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LASER EXCITATION OF SURFACE ELECTRONIC STATES FOR A ONE-DIMENSIONAL ETC(U)

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Laser Excitation of Surface Electronic
States for a One-Dimensional Semiconductor.

by

William C. Murphy and Thomas F. George

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Surface Science

University of Rochester
Department of Chemistry
Rochester, New York 14627

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Laser Excitation of Surface Electronic
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Abstract

The electronic band structure of a finite one-dimensional semiconductor is examined. The surface states are identified and the absorption cross-section for transitions to these states is calculated. The use of a laser to enhance surface charge density is discussed.

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1. Introduction

Recently, there has been much interest in the effects of laser radiation in solids.¹ Using experimental electronic band structures and time dependent perturbation theory, researchers have calculated the photon absorption rate for the electronic states in a variety of semiconductors. Much effort has also been devoted to the study of the effects of laser radiation on the phonons in solid surfaces.² However, the effect of a surface on photon-excited electrons has not been examined.

Studies³ of the effects of synchrotron radiation on metal surfaces have shown that photon-stimulated desorption can occur through electronic excitation. The details⁴ of this desorption process are involved, but the ultimate reason for the desorption is the shift of electronic charge and the resultant Coulombic repulsion.

For a semiconductor, the bulk electronic band structure consists of a number of valence and conduction bands. In addition to these bulk bands, a surface can introduce additional bands and local states.⁵ These surface bands correspond to a charge density localized in the surface region. Consequently, by using a laser to excite electrons to or from the surface band, we can effectively control the surface charge and, ultimately, the Coulombic repulsion or attraction of the surface for adsorbed species.

In the following section we will examine the effects of a surface on the band structure of a model system. Laser excitation of valence electrons to surface states in this model system will then be analyzed in the next two sections. Finally, a discussion

of the applications and limitations of this model for studying laser induced surface processes will be presented.

2. Model Band Structure

In an infinite crystalline solid, the energies of the electrons divide into bands of allowed states separated by energy gaps. For a finite crystal thin film calculations⁶ have demonstrated that energy states exist in these gaps. Furthermore, the charge associated with these states resides mostly in the surface region.

Heine⁷ and, later, Lundqvist⁵ have shown that truncating a linear chain produces similar effects on the energy gap. For this simple one-dimensional model, the dispersion relationship is given in Figure 1. In a semiconductor, the lower inverted-U curve would be the bulk valence band and the upper U-shaped curve would be the bulk conduction band. The curve in complex crystal momentum space is associated with the surface states. If we have a chain with lattice constant a parallel to the z axis in the region $z < a/2$, the interior wave function associated with these gap states is that of a damped oscillator:

$$\psi_{\kappa}(z) = C_s e^{-\kappa z} \cos\left(\frac{gz}{2} + \phi\right) \quad (1)$$

with corresponding energy

$$E(\kappa) = \left(\frac{g}{2}\right)^2 - \kappa^2 \pm (v_g^2 - g^2 \kappa^2)^{1/2} \quad (2)$$

where C_s is the normalization constant; κ is the imaginary part of the crystal momentum and ranges from 0 at the edges of the

gap to $-|V_g/g|$ near the center; g is the reciprocal lattice constant and equals $2\pi/a$; and V_g is the g component of the Fourier transform of the effective potential. The phase factor ϕ is given by

$$\sin 2\phi = \frac{g\kappa}{V_g} . \quad (3)$$

The interior bulk states would have the form

$$\psi_k(z) = C[e^{ikz} + \frac{E(k)-k^2}{V_g} e^{i(k-g)z}] \quad (4)$$

with corresponding energy

$$E(k) = \frac{1}{2}[k^2 + (k-g)^2 \pm ([k^2 - (k-g)^2]^2 + 4|V_g|^2)^{1/2}] . \quad (5)$$

The interior solution for both the surface states, equation (1), and the bulk states, equation (4), should be matched to the exterior solution of the form

$$\psi(z) = C_s e^{-qz} \quad (6)$$

with

$$q = \sqrt{V_0 - E(\kappa)} , \quad (7)$$

where V_0 is the work function plus the energy at the top of the valence band. For bulk states, C_s in equation (6) and $E(\kappa)$ in equation (7) should be replaced by C and $E(k)$, respectively.

To obtain the normalization constants, we assume the chain to be of length $L = Na$ and the charge density was symmetric about the center. Under these conditions we obtain

$$C = \{ [1 + (\frac{E(k)-k^2}{V_g})^2] L + \frac{e^{-qa}}{q} \}^{-\frac{1}{2}} \quad (8)$$

for the bulk states and

$$C_s = [e^{-\kappa a} \{ \frac{2\kappa \cos 2\phi - g \sin 2\phi}{(2\kappa)^2 + g^2} [1 + e^{\kappa L} (-1)^{N-1}] + \frac{[e^{\kappa L} - 1]}{2\kappa} + \frac{e^{-qa}}{q}]^{-\frac{1}{2}} \quad (9)$$

for the surface states.

In the following sections, we wish to examine laser excitation from the bulk valence band, equation (4), to the surface band, equation (1). The initial conditions are assumed to be a filled valence band, but empty surface and conduction bands. Since equation (4) represents electrons delocalized throughout the chain and equation (1) corresponds to electrons in the surface region, the excitation of electrons to the surface states will increase the surface charge.

3. Selection Rule

To determine the transition rate, we must evaluate an integral of the form

$$M \equiv \langle \kappa | \vec{A} \cdot \vec{p} | k \rangle, \quad (10)$$

where $\vec{A}$ is the vector potential of the laser radiation and $\vec{p}$ is the momentum operator of the electron. Under the dipole approximation and assuming the laser is polarized parallel to the chain, M becomes

$$M = -i \left(\frac{2\pi I}{137} \right)^{1/2} \frac{e^{i\omega t}}{\omega} \langle \kappa | \frac{d}{dz} | k \rangle, \quad (11)$$

where I is the laser intensity and ω is the angular frequency.

The wave functions of interest, equations (1) and (4), can be written in the form

$$\psi_k(z) = e^{-ikz} u_k(z) \quad (12)$$

$$\psi_k(z) = e^{-i(\frac{g}{2})z} e^{\kappa z} u_k(z), \quad (13)$$

where the functions $u_k(z)$ and $u_{-k}(z)$ have the periodicity of the lattice. The integral in equation (8) now becomes

$$M' \equiv \langle \kappa | \frac{d}{dz} | k \rangle = 2 \int_{\frac{a}{2} - \frac{L}{2}}^{\frac{a}{2}} e^{-i(k - \frac{g}{2})z} e^{\kappa z} [u_k(z) u'_k(z) - i u_k(z) u_k(z)]$$

$$= \sum_{\ell=0}^{N-1} \int_{-\frac{a}{2}(1+2\ell)}^{\frac{a}{2}(1-2\ell)} e^{-i(k - \frac{g}{2})z} e^{\kappa z} [u_k(z) u'_k(z) - i u_k(z) u_k(z)], \quad (14)$$

where we have assumed that for large L the contribution of the exponential tail, equation (6), to the integral will be negligible. If we change the variable of integration to $z' = z + \ell a$ and exploit the periodicity of $u_k(z)$ and $u_{-k}(z)$, we obtain

$$M' = \sum_{\ell=0}^{N-1} e^{i(k - \frac{a}{2})\ell a} e^{\kappa \ell a} \int_{-\frac{a}{2}}^{\frac{a}{2}} e^{-i(k - \frac{g}{2})z'} e^{\kappa z'} [u_k(z') u'_k(z') - i u_k(z') u_k(z')]$$

$$= \sum_{\ell=0}^{N-1} e^{i(k - \frac{a}{2})\ell a} e^{\kappa \ell a} \langle \kappa | \frac{d}{dz} | k \rangle_o, \quad (15)$$

where the subscript "o" indicates integration over the first unit cell. The transition probability is related to the square of

equation (15):

$$M'^2 = \sum_{m=0}^{N-1} \sum_{\ell=0}^{N-1} e^{i(k-\frac{g}{2})(\ell-m)a} e^{\kappa(\ell+m)a} \langle \kappa | \frac{d}{dz} | k \rangle_0. \quad (16)$$

If we change the range of summation, the sums in front of the integral become

$$S = e^{\kappa Na} \sum_{m=-\frac{N-1}{2}}^{\frac{N-1}{2}} \sum_{\ell=-\frac{N-1}{2}}^{\frac{N-1}{2}} e^{i(k-\frac{g}{2})(\ell-m)a} e^{\kappa(\ell+m)a}. \quad (17)$$

Changing the summation variable to $s = \ell + m$ and $d = \ell - m$, we get

$$S = e^{\kappa Na} \sum_{d=1-N}^{N-1} e^{i(k-\frac{g}{2})da} \sum_{s=1-N+d}^{N-1-d} e^{\kappa sa} \quad (18)$$

where the prime signifies a count with an increment of 2. If N is very large, the sum over s will only be significantly affected by large values of d . Therefore, for large N , we can approximate this function by

$$S \approx e^{\kappa Na} \sum_{d=-(N-1)}^{N-1} e^{i(k-\frac{g}{2})da} \sum_{s=-(N-1)}^{(N-1)} e^{\kappa sa}. \quad (19)$$

Taking the limit as N goes to infinity, the function takes the form

$$S \approx \frac{2\pi}{a} \delta(k-\frac{g}{2}) \frac{1}{1-e^{2\kappa a}}, \quad (20)$$

where $\delta(k-\frac{g}{2})$ is the Dirac delta function. Equation (16) can now be written

$$M'^2 \approx \frac{2\pi}{a} \delta(k-\frac{g}{2}) \frac{|\langle \kappa | \frac{d}{dz} | \frac{g}{2} \rangle_0|^2}{1 - e^{2\kappa a}}. \quad (21)$$

Consequently, the transition from a bulk to a surface state is

only permitted if the real part of the crystal momentum remains unchanged. This relationship is not too surprising since it is an exact restriction on laser-induced transitions between bulk bands.⁸ Furthermore, for our model, it confines us to the top of the valence band, where the density of states is a maximum (infinite) and the laser frequency needed for a transition is a minimum.

4. Transitions

To first order, the transition rate from a valence band to a surface band is

$$T = 2\pi \left(\frac{L}{\pi}\right) \left(\frac{2g}{V_g}\right) \int_0^g dk \int_0^g d\kappa \left| \langle \kappa | \vec{A} \cdot \vec{p} | k \rangle \right|^2 \delta(E(\kappa) - E(k) - \omega). \quad (22)$$

Using equations (11) and (21), this can be simplified to

$$T = \left(\frac{8\pi}{137}\right) \left(\frac{I g^2 L}{V_g \omega^2}\right) \int_0^g dk \frac{\left| \langle \kappa | \frac{d}{dz} \left| \frac{g}{2} \right\rangle_0 \right|^2}{1 - e^{2\kappa a}} \delta(E(\kappa) - E(k) - \omega). \quad (23)$$

After evaluating the integral over κ , the expression reduces to

$$T = \left(\frac{8\pi}{137}\right) \left(\frac{I g^2 L}{V_g \omega^2}\right) \frac{\left| \langle \kappa | \frac{d}{dz} \left| \frac{g}{2} \right\rangle_0 \right|^2}{1 - e^{2\kappa a}} \left| \frac{d\kappa}{dE(\kappa)} \right| \quad (24)$$

where κ in equation (24) refers to the state obeying the resonant condition:

$$\omega = V_g - \kappa^2 \pm (V_g^2 - g^2 \kappa^2)^{1/2}. \quad (25)$$

We now define the absorption cross-section, σ :

$$\sigma \equiv \frac{\omega T}{I}. \quad (26)$$

Using equations (1), (4), (8) and (9) in equation (24) and taking the limit as L goes to infinity, the cross-section becomes

$$\sigma = \frac{\left(\frac{\pi}{137}\right) \left(\frac{g^6}{V_g \omega \kappa^2}\right) \left(\frac{1-e^{\kappa a}}{1+e^{\kappa a}}\right) \left(\frac{\kappa \sin \phi + g \cos \phi}{\kappa^2 + g^2}\right)^2 \left|\frac{d\kappa}{dE(\kappa)}\right|}{\left(\frac{2\kappa \cos 2\phi - g \sin 2\phi}{(2\kappa)^2 + g^2} - \frac{1}{2\kappa}\right)} \quad (27)$$

With equation (2), we can also readily evaluate the derivative:

$$\left|\frac{d\kappa}{dE(\kappa)}\right| = \left|\left(\frac{1}{\kappa}\right) \frac{\sqrt{V_g^2 - g^2 \kappa^2}}{2\sqrt{V_g^2 - g^2 \kappa^2} \pm g^2}\right|. \quad (28)$$

Equations (27) and (28) constitute the cross-section for electronic transitions from the valence band to the surface band. Although this cross-section is quite complicated, we can readily deduce its behavior by analyzing the expressions at various limits.

If the exciting laser radiation is at the frequency near $\frac{1}{2} E_g$ where $\kappa = -|V_g/g|$, equation (28) will vanish, and thus

$$\sigma_{\omega \approx \frac{1}{2} E_g} = 0. \quad (29)$$

This is exactly what one would expect since this mid-gap energy is a branch point at which no surface states exist.

If the laser radiation is near a frequency of 0 or E_g , the cross-section becomes

$$\sigma_{\omega \rightarrow 0} = \left(\frac{\pi}{137}\right) \left(\frac{ag^2}{E_g}\right) \left(\frac{E_g + g^2}{E_g - g^2}\right)^2 \left|\frac{1}{\kappa}\right| \quad (30)$$

and

$$\sigma_{\omega \rightarrow E_g} = \left(\frac{\pi}{137}\right) \left(\frac{ag^4}{E_g [E_g + g^2]}\right) \left|\frac{1}{\kappa}\right|. \quad (31)$$

At both extremes, $\kappa \rightarrow 0$ and equations (30) and (31) diverge. This occurs because at the surface band edges the charge associated with the surface states becomes more and more delocalized throughout the lattice until at $\kappa = 0$, the charge is completely delocalized. At this point the surface states become bulk states, and instead of cross-sections, one should consider absorption coefficients.

Figure 2 illustrates the behavior of the cross-section over the entire frequency range. The values⁹ for the lattice constant, a , and the energy gap, E_g , were taken to be those of silicon.

5. Discussion

As pointed out in the Introduction, we are interested in using a laser to increase the surface charge density. Since the effective charge depth is $|1/(2\kappa)|$ (see equation (1)), we wish to excite states for large values of κ . For silicon at maximum κ , the surface charge depth is 1.39 lattice constants (1.39 a); consequently, the charge is confined to a region very near the surface. As we move away from this mid-gap region the charge depth increases as mentioned in the last section. Therefore we are mostly interested in laser frequencies near $\frac{1}{2}E_g$. As seen in figure 2, the cross-section is quite substantial near the mid-gap region and, subsequently, we could readily increase the surface charge by using a laser of moderate intensity. Even at $\omega = .15 E_g$ and $\omega = .85 E_g$, where

the cross-section has increased substantially, the charge depth is still only about 2 lattice constants. Consequently, we would expect a laser tuned to a frequency around $\frac{1}{2}E_g$ to be an effective controller of surface charge. Furthermore, since the energy gaps for most semiconductors are approximately 1 eV (for silicon, $E_g = 1.17$ eV), the frequency of interest will be in the infra-red region.

Since these results are based on a one-dimensional model, certain limitations should be pointed out. First of all, most common semiconductors have indirect band-gaps (the minimum in the conduction band is not over the maximum in the valence band).⁹ To excite states in these gaps would require phonon excitation in addition to electronic excitation. Furthermore, the form of the wave function in the indirect gap is not readily obtainable from a simple model.

Secondly, the various possible planes of a three-dimensional surface can lead to surface states between some bulk bands and not others. The surface charge will also be delocalized over the surface plane, in contrast to only on the other end atoms of a linear chain.

Finally, the effect of such phenomenon as surface relaxation and reconstruction are not included in our one-dimensional model. Surface states can also be modified by the presence of faults and adspecies.

Some of these problems, including effects of higher dimensions and the existence of adspecies, are the subject of continuing research. Dynamical processes such as laser-stimulated

desorption and adsorption are under consideration.

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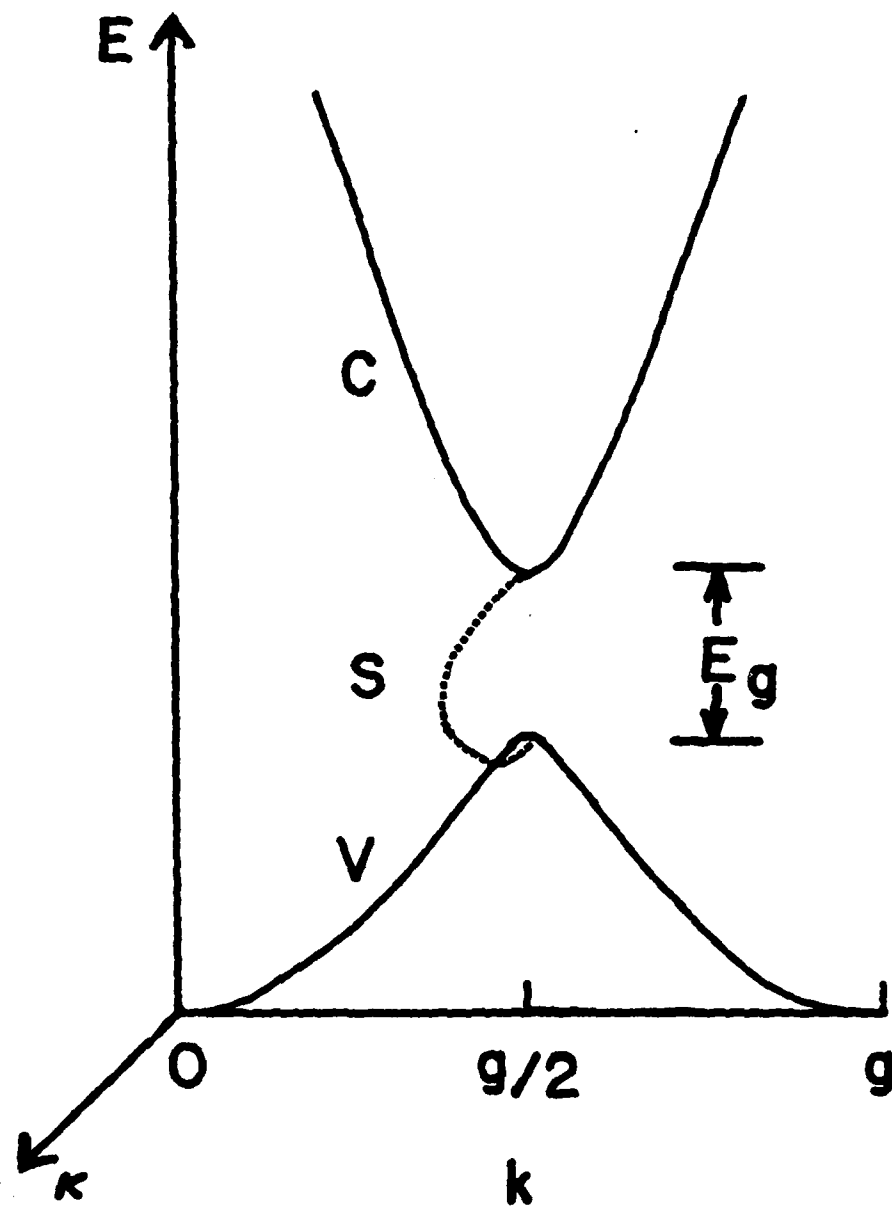
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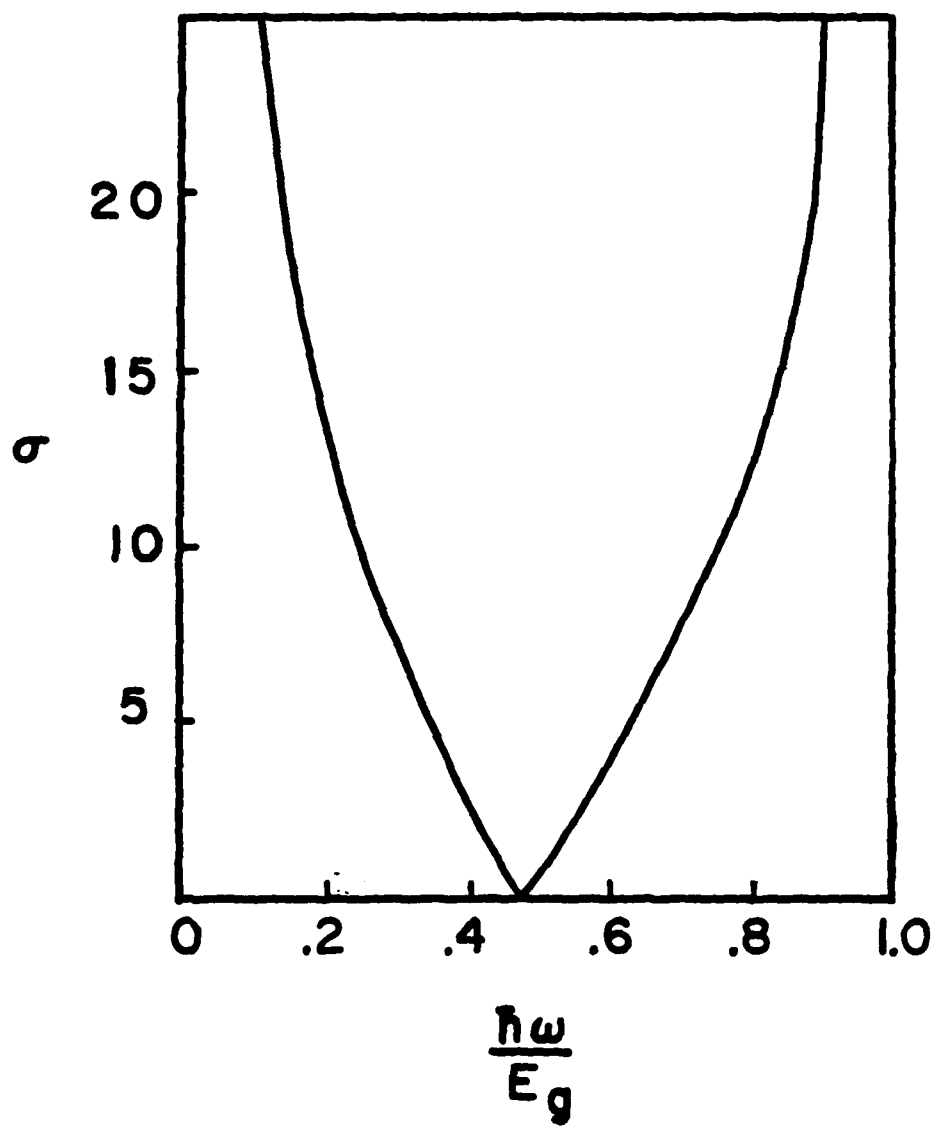
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Figure 1. Dispersion relationship in complex crystal momentum space ($k+ik$) for a finite linear chain. The valence, surface and conduction bands are labeled V, S and C, respectively.

Figure 2. Absorption cross-section for surface states, σ , in A^2 versus the frequency of the exciting laser radiation.





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