Validity of generalized scattering equations and corresponding inelastic-cross-section expressions for comprehensive electron diffraction conditions

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Generalized scattering equations for electron diffraction in a crystalline environment have recently been derived with the assumption that the zeroth-order Laue zone (ZOLZ) lies parallel to the crystal surface (or alternatively that the zone axis is parallel to the crystal surface normal) [L. J. Allen and T. W. Josefsson, Phys. Rev. B **52**, 3184 (1995)]. The contribution from higher-order Laue zones (HOLZ's) was not considered. We show that these scattering equations are in fact valid for general orientations of the ZOLZ with respect to the crystal surface, except for the specific case where the ZOLZ lies perpendicular to the surface. Furthermore, a widely applicable expression for the cross section for any type of inelastic scattering in a crystal is shown to be valid for any orientation of the ZOLZ and allows the inclusion of reflections in the HOLZ's.

I. INTRODUCTION

General dynamical equations for electrons incident on a crystal of finite thickness which are a generalization of those given by Yoshioka[1] have recently been derived.[2] These fundamental equations follow from a Schrödinger equation in integro-differential form containing a nonlocal kernel which represents the inelastic scattering. Based on this formulation, a general expression for the cross section for any type of inelastic scattering in a crystal was derived which has wide general applicability.[3–6]

Dudarev *et al.*[7] have addressed the problem of scattering of high energy electrons by solids on the basis of a kinetic equation for the one-particle density matrix. They pointed out that, unlike their approach, most theoretical approaches to the problem of diffraction and inelastic scattering of high-energy electrons by crystals have been developed in forms which are suitable for specific diffraction geometries. The fundamental scattering equations in Ref. 2 were derived for the specific diffraction conditions where the zeroth-order Laue zone (ZOLZ) lies parallel to the crystal surface, i.e., where the zone axis was parallel to the crystal surface normal. In addition the contribution from higher-order Laue zones (HOLZ's) was not considered. In this Brief Report we show that the fundamental scattering equations are in fact valid for any orientation of the ZOLZ with respect to the crystal surface, excepting for the case where the ZOLZ lies perpendicular to the crystal surface, provided that HOLZ contributions are ignored. However, to a good approximation (for all but the thinnest of crystals) the fast electron wave function obtained from the scattering equations is not greatly affected by the assumption that the crystal is infinite. The scattering equations then reduce to the well known Yoshioka[1] equations and hold for all ZOLZ orientations and also allow HOLZ reflections to be included.

A widely applicable expression[2] for the cross section for any type of inelastic scattering in a crystal is shown to be valid for all diffraction conditions, i.e., orientations of the ZOLZ and allows inclusion of HOLZ effects. Unlike the scattering equations, the derivation and use of the cross section expression depends critically on the fact that the crystal is of finite thickness.

II. THEORY

The generalized scattering equations for fast electrons incident on a crystal of finite thickness t are given by[2]

$$[K^2-(\mathbf{k}^i+\mathbf{g})^2]L^{ii}(t)C^i_{\mathbf{g}}+\frac{2m}{\hbar^2}\Bigg[L^{ii}(t)\sum_{\mathbf{h}\neq\mathbf{g}}V_{\mathbf{g}-\mathbf{h}}C^i_{\mathbf{h}}+\sum_{\mathbf{h}}W^{ii}_{\mathbf{g},\mathbf{h}}C^i_{\mathbf{h}}\Bigg]=0, \tag{2.1}$$

where $V_{\mathbf{g}-\mathbf{h}}$ and $W^{ii}_{\mathbf{g},\mathbf{h}}$ are coefficients related to the local crystal potential, and the nonlocal inelastic scattering potential, respectively. These scattering equations can be solved in a self-consistent way to obtain the coefficients in the standard Bloch state expansion of the elastic scattered electron wave function $\psi_0(\mathbf{r})$ in the solid, i.e.,

$$\psi_0(\mathbf{r})=\sum_i \alpha^i\sum_{\mathbf{g}} C^i_{\mathbf{g}}\exp[i(\mathbf{k}^i+\mathbf{g})\cdot\mathbf{r}], \tag{2.2}$$

where $\mathbf{g}$ and $\mathbf{h}$ are reciprocal lattice vectors and α^i is the amplitude of the ith Bloch state with wave vector $\mathbf{k}^i=\mathbf{K}+\lambda^i\hat{\mathbf{n}}$. The incident electron wave vector in the crystal is denoted by $\mathbf{K}$ and $\hat{\mathbf{n}}$ is an inwardly directed crystal surface normal. The complex quantities λ^i can be written as $\lambda^i=\gamma^i+i\eta^i$ where γ^i are the *anpassung* (matching coefficients) and η^i are the absorption coefficients. The thickness dependent term $L^{ii}(t)$ in the scattering equations is a special case of

$$L^{ij}(t)=\frac{\exp[i(\lambda^i-\lambda^{j*})t]-1}{i(\lambda^i-\lambda^{j*})t}. \tag{2.3}$$

The $L^{ij}(t)$ were obtained as a by-product of evaluating the integral

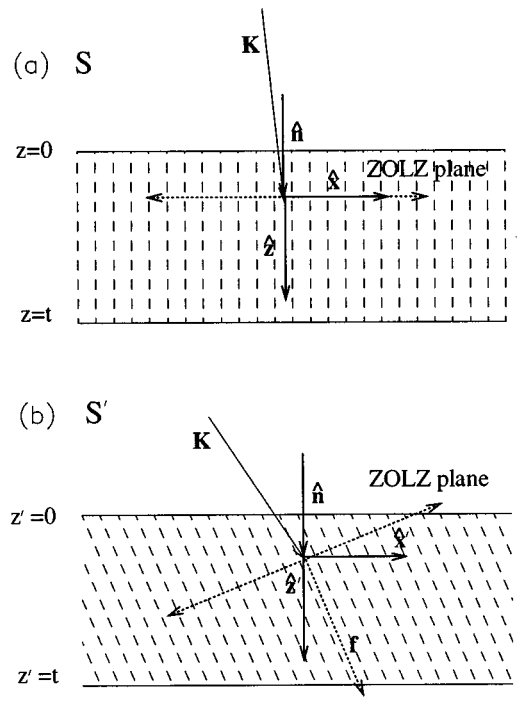

FIG. 1. Schematic mixed real-space and reciprocal-space diagrams showing the scattering geometries discussed in the text. The crystal is of thickness t with surface normal $\hat{\mathbf{n}}$. $\mathbf{K}$ is the wave vector of the incident electron beam (corrected for refraction). Real-space diffracting planes are indicated by the dashed lines. The ZOLZ plane is indicated by the dotted line. (a) The restricted diffraction geometry with the ZOLZ plane parallel to the crystal surface as defined in Ref. 2, and the associated coordinate system S. (b) The more general diffraction geometry is discussed here with the ZOLZ plane at some arbitrary angle to the crystal surface, and the associated coordinate system S'. Here $\mathbf{f}$ is a vector normal to the ZOLZ plane as discussed in the text.

$$I=\frac{1}{V}\int_V \exp[i(\mathbf{k}^i+\mathbf{g}-\mathbf{k}^{j*}-\mathbf{h})\cdot\mathbf{r}]d\mathbf{r}. \tag{2.4}$$

It was assumed in Ref. 2 that the crystal slab effectively extends to infinity along the surface of incidence while being of finite thickness t. In addition, it was assumed that all the reciprocal lattice vectors $\mathbf{g}$ and $\mathbf{h}$ pertinent to the diffraction conditions lie in a plane parallel to the crystal surface. This plane corresponds to the ZOLZ of a diffraction condition with zone-axis normal to the crystal surface. Any contribution from $\mathbf{g}$ and $\mathbf{h}$ vectors corresponding to reciprocal lattice points in HOLZ's was ignored.

The Cartesian coordinate system S, shown in Fig. 1(a), was used to evaluate the integral. In S, the unit vectors $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ define the xy plane in which all the ZOLZ reciprocal lattice points (and vectors) lie, and the diffraction zone axis defines the z direction. For the specific case considered in Ref. 2 the xy plane lies parallel to the crystal surface and the z direction parallel to the surface normal. Factoring the integral in Eq. (2.4) into components along the xy plane and z direction, we write

$$\begin{aligned} I&=\frac{1}{A}\int_A \exp[i(\mathbf{h}-\mathbf{g})\cdot\mathbf{r}_{xy}]d\mathbf{r}_{xy} \\ &\quad\times\frac{1}{t}\int_0^t \exp[i(\lambda^i\hat{\mathbf{n}}-\lambda^{j*}\hat{\mathbf{n}})\cdot\mathbf{z}]d\mathbf{z} \\ &=\delta_{\mathbf{h},\mathbf{g}}L^{ij}(t). \end{aligned} \tag{2.5}$$

Since t is much smaller than the surface dimensions A of the crystal we do not obtain a δ function in the z direction. This assumption is pivotal in the derivation of Eq. (2.1).

Evaluation of Eq. (2.5) for other diffraction conditions, where the zone axis is not parallel to the surface normal, is problematic because the $\mathbf{g}$ and $\mathbf{h}$ vectors have components in both the z direction and xy plane. To circumvent this we consider a new Cartesian coordinate system S' [see Fig. 1(b)], where the crystal surface normal $\hat{\mathbf{n}}$ now defines the z' direction and the crystal surface is parallel to the $x'y'$ plane. For any arbitrary $\mathbf{g}$ and $\mathbf{h}$ vectors pertinent to the diffraction conditions (i.e., corresponding to reciprocal lattice points lying in the ZOLZ) we can write

$$\begin{aligned} I&=\frac{1}{A}\int_A \exp[i(\mathbf{h}-\mathbf{g})\cdot\mathbf{r}_{x'y'}]d\mathbf{r}_{x'y'} \\ &\quad\times\frac{1}{t}\int_0^t \exp[i(\mathbf{k}^i+\mathbf{h}-\mathbf{k}^{j*}-\mathbf{g})\cdot\mathbf{z}']d\mathbf{z}' \\ &=\delta_{\mathbf{h}\cdot\mathbf{r}_{x'y'},\mathbf{g}\cdot\mathbf{r}_{x'y'}}\frac{1}{t}\int_0^t \exp[i(\lambda^i-\lambda^{j*}+h_{z'}-g_{z'})z']dz'. \end{aligned} \tag{2.6}$$

The surface Kronecker δ, $\delta_{\mathbf{h}\cdot\mathbf{r}_{x'y'},\mathbf{g}\cdot\mathbf{r}_{x'y'}}$ seems to impose a less stringent constraint than that in Eq. (2.5), namely that the integral is zero unless $h_{x'}=g_{x'}$ and $h_{y'}=g_{y'}$.

We now consider the implications of this surface Kronecker δ function in evaluating the above integral. We represent any arbitrary reciprocal lattice vectors $\mathbf{h}$ and $\mathbf{g}$ corresponding to points in the ZOLZ in the S' system as

$$\mathbf{h}=h_{x'}\hat{\mathbf{x}}'+h_{y'}\hat{\mathbf{y}}'+h_{z'}\hat{\mathbf{z}}', \tag{2.7}$$

$$\mathbf{g}=g_{x'}\hat{\mathbf{x}}'+g_{y'}\hat{\mathbf{y}}'+g_{z'}\hat{\mathbf{z}}', \tag{2.8}$$

and consider a vector normal to the ZOLZ plane given by

$$\mathbf{f}=f_{x'}\hat{\mathbf{x}}'+f_{y'}\hat{\mathbf{y}}'+f_{z'}\hat{\mathbf{z}}'. \tag{2.9}$$

Any two vectors $\mathbf{g}$ and $\mathbf{h}$ corresponding to points in the ZOLZ must satisfy

$$c\mathbf{f}=\mathbf{h}\times\mathbf{g}, \tag{2.10}$$

for some constant c. Now we impose the constraint that $h_{x'}=g_{x'}$ and $h_{y'}=g_{y'}$ required by $\delta_{\mathbf{h}\cdot\mathbf{r}_{x'y'},\mathbf{g}\cdot\mathbf{r}_{x'y'}}$ if the integral I is not to be zero. We obtain

$$\begin{aligned} cf_{x'}\hat{\mathbf{x}}'+cf_{y'}\hat{\mathbf{y}}'+cf_{z'}\hat{\mathbf{z}}'&=(h_{y'}g_{z'}-h_{z'}h_{y'})\hat{\mathbf{x}}' \\ &\quad+(h_{z'}h_{x'}-h_{x'}g_{z'})\hat{\mathbf{y}}', \end{aligned} \tag{2.11}$$

giving

$$cf_{x'}=h_{y'}(g_{z'}-h_{z'}), \tag{2.12}$$

$$cf_{y'}=h_{x'}(h_{z'}-g_{z'}), \tag{2.13}$$

$$cf_{z'}=0. \tag{2.14}$$

With the assumption (to be discussed shortly) that $f_{z'}\neq 0$, Eq. (2.14) implies $c=0$ and hence that $\mathbf{h}$ is parallel to $\mathbf{g}$. Then assuming that at least one of $h_{x'}$ or $h_{y'}\neq 0$ we see from Eq. (2.12) and Eq. (2.13) that

$$g_{z'}=h_{z'}, \tag{2.15}$$

and hence $\mathbf{h}=\mathbf{g}$ if the integral I is not to be zero. Hence Eq. (2.6) reduces to the same result that Eq. (2.5) does.

Now let us examine the assumption we made in coming to this conclusion. First we assumed $f_{z'}\neq 0$. If in fact $f_{z'}=0$ then the ZOLZ is perpendicular to the crystal surface (i.e., the zone axis is perpendicular to the crystal surface normal). Secondly we assumed that at least one of $h_{x'},h_{y'}\neq 0$. If $h_{x'}=h_{y'}=0$ then $\mathbf{h}=h_{z'}\hat{\mathbf{z}}'$ and $\mathbf{g}=g_{z'}\hat{\mathbf{z}}'$, once again corresponding to the case where the ZOLZ is perpendicular to the crystal surface. Hence, with this one exception, the scattering equations [Eq. (2.1)] are valid for general orientations of the ZOLZ with respect to the crystal surface. We point out that to a good approximation (except for very thin crystals) the fast electron wave function obtained from the scattering equations are not greatly affected by the assumption that the crystal is finite in thickness. For an infinite crystal the scattering equations reduce to the well known results of Yoshioka[1]

$$[K^2-(\mathbf{k}^i+\mathbf{g})^2]C^i_{\mathbf{g}}+\frac{2m}{\hbar^2}\left[\sum_{\mathbf{h}\neq\mathbf{g}} V_{\mathbf{g}-\mathbf{h}}C^i_{\mathbf{h}}+\sum_{\mathbf{h}} W_{\mathbf{g},\mathbf{h}}C^i_{\mathbf{h}}\right]=0. \tag{2.16}$$

In this case the integral in question [Eq. (2.5)] reduces to $\delta_{\mathbf{h},\mathbf{g}}$ for any $\mathbf{h}$ and $\mathbf{g}$ vectors which may each correspond to a reciprocal lattice point lying in the ZOLZ or any HOLZ.

The inelastic scattering cross section, discussed at length in Ref. 2, is given by

$$\sigma=NV_c\left\{\left[1-\sum_{i,j} B^{ij}(t)\sum_{\mathbf{g}} C^i_{\mathbf{g}}C^{j*}_{\mathbf{g}}\right]\mu_{\mathbf{0},\mathbf{0}} + \sum_{i,j} B^{ij}(t)\sum_{\mathbf{g},\mathbf{h}} C^i_{\mathbf{g}}C^{j*}_{\mathbf{h}}\mu_{\mathbf{h},\mathbf{g}}\right\}, \tag{2.17}$$

where $B^{ij}(t)=\alpha^i\alpha^{j*}L^{ij}(t)$ and the $\mu_{\mathbf{h},\mathbf{g}}$ are the inelastic scattering coefficients for the particular inelastic scattering process under consideration, e.g., inner-shell ionization, Rutherford backscattering. This important general result takes into account all other (background) inelastic scattering in the crystal leading to absorption from the dynamical Bragg-reflected beams (in practice mainly due to thermal diffuse scattering). There is a contribution to the cross section from all absorbed electrons as well as from the dynamical electrons. Contrary to the scattering equations, the assumption of finite thickness manifested in the $L^{ij}(t)$ is crucially important, and this has been previously emphasized.[2,8,9] The assumption of an infinitely thick crystal in this context is tantamount to assuming that the detectors in an experiment are inside the crystal and this leads to incorrect results.[9]

The derivation of the above cross section expression in Ref. 2, was made with the unnecessary assumption that the ZOLZ was parallel to the surface. This assumption need not have been made, as the $h_{z'}$ and $g_{z'}$ components that occur in Eq. (2.6) are removed by a subsequent further integration [over the initial states of the solid, Eq. (A24) in Ref. 2]. This integration is independent of any particular scattering geometry. The inelastic scattering cross section is therefore valid as it stands for any orientation of the ZOLZ and, without the limitation of the $\mathbf{h}$ and $\mathbf{g}$ vectors in Eq. (2.17) having to correspond to reciprocal lattice points in the ZOLZ only, contributions from HOLZ's are readily included.

III. CONLUSION

We have shown that recently derived generalized scattering equations for fast electrons incident on a crystal of finite thickness[2] are valid as they stand for general orientations of the diffraction zone axis with respect to the crystal surface (provided HOLZ effects are not included). The only exception is for the specific case where the ZOLZ lies perpendicular to the crystal surface. This restriction on the orientation of the ZOLZ and the inclusion of HOLZ reflections is removed if the thickness is considered to approach infinity so that Yoshioka's generalized scattering equations are recovered. Furthermore, a widely applicable expression for the cross section for inelastic scattering in a crystal[2] (where the finite thickness of the crystal is crucial) is shown to be valid for any orientation of the ZOLZ, and in addition contributions from HOLZ reflections may be included in the inelastic cross section expression. Therefore, in practice, the use of Yoshioka's fundamental scattering equations coupled with the inelastic cross section expression given by Eq. (2.17) makes possible the calculation of the *total* cross section of scattering for any diffraction condition.

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