequency dielectric constant ϵ_∞ of 3C SiC and, in particular, of silicon, displays a tendency to saturate in the limit of high pressures. We have described the nonlinear character of the hydrostatic pressure variation of ϵ_∞ of diamond, the cubic 3C phase of SiC, and silicon using a fourth-order polynomial expansion around zero pressure (see table 2). The absolute value of the theoretical dielectric constant differs from the experimental one as a consequence of the use of the LDA. For diamond, this deviation is less than 0.1%, whereas it amounts to about 8% in the case of silicon and of 3C silicon carbide. The reason for this discrepancy is still an open question [46]. However, there are two strong indications that the pressure dependence of the high-frequency dielectric constant is correctly described within the LDA. (i) The pressure dependence of the LO–TO splitting at the Γ point is related to that of the Born effective charge Z^B and of the high-frequency dielectric constant ϵ_∞ via the relation (2). Indeed, the pressure dependence of the LO–TO splitting is accurately reproduced within the LDA not only for 3C SiC but also for several heteropolar III–V semiconductors such as BN, AlN, and GaAs [47]. Recently, Sengstag *et al* calculated the volume dependence of the Born effective charge for several semiconductors, including also 3C SiC, using a method based on a Berry phase approach which directly yields macroscopic polarization differences [48]. Their value for the volume derivative at the equilibrium volume of the Born effective charge $\partial Z^B / \partial V$ for 3C SiC of -0.034 \AA^{-1} agrees very well with our result of -0.035 \AA^{-1} as well as with the experimental value of -0.038 \AA^{-1} [49]. Therefore, the accurate description of the pressure dependence of both the LO–TO splitting and the Born effective charge implies that of the dielectric constant. (ii) The underestimation of the absolute value of the high-frequency dielectric constant may be related to the failure of the LDA to reproduce the correct value of the bandgap [46, 50, 51]. Although the energy gap between occupied and unoccupied states in

semiconductors and insulators is consistently underestimated by 20–50% within the LDA compared with experiment, it has been shown that the pressure derivatives, or deformation potentials, of the bandgap are accurately calculated within the LDA and, moreover, are not sensitive to the use of different (pseudo-) potentials and exchange–correlation functionals [52–54]. For these reasons, it is more appropriate to use the present parameter-free approach than the previous semiempirical models [6, 44, 55] to produce reliable values for the pressure (volume) dependence of the high-frequency dielectric constant of homopolar and heteropolar semiconductors.

As can be seen from the figure 3 and the table 2 the static dielectric constant $\epsilon(0)$ of 3C SiC decreases more quickly with increasing hydrostatic pressure than the high-frequency dielectric constant $\epsilon(\infty)$; see also table 2. The logarithmic derivatives of $\epsilon(0)$ and $\epsilon(\infty)$ are both negative and amount to -0.229 and -0.189 , respectively. This behaviour can be explained in terms of the lattice polarization contribution. As the lattice expands, the restoring forces between the ions decrease. The lattice polarization is approximately inversely proportional to the forces acting between the ions and increases with decreasing pressure. Furthermore, the displacements of the ions are accompanied by a deformation of the electronic charge distribution. For the crystals under consideration this effect adds a considerable contribution to the resulting polarization. Therefore, the static dielectric constant decreases more quickly with increasing pressure than does the high-frequency dielectric constant.

3.3. The Born effective charge

The tensor of the Born effective charge $Z^B(\kappa)$ is formally related to the macroscopic polarization $\mathbf{P} = \mathbf{P}^{\text{ion}} + \mathbf{P}^{\text{el}}$ induced by a displacement $\mathbf{u}(\kappa)$ of the sublattice κ with electric boundary conditions corresponding to zero macroscopic electric field:

$$Z_{\alpha\beta}^B(\kappa) = V \left. \frac{\partial P_\alpha}{\partial u_\beta(\kappa)} \right|_{E=0} = Z(\kappa)\delta_{\alpha\beta} + V \left. \frac{\partial P_\alpha^{\text{el}}}{\partial u_\beta(\kappa)} \right|_{E=0}. \quad (6)$$

Whereas the ionic contribution to the polarization is given by the charge $Z(\kappa)$ of the κ th ion, the electronic contribution is determined within DFPT from the linear response of the electrons to an ionic displacement corresponding to a zone-centre phonon mode [20, 21].

For SiC the tensor $Z^B(\kappa)$ is diagonal and the two effective charges are related to each other via the acoustic sum rule $Z^B = Z^B(\text{Si}) = -Z^B(\text{C})$. The calculated Born effective charge Z^B of 3C SiC is shown as a function of external hydrostatic pressure in figure 4. Like the LO(Γ)–TO(Γ) splitting, the Born effective charge Z^B increases with increasing hydrostatic pressure. This behaviour indicates an increase of the bond ionicity with pressure, due to an effective charge transfer from the silicon ions towards the carbon ions. Thus 3C SiC becomes more ionic as a function of pressure, in contrast to the common behaviour of most of the III–V semiconductors [39, 45, 56, 57]. In the III–V compounds, the application of an external hydrostatic pressure gives rise to a decrease in the ionicity and, hence, an increase of the covalent character of the bonding. The increase of the LO(Γ)–TO(Γ) splitting as well as of the Born effective charge has been found experimentally for 3C SiC [6]. At high pressure the Born effective charge Z^B exhibits a strongly nonlinear behaviour. However, a decrease as observed experimentally in [40] in the case of 6H SiC is not found. The pressure dependence of the Born effective charge of 6H SiC was derived from experimental values of the LO(Γ)–TO(Γ) splitting using an incorrect volume dependence of the high-frequency dielectric constant, $\partial \ln \epsilon_\infty / \partial \ln V = 0.6$; see equation (3). To estimate the magnitude of the error caused by this rough approximation, we recalculated the Born

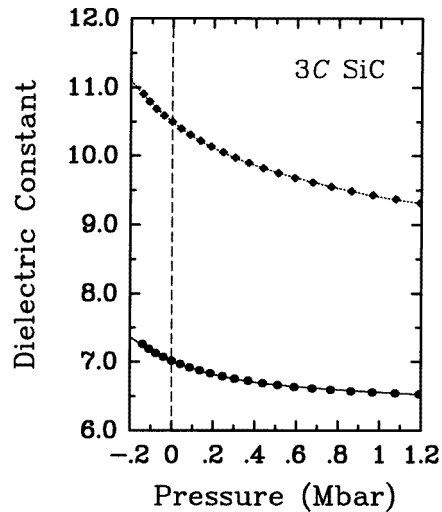


Figure 3. The pressure dependence of the static ϵ_0 (diamonds) and high-frequency ϵ_∞ (circles) dielectric constants of 3C SiC.

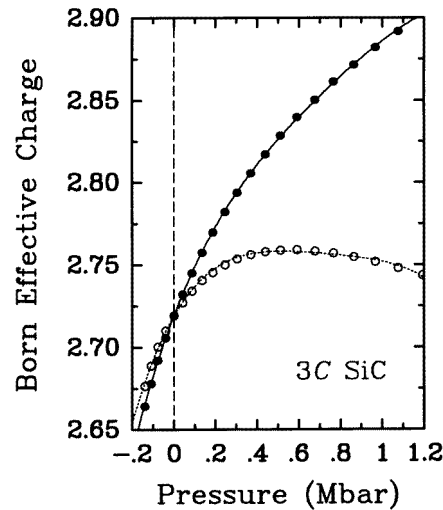


Figure 4. The calculated pressure dependence of the Born effective charge of 3C SiC. The solid line represents a least-squares fit with equation (1). The filled circles show the results of the self-consistent calculation within DFPT. The open circles (dotted line) denote the values of the Born effective charge estimated using an incorrect pressure dependence of ϵ_∞ ; see the text.

effective charge of 3C SiC from the theoretical LO(Γ)–TO(Γ) splitting using the same approximation for the volume dependence of ϵ_∞ . The so-estimated pressure dependence of Z^B for 3C SiC is compared in figure 4 with correct values of Z^B . The most significant result from figure 4 is that the saturation behaviour of the estimated Born effective charge at high pressure is unreliable. Therefore, we conclude that the unusual behaviour of the experimentally derived Born effective charge of 6H SiC may be related to the inadequate description of the pressure dependence of the high-frequency dielectric constant used in [40]. A detailed discussion of the pressure dependence of the structural, lattice-dynamical, and dielectric properties of 2H and 4H polytypes of SiC as well as of the influence of the polytypism on these properties will be given in a forthcoming publication [47].

The Born effective charge, which is defined in terms of lattice-dynamical properties [58], can be used as a measure of the *dynamic* ionicity of a crystal. A definition for the *static* ionicity of binary compounds has been given by Garcia and Cohen [11] in terms of the asymmetry of the ground-state valence charge density with respect to the bond centre. The pressure dependence of the static ionicity coefficient g in comparison with that of the Born effective charge Z^B of 3C SiC is shown in figure 5. As illustrated in this figure the static ionicity increases linearly for small pressures like Z^B but shows a weaker pressure dependence at ultrahigh pressures than Z^B . This behaviour of the static ionicity coefficient g is caused by a pressure-induced charge transfer from the cation Si to the anion C [59]. The static ionicity factor g and the Born effective charge Z^B of 3C SiC as functions of applied hydrostatic pressure exhibit very similar behaviour for pressures below 0.4 Mbar. Moreover, the experimental finding for 3C and 6H SiC, that their chemical bonds become more ionic for small pressures, is reproduced [6, 40]. This encourages us to use both the

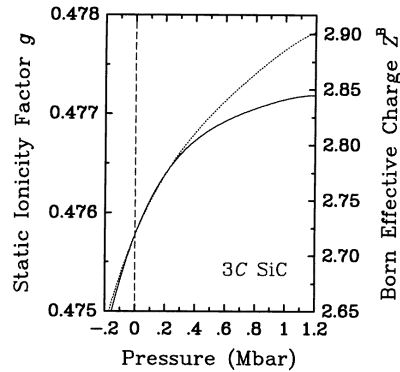


Figure 5. The pressure dependence of the static ionicity coefficient g (solid line) and of the Born effective charge Z^B (dotted line) of 3C SiC.

static and dynamic ionicity approaches as consistent ionic scales for crystals in equilibrium conditions as well as under small applied hydrostatic pressures.

4. Conclusions

We presented an application of the density-functional and density-functional perturbation theory to the *ab initio* calculation of pressure-dependent lattice-dynamical and dielectric properties of silicon carbide in the zinc-blende phase. In particular, we determined the pressure dependence of the optical zone-centre phonon frequencies, the high-frequency dielectric constant, and the Born effective charge. We demonstrated that the dependence of the dielectric constant on the volume (pressure) is insufficiently described by approximations which are commonly used in the literature, the validity of the latter being restricted to volumes (external hydrostatic pressures) close to the equilibrium one (zero pressure). A decrease of the Born effective charges at ultrahigh pressure as experimentally quoted for the hexagonal 6H SiC was not found for the 3C phase of SiC. We demonstrated that this unusual behaviour of the Born effective charges of 6H SiC might be mainly related to the incorrect description of the volume dependence of the dielectric constant. Moreover, the pressure-dependent properties of SiC have been compared with those of Si and C in the diamond structure. Finally, we analysed the ionicity of 3C SiC in its static (charge-asymmetry coefficient) as well as dynamic (Born effective charge) aspects and obtained a qualitative agreement between the two approaches.

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from the experimental LO–TO splitting the value $\partial Z^B/\partial V = -0.038 \text{ \AA}^{-1}$.

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