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Virtual scattering of high-energy electrons by plasmons and valence electrons in silicon

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Abstract

The contribution from plasmon and valence-electron scattering to the higher-order Fourier coefficients of the electron virtual scattering potential (EVSP) is shown to be non-negligible in contradiction to previous assumptions. In particular, calculations are made for this contribution to the EVSP $C^r_{0,\mathbf{g}}$(plasmon) for $\mathbf{g} = 111$ in silicon. In contrast to the elastic potential, $C^r_{0,\mathbf{g}}$(plasmon) is found to be highly energy dependent, varying by three orders of magnitude between 20 eV and 100 keV. Whilst it is found that the contribution from plasmon scattering is larger than that from tightly bound core states, the relative contribution to the total potential is smaller than that presently resolvable by electron experimental methods at 100 keV.

1. Introduction

It has long been thought that plasmon scattering does not give a contribution to the Fourier coefficients of the electron inelastic scattering potential (Humphreys & Hirsch, 1968; Radi, 1968*a*,*b*, 1970; Rez, 1978). However, recent work (Forsyth, Smith & Josefsson, 1997) has shown that the plasmon and valence-electron contribution to real inelastic scattering, $C^i_{0,\mathbf{g}}$(plasmon), is in fact larger than the corresponding core contribution from *K*-shell ionization, $C^i_{0,\mathbf{g}}(K$ shell), for $\mathbf{g} = 111$.

In addition to this 'real' inelastic scattering potential, there is a contribution to the elastic scattering potential from virtual scattering. In view of the non-zero size of the plasmon and valence-electron contribution to the inelastic potential, it would seem reasonable that the virtual scattering contribution to the elastic potential due to plasmons and valence electrons may also be larger than previously expected. We examine this in the present note, following on from the work of Rez (1978), which calculated the core contribution to virtual scattering.

As in previous work, our analysis is built on dielectric response theory using a non-local empirical pseudopotential band structure for silicon. Once the pseudopotential parameters are chosen (and these are fixed by the band structure), there are no adjustable parameters in the model. The model uses Bloch states rather than plane-wave states, allowing us to perform the first rigorous calculation of the higher-order Fourier coefficients of the plasmon and valence-electron contribution to the electron virtual scattering potential (EVSP). In this note, we examine in particular the contribution to the EVSP from virtual scattering for the 111 reflection in silicon, and show that it is also larger than the contribution from the tightly bound core states.

2. The electron virtual scattering potential

Virtual scattering refers to scattering from an initial state to an intermediate state, followed by further scattering back to the initial state, the net result contributing to the elastic scattering potential. On the basis of linear dielectric response theory, we may express the contribution to the EVSP from valence-electron and plasmon excitation in terms of the wave vector and frequency-dependent dielectric matrix $\varepsilon_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega)$ (see, for example, Josefsson & Smith, 1994) as

$$\begin{aligned} C^r_{0,\mathbf{g}}(\text{plasmon}) = {} & 4\pi e^2 \int_0^{E^0/\hbar} \mathrm{d}\omega \sum_{\mathbf{G}} \int_{\mathrm{BZ}} [\mathrm{d}\mathbf{q}'/(2\pi)^3] \\ & \times [-1/(2|\mathbf{q}'+\mathbf{G}||\mathbf{q}'+\mathbf{g}+\mathbf{G}|)] \\ & \times [\varepsilon^{-1}_{\mathbf{G},\mathbf{g}+\mathbf{G}}(\mathbf{q}',\omega) + \varepsilon^{*-1}_{\mathbf{g}+\mathbf{G},\mathbf{G}}(\mathbf{q}',\omega) - 2\delta_{0,\mathbf{g}}] \\ & \times \delta[\omega - (\mathbf{q}'+\mathbf{G})V + \hbar(\mathbf{q}'+\mathbf{G})^2 \end{aligned}$$

h, **g** and **G** are reciprocal-lattice vectors, $E^0 = \hbar\omega$ is the incident electron energy, V is the incident electron velocity and m is the electron mass. We note that $\varepsilon^{-1}_{\mathbf{0},\mathbf{g}}(\mathbf{q},\omega)$ is the $(\mathbf{0},\mathbf{g})$ element of the inverse of the entire dielectric matrix, containing all the elements $\varepsilon_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega)$.

For a centrosymmetric crystal structure such as silicon, assuming isotropic dielectric response and using the approximation $\varepsilon^{-1}_{\mathbf{0},\mathbf{g}}(\mathbf{q}'+\mathbf{G},\omega) = \varepsilon^{-1}_{\mathbf{G},\mathbf{G}+\mathbf{g}}(\mathbf{q}',\omega)$, we may express the higher-order Fourier coefficients of the EVSP in terms of the dielectric response as

$$\begin{aligned} C^r_{0,\mathbf{g}} = {} & (e^2/\pi V) \int_0^{E^0/\hbar} \mathrm{d}\omega \int_{q(\theta_{\min})}^{q(\pi)} (\mathrm{d}|\mathbf{q}|/|\mathbf{q}+\mathbf{g}|) \\ & \times \mathrm{Re}\{-\varepsilon^{-1}_{\mathbf{0},\mathbf{g}}(|\mathbf{q}|,\omega)\}. \end{aligned} \tag{2}$$

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

In this work, we adopt the random-phase approximation dielectric matrix obtained by Adler (1962) and Wiser (1963):

$$\begin{aligned} & \varepsilon_{\mathbf{h},\mathbf{g}}(\mathbf{q},\omega) \\ & = \delta_{\mathbf{g},\mathbf{h}} - (4\pi e^2/\Omega|\mathbf{q}+\mathbf{g}||\mathbf{q}+\mathbf{h}|) \\ & \quad \times \lim_{\alpha\to 0^+} \sum_{k,n,n'} \frac{f_0[E_{n'}(\mathbf{k}+\mathbf{q})] - f_0[E_n(\mathbf{k})]}{E_{n'}(\mathbf{k}+\mathbf{q}) - E_n(\mathbf{k}) + \hbar\omega + i\hbar\alpha} \\ & \quad \times \langle \mathbf{k}+\mathbf{q}, n' | \exp[i(\mathbf{q}+\mathbf{g})\cdot\mathbf{r}] | \mathbf{k}, n\rangle \\ & \quad \times \langle \mathbf{k}, n | \exp[-i(\mathbf{q}+\mathbf{h})\cdot\mathbf{r}] | \mathbf{k}+\mathbf{q}, n'\rangle, \end{aligned} \tag{3}$$

where Ω is the crystal volume, $f_0(E)$ is the Fermi function and n, n' are the band indices, labeling the Bloch states $|\mathbf{k}, n\rangle$ of energy $E_n(\mathbf{k})$ in the solid. A nonlocal empirical pseudopotential based on the work of Cohen & Chelikowsky (1989) is used to obtain the electronic states in (3).

3. Results

Details of the method of calculation are given in Forsyth, Josefsson & Smith (1996) and, for the sake of brevity, will not be discussed here. Fig. 1 shows the higher-order virtual dielectric loss function $\mathrm{Re}\{\varepsilon^{-1}_{0,\mathbf{g}}(\mathbf{q},\omega)\}$ for $\mathbf{g} = 111$. The actual mesh used for the integration in (2) is much finer than this, and extends to 100 eV.

Fig. 2 shows the magnitude of the Fourier coefficients $C^r_{0,\mathbf{g}}$(plasmon) for $\mathbf{g} = 111$ and $C^r_{0,0}$(plasmon) as functions of incident energy. $C^r_{0,\mathbf{g}}$, $\mathbf{g} = 111$, has the opposite sign to $C^i_{0,0}$. This is a geometrical effect, and is expected for a crystal with zinc-blende symmetry. Previous work (Rez, 1978) at 100 keV gives values for the contribution to the potential from virtual inelastic scattering in silicon for $\mathbf{g} = 111$ ranging from 7.6×10^{-5} to 2.2×10^{-4} eV. Note that these values included only single-particle excitations from tightly bound core states and ignored the contribution from plasmon excitation. The present work calculates the contribution from plasmon and valence-electron excitation *only*. According to previous work (Rez 1978), this value should be zero, however, we obtain a value of 3.0×10^{-4} eV for an incident energy of 100 keV, which is actually larger than the largest estimate (Rez, 1978) made for the core-excitation contribution to the EVSP.

However, even taking the largest value for the core contribution as calculated by Rez (1978) and adding it to the value for the plasmon and valence contribution as calculated here for 100 keV, we obtain a total potential of $C^r_{0,\mathbf{g}} = 5.2 \times 10^{-4}$ eV. A Dirac–Fock calculation of the elastic scattering potential using the parameters of Waasmaier & Kirfel (1995) gives a value for $V_{\mathbf{g}}$ of 5.48 eV at 100 keV for $\mathbf{g} = 111$. When compared with the elastic potential, the relative contribution $C^r_{0,\mathbf{g}}/V_{\mathbf{g}}$ is only 9.5×10^{-5}, while estimated experimental relative errors (Hewat & Humphreys, 1974) are 4×10^{-3}. Note that in contrast to the elastic potential (which is dependent on energy principally *via* the standard relativistic correction), $C^r_{0,\mathbf{g}}$ is highly energy dependent, and may be significant at low energies.

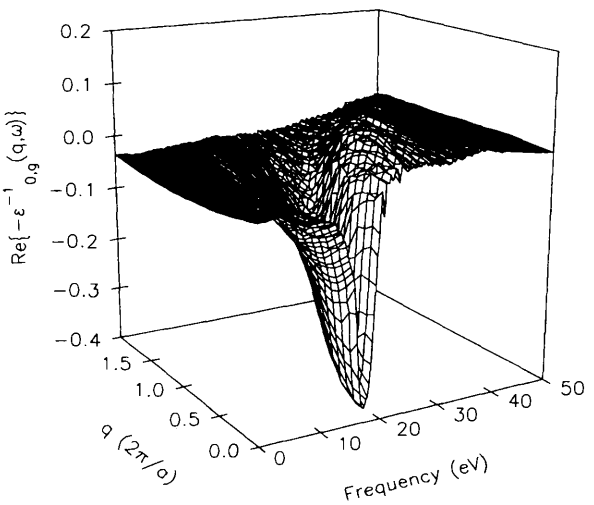

Fig. 1. The higher-order virtual loss function $\mathrm{Re}\{-\varepsilon^{-1}_{0,\mathbf{g}}(|\mathbf{q}|,\omega)\}$, $\mathbf{g} = 111$, for silicon as a function of wave vector and frequency.

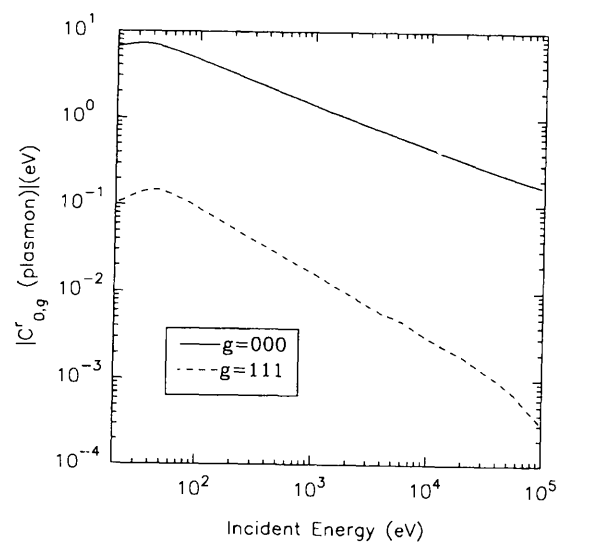

Fig. 2. Variation of $|C^r_{0,\mathbf{g}}(\text{plasmon})|$ with incident electron energy for $\mathbf{g} = 111$ and $\mathbf{g} = 000$. Note that $C^r_{0,0}$ is the virtual mean inner potential of Josefsson & Smith (1994). $C^r_{0,\mathbf{g}}$, $\mathbf{g} = 111$, has the opposite sign to $C^i_{0,0}$.

4. Conclusions

In conclusion, this work presents the first rigorous calculation of the $\mathbf{g} = 111$ Fourier coefficient of the contribution to the electron inelastic scattering potential due to virtual inelastic scattering by plasmon and valence-electron excitations in silicon. This is based on a nonlocal electronic band-structure-dependent dielectric matrix calculation. Previous work (Humphreys & Hirsch, 1968; Radi, 1968*a*,*b*, 1970; Rez, 1978; Whelan, 1965; Josefsson & Smith, 1993, 1994) has concentrated on the virtual inelastic mean inner potential $C^r_{0,0}$(plasmon), believing that the higher-order Fourier coefficients $C^r_{0,\mathbf{g}}$(plasmon) would be negligible. At 100 keV, it appears that the relative contribution to the electron scattering potential from virtual inelastic scattering due to the $\mathbf{g} = 111$ reflection is still smaller than the relative experimental error, even with the plasmon and single-particle excitation contribution included.

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References

Adler, S. (1962). *Phys. Rev.* **126**, 413–420.

Cohen, M. L. & Chelikowsky, J. R. (1989). *Electronic Structure and Optical Properties of Semiconductors*, 2nd ed. Berlin: Springer-Verlag.

Forsyth, A. J., Josefsson, T. W. & Smith, A. E. (1996). *Phys. Rev. B*, **54**, 14355–14361.

Forsyth, A. J., Smith, A. E. & Josefsson, T. W. (1997). *Phys. Rev. B.* Submitted.

Hewat, E. A. & Humphreys, C. J. (1974). *High Voltage Electron Microscopy*, edited by P. R. Swann, C. J. Humphreys & M. J. Goringe, pp. 52–56. London/New York: Academic Press.

Humphreys, C. J. & Hirsch, P. B. (1968). *Philos. Mag.* **18**, 115–122.
Josefsson, T. W. & Smith, A. E. (1993). *Phys. Lett.* A**180**, 174–178.
Josefsson, T. W. & Smith, A. E. (1994). *Phys. Rev. B*, **50**, 7322–7330.
Radi, G. (1968*a*). *Z. Phys.* **212**, 146–168.
Radi, G. (1968*b*). *Z. Phys.* **213**, 244–257.
Radi, G. (1970). *Acta Cryst.* A**26**, 41–56.
Rez, P. (1978). *Acta Cryst.* A**34**, 48–51.
Waasmaier, D. & Kirfel, A. (1995). *Acta Cryst.* A**51**, 416–431.
Whelan, M. J. (1965). *J. Appl. Phys.* **36**, 2099–2103.
Wiser, N. (1963). *Phys. Rev.* **129**, 62–69.