tions were superimposed, but no effort was made to form a contiguous cavity linking them together (in contrast to some earlier experiments). The first junction was operated as a Josephson junction. The second junction acted as a detector of the fields created in the first by observing the structure induced in its I-V characteristic by these fields. Since the geometery excludes the possibility of detection of the fields created by TC, the observed structure in the detector junction can be considered as a verification of the existence of coupling between the ac Josephson currents and the surface plasmons occurring for three coupled superconducting films.[3]

We conclude by pointing out two possible ways for distinguishing between the two coupling mechanisms. The different temperature behavior offers one possibility. The $d_m^{1/2}$ dependence of $\bar{c}_l$ ($\bar{c}_t$ is independent of d_m as long as $\lambda \ll d_m$) is another test which may be used.

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DIELECTRIC DEFINITION OF ELECTRONEGATIVITY

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A simple model is proposed for the static electronic dielectric constants of zincblende and wurtzite crystals in terms of the dielectric constants of diamond-type crystals. The model suggests a natural definition of electronegativity in (sp^3)-hybridized valence states.

A dielectric model for covalently bonded crystals has been proposed[1] which accounts for the observed x-ray charge distribution in diamond, and predicts the optical-pseudopotential form factors[2] in terms of self-consistent dielectric screening of ion-core form factors derived from values of the free-ion term.[3] The homopolar model Hamiltonian is the spherically averaged one used by Penn[4] which contains an energy gap E_{g0} at the Jones zone faces. The relation between E_{g0} and the homopolar static dielectric constant ϵ_{00} is[4]

$$\epsilon_{00} = 1 + (\hbar\omega_p/E_{g0})^2 A, \tag{1}$$

where $\hbar\omega_p$ is the plasma energy and A is a number of order unity.[4]

To treat heteropolar (sp^3)-bonded crystals, the homopolar model Hamiltonian has been generalized[5] from a two-dimensional to a four-dimensional space, yielding a relation similar to (1) but involving a charge transfer constant

$C_{\alpha\beta} = y_{\alpha\beta}E_{g0}$. The dimensionless parameter y measures the strength of heteropolar/homopolar interactions and the static heteropolar dielectric constant is given by

$$\epsilon_0 = 1 + (\hbar\omega_p)^2/[(E_{g0})^2 + (C_{\alpha\beta})^2]A. \tag{2}$$

It has been shown[5] that the heteropolar model explains heteropolar optical form factors[2] and the effective charge e^* which splits the frequencies of longitudinal and transverse optical lattice vibrations.

In this note we show that the assumption

$$C_{\alpha\beta} = \left(\frac{Z_\alpha e^2}{r_{\alpha 0}} - \frac{Z_\beta e^2}{r_{\beta 0}}\right) f_{\alpha\beta} \tag{3}$$

enables one to calculate ϵ_0 for all zinc-blende and wurtzite crystals given only ϵ_{00} as a function of nearest-neighbor spacing d in the diamond-type crystals C, Si, Ge, and grey Sn. This function, together with $E_{g0}(d)$, is shown in Fig. 1. Here for atom α the valence is Z_α, and $2r_{\alpha 0}$ is the nearest-neighbor spacing d of the diamond-type crystal belonging to the same row as atom α. The factor $f_{\alpha\beta}$ represents the effect of Thomas-Fermi screening. When α and β belong to the same row of the periodic table, then

$$f_{\alpha\beta} = f_{\alpha\alpha} = 0.9\exp(-k_s r_{\alpha 0}), \tag{4}$$

where k_s is the Thomas-Fermi screening radius for a free-electron gas,

$$k_s = 0.82 k_F r_s^{1/2}, \tag{5}$$

having the appropriate valence-electron density,

$$N/\Omega = (4\pi r_s^3/3)^{-1}.$$

When α and β belong to different rows, $f_{\alpha\beta}$ is the average of $f_{\alpha\alpha}$ and $f_{\beta\beta}$.

The preceding definitions constitute a simple, closed system from which we can calculate ϵ_0 for 15 zinc-blende and nine wurtzite crystals given only $\epsilon_{00}(d)$. The results obtained are shown in Table I.

In discussing these results we note from Fig. 1 that there is a break in the curves $\epsilon_{00}(d)$ or

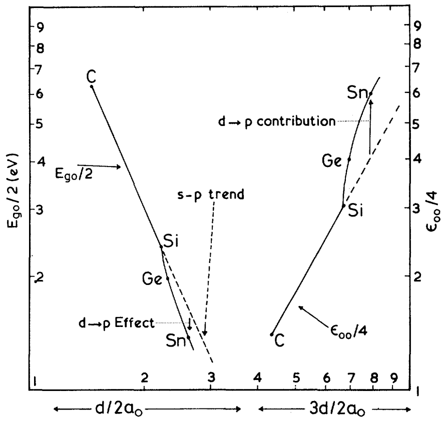

FIG. 1. Log-log plots of the parameters $\frac{1}{2}E_{g0}$ (in eV) and $\frac{1}{4}\epsilon_{00}$ against nearest-neighbor distance d in a.u. The s-p trend is separated from the effects of d core polarization.

Table I. Dielectric properties of diamond, zincblende, and wurtzite (*) crystals.

Crystal	$C_{\alpha\beta}$(eV)	y	ϵ_{00}	ϵ_0(calc.)	ϵ_0(exp.)
C	0	0	5.7	5.7	5.7
Si	0	0	12.0	12.0	12.0
Ge	0	0	16.0	16.0	16.0
Sn	0	0	23.8	23.8	23.8
SiC	4.0	0.50	8.6	7.1	6.7
BN	7.0	0.56	5.7	4.6	4.5
BeO*	6.3	1.28	6.3	3.0	3.0
AlP	3.1	0.65	11.9	8.7	
MgO	13.5	1.55	8.1	3.1	3.0
MgTe*	4.35	1.50	22.5	7.6	
GaP	3.6	0.75	12.0	8.1	8.4
AlN*	8.8	1.10	8.6	4.4	4.8
GaAs	3.0	0.71	16.0	11.0	10.9
ZnSe	5.9	1.41	16.0	6.0	5.8
ZnO*	13.5	1.90	8.8	2.7	4.6
ZnS*	6.2	1.29	12.0	4.7	5.1
ZnTe	4.4	1.30	19.7	7.9	8.3
AlSb	1.7	0.50	19.7	15.9	10.2
InP	3.5	0.93	17.0	9.6	9.6
AlAs	2.7	0.63	14.6	10.8	
GaN*	8.9	1.17	8.6	4.2	
InN*	9.0	1.50	10.2	3.8	
InSb	2.2	0.79	23.8	15.1	15.7
CdTe	4.4	1.58	23.8	7.5	7.1
CdS*	6.1	1.57	16.7	5.5	5.2
CdSe*	5.8	1.65	19.2	5.9	7.0
GaSb	1.9	0.55	19.7	15.4	14.4
InAs	3.2	0.93	19.2	10.8	12.2

$E_{g0}(d)$ near Si. The reason for this is that an important contribution to ϵ_{00} is made for third (Ge)- and fourth (Sn)-row atoms by $nd \rightarrow (n+1)p$ transitions.[6] Indeed (as shown by comparing the dashed and solid curves), one-third of the derivative of $E_{g0}(d)$ at Sn arises from these transitions, so that we may say that one-third of the forces responsible for the grey-Sn (covalent) → white-Sn (metallic) transition arise from d core electrons.[7-9] (The same forces are responsible for the large cohesive energies of Cu, Ag, and Au compared with the alkali metals.) Penn's model does not include any specific effect of this type; so we expect markedly better results when there are no d electrons in the core of α or β. It is worth noting, however, that the model includes some core-polarization effects through the use of observed nearest-neighbor distances. The d electron polarization gives rise to very poor results for $\epsilon_{00}-\epsilon_0$ in only two cases, ZnO and AlSb. In the remaining 16 crystals for which data are available, the average absolute error in $\epsilon_{00}-\epsilon_0$ is less than 10%. The contribution from d electron polarization could be treated more accurately by adding a parameter for the oscillator strength and using the observed values[6] for $nd \rightarrow (n+1)p$ thresholds, but we shall not do this here.

From the success of this simple model it is clear that, although E_{g0} is an inherently quantum-mechanical parameter which characterizes the strength of the covalent bond, the charge transfer constant $C_{\alpha\beta}$ is semiclassical in nature. For elements α and β belonging to the same row of the periodic table, $C_{\alpha\beta}$ is proportional to $(Z_\alpha - Z_\beta)$, which suggests that we can use $C_{\alpha\beta}$ to define an electronegativity scale for (sp^3)-hybridized valence states.

There are two electronegativity scales in common use. The ideal scale is additive:

$$X_{\alpha\beta} = X_{\alpha\gamma} + X_{\gamma\beta}, \qquad (6)$$

which implies that

$$X_{\alpha\beta} = X_\alpha - X_\beta. \qquad (7)$$

Mulliken's scale is based on the ionization energies of valence states of free atoms,[10] so that it automatically satisfies (6) and (7). It does not describe, however, the power of an atom to attract electrons as it is modified in a bonded condition. Pauling noticed that the α-β bond energy generally exceeds the mean of the α-α and β-β bond energies, and one would expect that the difference $\Delta_{\alpha\beta}$ should be proportional to $(Z_\alpha - Z_\beta)^2$ when α and β belong to the same row. Thus he defined $X_{\alpha\beta} \propto [\Delta_{\alpha\beta}]^{1/2}$ in general.

Because Pauling's definition already describes bonded atoms, it has been widely used. His original values for $\Delta_{\alpha\beta}$ were taken from a wide range of valence states and did not always satisfy (6) and (7). It has been found[10] that use of modern data for *mean* bond energies in polyatomic molecules $\alpha\beta_n$ yields values more consistent with (6) and (7). These are listed in Table II.

It is widely believed[11] that three factors influence acidity of α relative to β: electronegativity, bond energies, and dielectric factors, and that of these, bond energy is the *least* important. A definition of electronegativity based on the dielectric properties of the atoms in a definite valence state should therefore prove useful. The results of Table I show that (3) can be used to define electronegativity dielectrically. We eliminate the medium factor $f_{\alpha\beta}$ in order to satisfy (6) and (7) and therefore define

$$X_\alpha = 3.6(Z_\alpha / r_{\alpha 0}) f_{\alpha\alpha} + 0.5. \qquad (8)$$

The additive constant and the constant of proportionality have been chosen so that with $r_{\alpha 0}$ in atomic units, (8) gives $X_\alpha = 2.5$ and 3.0 in C and N, in agreement with Pauling.[12] Analysis of his values shows (although it is not stated explicitly) that he has chosen

$$X_\alpha \propto Z_\alpha / r_{\alpha 0}, \qquad (9)$$

i.e., Pauling omits the Thomas-Fermi screening factor. This is not inappropriate if one considers an average electronegativity for a wide variety of valence states, but for a *fixed* valence state [e.g., (sp^3)], there is little doubt that (8) is superior to (9). This becomes apparent immediately from Table I if one com-

Table II. Electronegativity values for tetravalent elements. The column BD is Ref. 13, and the other columns are discussed in the text.

Element	Atomic	Pauling	BD	Dielectric
C	2.63	2.5	2.50	2.50
Si	2.44	1.8	1.65	1.41
Ge	⋯	1.8	1.40	1.35
Sn	⋯	⋯	1.65	1.15

pares the value of $C_{\alpha\beta}$ for BN (where $f = 0.200$) with GaAs ($f = 0.135$) or InSb ($f = 0.120$).

The essential feature of (8) and (9) is the variation from C to Si to Ge to Sn; the remaining values for all other nontransition elements save H can be obtained by varying Z_α. We show in Table II electronegativity values for these elements based on Mulliken's atomic values,[10] Pauling's bond energies [which is really only Eq. (9)], a refined set of Pauling values,[13] and our dielectric definition (8). The refined value[13] for Sn in a IV valence state is much larger than our (sp^3) value; it must include substantial d hybridization. Otherwise the refined values are closer to ours than Pauling's original values are; apparently the more recent empirical studies have tended to include partially the screening factor $f_{\alpha\alpha}$, without recognizing its dielectric origin.

As an example of the superiority of our definition to the ad hoc chemical definitions, we cite the fact that Cl has only a small hydrogen-bond forming power, whereas nitrogen-hydrogen bonds are of medium strength. All the chemical scales of electronegativity predict that Cl is at least as electronegative as N, whereas our dielectric scale places Cl near B, which also forms very weak hydrogen bonds. Pauling attempts to explain this by noting[12] that Cl is larger than N, but as shown by (9), his scale already includes size effects. It does not include, however, changes in k_s, and these are what make Cl weakly electronegative compared with N.

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