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Electronic charge distribution in crystalline diamond, silicon, and germanium

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Cummings and Hart) structure factors, finding signif-
icant discrepancies for all except the OPW

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Fourier-synthesized total density map
G'max

pexpt(&~ Gmax) =) pexpt(G)e

G,

(4b)

approach used originally by Dawson and

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TABLE I. Parameters obtained by fitting Dawson's model [Eqs. (5) and (6)] to the observed structure factors of C, Si, and Ge. For C and Si we use Clementi s (Ref. 51) nonrelativistic orbital densities n„I,, while for Ge we use both Clementi s nonrelativistic (NR) orbital densities as well as the relativistic LDA results. In all cases we use A~ = 4 for all l values. An

asterisk indicates that quantity was held fixed during the fit. The value (= 2.435 for Si in Ref. 6 was in

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find that when the f' values are subtracted, using the
best estimate of the Debye-Wailer factor, there are fairly
large deviations between the three sets for the common
(hkt) 's.
One approach to the problem taken by Brown and
Spackman4s is to combine the sets in a somewhat ar-bitrary fashion. They use the MK set but replace the
(111) and add the (511) values by those of TS, scaled by the average ratios of the common reflections in the two sets. The analysis of Brown and Spackman did not con-sider the DHC set, which was not yet published at that
time.

We have adopted a different approach. We fit the
structure factors of TS and MK separately with the same

"standard" model (B,(, As, A4, Sj, where S is a scale
factor [Eq. (8)t, common to all I" 's of a set as in Ref. 45.
The residuals for MK were 1.5—2 times smaller than for

TS. Consequently, we adopted the set of MK as it is with no replacement or additions from scaled values of TS. However, we added to this set the DHC set, which has a few (hkt) in common with MK and a few higher-order reflections, which are important for the determination of B. Where common (hkl) exist, both were included in
the set. The fit of the standard model to this combined

set (using of course two S's; one for MK and one for DHC) did not show any systematic trends in the resid-uals, which remained all of the same average magnitude as for the MK set alone.

The number of I' 's in the measured set is 14, with some hkt having two measured values. The f' values used are those of Creaghss for the MK data (f' = —1.089), and those of DHC (Ref. 43) for their data (f' = 0.09). These

were fixed in all fits.

Since Ge is a rather heavy element (Z = 32), one might expect relativistic effects to be non-negligible in the cal-
culated charge densities and structure factors. This is
indeed born out by the comparison of the local-density-approximation (LDA) nonrelativistic (NR) atomic struc-ture factors with their relativistic (R) counterparts shown in Table II. Both sets of structure factors were calculated
from numerical integration of the local-density equa-
tions using the spin-polarized Ceperley-Alder e~change-correlations and the s p2 configuration. The corre-
sponding relativistic corrections for carbon and silicon
are negligible. Table II shows also the Hartree-Fock (HF) atomic results. s~ si We see that whereas the relativistic

correction n~(q) —nNR(q) is similar in the LDA and HF results, there are differences between the absolute values of n(q) as calculated by HF and the LDA. These reflect
the absence of correlation in the HF calculation and the

different nature of the exchange potential (local versus
nonlocal in the LDA and HF results, respectively). Since there are no tabulations of orbital-by-orbital Hartree-
Fock relativistic densities n„i(r) for Ge, the relativistic
fits were done using LDA orbitals.
Table I gives the results of the fit to the Ge data us-ing relativistic LDA orbitals. The table also gives the
results obtained with the conventional (nonrelativistic)
Clementi Hartree-Fock orbitals. We see that the statis-

tical quality of both Bts are nearly the same. However,
as shown below, the relativistic fit produces much bet-
ter agreement with (i) the measured forbidden (442) and
(622) reflections and (ii) the calculated crystal structure
factors. Thus, we will use below mostly the results of

TABLE II. Calculated free-atom germanium structure factors n(q) for momentum q = sine/A as obtained in the local-density approximation (LDA) and the Hartree-Fock (HF) approximation. We list the nonrelativistic (NR) values, as well as the relativistic (R) correction np(q) —nNp, (q).

0.05
0.10
0.15
0.20
0.25
0.30
0.35
0.40
0.50
0.60
0.70
0.80
0.90
1.00
1.10
1.20
1.30
1.40
1.50

LDA

nNR(q)
31.278
29.540
27.498
25.532
23.722
22.040
20.450
18.934
16.130
13.699
11.695
10.114
8.906
7.997
7.313
6.789
6.371
6.020
5.710

HF

nNR(q)
31.278
29.527
27.477
25.532
23.759
22.109
20.536
19.021
16.188
13.716
11.681
10.083
8.870
7.963
7.286
6.770
6.361
6.018
5.714

LDA

n~ (q) —nNR (q)
0.006
0.019
0.029
0.031
0.028
0.024
0.023
0.026
0.040
0.054
0.064
0.068
0.067
0.065
0.062
0.059
0.059
0.060
0.061

HF

nR (q) —nNR(q)
—0.002
0.007
0.027
0.035
0.032
0.027
0.024
0.026
0.039
0.054
0.064
0.068
0.067
0.065
0.062
0.060
0.058
0.058
0.060

Reference 51.
Reference 57.

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TABLE III. Dynamic (F) structure factors for Si in units of e/atom corresponding to the most converged set (column 1) in Table I. The experimental data (corrected for anomalous dispersion and nuclear scattering), including the estimated standard deviations o (in me/atom), are taken from Cummings and Hart (Ref. 1), except the (222) result taken from Alkire, Yelon, and Schneider (Ref. 7). The difference bFr is F,~~,(G) —Fe»t(G) (me/atom), while bF2 = F,„r(G) —F,„~t(C) (me/atom). The root-mean-square deviation for hF& is 12 me/atom and an unweighted R factor is 0.21%%uo. The dynamic F, &,(G) is obtained from the static p, ~,(G) using B = 0.4632 (Ref. 30).

111
220
311
222
400
331
422
333
511
440
444
551
642
800
660
555
844
880

(G)
[Eq. (12a)]
10.600
8.397
7.694
0.161
6,998
6.706
6.094
5,760
5.781
5.318
4.115
3.931
3.649
3.253
2.917
2.802
2.165
1.543

Dynamic, solid
Fe&cpt (&)
[Eq (1)]
10.6025
8.3881
7,6814
0.1820
6.9958
6.7264
6.1123
5.7806
5.7906
5.3324
4.1239
3.9349
3.6558
3.2485
2.9143
2.8009
2.1506
1.5325

2.9
2.2
1.9
1.0
1.2
2.0
2.2
2.1
2.7
2.0
1.8
3.4
5,4
3.4
1.6
2.1
2.4
2.6

—3
9

13 —21
2
—20
—18

—21 —10
—14

9 —4
—7
5
3
1
14
11

10.455
8.450
7.814
0.000
7.033
6.646
6.077
5.769
5.769
5.302
4.107
3.925
3.644
3.251
2.915
2.802
2.163
1.542

—148
62
133
—182

37 —80

—35 —12 —22

—30
—17
—10
—12
3
1
1
12
10

Dynamic, atoms

F-,(G)
[Eq. (15)]

This is compared with the local-density calculated struc-

ture factors F, &,(G) from Eqn25.67 Td Td ((G63aT BT 1 493.)] fromfactors

~,

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p, (G) is similar to the agreement with p (C) for the
first ll G values, for which theoretical results are avail-
able. Hence, the use of two diferent Debye-Wailer factors
in the model does not significantly affect this compari-
son. There are, however, systematic deviations between

p (G) and p, (G) for higher G values: while our R,

value remains 0.21'%%uo using either the first ll G or all 18
G values, our R increase from 0.16%%uo (11 G values) to
0.35% (18 C values).
Our values R, = 0.2l%%uo and R = 0.16'%%uo show im-
provement over all previous LDA calculations. Note in
particular the improvement over the pseudopotential re-

TABLE IV. Comparison between calculated and experimentally deduced static structure factors p(C) for Si in units of e/atom. We give two sets of experimental p(G): (i) The value p, (G) is extracted from F,„~t(G) using in Eq. (4a) a single B = 0.4632 from Spackman (Ref. 30). The experimental F „„t(G)(corrected for anomalous dispersion and nuclear scattering) are taken from Cummings and Hart (Ref. 1) except the (222) result taken from Alkire, Yelon, and Schneider (Ref. 7). (ii) The values p~(G) are the Fourier transforms of the static model-density fit by Deutsch (Ref. 6) to the same set of F,„~t,(G). This uses difFerent B values for core and valence (Table I). In the theoretical values we include only those pseudopotential results in which the core contribution has been added by the authors. We also give the R factors [Eq. (22)] and the number of data points (N) used in comparison. The calculations shown use a variety of approximations for the exchange-correlation potential, i.e.,

the

local-density approximation (LDA) (including the Xn), the Hartree-Fock (HF) approximation, or the nonlocal weighted-density
approximation (WDA). These Hamiltonians were approximately solved by a variety of band-structure methods: LAPW is the linearized augmented plane wave, OPW is the orthogonalized plane wave, LCAO is the linear combination of atomic orbitals, LCGO is the linear combination of Gaussian orbital, and PP is pseudopotential.

111
220
311
222
400
331
422
333
511
440
444
551
642
800
660
555
844
880

R,'Fp
&m~0
N

Expt,

G)

7) (18

7).220

0.

0.

0.

values).

0.
G)

844

LAPW

0.

0.

0.

5 7 3 0.

7). 551

(HF)

0.

7)

(18 7) 0. 0.

0.
7).

4

7) 0. 55 477)

7) 555

I).

555

0.

551

I).

I).

47

444

0.

(18 potential,

0.

551
LAPW

(LDA)

7) (18 88

I).

33

0.

0. 64220

7).

440

0.

555

wave,

555

0.I).220

0.

(18

(LDA)

0.

2 7 7).

0 555

0

I).

0.

400

880 880

0.

0.

880

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suits, e.g.

, Yin and Cohen~s giving R,

= 1.09% and
R~

= 1.02%, Zunger~s yielding R,

= 1.44% and

R
1.40%, and the improvement over the recent weighted-density calculation of Balbas et aL s R, = 1.53% and R~

= 1.47%. The recent Hartree-Fock calculation
of Pisani, Dovesi, and Orlando2s yields

a rather good
R,

= 0.24% and R~

= 0.36%.
The improvement in the present calculation over pre-vious pseudopotentiat results reflects our use of an all-electron representation whereby core and valence states are treated self-consistently as solid-state wave functions on equal footing. The improvement in the present

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GW data. We also notice that the scaled GW structure
factors are systematically larger than the calculated val-
ues by an average of 17 me/atom for C & (531).

Static structure factors for diamond

There are many previous calculations of the static
structure factors 7

and charge-density maps
of diamond. Table VI compares the experimental static p,»r, (G) deduced from the TTKS and the GW data with various previous theoretical calculated values.

Since for diamond B«„=B ~~,„«(Table I), there is no ambiguity in the choice of p, (G) vs p (G) as dis-cussed for Si. Hence, the experimental p,»r, (G) were
obtained from Eq. (4a) by using a single Debye-Wailer
factor from Table I. As was the case for the dynamic
structure factors, we find that all calculations agree much
better with the TTKS data than with the GW data. The Xn calculation of Ivey"2 produces the best R fac-tor (Rq = 0.80%%uo and R2 = 1.91%), while our calculation yields Rq = 0 95%%uo and R2 = 2.36%%uo. Our R factor for
the static structure factors in diamond is 5 times larger
than our R factor for Si (0.21%%uo). This is partly due to the fact that the denominator of B for diamond is only
s

that of Si, even though the rms value of bFq is only slightly larger than that of Si (for the TTKS data).

$. Dynamic structure factors for germanium

Table VII compares the experimental F,»r, (G) of Ge with the calculated values using B = 0.5654 A2 obtained in the relativistic fit (Table I). The rms error between theory and experiment is 170 me/atom and the R factor is 0.85%%uo. Solid-state effects are apparent by noting that the R factor of F,»t(G) relative to the superposition of free atom density is 1.10%%uo.

6. Static structure factors for germanium

Table VIII compares various theoretically calculated static p,~~,(G) with experiment. We use two experi-mental sets of p,„~t,. one derived from a model fit using
Clementi's nonretativistic Hartree-Pock free-atom struc-ture factors, and one using

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while Figs.

2

—4 show the deformation charge densities
Ep«„(r), Ap ~(r), and Ap«(r), respectively [Eq. (18)
for

G

—+ oo]. In this and in the subsequent figures we

show side-by-side line plots in the (ill) direction and contour plots in the (110) plane. The solid

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TABLE X. Values of the valence and total charge density [p,~(R) and pq, q(R) in e/A] and ~R~ (in units of v 3a, origin on the left atom) at various points (o., P, p, 6, and e) along the (ill) direction as indicated in Figs. 2 and 3. The second line for each entry lists the corresponding value obtained from the model fits to the measured structure factors. The negative p„~ value obtained for Ge from the model at point e gives an idea of the uncertainty in the model fit. We only list the model fit of TTKS data for diamond.

Calc.

Expt.

Calc.
Expt.

Calc.
Expt.

Calc.

Expt.

Calc.

Expt.

Calc.
Expt.

1.55
1.47

1.38
1.08

1.94
1.93

1.59
1.61

0.09
0.10

1.59
1.61

iRi

0.056
0.060

0.029
0.030

0.064
0.069

0.125
0.125

0.50
0.50

0.125
0.125

P

p,)(R)
0.31
0.30

p- ~(Rs)
0.01
0.13

p-~(R~)
0.57
0.58

p-~(R~)
0.56
0.58

p~a(R.)
0.02
0.02

pgog(Rp)
0.56
0.58

Si
[Rf

0.081
0.083

0.040
0.042

0.096
0.103

0.125
0.125

0.50
0.50

0.125
0.125

0.36
0.40

0.02
0.16

0.49
0.60

0.46
0.58

0.02 —0.01
0.49
0.61

Ge
fRf

0.077
0.079

0.044
0.042

0.091
0.092

0.

0.

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close to the amplitude of p»i(R~), indicating that the
core contribution is negligible there.
(v) The deformation density maps Epics (Fig. 4)

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0.

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E. Comparison of ab initio and model
charge densities: p„~

& -(a) p„(cate)-

The text surrounding Eqs. (4b)—(7a) highlights the
fundamental difFerence between a Fourier-synthesis ap-
proach [Eq. (4b), illustrated in Figs. 6 and 7) and the
model-density approach [Eqs. (5)—(7a)] of Dawson,
Stewart, 4 Coppens, Spackman, 4

and Deutsch:

the latter method is guaranteed, by construction, to yield a smooth function despite the use of a limited set of struc-
ture factors. We will next compare the results of the
model density to our fully converged results.

0

Q. p

I I I

-(b)

I I

p„(Madel')—

p~~] 2n silicon

Figure 8 compares our calculated p i(r, oo) [Eq. (17a)] for silicon with Deutsch's model valence density [Eq. (19a), see parameters in Table I (Ref. 92)]. The
agreement between theory and the experimentally de-
rived function is excellent. Table X gives a quantitative
comparison of p„(calc) and p„(model) at locations n, P, p, 6, and e of Fig. 8. The only significant discrepancy
exists in the inner-bond minima (point P), where our re-
sult shows a significantly lower amplitude than Deutsch's fit. Figure 6 demonstrated, however, that this feature is highly dependent on Fourier truncation. Our results and those of Deutsch are very difFerent from the elliptic,
single-peaked density obtained by Yang and Coppens
using a truncated Fourier series.

0+,
-2 0 2
Distance along (111) (A)

FIG. 9. Comparison of the ab initio calculated static va-lence charge density (a) p i [Eq. (17a)] of diamond with (b) the model density from fitting the TTKS data (solid line) and GW data (dashed line). The corresponding contour plot is for the TTKS data. Part (c) shows Spackman's results (Ref. 44) in which Stewart's orbital for 28 and Clementi's orbital for

2p are used. The contour step is 0.20e/A. See the text for quantitative comparisons.

8. p ~ in diamond

0.6 -(a)
0.4-

I I
I

I I

] p„(canc) i-

~0

p~

I

O. 6 Xb)
~0.~-

]p„(mcdci)]-

0.2

0

Si Si

I I I I I I I I

-2 0 2 4
Distance along (111) (A)

FIG. 8. Comparison of the ab initio calculated static va-
lence charge density p„ i of Si [Eq. (17a)] with Deutsch's fit
to the

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0.6 -(a)
0.4-

0.2

0

~ 0.6-(b)
0.4-

0.2-

I

~

&

I

y p„{Calc)—

I

'

I

p„{Model)

the local minima in the valence density, e.g., Martins and Zunger (see Fig. 5), Yin and Cohen, van 18{b) Camp, van Doren, and Devreese, and Nielsen and Martin. All-electron calculations have also observed

28 the local minimum, e.g., Pisani, Dovesi, and Orlan o, Methfessel, Rodriguez, and Andersen, Polatoglou and Methfessel and Weyrich. None of the previous cal-culation on Ge, except that of Martins r91 and Zunger (Fig. 5), seem to have found a local minimum in the valence density.

0.6-(c)

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8. Deformation mays for diamond

Figure 12 compares the ab initio calculated static den-
sity deformation map Apq t(r) [Eq. (18)] of diamond
(a) with two experimentally deduced static density de-formation plots: that obtained from the TTKS data [Fig. 12(b)] and from the GW data [Fig. 12(c)]. Ta-ble XI gives a quantitative comparison between the ab initio result and model Ap extracted from the TTKS data. The overall agreement is considerably better with the TTKS data, as noticed also in comparing calculated and measured structure factors (Sec. VI A3). Notice in particular that the amplitude of the GW results are too high at the bond center, and that Ap in this model has the wrong slope in the interstitial region. The shapes near the atomic sites are very different in the two model densities. This deserves further comment: we see from

Eq. (21) that if the orbital deformation parameter r„I is 1, AP is given just by

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(Fe„&t(C)

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a Fourier series (but no fit). In contrast, Fig. 15(c) is
significantly difFerent. This means that: (i) if one uses
a large Set of structure factors, the accurate result ob-
tained by Fourier summation [Fig. 7(f)] is difFerent from
that obtained from the model fit [Fig. 15(c)]. In other
words, there is a loss of information in fitting a structure factor set with high-C components to Dawson's model. This is related to the limited functional flexibility of the model near the core, discussed above, and is evidenced by the large fitting error we find in Fig. 15(c). (ii) If one uses a stnaller set of structure factors, a direct Fourier series produces noise [Fig. 7(e)], while fitting it to the model [Fig. 15(b)] filters out the noise. This reflects the fact that Dawson's model extrapolates the high-G com-ponents using a spatially smooth form. (iii) It follows from (i) and (ii) above that when a limited set of struc-ture factors is available experimentally, the best strategy is to fit it to Dawson's model (rather than Fourier sum it directly). To compare the resulting p„(r) with the-ory, one should either subject an equivalently small set of p, I,(G) to a model fit [Figs. 15(a) and 15(b)) or Fourier sum a converged set of p«i, (C) [Fig. 7(f)]. Subjecting a large set of p(C) to a

GT3Fog.GT3Fog. 15(c69 Tno uscussd (set)Tj1BT /Xi22 9.26 Tf9.78 5293 Tdltin9 33 518 u9Td7Fo BT /Xi1—BT /Xi22 9.26 Tf34f.11 67318.44 56bove,

to(G)

u9 T o49 T

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differ from that of diamond. The following features are
observed. (a)

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s R. J. WeiTf 112.35 Tf 61.78 606.473881.33 Td (Rev.)Tj EiTf 112.35 /Xi24 10.08 Tf 122 515 Td (R.)9 (R.)Tj ET BT /Xi24 8.69 Tf 141.11 220.44 T1 (J.)Tj ET BT

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