Inelastic plasmon and interband electron-scattering potentials for Si from dielectric matrix calculations

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Inelastic scattering of electrons in a crystalline environment may be represented by a complex non-Hermitian potential. Complete generalized expressions for this inelastic-electron-scattering-potential matrix, including virtual-inelastic scattering, are derived for outer-shell electron and plasmon excitations. The relationship between these expressions and the general anisotropic dielectric-response matrix of the solid is discussed. These generalized expressions necessarily include the off-diagonal terms representing effects due to departure from translational invariance in the interaction. Results are presented for the diagonal band-structure-dependent inelastic and virtual-inelastic scattering potentials for Si, from a calculation of the inverse dielectric matrix in the random-phase approximation. Good agreement is found with experiment as a function of incident energies from 10 eV to 100 keV. Anisotropy effects, and, hence, the interaction delocalization represented by the off-diagonal scattering-potential terms are found to be significant below 1 keV.

I. INTRODUCTION

Inelastic scattering of electrons, such as thermal diffuse scattering, inner-shell electron ionization, and valence electron excitations, plays a crucial role in the dynamics of quantitative electron diffraction-microscopy and electron spectroscopy.[1–3] This includes convergent beam electron diffraction, low energy electron diffraction,[4] and electron spectroscopy techniques[5–7] [such as Auger electron spectroscopy, x-ray photoemission spectroscopy, and electron energy loss spectroscopy (EELS)]. The inelastic electrons are a sensitive measure of many types of elementary excitations in the solid.[8] In this work, we are interested in the electronic excitations, interband single-particle and collective plasmon excitations.[9] These in turn depend strongly on the electronic band structure of the solid.

In a rigorous treatment of inelastic scattering based on the standard dynamical diffraction equations of Bethe[10] one obtains, in addition to the usual Hermitian elastic scattering potential, a complex non-Hermitian "correction matrix."[11] This correction matrix or inelastic scattering-potential matrix, is analogous to the Fourier components of the complex optical potential often used to phenomenologically characterize inelastic scattering.[12,13] For electronic excitations, simplified versions of the inelastic scattering-potential matrix have been related in various ways to the dielectric response of the solid.[14–17] The utility of the dielectric approach is apparent. It is, in principle, fully capable of representing, by use of empirical data or direct calculation,[5,18] the effects of valence-conduction electron and plasmon excitations.

Previous work in this area at medium to high incident energies[13,19,20] (10–100 keV), has concentrated mainly on determining the inelastic mean-inner potential (MIP), effectively the "head" of the inelastic potential matrix. This is due in part to the very delocalized nature of valence-conduction electron and plasmon excitations[20,21] at these energies. For large interaction delocalization the off-diagonal inelastic potential matrix terms are expected to be small and hence the diagonal terms (representing the MIP) to dominate. At lower incident energies (<1 keV) effects due to larger off-diagonal terms may be significant in scattering experiments.[4] Recent advances and developments in electron diffraction microscopy and electron spectroscopy work in the medium energy regimes,[22] are also sensitive to solid state bonding (band structure) effects.[12,23,24] Hence, it is now desirable to estimate quantitatively the effect of these off-diagonal terms.

In Sec. II, we give a general derivation of the complex inelastic and virtual-inelastic scattering potential matrix.[25] We discuss some properties of these inelastic potentials and their relationship to generalized dynamic form factor matrices. In Sec. III, we show how the full inelastic scattering-potential matrix may be expressed in terms of the frequency and wave-vector-dependent dielectric matrix[26,27] of the solid. The relationship between these expressions and other less general results[15,17] are also pointed out.

Various simplifications to the inelastic scattering potential are considered in Sec. IV. The effects of interaction delocalization and dielectric isotropy on the inelastic potential is discussed in some detail. Because of its significance to electronic excitations, particular emphasis is placed on the diagonal terms of the inelastic potential and how they reduce to the well-known results of Pines,[8] in the dielectric isotropy approximation. Finally, in Sec. V we present quantitative first-principles calculations of the band-structure-dependent inelastic and virtual-inelastic

mean-inner potential for Si. This is based on a random-phase approximation (RPA) calculation[19,28] of the dielectric response matrix, in turn based on an empirical pseudopotential band structure.[29]

II. THE INELASTIC SCATTERING-POTENTIAL MATRIX

It is instructive to briefly consider the origin of the inelastic scattering-potential matrix, as derived by Yoshioka.[11] We consider Schrödinger's equation for the incident electron and periodic solid, $H\Phi = E\Phi$,

$$H = \frac{-\hbar^2\nabla^2}{2m} + H_c + H' , \tag{1}$$

where H_c is the unperturbed solid Hamiltonian and H' the interaction Hamiltonian between the incident electron and the solid. The total wave function Φ of the system is expressed in terms of the exact "stationary" unperturbed solid wave functions, a_n such that

$$\Phi(\mathbf{r}, \mathbf{r}_1, ..., \mathbf{r}_n) = \sum_n \phi_n(\mathbf{r}) a_n(\mathbf{r}_1, ..., \mathbf{r}_n) , \tag{2}$$

where $\mathbf{r}$ is the coordinate of the incident electron and $\mathbf{r}_j$ the coordinate of the jth electron in the solid and $H_c a_n = E_n a_n$. $\phi_0(\mathbf{r})$ represents the elastic scattered wave (of energy $E - E_0$) and $\phi_n(\mathbf{r})$ the inelastic scattering of the electron (of energy $E - E_n$) resulting from a corresponding excitation in the solid to the state a_n. Integrating over all electron coordinates $\mathbf{r}_j$ in the solid (of volume Ω) one obtains

$$\nabla^2\phi_0(\mathbf{r}) + \left[k_0^2 - \frac{2m}{\hbar^2}H'_{0,0}(\mathbf{r})\right]\phi_0(\mathbf{r}) - \frac{2m}{\hbar^2}\sum_{m\neq 0} H'_{0,m}(\mathbf{r})\phi_m(\mathbf{r}) = 0 , \tag{3}$$

$$\nabla^2\phi_n(\mathbf{r}) + \left[k_n^2 - \frac{2m}{\hbar^2}H'_{n,n}(\mathbf{r})\right]\phi_n(\mathbf{r}) - \frac{2m}{\hbar^2}\sum_{m\neq n} H'_{n,m}(\mathbf{r})\phi_m(\mathbf{r}) = 0 . \tag{4}$$

$\mathbf{k}_n$, the wave vector for the scattered electron, is defined in terms of the total energy E or the energy E^0 relative to the ground state E_0 of the solid (the vacuum incident electron energy), $\hbar^2 {k_n}^2/2m = E - E_n = E^0 - (E_n - E_o)$. $\mathbf{k}_0$ is the elastic scattered wave vector (of energy E^0). The matrix elements $H'_{n,m}$,

$$H'_{n,m}(\mathbf{r}) = \langle a_n|H'(\mathbf{r})|a_m\rangle \equiv \int a_n^* H'(\mathbf{r}) a_m d\mathbf{r}_1, ..., d\mathbf{r}_n , \tag{5}$$

is the interaction term for a transition in the solid between the states n and m. We note that $H'_{0,0}$ is just the usual "elastic" crystal potential $-H'_{0,0}(\mathbf{r}) = V(\mathbf{r}) = \sum_{\mathbf{g}} V_{\mathbf{g}} \exp i(\mathbf{g}\cdot\mathbf{r})$ and $\mathbf{g}$ is a reciprocal lattice vector. We have used the standard sign convention that defines the crystal potential $V(\mathbf{r})$ as positive inside the crystal.

With the assumption that $H'_{n,m} \ll H'_{n,0}$, so that only excitations from the ground state contribute significantly to the scattering,[25] Eq. (4) reduces to

$$\nabla^2\phi_n(\mathbf{r}) + k_n^2\phi_n(\mathbf{r}) - \frac{2m}{\hbar^2}H'_{n,0}(\mathbf{r})\phi_0(\mathbf{r}) = 0, \qquad n \neq 0 . \tag{6}$$

The resultant out-going (the retarded) wave solution $\phi_n(\mathbf{r})$,

$$\phi_n(\mathbf{r}) = \frac{-m}{2\pi\hbar^2}\int_\Omega \frac{e^{ik_n|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}H'_{n,0}(\mathbf{r}')\phi_0(\mathbf{r}')d\mathbf{r}' \tag{7}$$

substituted into Eq. (3) yields

$$\nabla^2\phi_0(\mathbf{r}) + \left[k_0^2 - \frac{2m}{\hbar^2}H'_{0,0}(\mathbf{r})\right]\phi_0(\mathbf{r}) + \frac{2m}{\hbar^2}\int_\Omega A(\mathbf{r},\mathbf{r}')\phi_0(\mathbf{r}')d\mathbf{r}' = 0 , \tag{8}$$

where $A(\mathbf{r},\mathbf{r}')$ is the nonlocal kernel given by

$$A(\mathbf{r},\mathbf{r}') = \frac{m}{2\pi\hbar^2}\sum_n \langle a_0|H'(\mathbf{r})|a_n\rangle \times \langle a_n|H'(\mathbf{r}')|a_0\rangle \frac{e^{ik_n|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|} . \tag{9}$$

This term incorporates all the excitations from the initial state $|a_0\rangle$ to the final states $|a_n\rangle$ in the solid and hence represents the effect of inelastic scattering of the electrons. Expanding the wave function of the electron in the solid in Bloch form, $\phi_0(\mathbf{r}) = \sum_{\mathbf{h}} \chi_{\mathbf{h}} \exp i(\mathbf{K}+\mathbf{h})\cdot\mathbf{r}$, where $\mathbf{K}$ is the electron beam wave vector in the solid, one may show[11] by premultiplying by $\phi_0(\mathbf{r})$ in Eq. (8) and by integrating over the crystal volume Ω that the effect of the interaction $H'(\mathbf{r})$ on the elastic scattered wave $\phi_o(\mathbf{r})$ is given by

$$\frac{\hbar^2}{2m}[(\mathbf{K}+\mathbf{h})^2 - k_0^2]\chi_{\mathbf{h}} - \sum_{\mathbf{g}}(V_{\mathbf{h}-\mathbf{g}} + V'_{\mathbf{h},\mathbf{g}})\chi_{\mathbf{g}} = 0 . \tag{10}$$

Equation (10) is the basic equation of dynamical electron diffraction theory generalized to include inelastic scattering.[11] $V_{\mathbf{h}-\mathbf{g}}$ are the usual local crystal potential Fourier components that result in elastic scattering. The $V'_{\mathbf{h},\mathbf{g}}$ are the complex nonlocal inelastic scattering-potential matrix components that represent and incorporate the effects of excitations in the solid,

$$V'_{\mathbf{h},\mathbf{g}} = \frac{1}{\Omega}\int_\Omega\int_\Omega e^{-i(\mathbf{K}+\mathbf{h})\cdot\mathbf{r}} A(\mathbf{r},\mathbf{r}') e^{i(\mathbf{K}+\mathbf{g})\cdot\mathbf{r}'} d\mathbf{r}d\mathbf{r}' . \tag{11}$$

The total scattering potential may then be written in the standard form[12] as an effective complex "optical-potential," $V^s_{\mathbf{h},\mathbf{g}} = V_{\mathbf{h}-\mathbf{g}} + V'_{\mathbf{h},\mathbf{g}}$. The problem of inelastic scattering in electron diffraction is then reduced to solving Bethe's[10] equation with the correct inelastic scattering-potential matrix.

To solve for the $V'_{\mathbf{h},\mathbf{g}}$ in a real solid, we substitute the

Green's function relation[30]

$$\frac{1}{4\pi}\frac{e^{ik_n|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|} = \lim_{\delta\to 0^+}\int \frac{d\mathbf{K}'}{(2\pi)^3}\frac{e^{i\mathbf{K}'\cdot(\mathbf{r}-\mathbf{r}')}}{K'^2-k_n^2-i\delta}\,, \tag{12}$$

into $A(\mathbf{r},\mathbf{r}')$ and, generalizing the sum over the ground state $|a_0\rangle$ and final states $|a_n\rangle$ by the sum over multiple initial $|a_i\rangle$ and final states $|a_f\rangle$, we obtain [with $\hbar^2 k_f{}^2/2m = E - E_f = E^0 - (E_f - E_i)$]

$$A(\mathbf{r},\mathbf{r}') = -\lim_{\varepsilon\to 0^+}\int \frac{d\mathbf{K}'}{(2\pi)^3}\sum_{i,f}\frac{f_0(E_i)[1-f_0(E_f)]\langle a_i|H'(\mathbf{r})e^{i\mathbf{K}'\cdot\mathbf{r}}|a_f\rangle\langle a_f|H'(\mathbf{r}')e^{-i\mathbf{K}'\cdot\mathbf{r}'}|a_i\rangle}{E^0-E_{K'}-(E_f-E_i)+i\varepsilon}\,, \tag{13}$$

where $f_0(E)$ is the Fermi distribution. We thus obtain the inelastic potential matrix components $V'_{\mathbf{h},\mathbf{g}}$ as

$$V'_{\mathbf{h},\mathbf{g}} = -\frac{1}{\Omega}\lim_{\varepsilon\to 0^+}\int \frac{d\mathbf{K}'}{(2\pi)^3}\sum_{i,f}\frac{f_0(E_i)[1-f_0(E_f)]W^*_{f,i}(\mathbf{K}+\mathbf{h}-\mathbf{K}')W_{f,i}(\mathbf{K}+\mathbf{g}-\mathbf{K}')}{E^0-E_{K'}-(E_f-E_i)+i\varepsilon}\,, \tag{14}$$

where

$$W_{f,i}(\mathbf{q}) = \int_\Omega \langle a_f|H'(\mathbf{r})e^{i\mathbf{q}\cdot\mathbf{r}}|a_i\rangle d\mathbf{r}\,. \tag{15}$$

In this work, we are specifically interested in the solid state electronic excitation contribution to the scattering, and hence the interaction term $W_{f,i}$ reduces (with the Hartree-Fock approximation to the electron-electron interaction Hamiltonian)[16] to

$$W_{f,i}(\mathbf{q}) = \frac{4\pi e^2}{q^2}\langle a_f|e^{i\mathbf{q}\cdot\mathbf{r}}|a_i\rangle\,, \tag{16}$$

and the inelastic potential matrix becomes

$$V'_{\mathbf{h},\mathbf{g}} = -\frac{1}{\Omega}\lim_{\varepsilon\to 0^+}\int \frac{d\mathbf{K}'}{(2\pi)^3}\frac{(4\pi e^2)^2}{|\mathbf{K}+\mathbf{h}-\mathbf{K}'|^2|\mathbf{K}+\mathbf{g}-\mathbf{K}'|^2} \times\sum_{i,f}\frac{f_0(E_i)[1-f_0(E_f)]\langle a_i|e^{-i(\mathbf{K}+\mathbf{h}-\mathbf{K}')\cdot\mathbf{r}}|a_f\rangle\langle a_f|e^{i(\mathbf{K}+\mathbf{g}-\mathbf{K}')\cdot\mathbf{r}}|a_i\rangle}{E^0-E_{K'}-(E_f-E_i)+i\varepsilon}\,. \tag{17}$$

We replace the summation over the initial ground state and final states in Eq. (17) by a summation over the occupied (at $T=0$) valence states v and unoccupied (at $T=0$) conduction states c in the solid, and so obtain

$$V'_{\mathbf{h},\mathbf{g}} = -\frac{1}{\Omega}\lim_{\varepsilon\to 0^+}\int \frac{d\mathbf{K}'}{(2\pi)^3}\frac{(4\pi e^2)^2}{|\mathbf{K}+\mathbf{h}-\mathbf{K}'|^2|\mathbf{K}+\mathbf{g}-\mathbf{K}'|^2} \times\sum_{v,c}\frac{M^*_{c,v}(\mathbf{K}+\mathbf{h}-\mathbf{K}')M_{c,v}(\mathbf{K}+\mathbf{g}-\mathbf{K}')}{E^0-E_{K'}-(E_c-E_v)+i\varepsilon}\,, \tag{18}$$

where the matrix elements $M_{c,v}(\mathbf{q})$ are given by $M_{c,v}(\mathbf{q}) = \langle a_c|e^{i\mathbf{q}\cdot\mathbf{r}}|a_v\rangle$. Equation (18) is the total complex inelastic scattering-potential matrix due to inelastic electronic excitations. We may rewrite this expression as

$$V'_{\mathbf{h},\mathbf{g}} = \frac{1}{\Omega}\lim_{\varepsilon\to 0^+}\int \frac{d\mathbf{K}'}{(2\pi)^3}\frac{(4\pi e^2)^2}{|\mathbf{K}+\mathbf{h}-\mathbf{K}'|^2|\mathbf{K}+\mathbf{g}-\mathbf{K}'|^2} \times S_{\mathbf{h},\mathbf{g}}\left(\mathbf{K}-\mathbf{K}',\frac{E^0-E_{K'}}{\hbar}\right)\,. \tag{19}$$

$S_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega)$ is a generalized non-Hermitian dynamic form factor matrix that we define here as

$$S_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \sum_{v,c}\frac{-M^*_{c,v}(\mathbf{q}'+\mathbf{G}+\mathbf{h})M_{c,v}(\mathbf{q}'+\mathbf{G}+\mathbf{g})}{\hbar\omega-\hbar\omega_{c,v}+i\varepsilon}\,, \tag{20}$$

where we restrict $\mathbf{q}'$ (but not $\mathbf{q}$) to lie in the first Brillouin zone by translation through the reciprocal lattice vector $\mathbf{G}$ such that $\mathbf{q}=\mathbf{q}'+\mathbf{G}$. In addition, we have used the more convenient notation $\hbar\omega_{c,v} = E_c - E_v$ and $\hbar\omega = E^0 - E_{K'}$.

It is convenient to explicitly separate Eq. (19) into two parts: $V'_{\mathbf{h},\mathbf{g}} = V^r_{\mathbf{h},\mathbf{g}} + iV^i_{\mathbf{h},\mathbf{g}}$, using the standard relation

$$\lim_{\varepsilon\to 0^+}\frac{a}{(x-b)+i\varepsilon} = aP\frac{1}{(x-b)} - i\pi a\delta(x-b)\,, \tag{21}$$

where P denotes the principal part of the function at the singularity $x=b$. The $V^r_{\mathbf{h},\mathbf{g}}$ represents virtual-inelastic scattering[11,25] that may be interpreted as representing the effects of the induced dynamic polarization of the crystal. The $V^i_{\mathbf{h},\mathbf{g}}$ represents the "real" inelastic scattering. We then obtain

$$V^r_{\mathbf{h},\mathbf{g}} = \frac{1}{\Omega}\int \frac{d\mathbf{K}'}{(2\pi)^3}\frac{(4\pi e^2)^2}{|\mathbf{K}+\mathbf{h}-\mathbf{K}'|^2|\mathbf{K}+\mathbf{g}-\mathbf{K}'|^2} \times S^r_{\mathbf{h},\mathbf{g}}\left(\mathbf{K}-\mathbf{K}', \frac{E^0-E_{K'}}{\hbar}\right), \quad (22)$$

$$V^i_{\mathbf{h},\mathbf{g}} = \frac{1}{\Omega}\int \frac{d\mathbf{K}'}{(2\pi)^3}\frac{(4\pi e^2)^2}{|\mathbf{K}+\mathbf{h}-\mathbf{K}'|^2|\mathbf{K}+\mathbf{g}-\mathbf{K}'|^2} \times S^i_{\mathbf{h},\mathbf{g}}\left(\mathbf{K}-\mathbf{K}', \frac{E^0-E_{K'}}{\hbar}\right), \quad (23)$$

where we define the Hermitian dynamic form factor matrices as

$$S^r_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \sum_{v,c}\frac{-M^*_{c,v}(\mathbf{q}'+\mathbf{G}+\mathbf{h})M_{c,v}(\mathbf{q}'+\mathbf{G}+\mathbf{g})}{\hbar\omega-\hbar\omega_{c,v}}, \quad (24)$$

$$S^i_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \pi\sum_{v,c} M^*_{c,v}(\mathbf{q}'+\mathbf{G}+\mathbf{h}) \times M_{c,v}(\mathbf{q}'+\mathbf{G}+\mathbf{g})\delta(\hbar\omega-\hbar\omega_{c,v}). \quad (25)$$

For centrosymmetric crystal structures (such as for Si) the matrix elements are real and hence the dynamic form factor matrices, and both $V^r_{\mathbf{h},\mathbf{g}}$ and $V^i_{\mathbf{h},\mathbf{g}}$, are real quantities. This entails that $V^r_{\mathbf{h},\mathbf{g}}$ and $V^i_{\mathbf{h},\mathbf{g}}$ are the real and imaginary part, respectively, of the total inelastic potential matrix in Eq. (19). For a general noncentrosymmetric structure the matrix elements are complex and hence both $V^r_{\mathbf{h},\mathbf{g}}$ and $V^i_{\mathbf{h},\mathbf{g}}$ are in general complex.[17] Direct substitution easily verifies[11] that the components are individually Hermitian $V^r_{\mathbf{h},\mathbf{g}} = V^{r*}_{\mathbf{g},\mathbf{h}}$ and $V^i_{\mathbf{h},\mathbf{g}} = V^{i*}_{\mathbf{g},\mathbf{h}}$.

III. INELASTIC SCATTERING AND THE DIELECTRIC MATRIX

Direct calculation of the inelastic scattering potential via Eq. (22) and Eq. (23) is computationally highly demanding. One may obtain a more useful expression for $V^r_{\mathbf{h},\mathbf{g}}$ and $V^i_{\mathbf{h},\mathbf{g}}$ in terms of the dielectric response of the solid. The dielectric response of most solids is well studied both theoretically and experimentally, allowing calculation of $V^r_{\mathbf{h},\mathbf{g}}$ and $V^i_{\mathbf{h},\mathbf{g}}$ directly from any given model dielectric function or experimental data.[5,18]

Because of the periodic nature of the crystal lattice potential, the general dielectric response of a crystalline solid to an applied external field of frequency ω and wave vector $\mathbf{q}$, is to induce rapidly oscillating microscopic or local fields of frequency ω and wave vector $\mathbf{q}+\mathbf{g}$, where $\mathbf{g}$ is a reciprocal lattice vector. This dielectric response may be expressed in matrix form.[9,31] In order to relate the generalized dynamic form-factor matrix in Eq. (19) to the dielectric response of the solid, we consider the dielectric matrix formulation of Adler[26] and Wiser[27] in the random-phase approximation,

$$\epsilon_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \delta_{\mathbf{h},\mathbf{g}} - \frac{4\pi e^2}{\Omega|\mathbf{q}+\mathbf{g}||\mathbf{q}+\mathbf{h}|}\lim_{\alpha\to 0^+}\sum_{\mathbf{k},n,n'}\frac{f_0[E_{n'}(\mathbf{k}+\mathbf{q})]-f_0[E_n(\mathbf{k})]}{E_{n'}(\mathbf{k}+\mathbf{q})-E_n(\mathbf{k})+\hbar\omega+i\hbar\alpha} \times\langle\mathbf{k}+\mathbf{q},n'|e^{i(\mathbf{q}+\mathbf{g})\cdot\mathbf{r}}|\mathbf{k},n\rangle\langle\mathbf{k},n|e^{-i(\mathbf{q}+\mathbf{h})\cdot\mathbf{r}}|\mathbf{k}+\mathbf{q},n'\rangle, \quad (26)$$

where the Brillouin zone summation is over all possible transitions between the states $|\mathbf{k},n\rangle$ of energy $E_n(\mathbf{k})$ and $|\mathbf{k}+\mathbf{q},n'\rangle$ of energy $E_{n'}(\mathbf{k}+\mathbf{q})$.

Using the standard relations[32] between the RPA and the Hartree-Fock (HF) dielectric functions $\epsilon^{-1}_{\mathrm{HF}} = 1+4\pi\alpha$ and $\epsilon_{\mathrm{RPA}} = 1-4\pi\alpha$, where α is the polarizability of the solid, we may write [in the notation of Eq. (18)]

$$\epsilon^{-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \delta_{\mathbf{h},\mathbf{g}} + \frac{4\pi e^2}{\Omega|\mathbf{q}+\mathbf{g}||\mathbf{q}+\mathbf{h}|} \times\lim_{\varepsilon\to 0^+}\sum_{v,c}\frac{M^*_{c,v}(\mathbf{q}+\mathbf{h})M_{c,v}(\mathbf{q}+\mathbf{g})}{\hbar\omega-\hbar\omega_{c,v}+i\varepsilon} - \frac{M^*_{v,c}(\mathbf{q}+\mathbf{h})M_{v,c}(\mathbf{q}+\mathbf{g})}{\hbar\omega+\hbar\omega_{c,v}+i\varepsilon}. \quad (27)$$

In general, this inverse dielectric matrix is both complex and non-Hermitian. To proceed further we use the delta function relation of Eq. (21) to deconstruct the dielectric matrix into the sum of two complex Hermitian matrices: $\epsilon^{-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \epsilon^{r-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) + i\epsilon^{i-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega)$, where

$$\epsilon^{r-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \delta_{\mathbf{h},\mathbf{g}} + \frac{4\pi e^2}{\Omega|\mathbf{q}+\mathbf{g}||\mathbf{q}+\mathbf{h}|} \times\sum_{v,c}\frac{M^*_{c,v}(\mathbf{q}+\mathbf{h})M_{c,v}(\mathbf{q}+\mathbf{g})}{\hbar\omega-\hbar\omega_{c,v}} - \frac{M^*_{v,c}(\mathbf{q}+\mathbf{h})M_{v,c}(\mathbf{q}+\mathbf{g})}{\hbar\omega+\hbar\omega_{c,v}}, \quad (28)$$

$$\epsilon^{i-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = -\pi\frac{4\pi e^2}{\Omega|\mathbf{q}+\mathbf{g}||\mathbf{q}+\mathbf{h}|}\sum_{v,c} M^*_{c,v}(\mathbf{q}+\mathbf{h}) \times M_{c,v}(\mathbf{q}+\mathbf{g})\delta(\hbar\omega-\hbar\omega_{c,v}) - M^*_{v,c}(\mathbf{q}+\mathbf{h})M_{v,c}(\mathbf{q}+\mathbf{g})\delta(\hbar\omega+\hbar\omega_{c,v}). \quad (29)$$

We then have the following relations:

$$\epsilon^{r-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \frac{1}{2}[\epsilon^{-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) + \epsilon^{*-1}_{\mathbf{g},\mathbf{h}}(\mathbf{q},\omega)], \quad (30)$$

$$\epsilon^{i-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = -\frac{i}{2}[\epsilon^{-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) - \epsilon^{*-1}_{\mathbf{g},\mathbf{h}}(\mathbf{q},\omega)] \,. \tag{31}$$

With $\delta(\hbar\omega + \hbar\omega_{c,v}) \equiv 0$ for all $\omega > 0$ and noting that the second summand in Eq. (28) is small, we obtain the general relation between the dynamic form-factor matrices and the inverse dielectric matrix

$$S^r_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \frac{-\Omega|\mathbf{q}'+\mathbf{h}+\mathbf{G}||\mathbf{q}'+\mathbf{g}+\mathbf{G}|}{4\pi e^2} \times\frac{1}{2}[\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega) + \epsilon^{*-1}_{\mathbf{g}+\mathbf{G},\mathbf{h}+\mathbf{G}}(\mathbf{q}',\omega) -2\delta_{\mathbf{g},\mathbf{h}}] \,, \tag{32}$$

$$S^i_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) = \frac{\Omega|\mathbf{q}'+\mathbf{h}+\mathbf{G}||\mathbf{q}'+\mathbf{g}+\mathbf{G}|}{4\pi e^2} \times\frac{i}{2}[\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega) - \epsilon^{*-1}_{\mathbf{g}+\mathbf{G},\mathbf{h}+\mathbf{G}}(\mathbf{q}',\omega)] \,. \tag{33}$$

From these general noncentrosymmetric expressions we may, by direct substitution of a Hermitian dielectric matrix, trivially obtain the equivalent centrosymmetric forms. We note that $S^i_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega)$ in Eq. (33) is related to the mixed dynamic form factor of Kohl and Rose.[17] In addition the diagonal $\mathbf{g} = \mathbf{h}$ term in Eq. (33) is equivalent to the dynamic form factor of Saslow and Reiter.[15]

With the general expressions Eq. (32) and Eq. (33) we obtain by substitution into our defining equations [Eq. (22) and Eq. (23)] the general relation between the inelastic scattering-potential matrix and the inverse dielectric matrix, as

$$V^r_{\mathbf{h},\mathbf{g}} = 4\pi e^2 \int \frac{d\mathbf{K}'}{(2\pi)^3} \frac{-1}{2|\mathbf{K}+\mathbf{h}-\mathbf{K}'||\mathbf{K}+\mathbf{g}-\mathbf{K}'|} \times\left[\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}\left(\mathbf{K}-\mathbf{K}'-\mathbf{G},\frac{E^0-E_{K'}}{\hbar}\right) + \epsilon^{*-1}_{\mathbf{g}+\mathbf{G},\mathbf{h}+\mathbf{G}}\left(\mathbf{K}-\mathbf{K}'-\mathbf{G},\frac{E^0-E_{K'}}{\hbar}\right) - 2\delta_{\mathbf{h},\mathbf{g}}\right] , \tag{34}$$

$$V^i_{\mathbf{h},\mathbf{g}} = 4\pi e^2 \int \frac{d\mathbf{K}'}{(2\pi)^3} \frac{i}{2|\mathbf{K}+\mathbf{h}-\mathbf{K}'||\mathbf{K}+\mathbf{g}-\mathbf{K}'|} \times\left[\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}\left(\mathbf{K}-\mathbf{K}'-\mathbf{G},\frac{E^0-E_{K'}}{\hbar}\right) - \epsilon^{*-1}_{\mathbf{g}+\mathbf{G},\mathbf{h}+\mathbf{G}}\left(\mathbf{K}-\mathbf{K}'-\mathbf{G},\frac{E^0-E_{K'}}{\hbar}\right)\right] . \tag{35}$$

We transform the integration over $\mathbf{K}'$ to an integration over $\mathbf{q}'$ (restricted to the first Brillouin zone) such that $\mathbf{K}-\mathbf{K}' = \mathbf{q} = \mathbf{q}'+\mathbf{G}$ where $\hbar\mathbf{q}$ is the momentum transfer to the solid in the scattering process. In addition, we also integrate over all the related energy transfers $\hbar\omega$ to the solid $\hbar\omega = E^0 - E_{K'}$ up to a maximum of E^0 (the vacuum incident electron energy). The variables ω and $\mathbf{q}$ are related by energy and momentum conservation, $\hbar\omega = \hbar\mathbf{q}\cdot\mathbf{V} - \hbar^2q^2/m$, where $\mathbf{V}$ is the incident electron velocity. We then obtain the final expressions for the inelastic and virtual-inelastic scattering-potential matrix components (for a general crystal structure), in terms of the dielectric response as

$$V^r_{\mathbf{h},\mathbf{g}} = 4\pi e^2 \int_0^{E^0/\hbar} d\omega \sum_{\mathbf{G}} \int_{\mathrm{BZ}} \frac{d\mathbf{q}'}{(2\pi)^3} \times\frac{-1}{2|\mathbf{q}'+\mathbf{h}+\mathbf{G}||\mathbf{q}'+\mathbf{g}+\mathbf{G}|} \times[\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega) + \epsilon^{*-1}_{\mathbf{g}+\mathbf{G},\mathbf{h}+\mathbf{G}}(\mathbf{q}',\omega) - 2\delta_{\mathbf{h},\mathbf{g}}] \times\delta\left(\omega - (\mathbf{q}'+\mathbf{G})\cdot\mathbf{V} + \frac{\hbar(\mathbf{q}'+\mathbf{G})^2}{2m}\right) , \tag{36}$$

$$V^i_{\mathbf{h},\mathbf{g}} = 4\pi e^2 \int_0^{E^0/\hbar} d\omega \sum_{\mathbf{G}} \int_{\mathrm{BZ}} \frac{d\mathbf{q}'}{(2\pi)^3} \times\frac{i}{2|\mathbf{q}'+\mathbf{h}+\mathbf{G}||\mathbf{q}'+\mathbf{g}+\mathbf{G}|} \times[\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega) - \epsilon^{*-1}_{\mathbf{g}+\mathbf{G},\mathbf{h}+\mathbf{G}}(\mathbf{q}',\omega)] \times\delta\left(\omega - (\mathbf{q}'+\mathbf{G})\cdot\mathbf{V} + \frac{\hbar(\mathbf{q}'+\mathbf{G})^2}{2m}\right) . \tag{37}$$

For centrosymmetric structures this reduces to

$$V^r_{\mathbf{h},\mathbf{g}} = 4\pi e^2 \int_0^{E^0/\hbar} d\omega \sum_{\mathbf{G}} \int_{\mathrm{BZ}} \frac{d\mathbf{q}'}{(2\pi)^3} \times\frac{-1}{|\mathbf{q}'+\mathbf{h}+\mathbf{G}||\mathbf{q}'+\mathbf{g}+\mathbf{G}|} \times\mathrm{Re}[\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega) - \delta_{\mathbf{h},\mathbf{g}}] \times\delta\left(\omega - (\mathbf{q}'+\mathbf{G})\cdot\mathbf{V} + \frac{\hbar(\mathbf{q}'+\mathbf{G})^2}{2m}\right) , \tag{38}$$

$$V^i_{\mathbf{h},\mathbf{g}} = 4\pi e^2 \int_0^{E^0/\hbar} d\omega \sum_{\mathbf{G}} \int_{\mathrm{BZ}} \frac{d\mathbf{q}'}{(2\pi)^3} \times\frac{1}{|\mathbf{q}'+\mathbf{h}+\mathbf{G}||\mathbf{q}'+\mathbf{g}+\mathbf{G}|} \times\mathrm{Im}[-\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega)] \times\delta\left(\omega - (\mathbf{q}'+\mathbf{G})\cdot\mathbf{V} + \frac{\hbar(\mathbf{q}'+\mathbf{G})^2}{2m}\right) . \tag{39}$$

IV. EVALUATING THE INELASTIC SCATTERING-POTENTIAL MATRIX

Conduction electron and plasmon excitations in a solid are well known to be highly delocalized processes[20,21]

$|V^i_{\mathbf{h},\mathbf{h}}| \gg |V^i_{\mathbf{h},\mathbf{g}}|$, for $\mathbf{h} \neq \mathbf{g}$. The most significant inelastic scattering-potential elements for these processes are, therefore, the diagonal $V^i_{\mathbf{h},\mathbf{h}}$ and $V^r_{\mathbf{h},\mathbf{h}}$ terms. Specifically the components $V^i_{0,0}$ and $V^r_{0,0}$ correspond to the zeroth order Fourier coefficients of the inelastic and virtual-inelastic potential, respectively, that is the so-called inelastic mean-inner potential (the inelastic MIP) and the virtual-inelastic MIP. For $\mathbf{g} = \mathbf{h}$ Eq. (37) becomes

$$V^i_{\mathbf{h},\mathbf{h}} = 4\pi e^2 \int_0^{E^0/\hbar} d\omega \sum_{\mathbf{G}} \int_{\mathrm{BZ}} \frac{d\mathbf{q}'}{(2\pi)^3} \frac{1}{|\mathbf{q}'+\mathbf{h}+\mathbf{G}|^2} \times \mathrm{Im}[-\epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{h}+\mathbf{G}}(\mathbf{q}',\omega)] \times \delta\left(\omega - (\mathbf{q}'+\mathbf{G})\cdot\mathbf{V} + \frac{\hbar(\mathbf{q}'+\mathbf{G})^2}{2m}\right) . \quad (40)$$

In an entirely delocalized system $V^i_{\mathbf{h},\mathbf{g}} \rightarrow \delta_{\mathbf{h},\mathbf{g}} V^i_{\mathbf{h},\mathbf{g}}$, the crystal potential is switched off and the inverse dielectric matrix becomes diagonal[9] $\epsilon^{-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) \equiv \delta_{\mathbf{h},\mathbf{g}} \epsilon^{-1}_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega)$. We make the assumption that the off-diagonal terms of the inverse dielectric matrix are small relative to the diagonal terms, so that to a good approximation we may evaluate $V^i_{\mathbf{h},\mathbf{h}}$ from a diagonal inverse dielectric matrix. The departure of the dielectric matrix from diagonal form is a measure of the strength of the higher order coefficients of the crystal potential. Substituting $\epsilon^{-1}_{0,0}(\mathbf{q}'+\mathbf{G}+\mathbf{h},\omega) = \epsilon^{-1}_{\mathbf{h}+\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega)$ into Eq. (40) we obtain

$$V^i_{\mathbf{h},\mathbf{h}} \approx 4\pi e^2 \int_0^{E^0/\hbar} d\omega \int_{Q(\omega)} \frac{d\mathbf{q}}{(2\pi)^3} \frac{1}{|\mathbf{q}+\mathbf{h}|^2} \times \mathrm{Im}[-\epsilon^{-1}_{0,0}(\mathbf{q}+\mathbf{h},\omega)] \times \delta\left(\omega - \mathbf{q}\cdot\mathbf{V} + \frac{\hbar q^2}{2m}\right) , \quad (41)$$

where $\mathbf{q}(\equiv \mathbf{q}'+\mathbf{G})$ now runs over all allowed momentum transfers $Q(\omega)$. We note that local-field effects discussed in Sec. III are still included in the inverse dielectric matrix in Eq. (41) for all $\mathbf{q}$ within the first Brillouin zone. These fields have a dramatic influence on the calculated [0,0] component of the dielectric response for small wave vectors.[28,31] However, they *do not* affect the approximation that the off-diagonal terms of the dielectric matrix are small in comparison with the diagonal terms.

Equation (41) simplifies for the $\mathbf{h} = 0$ case and $V^i_{0,0}$, the inelastic MIP becomes

$$V^i_{0,0} \approx 4\pi e^2 \int_0^{E^0/\hbar} d\omega \int_{Q(\omega)} \frac{d\mathbf{q}}{(2\pi)^3} \frac{1}{q^2} \times \mathrm{Im}[-\epsilon^{-1}_{0,0}(\mathbf{q},\omega)] \times \delta\left(\omega - \mathbf{q}\cdot\mathbf{V} + \frac{\hbar q^2}{2m}\right) . \quad (42)$$

If we now in addition to the diagonal approximation consider the dielectric response to be isotropic, $\epsilon^{-1}_{0,0}(\mathbf{q},\omega) \equiv \epsilon^{-1}_{0,0}(|\mathbf{q}|,\omega)$ (this approximation is supported by both experiment[33] and previous calculations[28,34] for large $|\mathbf{q}|$), Eq. (42) simplifies considerably. By transforming to spherical polar coordinates $d\mathbf{q} = q^2 dq \sin(\theta) d\theta d\phi$, we may trivially integrate over the azimuthal angle ϕ, and using the standard delta function transformation

$$\delta[f(x)] = \frac{\delta(x - x_R)}{\frac{\partial f(x)}{\partial x}|_R} , \quad (43)$$

where R denotes the "root" of the function, we may readily integrate further over the polar or scattering angle θ to obtain, using

$$\delta\left(\omega - qV\cos\theta + \frac{\hbar q^2}{2m}\right) = \frac{\delta[\theta - G(\omega,q)]}{qV\sin\theta} , \quad (44)$$

$$G(\omega,q) = \cos^{-1}\left[\frac{\omega}{qV} + \frac{\hbar q}{2mV}\right] , \quad (45)$$

finally giving

$$V^i_{0,0} = \frac{e^2}{\pi V} \int_0^{E^0/\hbar} d\omega \int_{q(\theta_{\min})}^{q(\pi)} \frac{dq}{q} \mathrm{Im}[-\epsilon^{-1}_{0,0}(q,\omega)] . \quad (46)$$

The $|\mathbf{q}|$ integration is over the range defined by some minimum possible scattering angle $\theta_{\min} \rightarrow 0$ through to $\theta = \pi$, and the ω limits run over all the possible energy transfers to the solid. $|\mathbf{q}| \equiv q$ and ω are related by the scattering angle θ such that

$$\hbar q(\theta) = \sqrt{2}m \left[V^2 - \frac{\hbar\omega}{m} - V\left(V^2 - \frac{2\hbar\omega}{m}\right)^{1/2}\cos\theta\right]^{1/2} . \quad (47)$$

Equation (46) is the standard equation first derived by Pines[8] for the inelastic mean-inner potential in the isotropic approximation (see also Ritchie and Howie[18]). The above analysis may trivially be extended to include $V^r_{0,0}$ and an entirely analogous argument leads to the following expression for the isotropic virtual-inelastic MIP

$$V^r_{0,0} = \frac{e^2}{\pi V} \int_0^{E^0/\hbar} d\omega \int_{q(\theta_{\min})}^{q(\pi)} \frac{dq}{q} \mathrm{Re}[-\epsilon^{-1}_{0,0}(q,\omega) + 1] . \quad (48)$$

We now reconsider the nonzero diagonal components $V^i_{\mathbf{h},\mathbf{h}}$ and $V^r_{\mathbf{h},\mathbf{h}}$. The term $\mathrm{Im}[-\epsilon^{-1}_{0,0}(\mathbf{q},\omega)]$ is the so-called loss function of the solid. The loss function [see Fig. 1(a)] and hence the integrand of Eq. (46) is sharply peaked in the forward scattering direction at $|\mathbf{q}| = 0$. Analogously, the integrand of Eq. (41) is sharply peaked at $|\mathbf{q}+\mathbf{h}| = 0$, i.e., translated to $\mathbf{q} = -\mathbf{h}$. For incident electron energies above 1 keV the momentum transfer, or $\mathbf{q}$ integration in Eq. (41), ranges over an order of magnitude further than the low $\mathbf{h}$ vectors and hence the shift in the loss peak is relatively small. The result of this small shift in conjunction with the sharply peaked loss function is that to a good approximation, (for incident electron energies > a few hundred eV) $V^i_{\mathbf{h},\mathbf{h}} \approx V^i_{0,0}$ and from a similar argument $V^r_{\mathbf{h},\mathbf{h}} \approx V^r_{0,0}$.

To solve for the off-diagonal scattering-potential matrix components $V^i_{\mathbf{h},\mathbf{g}}$ for $\mathbf{h} \neq \mathbf{g}$, we cannot in general make use of the diagonal-isotropic dielectric-response approximation. We need to solve Eq. (36) and Eq. (37)

numerically in their general form. This is a computationaly demanding calculation that is not as yet practical. However, we can make some approximations in considering the terms $V^i_{0,\mathbf{h}}$, which are effectively the higher order Fourier coefficients of the inelastic scattering potential. For the centrosymmetric case we may write using the approximation $\epsilon^{-1}_{0,\mathbf{h}}(\mathbf{q}'+\mathbf{G},\omega)=\epsilon^{-1}_{\mathbf{G},\mathbf{G}+\mathbf{h}}(\mathbf{q}',\omega)$

$$V^i_{0,\mathbf{h}} \approx 4\pi e^2 \int_0^{E^0/\hbar} d\omega \int_{Q(\omega)} \frac{d\mathbf{q}}{(2\pi)^3} \frac{1}{|\mathbf{q}||\mathbf{q}+\mathbf{h}|} \times \mathrm{Im}[-\epsilon^{-1}_{0,\mathbf{h}}(\mathbf{q},\omega)] \times \delta\left(\omega - \mathbf{q}\cdot\mathbf{V} + \frac{\hbar q^2}{2m}\right) . \quad (49)$$

Furthermore, transforming to spherical polar coordinates and using the delta function relations of Eq. (44) and Eq. (45), we obtain

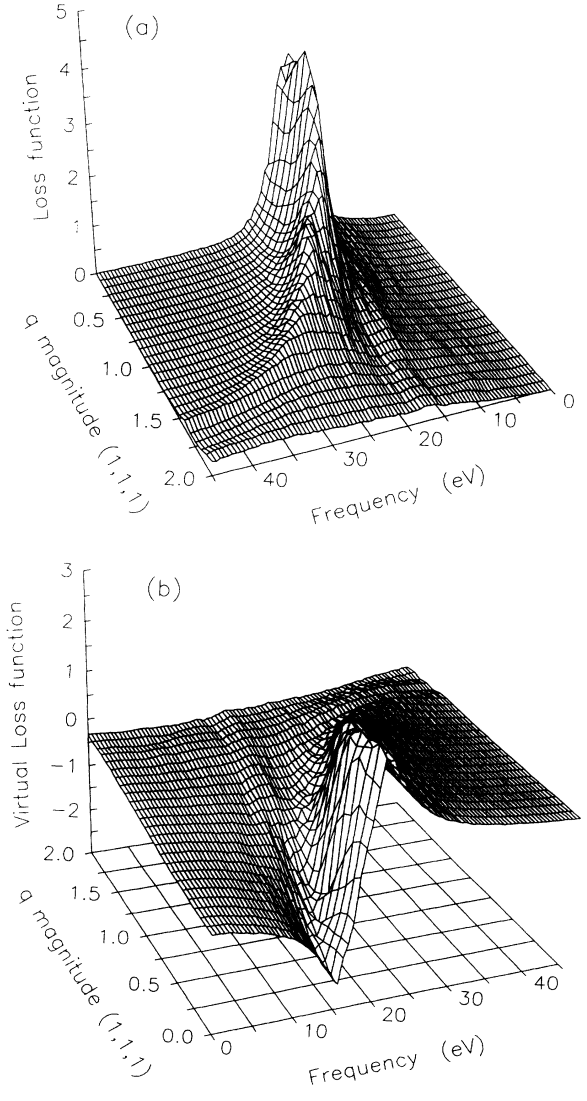

FIG. 1. (a) The loss function for Si as calculated in the (1,1,1) direction near the plasmon peak; and (b) the equivalent virtual-loss function for Si.

$$V^i_{0,\mathbf{h}} = \frac{e^2}{2\pi^2 V} \int_0^{E^0/\hbar} d\omega \int_{Q(\omega)} dq \int_{\theta_{\min}}^{\pi} d\theta \int_0^{2\pi} d\phi \frac{1}{|\mathbf{q}+\mathbf{h}|} \times \mathrm{Im}[-\epsilon^{-1}_{0,\mathbf{h}}(\mathbf{q},\omega)] \times \delta[\theta - G(\omega,q)] , \quad (50)$$

$$V^r_{0,\mathbf{h}} = \frac{e^2}{2\pi^2 V} \int_0^{E^0/\hbar} d\omega \int_{Q(\omega)} dq \int_{\theta_{\min}}^{\pi} d\theta \int_0^{2\pi} d\phi \frac{1}{|\mathbf{q}+\mathbf{h}|} \times \mathrm{Re}[-\epsilon^{-1}_{0,\mathbf{h}}(\mathbf{q},\omega)+1] \times \delta[\theta - G(\omega,q)] . \quad (51)$$

These expressions for the inelastic and virtual-inelastic scattering-potential matrix components, although simplified somewhat, are still numerically intensive calculations. For incident energies above a few hundred eV where the momentum transfer integration extends over very many reciprocal lattice vectors, we can say that these off-diagonal elements must be very small. This can be seen from physical arguments (the large delocalization of the interaction potential) and from calculations of the inverse dielectric matrix that show only small off-diagonal terms. We have previously found that these off-diagonal dielectric matrix terms are generally two orders of magnitude smaller than the diagonal terms. An indication that $V^i_{\mathbf{h},\mathbf{g}}$ for valence electron and plasmon excitations is likely to be orders of magnitude smaller than $V^i_{0,0}$, except at very low (< 500 eV) incident electron energies. This is further reinforced by both experiment[9,33] and our calculations in the next section that find the plasmon dispersion for Si to be only weakly anisotropic.

V. THE INELASTIC AND VIRTUAL SCATTERING POTENTIAL FOR SI

The loss function $\mathrm{Im}[-\epsilon^{-1}_{0,0}(\mathbf{q},\omega)]$ may be measured experimentally[9] by for instance EELS. A large peak in the loss function for Si [see Fig. 1(a) for the loss function calculated with $\mathbf{q}$ in the crystallographic $2\pi/a(1,1,1)$ direction] for small $|\mathbf{q}|$ near $\hbar\omega \approx 17$ eV, is a result of resonant interband transitions that may be interpreted as the plasmon excitation peak. This plasmon peak disperses close to quadratically for increasing $\mathbf{q}$ magnitude and at the same time decrease significantly in magnitude (an indication of the unsustainability of high momentum plasmons in the solid).

In Fig. 1(b), we show the equivalent "virtual-loss" function $\mathrm{Re}[-\epsilon^{-1}_{0,0}(\mathbf{q},\omega)+1]$ from Eq. (48), required to calculate the virtual MIP. The loss functions were calculated from Eq. (26) by inverting large (59×59) RPA dielectric matrices. These dielectric matricies in turn were calculated by summing over the 4 highest valence and 41 lowest conduction bands, and then many thousands of points[35] in the first Brillouin zone. The loss functions are numerically fully converged[28] and the detailed structure in these loss functions are due to the topology of the band structure we have used.

The inelastic MIP for Si has recently been calculated by the authors[19] for $\mathbf{q}$ in the crystallographic (1,0,0) direction, as a function of incident energies (10 eV–100 keV) and scattering angle θ. Results for both the inelas-

tic and the virtual-inelastic scattering MIP are shown in Fig. 2(a) (for $\theta_{min} \rightarrow 0$), calculated from Eq. (46) and Eq. (48). The results here are for **q** in the (1,1,1) direction. Excellent agreement is found with the experimental results[9,36–38] from 100 keV down to an incident energy as low as 10 eV. That the results agree with experiment so well at very low incident energies may perhaps seem fortuitous given the derivation of the general scattering equations in the weak scattering approximation and the neglect of exchange and correlation effects in the RPA.[9] However, the dielectric loss function itself (from which the MIP is obtained) is well known to be very accurate[19] over the range of energy and momentum transfers required for the MIP integrations. The good agreement is due to the fact that the calculated loss function models the experimental loss function so well at low energies.

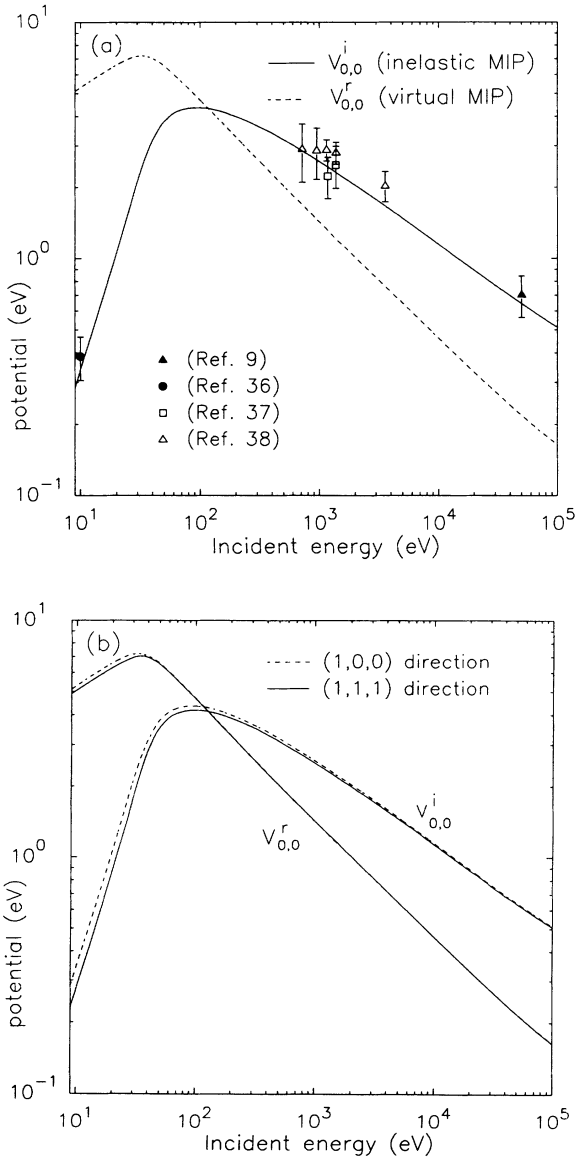

FIG. 2. (a) The calculated inelastic (solid line) and virtual (broken line) mean-inner potential for Si with q along (1,1,1). Experimental results for the inelastic MIP are as indicated. (b) The inelastic and virtual MIP for q along (1,0,0) (solid line) and (1,1,1) (broken line).

The dramatically different behavior of the two scattering potentials is evident. The virtual MIP, very large at low energies drops off markedly for increasing incident energies, and becomes small (negligible in comparison with the constant ≈14 eV elastic potential) in the high energy region. Below 1 keV virtual-inelastic scattering is quite significant, contributing an additional several eV (10%) to the elastic potential. The further fall in the virtual MIP below 30 eV is due to the decreasing number of possible interband excitations that are allowed. In contrast, the inelastic MIP increases rapidly from 10 eV (where it is less than 1/10 of the virtual MIP) as it approaches the plasmon excitation threshold, to a maximum above 50 eV (three times the plasmon energy peak of 16.9 eV). It then decreases slowly with increasing energy, in contrast to the more rapidly decreasing virtual-inelastic MIP.

In Fig. 2(b), we compare the inelastic and virtual-inelastic MIP's calculated by assuming dielectric isotropy with **q** in the (1,0,0) direction (dashed lines) and also the (1,1,1) direction (solid lines). It is clear that although evidence is found for a small dielectric anisotropy (and hence a nondiagonal inverse dielectric matrix) below 100 eV, this effect is negligible at higher incident energies.

We may conclude that accurate knowledge of the electronic structure of the solid is important for accurate calculations of the inelastic scattering-potential matrix below 1 keV. However, at higher incident energies only the diagonal components need be considered. The methods employed for the inelastic MIP calculations are described elsewhere,[28] and an analogous method was used here for the virtual MIP calculation. We note that great care has been taken to avoid numerical instabilities that arise in evaluating the inverse dielectric matrix that may yield inaccurate results.

VI. CONCLUSION

We have shown how the effects of inelastic and virtual-inelastic electron scattering in a crystalline environment, due to electronic valence-conduction electron excitations, may be related directly to the inverse dielectric-response matrix of the solid. The expressions for the complex inelastic scattering-potential matrix simplify to previous well-known approximations under certain conditions, such as dielectric isotropy or full excitation potential delocalization.

From a calculation of the inverse dielectric matrix in the random-phase approximation, based on accurate electronic band structure, we have calculated the diagonal elements of the inelastic scattering-potential matrix for Si as a function of incident electron energy. Excellent agreement with experiment was found in our calculations.

Calculations assuming dielectric isotropy in two different symmetry directions in reciprocal space show that the resulting inelastic scattering-potential matrix is highly isotropic, and hence very near diagonal (to several orders of magnitude) at incident energies above 1 keV. However,

at lower incident energies the diagonal inelastic scattering matrix becomes weakly anisotropic. This breakdown of the isotropic approximation in turn entails that the inelastic scattering-potential matrix becomes nondiagonal, and the full inelastic scattering-potential matrix must be calculated.

The band structure is responsible for the off-diagonal elements of the inelastic scattering-potential matrix and we have shown that these elements become significant below 1 keV. However, for higher incident energies we may to a very good approximation consider the inelastic scattering of electrons due to valence-conduction electron and plasmon excitations to be entirely delocalized, and hence the inelastic scattering-potential matrix to be diagonal. We have shown that accurate electronic band structure should be included in any calculation of the diagonal inelastic scattering-potential matrix to yield accurate results, particularly at low incident energies.

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