Absorptive Form Factors for High-Energy Electron Diffraction

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Abstract

The thermal diffuse scattering contribution to the absorptive potential in high-energy electron diffraction is calculated in the form of an absorptive contribution to the atomic form factor. To do this, the Einstein model of lattice vibrations is used, with isotropic Debye–Waller factors. The absorptive form factors are calculated as a function of scattering vector s and temperature factor M on a grid which enables polynomial interpolation of the results to be accurate to better than 2% for much of the ranges $0 \le Ms^2 \le 6$ and $0 \le M \le 2 \text{ Å}^2$. The computed values, together with an interpolation routine, have been incorporated into a Fortran subroutine which calculates both the real and absorptive form factors for 54 atomic species.

1. Introduction

Since the work of Hashimoto, Howie & Whelan (1962) and Humphreys & Hirsch (1968) it has become almost universal practice in electron-diffraction calculations to assume that the absorptive potential $V'(\mathbf{r})$ is one tenth of the real part $V(\mathbf{r})$. In terms of the Fourier coefficients of the potential this implies that the overall coefficient V_g^{tot} can be written

$$V_g^{\text{tot}} = V_g + iV_g' = (1 + 0 \cdot 1i)V_g \qquad (1)$$

where V_g and V_g' are the coefficients of $V(\mathbf{r})$ and $V'(\mathbf{r})$ respectively, for reciprocal-lattice vector g. This early work referred almost exclusively to primitive crystal lattices where V_g is a monotonically decreasing function of g. However, this has not stopped the same 10% rule being applied to complex structures where the V_g show no such systematic trend and it is by no means clear that the V_g' should follow the V_g. One example of this is for non-centrosymmetric crystals where both V_g and V_g' have an amplitude and a phase. The 10% rule assumes that the phases of the two are the same, but there is no reason in principle why this should be the case, and a phase difference can lead to interesting and observable effects (Gevers, Blank & Amelinckx, 1966; Bird, James & King, 1989; Bird, 1990). With electron microscopy becoming increasingly quantitative, both in high-resolution imaging (*e.g.* Cowley & Smith, 1987) and convergent-beam

diffraction (*e.g.* Zuo, Spence & O'Keeffe, 1988; Bird, James & Preston, 1987; Zuo, Spence & Høier, 1989) it is important to know whether or not it matters to include absorption more rigorously. This is difficult to assess unless we have a reliable method of including absorption which can be applied to a variety of materials and types of calculation. It is the construction of such absorptive potentials that is addressed in this paper.

A fully quantitative analysis of absorption is very difficult (*e.g.* Dederichs, 1972) and this is the basic reason why the 10% rule is still used. In principle one has to treat all the possible excitation processes (phonons, plasmons, single-electron transitions, *etc.*) which can scatter the fast electrons out of the Bragg beams and into the diffuse background. The resulting potentials are non-local and, unlike the corresponding real potential, cannot in general be constructed from individual atomic contributions. Instead, one would have to calculate separate absorptive potentials for every material of interest, which would be based on, for example, first principles calculations of the electronic structure or the phonon spectrum. This is clearly unrealistic in practice. However, there is a number of simplified models of diffuse and inelastic scattering processes which do lead to a formulation of the absorptive potential in terms of atomic contributions (*e.g.* Radi, 1970). One is the Einstein model for thermal diffuse scattering (TDS). Here, instead of using a realistic phonon model which includes correlation between atomic motions, it is assumed that each atom vibrates independently of the others. In this paper we follow the standard view (*e.g.* Radi, 1970; Dederichs, 1972) that TDS is usually the most significant absorption process, particularly for the important $V_{g \ne 0}'$. Electronic scattering processes also contribute to these coefficients, particularly at lower temperatures (Radi, 1970), but their major contribution is to V_0'. This coefficient has little effect on the form of an image or a diffraction pattern because it uniformly attenuates all the Bragg beams, and absolute intensities are rarely important. As far as realistic simulations are concerned, all we need are the $\mathbf{g} \ne \mathbf{0}$ coefficients, and these are dominated by TDS.

Within the Einstein model, and using isotropic Debye–Waller factors, the TDS contribution to the absorptive potential coefficients can be written (Hall

& Hirsch, 1965; Radi, 1970; Buxton, 1978)

$$V'_g = -\frac{\hbar^2}{2m_0}\frac{2h}{\beta m_0 c}\frac{4\pi}{\Omega}\sum_\kappa \exp{(-i\mathbf{g}\cdot\mathbf{r}_\kappa)}$$

$$\times \int d^2\mathbf{s}' f_\kappa(|\mathbf{s}'|)f_\kappa(|\mathbf{s}-\mathbf{s}'|)\{\exp{(-M_\kappa s^2)}$$

$$-\exp{(-M_\kappa s'^2)}\exp{[-M_\kappa(\mathbf{s}-\mathbf{s}')^2]}\}, \quad (2)$$

where m_0 is the electron rest mass, β is the velocity ratio v/c, κ labels the atoms within the unit cell with position vector $\mathbf{r}_\kappa$, elastic form factor f_κ and temperature factor M_κ, and Ω is the unit-cell volume. The equivalent formula for the Fourier components of the real potential is

$$V_g = -\frac{\hbar^2}{2m_0}\frac{4\pi}{\Omega}\sum_\kappa \exp{(-i\mathbf{g}\cdot\mathbf{r}_\kappa)}f_\kappa(s)\exp{(-M_\kappa s^2)},$$
$$(3)$$

where $s = |\mathbf{s}|$. In (2) and (3) the arguments of the form factors are written as $\mathbf{s}$ to make these consistent with the notation of the standard tabulation (Doyle & Turner, 1968). $\mathbf{s}$ and $\mathbf{g}$ are related by

$$\mathbf{s} = \mathbf{g}/4\pi \quad (4a)$$

and with Debye–Waller factors in the form $\exp{(-M_\kappa s^2)}$, M_κ is related to the mean-squared thermal vibration amplitude $\overline{u_\kappa^2}$ by

$$M_\kappa = 8\pi^2\overline{u_\kappa^2}. \quad (4b)$$

By comparison of (2) and (3) it follows that we may define an absorptive form factor $f'_\kappa(s, M)$ as

$$f'_\kappa(s, M) = \frac{2h}{\beta m_0 c}\int d^2\mathbf{s}'\, f_\kappa(|\mathbf{s}'|)f_\kappa(|\mathbf{s}-\mathbf{s}'|)$$

$$\times\{1 - \exp{[-2M_\kappa(s'^2 - \mathbf{s}.\mathbf{s}')]}\}, \quad (5a)$$

which, with the change of variables $\mathbf{s}' \to \mathbf{s}/2 + \mathbf{s}'$, may be written in the more symmetrical form

$$f'_\kappa(s, M) = \frac{2h}{\beta m_0 c}\int d^2\mathbf{s}'\, f_\kappa\left(\left|\frac{\mathbf{s}}{2}+\mathbf{s}'\right|\right)f_\kappa\left(\left|\frac{\mathbf{s}}{2}-\mathbf{s}'\right|\right)$$

$$\times\{1 - \exp{[-2M_\kappa(s'^2 - s^2/4)]}\}. \quad (5b)$$

Combination of (3) and (5) gives the total potential coefficient

$$V_g^{tot} = -\frac{\hbar^2}{2m_0}\frac{4\pi}{\Omega}\sum_\kappa \exp{(-i\mathbf{g}\cdot\mathbf{r}_\kappa)}[f_\kappa(s)$$

$$+ if'_\kappa(s, M)]\exp{(-M_\kappa s^2)}. \quad (6)$$

Note that we have chosen to extract the Debye–Waller factor $\exp{(-M_\kappa s^2)}$ from our definition of the absorptive form factor (5). This has no physical significance – it is purely to provide the convenient form of (6), where the familiar elastic form factor has been replaced by an overall form factor $(f + if')_\kappa$. It is also important to note that V^{tot} depends on the accelerat-

ing voltage through the $1/\beta$ factor in (5). This is in addition to the overall factor of γ $(=m/m_0)$ which multiplies the potential coefficients in dynamical diffraction calculations and which takes account of the relativistic increase of electron mass (*e.g.* Humphreys, 1979).

Equations (5) and (6) provide a prescription for including TDS absorption in any diffraction calculation because the form factors are all atomic properties and are therefore transferable between different materials. The absorptive form factor (5) is a function of only two variables, s and M, and it is therefore feasible to tabulate it for all atomic species. Previous workers have applied the equivalent of (5) either to specific materials (*e.g.* Humphreys & Hirsch, 1968; Radi, 1970) or to provide an approximate parameterization of the absorptive potential (Buxton & Loveluck, 1977; Ichimiya, 1985; Ichimiya & Lehmpfuhl, 1988). Here, we aim for generality by computing f'_κ at a number of selected values of s and M which enables accurate interpolation to be carried out for the wide ranges of s and M which are needed in practice. It is important to note that this does not imply that we are producing fully quantitative values for V'_g. Our analysis is confined to the use of the Einstein model, and within this, the integrals and interpolation are performed accurately. However, the Einstein model neglects both electronic effects, which contribute to V'_g at the few percent level (Radi, 1970), and phonon effects, which can make V'_g dependent on orientatation as well as alter its magnitude. The difficulty is that all such effects will lead to absorptive contributions which are not transferable; the Einstein model might not be truly quantitative, but it does give rise to generally usable results.

2. Computation

There are two aspects to the computation of the integral (5b) for any values of s and M. The first is the evaluation of the integrand, which involves knowledge of the elastic form factors f_κ over a sufficiently large range to ensure that the integral has converged. The second is the evaluation of the integral itself. We shall consider these points in turn.

Doyle & Turner (1968) give the values of $f_\kappa(s)$ for 54 atoms over a range $0 \le s \le 6$ Å^{-1}. We are interested in evaluating the absorptive form factor over a similar range of s values and test runs show that to achieve an accuracy of better than 99% it is necessary to extend the form factors out to s values of about 30 Å^{-1}. The tails of the form factors do not make a particularly large contribution to the final result, but they are included to make it as accurate as possible. Tails are added to the tabulated form factors by assuming that, at large s, f_κ has a Lorentzian form

$$f_\kappa(s) = 0.0239\, Z_\kappa/(s^2 + \alpha_\kappa), \quad (7)$$

where Z_κ is the atomic number of species κ and α_κ is a fitting parameter. The constant in (7) arises from the condition that as $s \to \infty$ the form factors just represent Fourier transforms of the nuclear Coulomb potential. The α_κ are determined in one of two ways. (i) A cubic spline fit (using subroutines $E01BAF$ and $E02BBF$ of the NAG mathematical library) is used on the raw Doyle & Turner (1968) values to provide both the value of f_κ and its derivative df_κ/ds for any s. The range of s between 3·5 and 6 Å^{-1} is then scanned to see if a value, s_0, can be found at which, for some α_κ, both f_κ and df_κ/ds match the value and derivative of (7). If so, this value of α_κ is used in (7) to generate f_κ for $s > s_0$. For $0 \le s \le s_0$, the value from the spline fit is used. This procedure provides a tail which has the correct functional form at large s and which fits smoothly onto the tabulated function. (ii) In some cases this procedure did not provide a matching point within the desired range. In this case α_κ was determined simply by matching the tabulated f_κ to the limiting form (7) at $s = 5$ Å^{-1}. Equation (7) is then used for $s > 5$, and the spline fit for $s \le 5$. The match here will be less smooth, but as the tails do not make a particularly large contribution to the whole integral, this will not introduce a significant error.

With the integral written in the symmetrical form (5b) it is clear that the integrand has two mirror planes in the two-dimensional s' plane, parallel and perpendicular to s. It is therefore necessary only to evaluate the integral in one quadrant of the s' plane. The form of the integrand for Ga is shown in Fig. 1 for a number of values of s and M. Values of f_κ are obtained either from a cubic spline fit or the analytical tail (7) as discussed above. The shape of the integrand is similar for other atomic species. When Radi (1970) evaluated the integral he attempted to perform the azimuthal integration analytically. Unfortunately he did this incorrectly (Buxton, private communication), because in his contour integration he writes $\cos \theta = (z + z^*)/2$ [instead of the correct $\cos \theta = (z + z^{-1})/2$] which is invalid because the complex conjugate z^* is not an analytical function of z. We have not attempted to correct Radi's method, because with modern high-speed computers there is little difficulty in performing the full two-dimensional integral many times for different values of s and M. We use the general-purpose two-dimensional integrating subroutine $D01DAF$ of the NAG library to perform the integrals. As can be seen from (5b) and Fig. 1, the integrand passes through zero on the circle $|s'| = |s/2|$. At this point the integrand can be rapidly varying, which may cause problems in the integrating routine. To circumvent this, we split the integral into two parts $0 \le |s'| \le |s/2|$ and $|s/2| \le |s'| \le 30$ Å^{-1}, where, as mentioned above, the upper limit ensures convergence to better than 99%. With this division, no problem is encountered in the integration, and a relative

(a)

(b)

(c)

(d)
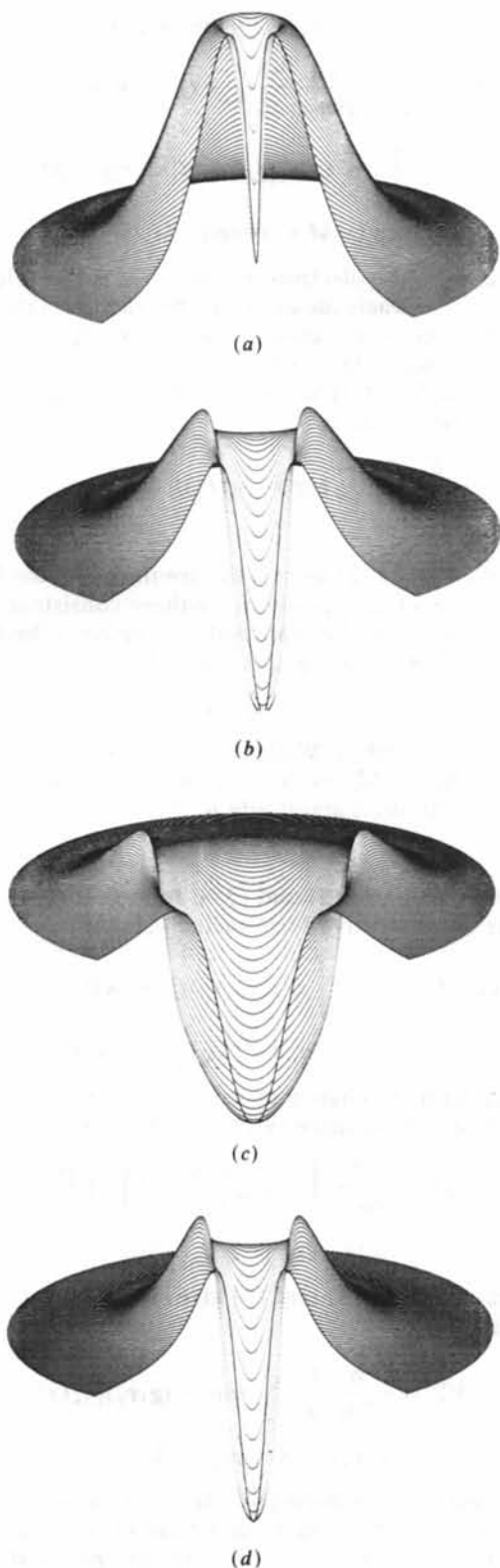
Fig. 1. The integrand of (5b) for Ga. (a) $s = 0·0$, $M = 0·7$; (b) $s = 0·5$, $M = 0·7$; (c) $s = 1·5$, $M = 0·7$; (d) $s = 0·5$, $M = 2·0$. The edge of the disc corresponds to $|s'| = 2$ Å^{-1}. An angular cut of 80° is used to show the 'inside' of the function.

Table 1. *The elastic (real) and absorptive (abs.) potential coefficients for four f.c.c. metals at an accelerating voltage of* 100 kV

For comparison, the results of Radi (1970) and Buxton & Loveluck (1977) are also shown. Thermal vibration amplitudes and lattice parameters are the same as those used by Radi (1970).

G	Al				Cu			
	Real	Abs.	Radi	B+L	Real	Abs.	Radi	B+L
000	17·112	0·164	0·162	0·150	22·807	0·742	0·746	0·659
111	6·141	0·145	0·149	0·132	11·752	0·679	0·708	0·599
200	5·113	0·140	0·144	0·127	10·545	0·660	0·696	0·580
220	3·228	0·122	0·129	0·110	7·767	0·592	0·641	0·515
311	2·619	0·112	0·119	0·100	6·573	0·547	0·602	0·472
222	2·476	0·109	0·115	0·097	6·257	0·533	0·585	0·458
400	2·060	0·097	0·104	0·086	5·257	0·482	0·535	0·410
331	1·843	0·089	0·097	0·078	4·697	0·448	0·500	0·377
442	1·579	0·078	0·085	0·068	3·986	0·397	0·446	0·329
440	1·293	0·064	0·070	0·054	3·204	0·330	0·375	0·266
620	1·099	0·052	0·058	0·043	2·669	0·276	0·314	0·215
444	0·955	0·042	0·048	0·034	2·286	0·231	0·265	0·175
642	0·843	0·035	0·039	0·027	2·001	0·195	0·224	0·142
733	0·725	0·026	0·030	0·020	1·706	0·154	0·179	0·105
662	0·650	0·021	0·024	0·015	1·522	0·127	0·149	0·082
664	0·571	0·015	0·018	0·010	1·331	0·099	0·117	0·058
933	0·513	0·011	0·013	0·007	1·195	0·078	0·093	0·040

G	Ag				Au			
	Real	Abs.	Radi	B+L	Real	Abs.	Radi	B+L
000	24·462	1·308	1·344	1·166	29·827	2·859	3·042	2·495
111	14·361	1·207	1·296	1·068	19·048	2·683	2·976	2·325
200	12·953	1·176	1·254	1·039	17·274	2·628	2·867	2·273
220	9·514	1·065	1·167	0·932	12·889	2·426	2·730	2·081
311	7·992	0·992	1·094	0·862	10·960	2·290	2·589	1·952
222	7·591	0·970	1·067	0·840	10·455	2·247	2·529	1·911
400	6·329	0·886	0·986	0·760	8·868	2·087	2·385	1·759
331	5·630	0·829	0·928	0·707	7·991	1·976	2·273	1·656
422	4·758	0·746	0·842	0·628	6·895	1·809	2·109	1·499
440	3·823	0·633	0·721	0·522	5·702	1·576	1·861	1·283
620	3·209	0·540	0·616	0·436	4·896	1·379	1·633	1·101
444	2·777	0·463	0·535	0·366	4·311	1·209	1·457	0·946
642	2·456	0·399	0·462	0·307	3·864	1·062	1·289	0·813
733	2·128	0·325	0·380	0·241	3·392	0·890	1·095	0·659
662	1·925	0·275	0·323	0·198	3·090	0·771	0·952	0·555
664	1·714	0·221	0·263	0·151	2·766	0·637	0·801	0·438
933	1·562	0·180	0·215	0·116	2·526	0·534	0·673	0·350

accuracy of better than 10^{-4} is obtained with typically 1000 function calls. An alternative approach to the evaluation of the integral was taken by Buxton & Loveluck (1977). They used Doyle & Turner's (1968) parameterization of the elastic form factors in terms of a sum of four Gaussians to perform the integral analytically. However, this parameterization is valid only for $s < 2$ Å^{-1}, so the influence of the tails of the form factors will be seriously underestimated. Nevertheless, comparison of their results with our 'exact' integration provides a useful measure of the relative contributions of different parts of the integrand.

3. Interpolation

For any given material it is possible to evaluate $(5b)$ at the relevant values of s and construct the absorptive potential as in (6). This has been done for a number of elements to compare our results with those of Radi (1970) and Buxton & Loveluck (1977). The results are presented in Table 1. The Debye-Waller factors

used are the same as those of Radi. Our results are similar to his, which is surprising given that his azimuthal integration is performed incorrectly. Table 1 also shows that the Buxton & Loveluck (1977) parameterization of $f'_\kappa(s)$ does not provide a particularly good approximation to the full integral, particularly for large g. Further discussion on the absorptive potential, including its ratio to the elastic potential, its variation with g and its temperature dependence may be found in the following paper (Bird, 1990).

However, the primary purpose of this work is not to calculate the absorptive potential for certain special cases, but to provide the means of constructing it for any material. To this end, we have attempted to find a grid of s and M values on which f'_κ may be calculated using $(5b)$ and from which it can be evaluated for any s and M by interpolation. For ease of use it is clear that the number of grid points required to do this must be kept as small as possible, while keeping the desired accuracy in the interpolation scheme. It is therefore important to work with the smoothest

Table 2. *Grids of f'_κ/f_κ for light (C), medium (Ga) and heavy (Au) atoms*

Results are expressed as percentages. These are the values used in the polynomial interpolation.

Element: C

M Ms^2	0.05	0.15	0.30	0.70	1.30	2.00
0.000	0.05	0.11	0.18	0.32	0.46	0.59
0.005	0.13	0.17	0.23	0.35	0.49	0.61
0.025	0.41	0.39	0.40	0.48	0.58	0.68
0.070	0.81	0.79	0.75	0.74	0.78	0.85
0.200	1.42	1.45	1.42	1.31	1.26	1.25
0.500	1.88	2.04	2.05	1.98	1.88	1.82
1.200	1.32	1.70	1.83	1.94	1.95	1.93
2.000	−0.95	−0.42	0.09	0.49	0.68	0.79
3.500	−9.38	−8.84	−7.05	−5.85	−4.94	−4.50
6.000	−43.25	−42.63	−39.54	−32.75	−29.67	−27.81

Element: Ga

M Ms^2	0.05	0.15	0.30	0.70	1.30	2.00
0.000	0.35	0.73	1.12	1.78	2.37	2.83
0.005	0.98	1.30	1.60	2.12	2.62	3.02
0.025	2.13	2.52	2.84	3.20	3.49	3.72
0.070	3.99	4.06	4.34	4.74	4.91	4.98
0.200	7.28	7.03	6.89	7.06	7.22	7.23
0.500	10.01	10.25	10.04	9.71	9.65	9.64
1.200	8.03	9.21	9.86	10.41	10.71	10.91
2.000	−2.48	0.35	2.30	4.69	6.64	7.98
3.500	−43.31	−36.40	−30.36	−28.83	−16.37	−11.13
6.000	−208.56	−190.34	−172.11	−144.76	−126.54	−108.31

Element: Au

M Ms^2	0.05	0.15	0.30	0.70	1.30	2.00
0.000	1.24	2.53	3.74	5.72	7.42	8.69
0.005	2.56	3.55	4.57	6.27	7.81	8.99
0.025	5.79	6.38	6.97	8.08	9.18	10.06
0.070	10.24	10.68	10.82	11.16	11.62	12.08
0.200	18.16	17.79	17.77	17.20	16.63	16.33
0.500	25.77	25.46	25.03	24.57	23.46	22.48
1.200	21.97	24.75	25.99	27.17	27.67	27.36
2.000	−2.28	5.17	10.27	16.62	20.75	22.82
3.500	−99.82	−79.18	−62.68	−41.23	−23.18	−12.06
6.000	−505.29	−440.53	−387.83	−312.54	−252.30	−199.60

possible functions. Two refinements help here. First, we work with the ratio $f'_\kappa(s, M)/f_\kappa(s)$ rather than the raw f'_κ values as it is smoother, particularly for the more significant smaller values of s (*e.g.* Humphreys & Hirsch, 1968; Radi, 1970). Second, we work with an (Ms^2, M) grid rather than the simpler (s, M) grid. Again, this provides a smoother function for interpolation and prevents the inclusion of large Ms^2 values for which the $\exp(-Ms^2)$ factor in (6) makes the potential coefficient extremely small. After extensive testing we settled on a grid with the values

$$Ms^2 = 0.0, \ 0.005, \ 0.025, \ 0.07, \ 0.2,$$
$$0.5, \ 1.2, \ 2.0, \ 3.5, \ 6.0, \tag{8}$$
$$M = 0.05, \ 0.15, \ 0.3, \ 0.7, \ 1.3, \ 2.0 \ \text{Å}^2.$$

$M = 0$ is not included because f'_κ is zero there. For each element, 60 values of f'_κ must therefore be calculated; three examples are given in Table 2. With a two-dimensional polynomial interpolation procedure (see below) this grid nearly always provides f'_κ to an

accuracy of better than 2% for the lower Ms^2 values ($\lesssim 2.0$) and better than 6% for the less-significant higher Ms^2 values (between about 2.0 and 6.0) over the whole range of values $0 \le M \le 2$ Å^2 and for all atomic species. These ranges were chosen to cover all the values which are likely to be found in practice. Because we tabulate the ratio f'_κ/f_κ it is necessary to know f_κ in order to find f'_κ – this is obtained from cubic interpolation of the Doyle & Turner (1968) values. This also provides a further restriction that s in f'_κ must be less than 6 Å^{-1}. We emphasize that the 2–6% error here is only the relative error between the interpolated values and the full integration of (5b). It should not be taken to imply that the resulting absorptive potential coefficients (which, as discussed in the *Introduction*, ignore phonon and electronic-scattering effects) have this absolute accuracy. Given all the uncertainties in the basis of the calculations, this accuracy in the interpolation is more than adequate. The grid (8) gives a significantly higher percentage error only near the range of values where f'_κ goes through zero (*e.g.* Humphreys & Hirsch, 1968). For most species, this occurs near $Ms^2 \simeq 2.0$ (see Table 2) and although the percentage error can become quite large, the absolute error remains small because f'_κ is itself very small.

A two-dimensional polynomial interpolation procedure is used to obtain f'_κ values from the computed grid (see, for example, Press, Flannery, Teukolsky & Vetterling, 1986). A bi-cubic interpolation is illustrated in Fig. 2. It involves five one-dimensional cubic
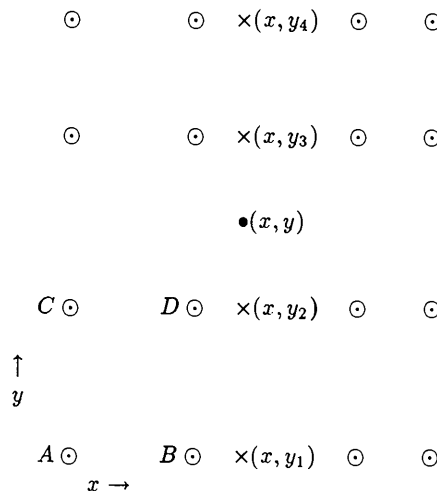

Fig. 2. The two-dimensional cubic interpolation procedure. The function is known on the grid points ⊙. To determine its value at (x, y) we use equation (9) on the constant y lines to evaluate it at the points (x, y_i), $i = 1, 2, 3, 4$. A fifth one-dimensional interpolation is then used on these four points. The same method works at the edge of the grid, but is less accurate because (x, y) does not lie in the centre of the nine rectangles. In the case of a bi-quadratic interpolation there are four possible choices of nine grid points, corresponding to the bottom left of the 3×3 array being points A, B, C, D respectively.

interpolations, for which a function $h(x)$ at any point x is given by

$$h(x) = \frac{(x-x_2)(x-x_3)(x-x_4)}{(x_1-x_2)(x_1-x_3)(x_1-x_4)} h(x_1)$$

$$+ \frac{(x-x_1)(x-x_3)(x-x_4)}{(x_2-x_1)(x_2-x_3)(x_2-x_4)} h(x_2)$$

$$+ \frac{(x-x_1)(x-x_2)(x-x_4)}{(x_3-x_1)(x_3-x_2)(x_3-x_4)} h(x_3)$$

$$+ \frac{(x-x_1)(x-x_2)(x-x_3)}{(x_4-x_1)(x_4-x_2)(x_4-x_3)} h(x_4), \qquad (9)$$

where the x_i are the grid points nearest to x (Fig. 2). A similar procedure is possible for quadratic interpolation; there is a slight problem here in that four possible two-dimensional quadratic interpolations can be carried out for any point (x, y) (Fig. 2). It is also of course possible to mix different orders of polynomial in the different directions. The best interpolation procedure was found using trial and error by comparing the various possible schemes with the results from the exact integration over a wide range of M and s values. Quadratic interpolation was found to be best for values of Ms^2 up to 2·0 while cubic interpolation is more accurate for $2·0 \leq Ms^2 \leq 6·0$. Perhaps surprisingly, where quadratic or cubic interpolation works best in the Ms^2 direction, the same order of polynomial also gives the best fit in the M direction. We therefore use bi-quadratic interpolation for $0 \leq Ms^2 < 2$ and bi-cubic interpolation for $2 \leq Ms^2 \leq 6$. For the quadratic interpolations, the four possible results (or fewer, for points near the edge of the grid) are averaged to produce the final result. This interpolation scheme is straightforward to implement, and the final number of data points needed for each atom becomes 27 Doyle & Turner (1968) values for f_κ and 60 computed values of f'_κ/f_κ. We also need the grid points (8) and the s values used by Doyle & Turner, but these are the same for every atom. This relatively small amount of data makes it quite feasible to distribute generally usable values for both the elastic and absorptive form factors.

4. Subroutine *ATOM*

We have chosen to distribute our results in the form of a Fortran subroutine which, when given the name of an atom, the value of s, the temperature factor and the accelerating voltage will return the elastic and absorptive form factors multipled by the Debye–Waller factor $\exp(-M_\kappa s^2)$. These can then be combined to form overall potential coefficients using (6). The accelerating voltage is involved because f'_κ depends on β – the form factors do not include the extra factor of γ which is discussed after (6). The subroutine contains all the data required for interpo-

lation in the form of DATA statements, so no external file handling is required. This makes the subroutine rather lengthy both in its source form and when compiled, so every effort has been made to minimize its size. As part of this, we store values of $f'_\kappa(s, M) \exp(-Ms^2)/f_\kappa(s)$ (rather than the f'/f values which are used for interpolation) to reduce the magnitude of the larger Ms^2 points (Table 2). The resulting flattened data are then scaled with additive and multiplicative factors to make it fit into the range 0 to 999. All the data can then be stored to sufficient accuracy with only three significant figures. This flattening enhances the accuracy with which the majority of points are stored, at the expense of a small degradation for the less-significant higher Ms^2 values. Polynomial fits were also tried on the flattened data, but these required considerably more data for an equally good fit. The interpolation routine is included in the subroutine, so it is fully self-contained. It is written in standard Fortran 77 and has been tested running under several different operating systems. The specification of the subroutine is shown in Fig. 3. At present it contains all the elements which appear in Doyle & Turner (1968); later versions will be extended to cover all atoms. The size of the source code is 45 kbyte.

Subroutine *ATOM* will be made freely available both *via* electronic mail and on floppy disk. There

```
C -----------------------------------------------------------------
C --------------- ATOM - Atomic Form Factors Subroutine ------------
C -----------------------------------------------------------------
C
C       CALL ATOM( ELNAME, M, S, KVOLTS, FE, FA, STATUS )
C
C In:ELNAME - CH*2   - Element name (upper case only)
C        M - D.P.   - 8.(pi^2).u^2; u^2 = mean square atomic vibration
C        S - D.P.   - Scattering vector (as in Doyle and Turner)
C    KVOLTS - D.P.   - Accelerating voltage in KVolts
C    STATUS - INT.   - Error control
C
C Out:    FE - D.P.   - Elastic part of atomic form factor * exp(-Ms^2)
C         FA - D.P.   - Imag part of form factor (inc. 1/beta) * exp(-Ms^2)
C    STATUS - INT.   - Returned status
C
C Note: If Ms^2 > 6.0 or s > 6.0 both FE and FA return 0.0D0
C        If M < 0.05 then FA returns 0.0D0
C
C ELNAME: Element name, in upper case, left justified,
C          e.g. 'GA', 'AS', 'S ', 'P '
C
C KVOLTS: Accelerating voltage in KeV
C          If KVOLTS = 0.0D0, BETA is taken as 1
C
C STATUS: If STATUS is passed with a negative value, then should an
C          error occur, the subroutine will write the relevent error
C          message to unit -STATUS and execution will be terminated.
C          If STATUS is positive, a code number is returned in STATUS
C          and even if an error has occured, execution will not be
C          terminated. The codes are:
C
C          0 - Successful
C          1 - Unknown element
C          2 - M out of range (M < 0 or M > 2)
C          3 - S less than zero
C          4 - KVOLTS out of range (KVOLTS < 0.0 or KVOLTS > 10.^4)
C
C -----------------------------------------------------------------
```

Fig. 3. The specification of subroutine *ATOM*.

will be no charge for electronic mail distribution – contact should be made with D.Bird@uk.ac.bath.gdr. Distribution by floppy disc will be in the form of $5\frac{1}{4}$ in discs on PCs running under DOS. In this case there will be a handling charge of US \$20.

5. Discussion

Subroutine *ATOM* provides a simple means of constructing elastic and absorptive potentials from individual atomic contributions. How useful are the results likely to be? For the elastic potential, the use of atomic potentials is known to be an excellent approximation. Even for the lower-order V_g, where effects due to bonding can be observed, the difference between the actual and neutral-atom potential coefficients is very small (*e.g.* Zuo, Spence & O'Keeffe, 1988). For the absorptive potential, the use of atomic potentials is certain to be a poorer approximation, but it is very difficult to be quantitative about this. It also has to be recognized that TDS is only one contribution to V_g', and that the use of isotropic Debye–Waller factors introduces further inaccuracy. (The question of what is the best average Debye–Waller factor to use in any given situation when working with anisotropic crystals is rather difficult and we do not attempt to address it here). Our view is that the calculations presented here are about the best that can be done in a general fashion. We expect that they will reliably reflect trends in the absorptive potential as a function of s and M, and will show the correct behaviour of the ratio of elastic to absorptive potential for light and heavy atoms. In this way our results should provide a much better guide to the form of the absorptive potential than any *ad hoc* rule. Given that they can be incorporated into diffraction calculations in a very straightforward way, we suggest that they should be used, if only to see whether the inclusion of absorption in a rather more rigorous fashion makes any significant difference to the final results.

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Absorption in High-Energy Electron Diffraction from Non-Centrosymmetric Crystals

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Abstract

The influence of absorptive potentials on high-energy electron diffraction amplitudes is analysed, with an emphasis on effects which are specific to diffraction from non-centrosymmetric crystals. It is shown that the phase difference which then exists between the elastic and absorptive potential coefficients can give rise to a significant asymmetry between +g and −g reflections. This phase difference is calculated for a number of important III–V semiconductors. In many diffraction calculations it is assumed that the phase difference is zero – it is argued here that such calculations cannot be truly quantitative. The inclusion of absorption by perturbation theory is shown to be valid, but only if the change in the eigenvectors as well as the eigenvalues is taken into account. It is shown that within two-beam theory the +g, −g