In fact, formula (62) can be used to describe qualitatively the acoustic attenuation for the entire frequency range[21] $\hbar\omega_{\mathbf{q}\lambda}<4\Delta$.

ACKNOWLEDGMENTS

I wish to thank Dr. M. Pomerantz for many helpful discussions on the subject and for the use of his experimental data. Thanks are also due to Dr. P. Marcus and Dr. P. Miller for their critical reading of the manuscript.

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[21] A phenomenological formula for the total acoustic attenuation coefficient of the form

$$\alpha_\omega=\frac{2}{3}\frac{N_3}{\rho c^3}\frac{\Xi^2}{k_BT}\frac{\omega^2\tau}{1+\omega^2\tau^2}$$

has been derived by M. Pomerantz (Ref. 1). N_3 is the number of impurity atoms per unit volume in the triplet states and τ is a relaxation time to be adjusted to fit the experimental data. One sees immediately that this expression is similar to the second term in Eq. (62) (multiplied by N_i, the impurity concentration) if τ is identified with the life time of a triplet state $(\Gamma/\hbar)^{-1}$. The above formula is no longer valid when $\hbar\omega$ is close to 4Δ.

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Theory of the Third-Order Elastic Constants of Diamond-Like Crystals

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A calculation of the third-order elastic constants of silicon, germanium, and other diamond-like crystals is presented which is based on a previously published method of setting up a suitable form for the elastic strain energy of a crystal. The six third-order constants are calculated in terms of three anharmonic first- and second-neighbor force constants and the two previously determined harmonic force constants. The experimental values of the six coefficients are well fitted by the theoretical expressions involving these three anharmonic force constants. The valence-electron interactions are discussed in the light of the values deduced for these force constants.

I. INTRODUCTION

IN a previous article,[1] hereafter referred to as I, the problem of writing down a form for the elastic strain energy of a crystal which exhibits the necessary invariance properties was considered. It was shown that one can write such a strain energy as a function of various scalar products between the vectors representing the relative positions of the nuclei. This method is basically equivalent to the Born-Huang method[2] of imposing the invariance requirements but the physical significance of the different contributions is now more apparent. In this present article, we shall use this form for the anharmonic strain energy as a starting point for a calculation of the third-order elasticity coefficients of crystals of the diamond type. Such a calculation can shed considerable light on the nature of the interatomic interactions in the solid state.

Interest in the third-order elasticity of crystalline solids has arisen only fairly recently and the number of solids for which measurements have been made is still small. The present experimental method[3–7] is to measure the changes in ultrasonic velocity (and thus in the effective elastic constants) under various types of applied static stress. At this time, measurements on silicon,[3,4] germanium,[3,5,7] quartz,[6] sodium chloride,[7] magnesium oxide,[7] and fused SiO_2[7] have been published. Formal theoretical treatments of third-order elasticity based on the Born-Huang formalism have been given by Leibfried and Ludwig[8] and Srinivasan[9] and a few calculations of third-order elasticity of certain primitive cubic crystals have been published.[10] The present work is based on the alternative formalism presented in I which has several advantages over the Born-Huang approach, especially when anharmonicity is included. Availability of experimental data is, of course, one reason why the diamond type of crystal was chosen for the present calculation. The other reasons are (a) that the results of previous work[1,11,12] suggest that only first- and second-neighbor

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interactions are of appreciable importance in this type of crystal and (b) that the origin of their second-order elasticity is now fairly well understood.[1]

The next section of this article will be concerned with considerations regarding the calculation of third-order elasticity which are general to all types of nonpiezoelectric crystal and this will lay the groundwork for the actual calculation, which is described in Sec. II. In the final section, we shall compare the theoretical results with the experimental data and discuss the origin of the different contributions. Several alternative definitions of the third-order elasticity coefficients have been proposed but the "thermodynamic" definition introduced by Brugger[13] is the most useful and this will be used throughout the present article.

II. GENERAL CONSIDERATIONS

The basis of the method of calculation which was used in I and will be used here is a comparison of the macroscopic and microscopic expressions for the strain-energy density and thus we begin by discussing the general form of these expressions when anharmonic terms are included.

The third-order elastic coefficients of a crystal are a measure of the anharmonicity of the forces in the crystal and are, of course, closely related to the Grüneisen parameter. It is usual to define the third-order elasticity coefficients as the coefficients of the cubic terms in a Taylor-series expansion of the macroscopic elastic strain energy in terms of some set of strain variables. For example, Brugger[13] defines them as the third derivatives (at constant entropy and zero strain) of the internal energy density with respect to the strain variables[13]

$$\eta_J=(x^a_{,b}x_{a,c}-\delta_{bc})/(1+\delta_{bc}),$$

where $x^a_{,b}=\partial x^a/\partial X^b$ if $\mathbf{x}$ is the position vector of a point in the deformed continuum and $\mathbf{X}$ is its position in the undeformed solid. The label J ranges from 1 to 6; a, b, c range from 1 to 3, and the summation convention has been used; the relation between J and bc is the usual one[2] introduced by Voigt. Hence, the internal energy per unit undeformed volume U' is[13]

$$U'=\tfrac{1}{2}\sum_{J}c_{JJ}\eta_J^2+\sum_{J<K}c_{JK}\eta_J\eta_K+\tfrac{1}{6}\sum_{J}c_{JJJ}\eta_J^3+\tfrac{1}{2}\sum_{J\neq K}c_{JJK}\eta_J^2\eta_K+\sum_{J<K<L}c_{JKL}\eta_J\eta_K\eta_L, \quad (1)$$

where the c_{JK} are the second-order elasticity coefficients and the c_{JKL} are the third-order coefficients.

It is often convenient to rewrite the above expansion in terms of the displacement gradients $u^a_{,b}=\partial u^a/\partial X^b=x^a_{,b}-\delta_{ab}$. In this case, the macroscopic energy density is conveniently rewritten as the sum of four contributions

$$U'=U'_1+U'_2+U'_3+U'_4$$

because of the fact that η_J contains terms both linear and quadratic in the $u^a_{,b}$ (for example, $\eta_1=u^1_{,1}+\frac{1}{2}u^i_{,1}u_{i,1}$ and $\eta_4=u^2_{,3}+u^3_{,2}+u^i_{,2}u_{i,3}$). Thus U'_1 contains the contribution to the energy density which is quadratic in the $u^a_{,b}$ and therefore involves only the c_{JK}. The terms U'_2 and U'_3 are cubic in the $u^a_{,b}$ with U'_2 involving only the c_{JK} and U'_3 involving the c_{JKL}. The term U'_4 contains the contributions of higher order in the $u^a_{,b}$ and will be of no further interest here.

It was demonstrated in I how the *microscopic* strain energy can be written as a series expansion in terms of the variables

$$\lambda_{mn}(l)=(\mathbf{x}_m(l)\cdot\mathbf{x}_n(l)-\mathbf{X}_m\cdot\mathbf{X}_n)/2a_0,$$

where the $\mathbf{x}_m(l)$ are the positions, relative to a reference nucleus in cell (l), of nuclei in unit cells adjacent to cell (l); a_0 is a unit cell dimension. For example, for primitive structures, the microscopic strain-energy density $U(l)$ in the bulk of a large crystal is given by

$$\Omega U(l)=\tfrac{1}{2}\sum_{l'}\sum_{m,n;m'n'}B_{mnm'n'}(l-l')\lambda_{mn}(l)\lambda_{mn}(l')+\tfrac{1}{6}\sum_{l',l''}\sum C_{mnm'n'm''n''}(l,l',l'')\times\lambda_{mn}(l)\lambda_{m'n'}(l')\lambda_{m''n''}(l'')+\cdots, \quad (2)$$

where Ω is the unit cell volume and where $\mathbf{x}_1(l)$, $\mathbf{x}_2(l)$, and $\mathbf{x}_2(l)$ are the positions of the nuclei in three mutually adjacent cells adjacent to cell (l) relative to the nucleus in cell (l). The sets of coefficients $\{B\}$ and $\{C\}$ must be invariant under the operations of the relevant space group and $\{B\}$ must also be positive definite. As pointed out in I, the λ_{mn} are closely related to the macroscopic strain variables η_J and can be considered as the close microscopic analogies to the η_J. For example, in primitive structures we have the simple relation

$$\lambda_{mn}(l)=\frac{1}{a_0}\sum_{a,b}\eta_{ab}(l)X_m^aX_n^b. \quad (3)$$

One result of this equivalence is that $\lambda_{mn}(l)$ contains terms both linear and quadratic in the components of the displacements of the nuclei, $u_m^a(l)$, which are the microscopic equivalents of the $u^a_{,b}$ and can be expressed in terms of them. The expansion (2) when expressed in terms of the $u_m^a(l)$ can again be written as the sum of four terms. The term U_1 is quadratic in the u_m^a and involves only the B coefficients, while U_2 and U_3 are cubic in the u_m^a, U_2 involving only the B coefficients and U_3 involving only the C coefficients. The calculational method for the second-order elastic coefficients which was utilized in I consists of comparing the microscopic and macroscopic expressions for $U_1(u^a_{,b})$ so that the relationship between the c_{JK} and the B's is obtained. Because of the close analogy between the expansions (1) and (2), we can obtain the desired relationship between the c_{JKL} and the C's for primitive structures merely by comparing the microscopic and macroscopic

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[13] K. Brugger, Phys. Rev. **133**, A1611 (1964).

expressions for U_3. Because of the analogy, it is not necessary to calculate U_2; if we do so and compare the macroscopic and microscopic expressions for U_2, we merely obtain the relationship between the c_{JK} and the B's already derived in I by comparing the two forms of U_1. Thus, the form (2) has this further advantage over the usual expansion [e.g., Eq. (5) of I] in terms of the $u_m{}^a$ other than those already mentioned in I and which is a direct consequence of the very similar way in which the η_J and $\lambda_{mn}(l)$ are defined.

The above discussion requires some modification for the case of nonprimitive crystals. Firstly, the relationship (3) between η_J and λ_{mn} is no longer so simple for nonprimitive structures because $u_m{}^a$ now contains a term due to the internal strain between the sublattices. The internal strain component $\xi_{pq}{}^a$ between the pair pq depends on the local "external" strain $u^b{}_{,c}$ and can be written in the form

$$\xi_{pq}{}^a = A_{pq}{}^a(bc)u^b{}_{,c} + \nu_{pq}{}^a\,,$$

where $\nu_{pq}{}^a$ contains terms quadratic and higher in the $u^b{}_{,c}$. This relationship is, of course, determined by using the conditions

$$(\partial U/\partial \xi_{pq}{}^a) = 0\,,$$

as carried out, for example, in I; the ν term is zero in the harmonic approximation. It is interesting to note that $\nu_{pq}{}^a$ does not contribute to U_2. Its contribution would be

$$\nu(\partial U/\partial \xi)_{\nu=0}$$

and the derivative is zero, apart from some higher order terms which contribute only to U_4, because of the requirement $\partial U/\partial \xi = 0$. The $\nu_{pq}{}^a$ do not, of course, contribute to U_3. Because of the internal strain, there is no longer complete cancellation of the terms in U_2 and U_2' and it becomes necessary to compare $U_a = U_2 + U_3$ with $U_2' + U_3'$, to obtain the desired relations for the c_{JKL}.

To summarize, we shall calculate the microscopic $U_a(u_m{}^a, \xi_{pq}{}^a)$ and need use only the linear part

$$\xi_{pq}{}^a(\text{lin}) = A_{pq}{}^a(bc)u^b{}_{,c}$$

of the internal strain; this has already been calculated in terms of the B coefficients for diamond-like crystals in I. We can thus obtain the microscopic $U_a = U_a(e_J)$, (where e_J is the linear part of η_J and is identical with the usual infinitesimal strain variable), which involves the C coefficients and, in nonprimitive structures, also the B coefficients via the $A_{pq}{}^a(bc)$. This will be compared with the macroscopic U'_a, which is

$$U'_2 + U'_3 = \tfrac{1}{6}\sum_J c_{JJJ}e_J{}^3 + \tfrac{1}{2}\sum_{J\neq K} c_{JJK}e_J{}^2 e_K$$
$$+ \sum_{J<K<L} c_{JKL}e_J e_K e_L + \tfrac{1}{2}\sum_J^3 c_{JJ}e_J{}^3 + \tfrac{1}{2}\sum_{J\neq K}^3 c_{JK}e_J e_K{}^2,$$

plus terms which cannot be written as products of the e_J, and expressions for the c_{JKL} in terms of the C coefficients will thereby be obtained.

III. THIRD-ORDER ELASTICITY OF DIAMOND-LIKE CRYSTALS

The diamond type of crystal belongs to the cubic class and thus, because of symmetry, there are only six distinct third-order coefficients. In this case, the macroscopic U'_a reduces to

$$\begin{aligned} U'_a = {} & \tfrac{1}{6}c_{111}(e_1{}^3+e_2{}^3+e_3{}^3) + \tfrac{1}{2}c_{112}[e_1{}^2(e_2+e_3)+e_2{}^2(e_3+e_1)+e_3{}^2(e_1+e_2)] + c_{123}e_1e_2e_3 \\ & + \tfrac{1}{2}c_{144}(e_1e_4{}^2+e_2e_5{}^2+e_3e_6{}^2) + \tfrac{1}{2}c_{166}[e_1{}^2(e_5{}^2+e_6{}^2)+e_2(e_6{}^2+e_4{}^2)+e_3(e_4{}^2+e_5{}^2)] + c_{456}e_4e_5e_6 \\ & + \tfrac{1}{2}c_{11}(e_1{}^3+e_2{}^3+e_3{}^3) + \tfrac{1}{2}c_{12}[e_1{}^2(e_2+e_3)+e_2{}^2(e_3+e_1)+e_3{}^2(e_1+e_2)]. \qquad (4) \end{aligned}$$

We now turn to the question of the microscopic energy density. The most important forces between atoms in diamond-like crystals are apparently short-range forces due mainly to shell-shell and shell-core interactions. For example, Cochran[11] obtained a good fit between theoretical and experimental phonon dispersion and elasticity data with the shell-model and only short-range forces. Similarly, it was pointed out in I that the shell-model reduces to a rigid-ion formulation if only the elasticity is to be considered and excellent agreement was obtained between theory and experiment using only a nearest-neighbor interaction and a noncentral second-neighbor interaction.[1] The long-range quadrupolar interaction introduced by Lax is small for more distant neighbors[14] and can probably be ignored. The shell-shell and shell-core effects give only small contributions[12] for third and more distant neighbors. Thus, as in I, we shall include interactions out to second neighbors only.

There is only one purely first-neighbor third-order interaction[1] and this is the central one in $\lambda_{mm}{}^3(l)$. We can expect this interaction to be an important one since most of the anharmonicity will be in the first-neighbor shell-shell interaction. This contribution is the third-order equivalent of the first-neighbor central harmonic interaction represented by α in I. Similarly, we shall include a second-neighbor term in $\lambda_{mn}{}^3(l)(m\neq n)$, which is the anharmonic equivalent of the harmonic term represented by β in I and represents anharmonicity in the forces resisting changes in the angle between "bonds." We shall also include a third second-neighbor interaction, the terms in $\lambda_{mm}\lambda_{mn}{}^2$. The physical significance of these three types of interaction, represented by γ', δ', ϵ', respectively, will be discussed in greater detail in the final section. The energy density term U_1 has already

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[14] M. Lax, in *Lattice Dynamics*, edited by R. F. Wallis (Pergamon Press, Inc., New York, 1965), p. 179.

been calculated in I and the values of α,β have already been determined there by comparison with experiment.

We write the macroscopic strain-energy density as

$$U=\frac{1}{2}\frac{\alpha}{64a_0^5}\sum_{i=1}^{4}(x_{0i}^2-3a_0^2)^2+\frac{1}{2}\frac{\beta}{32a_0^5}\sum_{i,j<i}^{4}(\mathbf{x}_{0i}\cdot\mathbf{x}_{0j}+a_0^2)^2$$
$$+\frac{1}{6}\frac{\gamma'}{16a_0^6}\sum_{i=1}^{4}(x_{0i}^2-3a_0^2)^3+\frac{1}{6}\frac{\delta'}{8a_0^6}\sum_{i,j<i}^{4}(\mathbf{x}_{0i}\cdot\mathbf{x}_{0j}+a_0^2)^3$$
$$+\frac{1}{6}\frac{\epsilon'}{16a_0^6}\sum_{i\neq j}^{4}(x_{0i}^2-3a_0^2)(\mathbf{x}_{0i}\cdot\mathbf{x}_{0j}+a_0^2)^2,$$

where the atomic labeling is as in Fig. 1 and Ref. 1 and the required symmetry has been imposed on the C's. We have $C_{mmmmmm}(0)=\gamma'$, $C_{mnmnmn}(0)=(6/5)\delta'$, and $C_{mmmnmn}(0)=(6/5)\epsilon'$ and the primes[1] on the summations are dropped because of symmetry, as discussed in I. Now $(x^2{}_{01}-3a_0^2)$ is $2a_0^2[e_1+e_2+e_3+e_4+e_5+e_6+(u'+v'+w')/a_0]+0(e^2)$, for example, as given in I, where u', v', w' are the components of the internal strain difference between the two lattices. The linear parts of these components are $u'=-a_0\zeta e_4$, etc.,[1] where $\zeta=(\alpha-\beta)/(\alpha+\beta)$, as derived in I. Thus, we obtain

$$\begin{aligned}U_a=&[(\gamma'-\delta'+3c')/3+(\alpha+3\beta)/8a_0](e_1^3+e_2^3+e_3^3)+2(\gamma'+3\delta'-\epsilon')e_1e_2e_3\\&+[\gamma'-\delta'+\epsilon'/3+(\alpha-\beta)/8a_0][e_1^2(e_2+e_3)+e_2^2(e_3+e_1)+e_3^2(e_1+e_2)]+2\gamma'(1-\zeta)^3e_4e_5e_6\\&+[\gamma'(1-\zeta)^2+\delta'(1+\zeta)^2+\tfrac{1}{3}\epsilon'(1+\zeta)(3\zeta-1)+\zeta^2(\alpha-\beta)/8a_0](e_1e_4^2+e_2e_5^2+e_3e_6^2)\\&+[\gamma'(1-\zeta)^2-\delta'(1+\zeta)^2+\tfrac{1}{3}\epsilon'(1+\zeta)(3-\zeta)+\zeta^2(\alpha-\beta)/8a_0][e_1(e_5^2+e_6^2)+e_2(e_6^2+e_4)^2+e_3(e_4^2+e_5^2)].\end{aligned}$$

Upon comparing this expression with Eq. (4) for the macroscopic U_a, we obtain

$$\begin{aligned}c_{111}&=\gamma-\delta+9\epsilon,\\c_{112}&=\gamma-\delta+\epsilon,\\c_{123}&=\gamma+3\delta-3\epsilon,\\c_{144}&=\gamma(1-\zeta)^2+\delta(1+\zeta)^2+\epsilon(1+\zeta)(3\zeta-1)+c_{12}\zeta^2,\\c_{166}&=\gamma(1-\zeta)^2-\delta(1+\zeta)^2+\epsilon(1+\zeta)(3-\zeta)+c_{12}\zeta^2,\\c_{456}&=\gamma(1-\zeta)^3,\end{aligned}\tag{5}$$

where $\gamma=2\gamma'$, $\delta=2\delta'$, $\epsilon=\frac{2}{3}\epsilon'$, and $c_{12}=(\alpha-\beta)/4a_0$.

IV. DISCUSSION OF RESULTS

The validity of the model chosen here for the anharmonic interactions in diamond-like crystals depends in part on Eq. (5) giving a reasonable description of the third-order elasticity of such materials. Now the constant ζ has already been calculated in I and is not available for adjustment to fit the experimental data. Thus, we have three adjustable parameters to fit six coefficient values. With this small number of interactions we cannot expect highly accurate agreement with experiment. However, such accurate agreement was not the aim of this article since the experimental results[3–5,7,15] reported by different workers show appreciable scatter and an accurate fit to one set of data would be undesirable. Instead, we aim to obtain theoretical values of the six constants which reproduce the general trend of differences between their experimental values, using a small number of anharmonic force constants. We shall see that this goal has been achieved remarkably well.

We shall use the recent data on silicon and germanium by McSkimin and Andreatch[3] for the comparison of our theoretical results with experiment. Other, earlier data is also given in Ref. 3. More recent data on germanium is available in Ref. 7. Because we have three constants and six elasticity coefficients, Eqs. (5) predict three relations between the coefficients. However, instead of using this approach, we have tried to obtain an approximate fit to the experimental data by trial and error and the resultant fit is exhibited in Table I. It will be noted that the agreement obtained is remarkably good for the use of only three adjustable parameters, which suggests that the anharmonic force-constant model used is a fairly realistic one. The introduction of additional interactions into the strain energy will, of course, allow a much better fit but such a fit might then not be significant physically. The present agreement is about as good as the agreement between the experimental results of

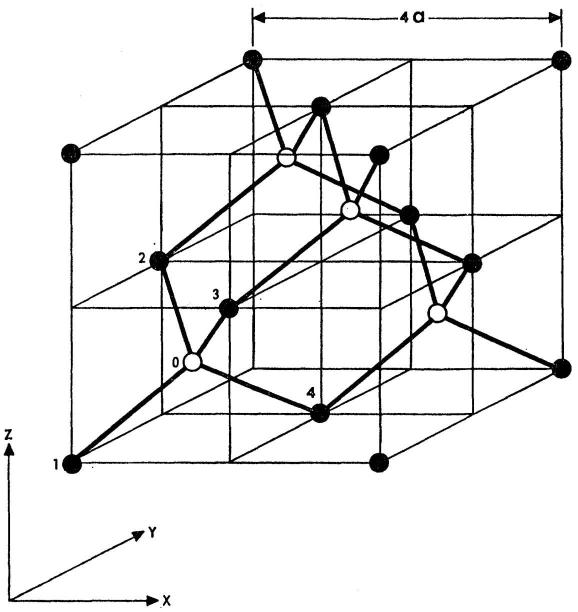

FIG. 1. The crystal model. The open and filled circles represent the atoms on the two different sublattices.

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[15] J. J. Hall (private communication).

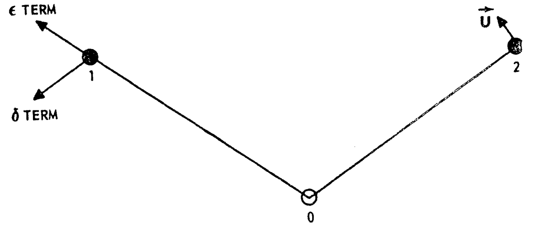

FIG. 2. Anharmonic forces on atom 1 due to a small rotation of the bond 02, as deduced by comparing theoretical and experimental results.

different workers[3–5,7,15] and the use of additional interactions would appear to be premature at this time.

It is of interest to examine the physical nature of the interactions which have been included in this work. The γ interaction is, of course, the purely first-neighbor central term and the large magnitude of γ (see Table I) is not surprising in view of the expectation[8] that most of the anharmonicity lies in the interaction between the valence electrons associated with nearest neighbors. The physical nature of the two second-neighbor interactions is not so obvious and thus we shall now discuss them in more detail. We calculate the anharmonic force on nucleus 1 due to displacements of nucleus 2 only:

$$F_{12}=\left(-\frac{\partial U(l)}{\partial \mathbf{x}_{01}}\right)_2=-\frac{(\mathbf{u}_{02}\cdot\mathbf{X}_{01})}{32a_0^6}\{\epsilon(\mathbf{u}_{02}\cdot\mathbf{X}_{01})\mathbf{X}_{01}+[2\epsilon(\mathbf{u}_{02}\cdot\mathbf{X}_{02})+\delta(\mathbf{u}_{02}\cdot\mathbf{X}_{01})]\mathbf{X}_{02}\}.$$

If

$$\mathbf{u}_{02}\cdot\mathbf{X}_{02}=0,\quad\text{then}\quad \mathbf{F}_{12}=-\frac{(\mathbf{u}_{02}\cdot\mathbf{X}_{01})^2}{32a_0^6}(\delta\mathbf{X}_{02}+\epsilon\mathbf{X}_{01}).$$

The forces represented by this expression are shown in Fig. 2, where the negative value of ϵ has been taken into account. We see that, roughly speaking, the δ term is the expected rotational response due to the tendency to maintain the bond angle at its equilibrium value, whereas the ϵ interaction is a less obvious one in which a rotation of bond "02" tends to push atom "1" out radially. The effect can be qualitatively understood in terms of the molecular orbital picture. A change in the bond angle results in a change in the *s-p* mixing for the orbitals associated with atom "0" and it is this which causes the tendency of the second neighbor to move outwards away from the central atom. However, the detailed nature of this process is not at present fully understood. Nevertheless, it is clear that this interaction is primarily a direct second-neighbor effect and cannot be ascribed to the coupling of first-neighbor shell-shell terms. It is important in that it accounts for the large difference between c_{111} and c_{112} and about half of the large difference between c_{112} and c_{123}. Most of the difference between c_{144} and c_{166} is due to the δ interaction while the general strong negative values of the elastic constants are due to the very important nearest-neighbor γ term.

TABLE I. Theoretical fits to the experimental data of Ref. 3. All quantities in units of 10^{12} dyn/cm^2. The values of α, β (and thus ζ) computed in Ref. 1 have been used (i.e., $\zeta=0.55$ for silicon and 0.53 for germanium).

	Silicon		Germanium	
	Theory	Experiment	Theory	Experiment
c_{111}	−8.21	−8.25	−7.38	−7.10
c_{112}	−4.45	−4.51	−3.54	−3.89
c_{123}	−0.69	−0.64	−0.26	−0.18
c_{144}	+0.14	+0.12	−0.10	−0.23
c_{166}	−3.43	−3.10	−3.08	−2.92
c_{456}	−0.33	−0.64	−0.28	−0.53
γ	−3.51		−2.72	
δ	+0.47		+0.34	
ϵ	−0.47		−0.48	

We have also investigated the replacement of the ϵ contribution to the strain energy by some of the other second-neighbor interaction terms but these give results which are appreciably inferior in reproducing the differences in the experimental values of the third-order constants. When the experimental values of the macroscopic constants are more accurately known, it will be convenient to introduce one or two of these interatomic interactions in addition to those included in the present calculation. At the present time, however, we have a reasonably good fit to nine pieces of data (three second-order and six third-order elastic constants) with five adjustable parameters, two harmonic and three anharmonic atomic force constants. Furthermore, these five interactions all contribute in a physically reasonable way.

Finally, it should be noted that the theoretical analysis presented here is also directly applicable to the III-V compounds with the analogous zinc blende structure. The long-range Coulomb effects due to the ionicity should not be very anharmonic and thus the present short-range anharmonic forces model should be a reasonable one for this case, too. However, the parameter ζ is to some extent adjustable in this case (unless it is directly measured), since the long-range Coulomb effects do contribute to the harmonic forces and the model used in I is less valid for zinc blende structures, as shown in Table II of I. It will be of interest to compare the present theoretical results with the experimental data for these solids when the latter become available.

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