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Optical conductivity in disordered alloys: an approach via the augmented space recursion

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Abstract

We present here a calculation of the configuration averaged optical conductivity of random binary alloys CuAu and AgAu. Our formulation is based on the augmented space formalism proposed by Mookerjee (1973 *J. Phys. C: Solid State Phys.* **6** 1340) and the optical conductivity is obtained directly through a recursive procedure suggested by Viswanath and Müller (1993 *The User Friendly Recursion Method (Troisième Cycle de la Physique, en Suisse Romande)*).

1. Introduction

In an earlier work [1] we developed a methodology for the calculation of the configuration averaged optical conductivity of a disordered alloy based on the augmented space formalism developed by one of us [2]. The method was based on the Kubo–Greenwood formula and we showed that disorder scattering renormalizes both the Green functions and the current terms.

In linear response theory, at zero temperature, the generalized susceptibility of a disordered alloy is given by the Kubo formula:

$$\langle \mathbf{j}^\mu(t) \rangle = \int_{-\infty}^{\infty} \chi^{\mu\nu}(t-t') A^\nu(t')$$

where $A^\nu(t)$ is the vector potential and

$$\chi^{\mu\nu}(t-t') = (i/\hbar) \Theta(t-t') \langle [\mathbf{j}^\mu(t) \mathbf{j}^\nu(t')] \rangle.$$

\mathbf{j}^μ is the current operator and Θ is the Heaviside step function. If the underlying lattice has cubic symmetry, $\chi^{\mu\nu} = \chi \delta_{\mu\nu}$. The fluctuation dissipation theorem then relates the imaginary part of the Laplace transform of the generalized susceptibility to the Laplace transform of a correlation function:

$$\chi''(\omega) = (1/2\hbar) (1 - \exp\{-\beta\hbar\omega\}) S(\omega)$$

where

$$S(\omega) = \int_0^{\infty} dt \exp\{i(\omega + i\delta)t\} \text{Tr} \{ \mathbf{j}^{\mu}(t) \mathbf{j}^{\mu}(0) \}. \quad (1)$$

Since the response function is independent of the direction label μ for cubic symmetry, in the following we shall drop this symbol. In the case of other symmetries we have to generalize our results for different directions. Our goal will be, given a quantum 'Hamiltonian' \mathbf{H} , to obtain the correlation function

$$S(t) = \langle \phi | \mathbf{j}(t) \mathbf{j}(0) | \phi \rangle.$$

We shall now determine the correlation directly via the recursion method as described by Viswanath and Müller [3]. In order to simplify the expressions for the dynamical quantities as produced by the Hamiltonian, we consider henceforth the modified Hamiltonian $\bar{\mathbf{H}} = \mathbf{H} - E_0 \mathbf{I}$, whose band energy is shifted to zero. Let

$$\langle \psi(t) | = \langle \phi | \mathbf{j}(t).$$

The time evolution of this *bra* is governed by the Schrödinger equation

$$-i \frac{d}{dt} \{ \langle \psi(t) | \} = \langle \psi(t) | \bar{\mathbf{H}}. \quad (2)$$

We shall now generate an orthogonal basis $\{ \langle f_k | \}$ for representation of equation (2). We shall do this in the following way:

(i) We begin with initial conditions:

$$\langle f_{-1} | = 0; \quad \langle f_0 | = \langle \phi | \mathbf{j}(0).$$

(ii) We now generate the new basis members using a three-term recurrence relationship:

$$\langle f_{k+1} | = \langle f_k | \bar{\mathbf{H}} - \langle f_k | \alpha_k - \langle f_{k-1} | \beta_k^2 \quad k = 0, 1, 2 \dots$$

where

$$\alpha_k = \frac{\langle f_k | \bar{\mathbf{H}} | f_k \rangle}{\langle f_k | f_k \rangle} \quad \beta_k^2 = \frac{\langle f_k | f_k \rangle}{\langle f_{k-1} | f_{k-1} \rangle}.$$

We now expand the bra $\langle \psi(t) |$ in this orthogonal basis:

$$\langle \psi(t) | = \sum_{k=0}^{\infty} \langle f_k | D_k(t). \quad (3)$$

Substituting equation (3) into (2) and using orthogonality of the basis, we get

$$-i \dot{D}_k(t) = D_{k-1}(t) + \alpha_k D_k(t) + \beta_{k+1}^2 D_{k+1}(t) \quad (4)$$

with $D_{-1}(t) = 0$ and $D_k(0) = \delta_{k,0}$. We shall now show that the pair of sequences generated by us, namely, $\{ \alpha_k \}$ and $\{ \beta_k^2 \}$, are sufficient for us to generate the correlation function. We note first that

$$D_0(t) = \langle \psi(t) | f_0 \rangle = S(t). \quad (5)$$

Let us define the Laplace transform

$$d_k(z) = \int_0^{\infty} dt \exp(-izt) D_k(t).$$

Putting this back in equation (4) we get

$$(z - \alpha_k) d_k(z) - i \delta_{k,0} = d_{k-1}(z) + \beta_{k+1}^2 d_{k+1}(z) \quad k = 0, 1, 2 \dots \quad (6)$$

This set of equations can be solved for $d_0(z)$ as a continued fraction representation:

$$d_0(z) = \frac{i}{z - \alpha_0 - \frac{\beta_1^2}{z - \alpha_1 - \frac{\beta_2^2}{z - \alpha_2 - \dots}}}. \quad (7)$$

The structure function, which is the Laplace transform of the correlation function can then be obtained from the above:

$$S(\omega) = \lim_{\delta \rightarrow 0} 2 \operatorname{Re} d_0(\omega + i\delta). \quad (8)$$

2. Augmented space formulation

The augmented space method was first proposed by one of us [2] as a feasible technique for carrying out configuration averaging in disordered systems. In particular, it has been applied extensively for studying the electronic structure of disordered alloys. Readers are referred to the article [4] for details. Here we shall discuss the salient features of this method for application to configuration averages of correlation functions.

The problem addressed involves the configuration averaging of a function of many independent random variables, e.g. $\langle\langle f(\{n_R\}) \rangle\rangle$ where R labels a set of lattice points. The first step is to associate with each random variable n_R an operator \mathbf{M}^R such that the spectral density of this operator is the probability density of the random variable:

$$p(n_R) = -(1/\pi) \langle \uparrow | (n_R \mathbf{I} - \mathbf{M}^R) | \uparrow \rangle.$$

For example, if n_R takes the values 0 and 1 with probabilities x and $y = 1 - x$, then the operator \mathbf{M}^R is of rank 2 and acts on the configuration space of n_R , ϕ^R , spanned by the eigenstates $|0\rangle$ and $|1\rangle$ of \mathbf{M}^R . The state $|\uparrow\rangle = \sqrt{x}|0\rangle + \sqrt{y}|1\rangle$ and the representation of \mathbf{M}^R in the basis $\{|\uparrow\rangle, |\downarrow\rangle = \sqrt{y}|0\rangle - \sqrt{x}|1\rangle\}$ is

$$\begin{pmatrix} x & \sqrt{xy} \\ \sqrt{xy} & y \end{pmatrix}.$$

The augmented space theorem then states that

$$\langle\langle f(\{n_R\}) \rangle\rangle = \langle\langle \emptyset | \tilde{\mathbf{f}}(\{\mathbf{M}^R\}) | \emptyset \rangle\rangle \quad (9)$$

where $\tilde{\mathbf{f}}$ is an operator of rank 2^N obtained by replacing each n_R by \mathbf{M}^R . This operator acts on the configuration space of the set of random variables, $\Phi = \prod^{\otimes} \phi^R$. A state in this space is given by a pattern of configurations at each site, i.e. sequences like $\{\uparrow\uparrow\downarrow\dots\} = \{\mathcal{C}\}$ which can be uniquely represented by the sequence of sites where we have (say) a \downarrow configuration. This sequence is called a *cardinality sequence*. The empty cardinality sequence, $\{\emptyset\}$, is the pattern of \uparrow s everywhere.

If we go back to equations (2)–(5), we notice that for a disordered binary alloy, $S(t) = S[\tilde{\mathbf{H}}(\{n_R\})]$. The augmented space theorem then states that

$$\langle\langle S(t) \rangle\rangle = \langle\langle \phi | \tilde{\mathbf{j}}(t) \tilde{\mathbf{j}}(0) | \phi \rangle\rangle = \langle \phi \otimes \{\emptyset\} | \tilde{\mathbf{j}}(t) \tilde{\mathbf{j}}(0) | \phi \otimes \{\emptyset\} \rangle = S[\tilde{\mathbf{H}}(\{\mathbf{M}^R\})]$$

where the augmented space Hamiltonian and the current operators are constructed by replacing every random variable n_R by the corresponding operator \mathbf{M}^R .

The recursion may now be modified step by step in the full augmented space:

$$\langle \psi(t) | = \langle \phi \otimes \{\emptyset\} | \tilde{\mathbf{j}}(t).$$

The time evolution of this bra is governed by the Schrödinger equation

$$-i \frac{d}{dt} \{ \langle \psi(t) | \} = \langle \psi(t) | \tilde{\mathbf{H}}. \quad (10)$$

As before, we shall generate the orthogonal basis $\{ \langle f_k | \}$ for representation of equation (10):

(i) We begin with initial conditions:

$$\langle f_{-1} | = 0; \quad \langle f_0 | = \langle \phi \otimes \{ \emptyset \} | \tilde{\mathbf{j}}(0).$$

(ii) The new basis members are generated using a three-term recurrence relationship:

$$\langle f_{k+1} | = \langle f_k | \tilde{\mathbf{H}} - \langle f_k | \tilde{\alpha}_k - \langle f_{k-1} | \tilde{\beta}_k^2 \quad k = 0, 1, 2 \dots$$

where

$$\tilde{\alpha}_k = \frac{\langle f_k | \tilde{\mathbf{H}} | f_k \rangle}{\langle f_k | f_k \rangle} \quad \tilde{\beta}_k^2 = \frac{\langle f_k | f_k \rangle}{\langle f_{k-1} | f_{k-1} \rangle}. \quad (11)$$

We now expand the bra $\langle \psi(t) |$ in this orthogonal basis:

$$\langle \psi(t) \otimes \{ \emptyset \} | = \sum_{k=0}^{\infty} \langle f_k | \tilde{D}_k(t).$$

Continuing exactly as in the last section we get

$$\tilde{D}_0(t) = \langle \psi(t) \otimes \{ \emptyset \} | f_0 \rangle = \langle \langle S(t) \rangle \rangle. \quad (12)$$

Taking Laplace transforms and using the three-term recurrence,

$$\tilde{d}_0(z) = \frac{i}{z - \tilde{\alpha}_0 - \frac{\tilde{\beta}_1^2}{z - \tilde{\alpha}_1 - \frac{\tilde{\beta}_2^2}{z - \tilde{\alpha}_2 - \dots}}}. \quad (13)$$

The configuration averaged structure function, which is the Laplace transform of the averaged correlation function, can then be obtained from the above:

$$\langle \langle S(\omega) \rangle \rangle = \lim_{\delta \rightarrow 0} 2 \operatorname{Re} \tilde{d}_0(\omega + i\delta). \quad (14)$$

The equations (11)–(14) will form the basis of our calculation of the configuration averaged correlation function.

3. The Hamiltonian and current operators in augmented space

Let us begin with a Hamiltonian for a random binary alloy represented in the basis of a tight-binding linear muffin-tin orbital method (TB-LMTO) $\{ | \chi_R \rangle \}$ [5]. For the sake of notational clarity we have suppressed the angular momentum index L . In what follows, we may consider R to be a composite index RL .

$$\mathbf{H} = \sum_R C_R \mathbf{P}_R + \sum_R \sum_{R'} \Delta_R^{1/2} S_{RR'} \Delta_{R'}^{1/2} \mathbf{T}_{RR'}$$

where \mathbf{P}_R and $\mathbf{T}_{RR'}$ are projection and transfer operators on the space spanned by the TB-LMTO basis. C_R and $\Delta_R^{1/2}$ are the TB-LMTO potential parameters and $S_{RR'}$ is the structure matrix. For a binary alloy the former are random and may be given by

$$C_R = C_A n_R + C_B (1 - n_R) = C_B + (C_A - C_B) n_R \\ \Delta_R^{1/2} = \Delta_A^{1/2} n_R + \Delta_B^{1/2} (1 - n_R) = \Delta_B^{1/2} + (\Delta_A^{1/2} - \Delta_B^{1/2}) n_R.$$

Here the random variables n_R take values 0 and 1 with probabilities x and y , as before. The augmented space theorem then builds up the augmented Hamiltonian by replacing the binary random variables $\{n_R\}$ by the corresponding operators $\{\mathbf{M}^R\}$ described in the earlier section:

$$\begin{aligned}\tilde{\mathbf{C}}_R &= \langle\langle C \rangle\rangle \mathbf{I} + (y-x)(C_A - C_B) \mathbf{P}_R^\downarrow + \sqrt{xy}(C_A - C_B) \mathbf{T}_R^{\downarrow\uparrow} \\ \tilde{\Delta}_R^{1/2} &= \langle\langle \Delta^{1/2} \rangle\rangle \mathbf{I} + (y-x)(\Delta_A^{1/2} - \Delta_B^{1/2}) \mathbf{P}_R^\downarrow + \sqrt{xy}(\Delta_A^{1/2} - \Delta_B^{1/2}) \mathbf{T}_R^{\downarrow\uparrow}\end{aligned}\quad (15)$$

where the configuration space operators are

$$\begin{aligned}\mathbf{P}_R^\downarrow |\{C\}\rangle &= \delta(R \in \{C\}) |\{C\}\rangle \\ \mathbf{T}_R^{\downarrow\uparrow} |\{C\}\rangle &= |\{C\} \pm R\rangle.\end{aligned}$$

The augmented space Hamiltonian is then given by

$$\tilde{\mathbf{H}} = \sum_R \tilde{\mathbf{C}}_R \otimes \mathbf{P}_R + \sum_R \sum_{R'} \tilde{\Delta}_R^{1/2} S_{RR'} \tilde{\Delta}_{R'}^{1/2} \otimes \mathbf{T}_{RR'}.\quad (16)$$

Next we look at the expression for the current operator in the TB-LMTO basis:

$$|\chi_R\rangle = |\phi_R\rangle + \sum_{R'} h_{RR'} |\dot{\phi}_{R'}\rangle.$$

The dot refers to the derivative with respect to energy. In this basis, the matrix elements of the current operator can be written as

$$\begin{aligned}J_{RR'}^\mu &= \langle \chi_{R'} | \mathbf{j}^\mu | \chi_R \rangle \\ &= e \left[V_{RR'}^{(1),\mu} \delta_{RR'} + \sum_{R''} V_{RR''}^{(2),\mu} h_{R''R'} + \sum_{R''} h_{RR''} V_{R''R'}^{(3),\mu} + \sum_{R''} \sum_{R'''} h_{RR'''} V_{R''R'''}^{(4),\mu} h_{R''R'} \right]\end{aligned}\quad (17)$$

where

$$\begin{aligned}V_{RR'}^{(1),\mu} &= \langle \phi_{R'} | \mathbf{v}^\mu | \phi_R \rangle; & V_{RR'}^{(2),\mu} &= \langle \dot{\phi}_{R'} | \mathbf{v}^\mu | \phi_R \rangle \\ V_{RR'}^{(3),\mu} &= \langle \phi_{R'} | \mathbf{v}^\mu | \dot{\phi}_R \rangle; & V_{RR'}^{(4),\mu} &= \langle \dot{\phi}_{R'} | \mathbf{v}^\mu | \dot{\phi}_R \rangle.\end{aligned}$$

The technique for calculating these matrix elements has been described in detailed by Hobbs *et al* [6]. We have also used this technique in our earlier paper [1] and we shall use it here as well. Readers are referred to these two papers for details.

Ideally the next step would be to calculate J_{AA}^μ , J_{BB}^μ , J_{AB}^μ , J_{BA}^μ as the current terms for two sites when they are occupied by atom pairs AA, BB, AB and BA embedded in the disordered medium. A simpler first step would be to obtain these current terms from the pure A and B and from the ordered AB alloy. In general, the current operator can be written as

$$\mathbf{j}^\mu = \sum_R J^\mu(0) \mathbf{P}_R + \sum_R \sum_{R'} J^\mu(\chi) \mathbf{T}_{RR'}\quad (18)$$

where $\chi = R - R'$.

In a disordered alloy, the current representations are random:

$$J^\mu(0) = J_{AA}^\mu(0) n_R + J_{BB}^\mu(0) (1 - n_R)$$

and

$$\begin{aligned}J^\mu(\chi) &= J_{AA}^\mu(\chi) n_R n_{R'} + J_{BB}^\mu(\chi) (1 - n_R) (1 - n_{R'}) \\ &\quad + J_{AB}^\mu(\chi) n_R (1 - n_{R'}) + J_{BA}^\mu(\chi) (1 - n_R) n_{R'}.\end{aligned}$$

Using the augmented space theorem, we replace the random variable n_R by an operator M^R in the expressions for $J^\mu(0)$, $J^\mu(\chi)$; we get the current operator in the augmented space:

$$\begin{aligned} \tilde{\mathbf{j}}^\mu = & \langle J^\mu(0) \rangle \mathbf{I} \otimes \mathbf{P}_R + J_1^\mu(0) \mathbf{P}_R^\downarrow \otimes \mathbf{P}_R + J_2^\mu(0) \mathbf{T}_R^{\uparrow\downarrow} \otimes \mathbf{P}_R \\ & + \langle J^\mu(\chi) \rangle \mathbf{I} \otimes \mathbf{T}_{R,R'} + (y-x) J_1^\mu(\chi) \mathbf{P}_{R'}^\downarrow \otimes \mathbf{T}_{R,R'} + (y-x) J_2^\mu(\chi) \mathbf{P}_R^\downarrow \otimes \mathbf{T}_{R,R'} \\ & + (y-x)^2 J_3^\mu(\chi) \mathbf{P}_R^\downarrow \otimes \mathbf{P}_{R'}^\downarrow \otimes \mathbf{T}_{R,R'} + \sqrt{xy} \left\{ J_1^\mu(\chi) \mathbf{T}_{R'}^{\uparrow\downarrow} \otimes \mathbf{T}_{R,R'} \right. \\ & \left. + J_2^\mu(\chi) \mathbf{T}_R^{\uparrow\downarrow} \otimes \mathbf{T}_{R,R'} \right\} \\ & + \sqrt{xy}(y-x) J_3^\mu(\chi) \left(\left[\mathbf{P}_R^\downarrow \otimes \mathbf{T}_{R'}^{\uparrow\downarrow} + \mathbf{T}_R^{\uparrow\downarrow} \otimes \mathbf{P}_{R'}^\downarrow \right] \otimes \mathbf{T}_{R,R'} \right) \\ & + xy J_3^\mu(\chi) \mathbf{T}_R^{\uparrow\downarrow} \otimes \mathbf{T}_{R'}^{\uparrow\downarrow} \otimes \mathbf{T}_{R,R'} \end{aligned} \quad (19)$$

where

$$\begin{aligned} \langle J^\mu(0) \rangle &= x J_{AA}^\mu(0) + y J_{BB}^\mu(0) & J_1^\mu(0) &= (y-x)[J_{AA}^\mu(0) - J_{BB}^\mu(0)] \\ J_2^\mu(0) &= \sqrt{xy}[J_{AA}^\mu(0) - J_{BB}^\mu(0)] \\ \langle J(\chi) \rangle &= x^2 J_{AA}^\mu(\chi) + xy (J_{AB}^\mu(\chi) + J_{BA}^\mu(\chi)) + y^2 J_{BB}^\mu(\chi) \\ J_1^\mu(\chi) &= x (J_{AA}^\mu(\chi) - J_{AB}^\mu(\chi)) + y (J_{BA}^\mu(\chi) - J_{BB}^\mu(\chi)) \\ J_2^\mu(\chi) &= x (J_{AA}^\mu(\chi) - J_{BA}^\mu(\chi)) + y (J_{AB}^\mu(\chi) - J_{BB}^\mu(\chi)) \\ J_3^\mu(\chi) &= J_{AA}^\mu - J_{AB}^\mu - J_{BA}^\mu + J_{BB}^\mu. \end{aligned}$$

This augmented current operator is used to construct the starting state of the recursion as described in the last section.

4. Results and discussion

For application of our methodology we have chosen two alloy systems: disordered CuAu (50–50) and disordered AgAu (50–50). Our choice was governed by two considerations: first, we had studied these alloys in an earlier work with a different approach [1] and it was of interest to compare the two approaches; and secondly, we have experimental data available for both of these alloys and it was of interest to compare our theoretical estimates with experiment. We have begun our study with a self-consistent TB-LMTO-ASR calculation on AgAu and CuAu 50–50 alloys. We have minimized the energy with respect to the variation in the average lattice constant for both the alloys. We have calculated the current terms from the basis functions and potential parameters obtained from a TB-LMTO calculation for the ordered 50–50 alloys. Ideally we should have embedded AA, BB and AB pairs in the disordered medium and obtained the current expressions from such a calculation. However, in our earlier work [1] we have shown that the effect of randomness in the current terms is small. This gives us the confidence to proceed with these estimates.

To check the density of states for the disordered alloys we have used the TB-LMTO-ASR programme developed by us [7]. The density of states has been shown in our earlier work [1]. The main feature of interest is that the filled d states lie about 2 eV below the Fermi level for both of these alloys.

In figure 1 we show the continued fraction coefficients for a few different angular momentum components. The coefficients do show a tendency of showing oscillatory convergence, but within the maximum ten steps that we were able to take, it was not possible to accurately locate the converged values. We therefore chose not to use the square-root terminator, but opted for the Beer–Pettifor terminator [8].

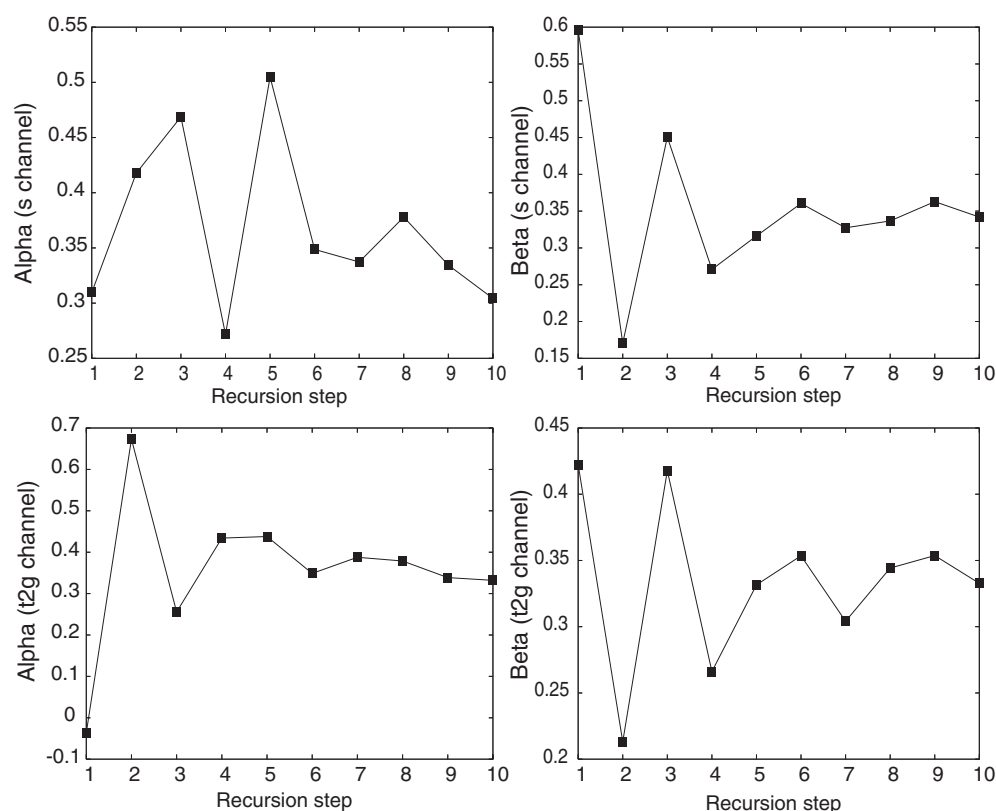


Figure 1. Continued fraction coefficients for the s and t_{2g} channels for CuAu alloy.

Experimental data on disordered CuAu (50–50) are available [9]. The authors have reported high resolution optical data for both ordered and disordered CuAu for photon energies between 1.2 and 6 eV. In figure 2, we also show the experimental data for $\epsilon_2(\omega)$, as dotted lines, and compare them with the theoretical data in figure 2 (bold lines). Drude-like behaviour is clearly seen up to photon energies of ~ 2.1 eV in theory and around 2 eV in the experiment. Subsequently $\epsilon_2(\omega)$ rises again because of transitions from the d states. Both the theory and experiment show a decrease around 6 eV. The experimental curve shows a sharper dip and the subsequent peak is at a lower energy.

We have experimental data on AgAu (50–50) [10], whose density of states closely resembles that of CuAu. The inter-band contribution to the imaginary part of the dielectric function $\epsilon'_2(\omega)$ may be obtained from the optical conductivity data, by subtracting away the Drude contribution and dividing the result by ω : $\epsilon'_2(\omega) = (\sigma(\omega) - \sigma^D(\omega))/\omega$. Below the onset of the inter-band transitions, this quantity vanishes. It shows a shoulder around 2.5 eV and then reaches a maximum at around 3.5 eV before decreasing and showing a feature around 5.5 eV.

As in the coherent potential approximation (CPA) estimates of Banhart [10] there is a $\simeq 0.5$ eV shift outwards in the theoretical estimate. In figure 3 we have shifted our theoretical data by 0.5 eV downwards for comparison. Banhart attributes this shift to the foundations of the electronic theory used: local density approximation which cannot reproduce excited states properly. This had been discussed also by Fehrenbach and Bross [12]. To this we shall

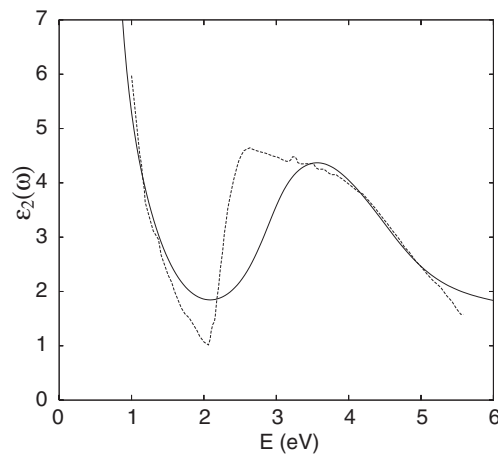


Figure 2. Averaged optical conductivity showing a Drude-like behaviour at low photon energies. Theoretical estimates are shown as full lines and experimental data for $\epsilon_2(\omega)$ for disordered CuAu taken from Scott and Muldrew [9] are shown as dotted lines.

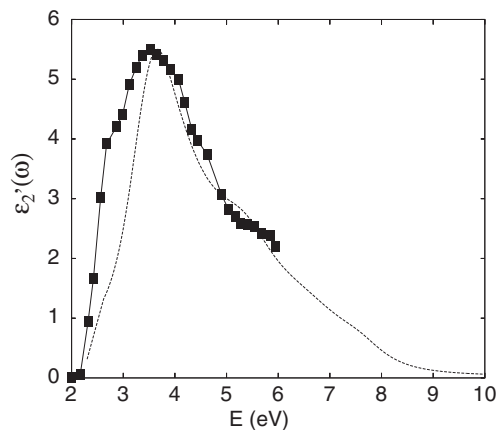


Figure 3. Intra-band contribution to $\epsilon_2'(\omega)$ for AgAu alloy, theoretical (full lines) and experimental [11] (points).

add that it is doubtful whether the energy linearized TB-LMTO can accurately reproduce the large energy window of our calculation. It may be useful in future to base our calculations on the newly developed NMTO formalism [13]. In addition, the termination procedure of the recursion requires further refinement, as the convergence of the continued fraction coefficients is not as rapid as in density of states calculations. Banhart notes that the CPA neglects all short-ranged order. However, since our calculations in the augmented space recursion do include correlated scattering from different sites, we cannot attribute the discrepancy to the CPA.

Apart from the energy shift, the experimental data are in reasonable agreement with our theory. The general shape with a maximum is clearly reproduced. The feature shown in the experimental data around 5.5 eV is also seen in our theoretical estimate, slightly shifted in energy. The shoulder in the experimental data just above 2 eV is not properly reproduced in the theoretical curve. The CPA estimate of Banhart [10] also does not seem to show such a prominent shoulder. Is it possible that the experimental alloy had short-ranged order?

It is known that short-ranged order can influence optical properties [14]. The augmented state formalism can handle short-ranged order [15] and we shall study this effect in a future communication.

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