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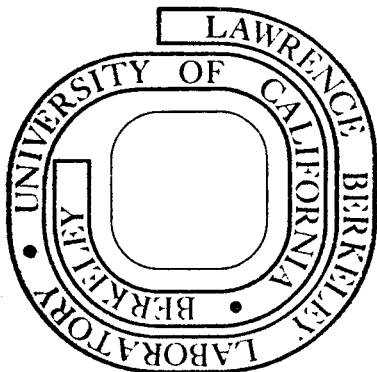
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New Insight into the Optical Properties of Amorphous Ge and Si*

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Abstract

A short range disorder model, unlike present long range disorder theories, is able to account well for both the density of states and the optical properties of amorphous Ge and Si. Our results indicate that the imaginary part of the dielectric function for amorphous Ge and Si has the same form as an averaged gradient matrix element as a function of energy. This conclusion should be valid for all tetrahedrally bonded amorphous solids.

The imaginary part of the dielectric function $\epsilon_2(E)$ and the density of states $n(E)$ for amorphous Ge and Si obtained from experiment¹⁻³ exhibit properties which are in many cases quite novel and cannot be obtained from a simple averaging of the crystalline spectra.

The theoretical attempts⁴⁻⁹ to explain the amorphous optical data have all assumed, until now, that long range disorder (LRD) is of primary importance. They have taken the simple crystalline band structure as a

starting point and have applied various modifications to introduce the effect of LRD. In some cases^{4,8} complete \underline{k} non-conservation was considered in the sense of a non-direct transition model and in other cases^{5-7,9} partial \underline{k} non-conservation was proposed which enabled the introduction of some type of short range order parameter. Although all these theories give good agreement with the amorphous $\epsilon_2(E)$ data, none of these theories predicts the correct trend to $n(E)$ for the amorphous case. This problem was discussed in detail in a previous paper¹⁰ where we studied $n(E)$ for Ge and Si in the diamond (FC-2), wurtzite (2H-4), Si III (BC-8) and Ge III (ST-12) structures using the Empirical Pseudopotential Method (EPM). The trends observed with the increasing complexity of the crystal structures suggested that the amorphous $n(E)$ results could be explained by a short range disorder (SRD) model defined as a system with a connected network of bonds but with deviations in the bond angles and odd numbered rings of bonds. It is precisely the lack of this SRD that brings about inconsistencies in the predictions of present LRD theories.

The results of the aforementioned LRD theories lead us to suspect that the $\epsilon_2(E)$ function may not be a good judge of the microscopic structural aspects of the amorphous state and that one definitely needs a theory that will be able to account for both $n(E)$ and $\epsilon_2(E)$ of the amorphous phase.

In this letter we shall show that a SRD model can indeed also account for the amorphous $\epsilon_2(E)$. In particular we shall find that the amorphous $\epsilon_2(E)$ spectrum should have the same form as an averaged gradient matrix element as a function of energy. This is quite interesting since the amorphous

$\epsilon_2(E)$ is not well understood.

There are two features of the amorphous $\epsilon_2(E)$ spectrum which must be explained. First we must ask why we have only one hump in the spectrum and secondly what determines the position in energy of this hump. We shall attempt to account for these features in the following analysis. The crystalline $\epsilon_2(E)$ can be written as

$$\epsilon_2(E) = C \cdot J(E) \cdot \sum_{\mathbf{k}} \sum_{c,v} \delta(E_c(\mathbf{k}) - E_v(\mathbf{k}) - E) |\langle \psi_c(\mathbf{k}) | \vec{r} | \psi_v(\mathbf{k}) \rangle|^2 / J(E) \quad (1)$$

where C is a constant, the sum of \mathbf{k} is over the whole Brillouin Zone (BZ), c and v represent conduction and valence band states respectively, $\psi(\mathbf{k})$ is a Bloch state, and $J(E)$ is the joint density of states given by:

$$J(E) = \sum_{\mathbf{k}} \sum_{c,v} \delta(E_c(\mathbf{k}) - E_v(\mathbf{k}) - E) \quad (2)$$

Equation (1) is just an expression for an averaged dipole matrix element $P(E)$ multiplied by the joint density of states $J(E)$. If we now incorporate the constant C into $J(E)$ we can write:

$$\epsilon_2(E) = J(E) \cdot P(E) \quad (3)$$

This is a physically reasonable expression and could be used to study the amorphous phase, since it is essentially the number of states accessible for transitions at an energy E , multiplied by an average probability for those transitions. When one does band structure calculations, however, it is easier to calculate an associated average matrix element $M(E)$ obtained by

a weighted averaging of $|\langle \psi_c(\underline{k}) | \vec{\nabla} | \psi_v(\underline{k}) \rangle|^2$. Then Eq. (3) can be written as:

$$\epsilon_2(E) = J(E) \cdot \frac{M(E)}{E^2} \quad (4)$$

Eq. (3), or equivalently Eq. (4), can now be used to make a model of the amorphous $\epsilon_2(E)$ by speculating the form of $J(E)$ and $P(E)$ in the amorphous phase. In this phase we would almost certainly expect $J(E)$ to be a smooth monotonically increasing function of energy, at least up to 6 eV, without any sharp structure from specific localized regions in the BZ. Similarly, we would expect the average dipole matrix element $P(E)$ to be a smooth monotonically decreasing function of energy for the most part. This expectation is quite different from the suggestion of Maschke and Thomas⁸ that the averaged gradient matrix element $M(E)$ should be a decreasing function of energy. It is much safer to expect that $P(E)$ is a decreasing function of energy since it is proportional to the probability of a transition at an energy E and must satisfy a more stringent sum rule than $M(E)$.

The product of $P(E)$ and $J(E)$ would then give a one hump structure as a function of energy so that the shape of the amorphous $\epsilon_2(E)$ comes out of this simple model quite naturally. To examine this in more detail, we have calculated $\epsilon_2(E)$, $J(E)/E^2$ and $M(E)$, and $J(E)$ and $M(E)/E^2$ as functions of energy for Ge and Si in the FC-2, 2H-4, BC-8, and ST-12 structures using the EPM and the Gilat-Raubenheimer integration scheme.¹¹ The results for Si are shown in Fig. 1 and are similar to those for Ge which will be presented in a more detailed paper.¹² For each row the product of the two curves in the second and third columns gives the $\epsilon_2(E)$ spectrum in the first column.

In the cases of the 2H-4 and ST-12 structures we show the weighted average of the parallel and perpendicular components of $\epsilon_2(E)$. We are interested in observing trends as we go from FC-2 down the columns to more and more locally disordered and complicated crystal structures. For the moment let us concentrate on the third column in the figure. We notice that with the increasing complexity of the crystal structures, $J(E)$ gradually loses the sharp structure prominent in the FC-2 case which was caused by the simplicity and symmetry of this band structure. When we reach ST-12, $J(E)$ is almost a smooth and featureless spectrum which would compare well with what we expected for the amorphous case. In addition the average dipole matrix element $M(E)/E^2$ for ST-12 is for the most part a smooth decreasing function of energy apart from some small wiggles at low energy which are not in the energy region of the peak in $\epsilon_2(E)$. If we now examine the $\epsilon_2(E)$ spectra we notice that it is precisely the ST-12 structure that has the qualities of the superimposed amorphous $\epsilon_2(E)$ spectrum obtained by Pierce and Spicer². The agreement between the ST-12 spectra and the amorphous spectra is quite encouraging and shows that the kind of SRD which accounted for the amorphous $n(E)$ also accounts for the important features of the amorphous $\epsilon_2(E)$ spectrum. The discrepancy in magnitude of the $\epsilon_2(E)$ curves is irrelevant in this discussion and is caused in part by the differences in bulk density of the ST-12 and amorphous structures.

An interesting feature that comes out of this analysis is that $J(E)/E^2$ should look something like a step function in the amorphous case since $J(E)$ is such a smooth polynomial-like increasing function of energy. This then

suggests that the average gradient matrix element $M(E)$ must contain most of the information about $\epsilon_2(E)$. This is shown in Fig. 1 as we go down the second column where we have plotted $J(E)/E^2$ and $M(E)$. In the FC-2 case the $\epsilon_2(E)$ spectrum looks mostly like $J(E)/E^2$ while $M(E)$ modulates the $J(E)/E^2$ spectrum. In the 2H-4 structure we find that the form of the $\epsilon_2(E)$ spectrum is now shared between $J(E)/E^2$ and $M(E)$, where $M(E)$ contributes most of the first peak and $J(E)/E^2$ contributes the second peak. When we examine the BC-8 case we find that the $\epsilon_2(E)$ spectrum now looks mostly like $M(E)$ while $J(E)/E^2$ just modulates the $M(E)$ spectrum. Finally in the ST-12 structure we find that $J(E)/E^2$ is a relatively featureless step-like function of energy and again $\epsilon_2(E)$ looks like $M(E)$.

Therefore we can safely conclude from this that the average gradient matrix element $M(E)$ determines the position in energy of the hump in the amorphous $\epsilon_2(E)$, and most important, when one measures the amorphous $\epsilon_2(E)$ spectrum one is essentially just measuring the average matrix element $M(E)$.

This result should be applicable to all tetrahedrally bonded amorphous solids since the forms of $J(E)/E^2$ and $M(E)$ are similar in all these cases.

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Figure Caption

Fig. 1. Imaginary part of the dielectric function ϵ_2 , average gradient matrix element M , associated joint density of states J/E^2 , average dipole matrix element M/E^2 , and joint density of states J for Si in the FC-2, 2H-4, BC-8, and ST-12 structures. For each row the product of the two curves in the second and third columns gives the ϵ_2 spectrum in the first column. The ϵ_2 for the 2H-4 and ST-12 structures was obtained by averaging over parallel and perpendicular polarizations. The matrix element M is in units of $\left(\frac{2\pi}{a}\right)^2$ where a is the smallest lattice constant of each crystal and J in the figure is in units of $\left(\frac{a}{2\pi}\right)^2 (\text{eV})^2$. The amorphous ϵ_2 curve was obtained from Pierce and Spicer (Ref. 2).

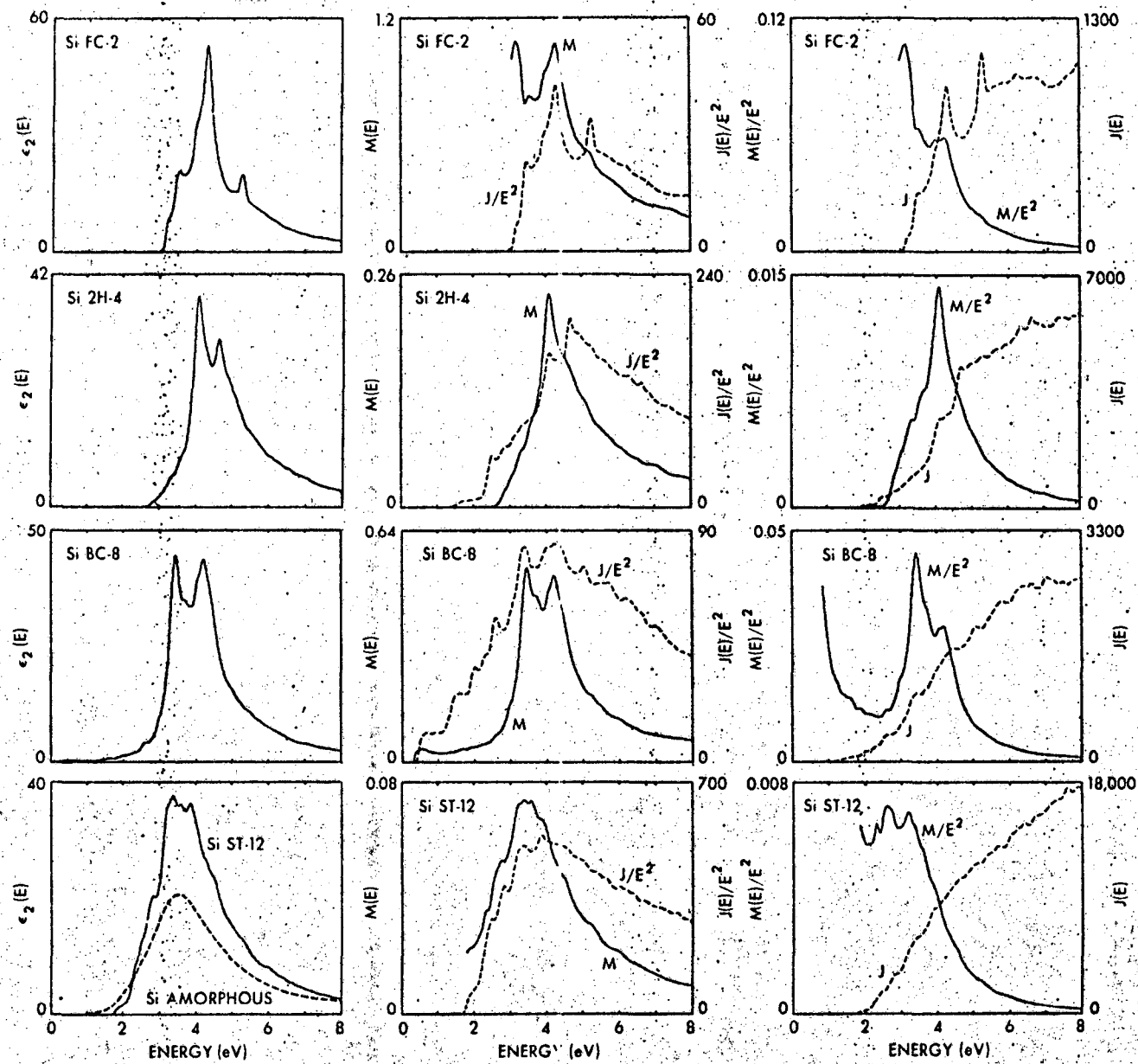


Fig. 1.

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