JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 17, SUPPLEMENT B-II, 1962
PROCEEDINGS OF INTERNATIONAL CONFERENCE ON MAGNETISM AND CRYSTALLOGRAPHY, 1961, VOL. II

Inelastic Interactions in Dynamic Electron Scattering

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A general method is proposed for incorporating the treatment of interactions between elastically and inelastically scattered waves in the various theories of dynamic diffraction of electrons. The applications are discussed mostly in relation to Cowley and Moodie's short wavelength-approximation; results in terms of a form factor are given for the atoms $Z=9$ and $Z=14$.

1. Introduction

In recent years some interest has been taken in absorption effects in electron micrograph fringes (Hashimoto, Howie and Whelan[1]) and dynamic fine structure (Honjo and Mihama[2]). These effects have been attributed to interactions between the elastically and inelastically scattered waves. A treatment of this interaction in the two beam case has previously been given by Yoshioka[3], who performed some calculations based on the Thomas-Fermi potential. In this paper a general method for treatment of inelastic interactions within the framework of the various theories of dynamic scattering is proposed. Results of calculations based on Hartree-Fock one-electron scattering factors are given for two atoms.

2. General theory

In the Schrödinger case, neglecting nuclear motion, the scattering of an electron by a system of electrons and nuclei is given by the Coulomb interaction

$$U(\boldsymbol{r}, \boldsymbol{r}_j)=(2/a_0)(U_N(\boldsymbol{r}) -\sum_j|\boldsymbol{r}-\boldsymbol{r}_j|^{-1}).$$

Here $\boldsymbol{r}$ and $\boldsymbol{r}_j$ are coordinates of the incident and object electrons respectively, U_N is the potential from the nuclei and $a_0=2/me^2$, where m is the relativistic mass of the incident electron. On expanding the wave function, Φ, of the system of object plus incident electron in energy eigenfunctions, φ_n, of the object, $\Phi=\sum_n\psi_n(\boldsymbol{r})\varphi_n(\boldsymbol{r}_j)$, we obtain (see e.g. Mott and Massey[4])

$$\{\Delta_r{}^2+k_0{}^2+U(\boldsymbol{r},\boldsymbol{r}_j)\}\Phi(\boldsymbol{r},\boldsymbol{r}_j) =\sum(k_0{}^2-k_n{}^2)\psi_n(\boldsymbol{r})\varphi_n(\boldsymbol{r}_j)\equiv K\Phi \qquad (1)$$

where ψ_n is interpreted as the amplitude of the wave corresponding to transition of the object from the initial state, 0, to the final state, n, and k_n is the length of the wave vector in vacuum corresponding to this wave; k_0 is the length of the wave vector of the incident wave. The determination of ψ_0, the amplitude of elastic scattering, is normally reduced to a potential scattering problem by multiplication of (1) with $\bar{\varphi}_0$, the complex conjugate of φ_0, and integration over all coordinates of the object under the assumption that the non-diagonal elements, $U_{0n}(\boldsymbol{r})=\int\bar{\varphi}_0\varphi_nU d\tau_{\boldsymbol{r}_j}$, of U are negligible. In the resulting equation U_{00} appears as the scattering potential. The effects of the inelastically scattered waves can be introduced in the solution, Ψ_{pot}, of this problem by iteration (see Yoshioka[3]).

Here we shall adopt a more direct approach. The inelastic interactions arise through the non-diagonal elements of U. The operator K, on the right hand side, has only diagonal elements, and these will, for the important values of $\boldsymbol{r}$, be smaller than the corresponding elements of U. Let us therefore, at first, neglect the right hand side of (1), postponing the discussion of the validity of this approximation. The resulting equation is of potential scattering type when regarding the $\boldsymbol{r}_j$ as parameters. On taking the incident wave $\exp(ik_0z)$ as the solution of the corresponding free space equation, the elastically scattered wave is given as:

$$\psi_0(\boldsymbol{r})=\Phi_{00}(\boldsymbol{r})=\int\bar{\varphi}_0(\boldsymbol{r}_j)\varphi_0(\boldsymbol{r}_j)\Phi(\boldsymbol{r},\boldsymbol{r}_j)d\tau_{\boldsymbol{r}_j}\,. \qquad (2)$$

A pictorial representation of this equation is to regard ψ_0 as a superposition of waves scattered by all configurations, $\boldsymbol{r}_j$, of the object, each configuration given the weight

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$\bar{\varphi}_0\varphi_0$. The amplitudes ψ_n, of the inelastically scattered waves are given by the non-diagonal elements Φ_{n0}, and the total intensity (elastic plus inelastic) by the weighted sum of the intensity scattered by all configurations.

Equation (1), with the right hand side set equal to zero, can be treated by any of the methods for treatment of dynamic diffraction of electrons, although special care is required when using methods depending upon the symmetry or periodicity of the potential, as $U(\boldsymbol{r},\boldsymbol{r}_j)$ does not have the symmetry of $U_{00}(\boldsymbol{r})$.

3. Applications

We shall discuss here mainly the application of the above formulation in connection with the "phase object approximation" *(Cowley and Moodie[5]); the results can usually be readily carried over to other theories. For potential scattering the phase object approximation can be written:

$$f_{\mathrm{pot}}(\boldsymbol{s}) = (ik/2\pi)\int \exp(i\boldsymbol{\rho s}) \times\left\{1-\exp\left[(i/2k)\int U(\boldsymbol{r})dz\right]\right\}df_{\rho} \qquad (3)$$

where $\boldsymbol{s}$ is 2π times the scattering vector and $\boldsymbol{r}=\boldsymbol{\rho}+(z/k)\boldsymbol{k}_0$.

On neglecting the right hand side of equation (1) and using (2), one obtains the corresponding expression including inelastic interactions:

$$f_0(\boldsymbol{s})=(ik/2\pi)\iint \bar{\varphi}_0\varphi_0\exp(i\boldsymbol{\rho s}) \times\left\{1-\exp\left[(i/2k)\int U(\boldsymbol{r},\boldsymbol{r}_j)dz\right]\right\}df_{\rho}d\tau_{\boldsymbol{r}_j}. \qquad (4)$$

To illustrate the difference between expressions (3) and (4), consider scattering from an atom. Introducing the Fourier expansion for $U(\boldsymbol{r},\boldsymbol{r}_j)$, and φ_0 in the form of a Slater determinant of one-electron wave functions, φ_α, a power series in $(1/k)$ is obtained for $f_0(\boldsymbol{s})$, which by comparison with the potential scattering expression gives, to the second order in $1/k$, the additional elastic scattering amplitude due to inelastic interactions:

$$(1/2a_0)(f_0(\boldsymbol{s})-f_{\mathrm{pot}}(\boldsymbol{s}))=(i/2\pi ka_0) \times\int \frac{\sum_{\alpha}(f_\alpha(\boldsymbol{s})-f_\alpha(\boldsymbol{t})f_\alpha(\boldsymbol{s}-\boldsymbol{t}))-\sum_{\alpha\neq\beta}f_{\alpha\beta}(\boldsymbol{t})f_{\beta\alpha}(\boldsymbol{s}-\boldsymbol{t})}{t^2(\boldsymbol{s}-\boldsymbol{t})^2}df_t \qquad (5)$$

* The term "phase object approximation" was introduced by Cowley and Kuwabara[6].

where

$$f_{\alpha\beta}=\int\varphi_\alpha(\boldsymbol{r})\varphi_\beta(\boldsymbol{r})\exp(i\boldsymbol{sr})d\tau_{\boldsymbol{r}}, \quad f_\alpha=f_{\alpha\alpha}$$

and df_t is a surface element, normal to $\boldsymbol{k}_0$, in reciprocal space. Higher order terms can be written down quite readily, but it is found that the majority of these terms can be taken into account by introducing the Fourier transform of (5) as an increment to the potential in the ordinary potential scattering expression. For a system consisting of several atoms, each atom will give a contribution of the form (5), multiplied with a geometric phase factor, neglecting then exchange terms involving electrons on different atoms.

According to this argument we may introduce the resulting imaginary potential terms in e.g. the two beam theory. A direct proof for this case can be given, however, by applying the two beam treatment to equation (1) remembering that $U(\boldsymbol{r},\boldsymbol{r}_j)$ has a non-periodic component which must be expressed as a continuum in addition to the Fourier series for $U_{00}(\boldsymbol{r})$. Formally this continuum will give rise to weak beams which will contribute to the "dynamic potentials". This method is, of course, directly applicable also to a non-periodic component of $U_{00}(\boldsymbol{r})$, due to e.g. disorder.

The expression (5) breaks down at $s=0$, due to the neglect of the right hand side of (1). An expression valid also in this region can be obtained by approximating the elements, $k_0^2-k_n^2$, of K by an average value $2k\Delta k$ and adding K to the potential operator in (4). The implication of the resulting factor $\exp(i\Delta kz)$ in the expression for the wave-function can be examined by a method developed by Schiff[7]. We find that the equation (5) must be re-interpreted as a volume integral in reciprocal space, and a factor

$$\delta(\Delta k-\zeta)+(i/\pi)(\Delta k-\zeta)^{-1} \qquad (6)$$

where ζ is the z-component of the vector $\boldsymbol{t}$, is included under the integral sign. The first term in (6) results in a term which converges rapidly to (5) as s increases beyond Δk; the second term gives rise to a real term important only when $s<\Delta k$.

4. Calculations

In order to calculate the difference (5) for an atom it can be shown that the scheme

Table I. Inelastic contribution to the atomic scattering amplitude.
Incident electron energy 40 keV.

a) Fluorine

s	$1s$	$2s$	$2p$	$2s2p$	$2p2p$	Δf	$\Delta\eta$	η_{pot}*
0 (real part)	$.2\times10^{-3}$	3.3×10^{-3}	3.9×10^{-3}	1×10^{-3}	$.1\times10^{-3}$	.02		
0 (im. part)	.5	11.0	13.0	3.4	.2	.05	.11	.09
1	.2	2.7	3.0	.3	.1	.02	.04	.09
2	.15	1.2	1.3	.08	.03	.008	.02	.10
4	.10	1.5	.22	—	.01	.0015	.006	.14

* after Ibers and Hoerni[11].

b) Silicon

s	$1s$	$2s$	$2p$	$3s$	$3p$	$2s2p$	$3s3p$	$3p3p$	Δf	$\Delta\eta$
0 (real part)	—	1×10^{-3}	1×10^{-3}	8×10^{-3}	29×10^{-3}	—	6×10^{-3}	—	.06	
0 (im. part)	—	4	4	28	93	1×10^{-3}	21	1×10^{-3}	.18	.12
1	$.1\times10^{-3}$	1.2	1.4	4.8	3.5	.1	.3	.3	.03	.02
2	.1	.8	.7	1.0	−.3	.10	−.13	—	.007	.01
4	.06	.32	.32	−.19		.03	−.03	—	.002	.006

given by Freeman[8] for computing incoherent X-ray scattering factors can be utilized, upon substitution of the two different sums in (5) for direct and exchange terms respectively, and treating the angular parts by a Fourier-Bessel transform technique. The one-electron form factors necessary for the calculations were taken from Freeman[9] (see also Dawson[10]). Results in terms of the increment, $\Delta\eta$, to the argument η, of the atomic scattering amplitude are given in Table for the atoms $Z=9$ and $Z=14$, and the incident electron energy 40 keV at four values of $s=4\pi\sin\vartheta/\lambda$. The units for the one-electron contributions are chosen such that the kinematic scattering amplitude is $(Z-F)/s^2$; the results can easily be converted into other energies by multiplication with $(k_{40keV}/k)\,(m/m_{40keV})$. To obtain values at $s=0$, Δk_{av} was set equal to 0.02 Å^{-1} for F and 0.01 Å^{-1} for Si.

The author wishes to express his gratitude to Dr. J.M. Cowley and Mr. A.F. Moodie for frequent stimulating and helpful discussions. Financial support from the Commonwealth Scientific and Industrial Research Organization and the Royal Norwegian Council for Scientific and Industrial Research is gratefully acknowledged.

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