Identification of Satellites Due to Resonant Excitation And Shakeoff in the C KVV Auger Lineshape of Polyethylene

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IDENTIFICATION OF SATELLITES DUE TO RESONANT EXCITATION AND SHAKEOFF
IN THE C KVV AUGER LINESHAPES OF POLYSTYRENE

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A quantitative interpretation of the C KVV Auger lineshape of polystyrene reveals the presence of satellites arising from resonant excitation into a core excitonic state, and from shakeoff of a valence electron during the initial core ionization. The intensity, energy, and shape of these satellites reveal much about localization and screening processes in one-dimensional polymers.

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1. Introduction

A quantitative interpretation of the C KVV Auger lineshape of polystyrene reveals the presence of three satellite contributions. Identification of the two at high kinetic energy indicates they arise from resonant excitation into a core excitonic state; the one at lower energy arises from shakeoff of a valence electron during the initial core ionization. This is the first identification of such satellites in an extended covalently bonded solid.

These satellite contributions directly reflect the localized nature of a conduction or valence bond electron in the presence of a core hole, and thus provide direct information on the screening processes in one-dimensional (1-D) polymers. Similar interpretations [1,2] of the C KVV Auger lineshapes of graphite and diamond (the comparable 2-D and 3-D carbon systems) do not reveal such satellites. On the other hand, the Auger lineshapes of several gas phase molecules, such as cyclohexane and benzene, reveal similar satellites [3].

2. The KVV Lineshape

2.1 Experimental

The experimental Auger lineshapes of polystyrene has been reported previously. Deyan and Pepper [4] recorded the data as dH(E)/dE using electron excitation and rastered the beam to eliminate beam damage effects, which were clearly observed when the beam was not rastered. Keiber et al [5] recorded the data as N(E) utilizing Mg K\alpha X-rays, which resulted in no observable damage to the sample. The apparent reduced beam damage under X-ray excitation probably arises because of the reduced beam currents. This is evident since electron and photon damage cross sections are generally very similar [6], and further, large numbers of secondary electrons of sufficient

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energy to initiate damage are produced by the photon beam in any event.

Consistent with the above, the use of Mg x-rays is not expected to
significantly reduce the amplitude of the resonantly excited satellites, since
high energy photons produce large numbers of secondary electrons with
energies well above the C 1s level at 284.6 eV [6]. Studies in condensed
bromine reveal that the resonantly excited satellite is still present with high
energy x-ray radiation, although the resonant satellite does not appear with
low photon energies [7].

Fig. 1a compares the deconvolved spectra obtained by Dayan and Pepper
[4] and Kelber et al. [5]. Similar background subtraction and deconvolution
procedures were utilized to remove the effects of secondary electrons and
loss processes [1,4,5]. The absolute kinetic energy of the primary peak in the
two reported spectra differs by 6 eV, indicating problems exist with sample
charging and energy calibration. Since Dayan and Pepper utilized a 700 A
film on a metalized glass disk, while Kelber et al. used a bulk slab, charging
is expected to be less of a problem in the former case; however, Kelber et
al. did attempt to account for the charging shift in their analysis. Thus
considerable uncertainty exists in the absolute energy scale for polyethylene,
a common problem with insulating samples. Ignoring the energy scale, the
differences in shape between the two spectra are small; what differences are
present occur primarily at lower energy where the uncertainties in the loss
deconvolution process are greatest.

The previous interpretations reported for the polyethylene lineshape,
although highly qualitative, differ sharply in their conclusions [4,5]. This
arises because the interpretations depend on the whether the emphasis was
placed on the experimental lineshape or the energy scale. Kelber et al. [5],
whose polyethylene lineshape was found to be essentially energy aligned with
the alake lineshapes, concluded that the two-hole Auger final state was
highly localized. Dayan and Pepper [4], who emphasized the lineshape over the
energy scale, found that a simple self-fold of the DOS (i.e., a completely
delocalized picture) reproduced some of the features of the experimental
lineshape, but did not have the proper width. They further reasoned that a
highly localized Auger final state would further narrow the theoretical
lineshape making the agreement even worse. Thus they concluded that other
processes must also be occurring, but they did not elaborate.

In this work, we place the emphasis on the spectral lineshape, which is
known with much greater certainty than the absolute energy scale. We agree
with the conclusions of Dayan and Pepper that a highly localized Auger final
state is not consistent with the lineshape, and that other contributions
beyond the normal Auger contribution are present. We have interpreted the
polyethylene lineshape in the context of our previous interpretations of the
diamond and graphite lineshapes [1,2], and concurrent interpretations of the
gas phase alkane and alkene lineshapes [3]. We find the polyethylene
lineshape to be consistent with all of the above, and based on our
interpretation, the absolute energy scale of Dayan and Pepper is the more
correct one. Thus in Fig. 1a the two-electron binding energy scale is
obtained by subtracting 280.1 eV (284.6 minus 4.5 for the analyzer work
function) from the Dayan and Pepper kinetic energy scale [4], and the
lineshape of Kelber et al. is energy aligned with that of Dayan and Pepper.

2.2 Theoretical

Polyethylene has been previously studied by electron energy loss
spectroscopy (EELS) [9], photoemission (both XPS and UPS) [6,10,11], UV
absorption spectroscopy [11,12], and by semiempirical and ab-initio theoretical
calculations [13,14]. The existence of the valence and core excitation level has
been clearly established [9,11]; however, the s and p components of the one
electron density of states (DOS) have not been accurately determined. We
interpret the Auger lineshape utilizing a bulk DOS (N(E)) obtained semi-
empirically from XPS data by a procedure similar to that described previously
[15]. It is also necessary to separate the p carbon-carbon (pcc) from the p
carbon-hydrogen (pch) bonding DOS, since Auger contributions involving these
orbitals experience different localization effects [16]. This was accomplished
by identifying each of the features in the DOS with orbitals in cyclohexane,
where the bonding character is clearly evident [3]. The resultant semi-
empirical DOS, having a charge distribution of σ^4pcc^4, is compared with
the well-known theoretical results of McCubbin and Mann [13] in Fig. 2a. The
semi-empirical results contain an effective broadening of 1-2 eV, which
apparently arises from spectrometer resolution and intermolecular
interactions, but the agreement otherwise is very good.

Final state hole-hole correlation effects are included in the lineshape
using the Cini expression [17].

\[
C[N(E)N(E)] = \frac{N(E)N(E)}{[1 - \Delta U N(E)]^2 + (\Delta U N(E))^2}
\]

where \( I(E) \) is the Hilbert transform,

\[
I(E) = \int N(E-\gamma N(E)/\gamma - E) \, d\gamma
\]

and \( C[N(E)N(E)] \) represents the Cini distortion of the DOS self-fold, \( N(E)N(E) \).

In Fig. 2b, eq. (1) has been applied individually to the σσ, σpcc, σpcc,
pccpcc, pccpcpcc and pccpccpcc components [1,3]. The quantity \( \Delta U \) equals \( U_\text{H} - U_\text{S} \) where \( U_\text{H} \) and \( U_\text{S} \) are the effective Coulomb repulsions of two holes in
the same orbital, and in nearest neighbor orbitals respectively [3,18]. In this
case the effective orbital can be pictured as a cluster orbital, consisting of
four sp^3 bond orbitals surrounding a single C atom [1,18] (i.e. a methyl group
orbital).

Although the \( \Delta U \)'s can be treated as variable parameters, we assigned
them the values which provided an optimal fit in a similar interpretation of
the cyclohexane Auger spectrum; namely 3 eV for all of the contributions
except for the pccpcc contribution which has a \( \Delta U \) of 1.2 eV [3]. The
cyclohexane gas phase lineshape exhibits more structural features and thus
allows for more certainty in determining the optimal \( \Delta U \)'s. We assume that
these \( \Delta U \)'s do not change on going to polyethylene, since both systems are
essentially continuous alkane chains.

The resultant total lineshape and six individual components are shown in
Fig. 2b. The magnitude of the hole-hole correlation effects can be seen by
comparison with the DOS self-fold, as shown in Fig. 2b. The \( \Delta U \)'s reflect the
larger localization in the C-H bond orbitals, as expected. The σ and σ components contain elements of both C-C and C-H bond character, and thus
these \( \Delta U \)'s are also somewhat larger than for the pccpcc contribution [3].
The Auger lineshapes for methane and ethane suggest that \( U_\text{H} \) for a methyl
group is around 11 - 12 eV and \( U_\text{S} \) in the ethane molecule is around 8 - 9 eV
[3]. The optimal \( \Delta U \) values of 3 eV used for cyclohexane and polyethylene in
the Cini expression are reasonably consistent with these \( U_\text{S} \).

Comparison of the theoretical and experimental lineshapes in Fig. 1a
reveals that the normal Auger process, kvv, accounts for only 70% of the
total intensity. The difference spectrum (experiment minus theory) in Fig. 1b
shows two main contributions, the lower one centered around 36 eV, the upper
one around 10 eV. The shape of the lower contribution is surprisingly similar
to the theoretical Auger lineshape, the upper one is similar to the
theoretical DOS self-fold. These theoretical lineshapes are aligned in energy
and compared with the difference contributions in Fig. 1b. The three features
in the lower contribution are reproduced, except the feature around 30 eV is
underestimated. The three sharp features in the upper contribution are
present, but much less sharp in the DOS self-fold. These comparisons indicate that the bottom contribution can be identified as an initial state shake/Auger satellite, and the top contribution as a resonant excitation satellite.

3. The resonant satellite

A resonant Auger satellite arises when Auger decay occurs in the presence of a localized electron, which can be created by resonant excitation into an excitonic state upon creation of the core hole. The existence of an excitonic state in polyethylene is well established, apparently arising from the ε* antibonding orbitals [9]. As indicated by the EELS spectrum shown in Fig. 3a [4], mimic the X-ray absorption spectrum (XAS)], the excitonic state has a binding energy of 1.6 eV relative to the vacuum level, or -2.9 eV relative to the Fermi level [9].

The generally good agreement between the energies of the sharp peaks between 0 and 25 eV in the difference spectrum (Fig. 1b) and the DOS self-fold indicates that the excitonic electron very effectively screens the two Auger *valence holes, reducing the correlation effects to zero. The poor resolution of the features in N(E) vs N(E), compared to that in the difference spectrum, suggests that the empirical DOS, N(E), may be too broad as already suggested above. The sharper features in the resonant satellite, compared to that in the normal Auger lineshape, probably reflects a smaller Franck-Condon envelope for the satellite. This is expected, since the lack of hole-hole correlation effects in the satellite means there will be less bond lengthening in the satellite final state due to the effects of the "coulomb explosion" [19].

The energy of the initial state in the resonant Auger process is

$$E_e + E_r + E_g + U_{pol} = 2U_{pol}$$

(4)

where we have assumed pair-wise additivity, and $E_c$ and $E_r$ are the binding energies of the Auger holes and the U's are the corresponding hole-hole repulsions. The Auger energy $E_{Aug}$ is the difference (initial - final) giving

$$E_{Aug} = E_e - E_r - E_g + (-U_{pol} - U_{pol} + 2U_{pol})$$

(5)

where the term in parentheses is the shift, $\delta$, of the resonant Auger satellite relative to that given by the one electron theory.

An estimate of $U_{pol}$ can be obtained from Fig. 3a, where we applied eq. (1) [i.e. evaluated C(N(E))] to the theoretical DOS to account for the presence of the core hole. In defect theory, this is called the "central cell tight binding approximation" [20] and $U_{pol}$ is the polarization energy due to the increased nuclear charge on the defect or donor atom. $U_{pol}$ was treated as a parameter and assigned the value which provided the optimal agreement between C(N(E)) and the EELS spectrum, which according to the final state rule should reflect the conduction band DOS in the presence of a core hole [21].

As shown in Fig. 3a, excellent agreement was obtained when $U_{pol}$ is 10 eV.

The required shift, $\delta$, of the DOS self-fold in Fig. 1b is 5 eV. Eq. 5 indicates that $U_{pol}$ must then be 7.5 eV if $U_{pol}$ is zero. $U_{pol}$ is probably near zero, since the kink lineshape, although exhibiting some correlation effects, reflects primarily delocalized valence holes. The existence of a valence excitonic state is well established [7 - 12]. The relative values of $U_{pol}$ and $U_{pol}$ are consistent with the observation that core excitons are generally more localized than valence excitons.

A high energy shoulder near the Fermi level appears in the difference spectra which is not reproduced by the DOS self-fold. This can be identified as a third satellite, arising also from a resonant electron excitation, but one in which the excitonic electron participates in the Auger decay. This
satellite lineshape can be approximated simply by the one electron DOS, as shown in Fig. 1b. The initial state of such a process is the same as in eq. (3). The final state has a one hole energy of $E_h$, giving an Auger energy of

$$E_{Aug} = E_h - E_e - (E_e + U_h),$$

(7)

where the term in parentheses is the shift required in H(E). In this case $E_h$ is equal to the exciton binding energy relative to the Fermi level. The shift in Fig. 1b was 3 eV, which is in excellent agreement with the exciton binding energy ($\approx 3$ eV) as determined by EELS [9].

The intensities of the ke-ve and ke-v satellites are 10.7% and 2.6%, respectively, relative to the total intensity. This intensity depends on the electron excitation energy and the secondary cascade process, so that their absolute intensities are not very interesting. It should be pointed out, however, that by utilizing synchrotron radiation tuned to the exact resonant energy, one could obtain experimentally just the resonant contribution. This process has been called de-excitation electron spectroscopy (DES), and has been reported for both gas phase and chemisorbed Cu, where the 2p level is resonantly populated [21].

Although their individual intensities are not of interest, the ratio of intensities, I(ke-v)/I(ke-ve), indicates something about the character of the excitonic level. The atomic Auger matrix elements per electron are essentially the same, for the s, p, and d contributions in keV spectra [11]. Therefore, we can estimate what the ratio of intensities should be, based purely on the ratio of local electron densities, assuming a completely localized excitonic level. With an initial state charge distribution of sp², I(ke-v)/I(ke-ve) should be 0.5, compared to 0.25 found experimentally. This suggests that although the excitonic level may be localized in time, it must be of a more diffuse nature spatially. The factor of two reduction suggests that the core exciton spends only half of its time on the methyl group with the core hole, the other half of the time presumably on neighboring methyl groups.

4. The shake satellite

We indicated above that the satellite centered at 36 eV can be attributed to an initial state shake/Auger process. Such a satellite arises when Auger decay occurs in the presence of a localized valence hole, which was created via the shake-off process during the initial ionization [23]. The satellites are most often seen in gas phase spectra, when the localization of the valence hole is assured. They are rarely seen in the spectra of solids, since the valence hole normally propagates away from the core hole before the Auger decay [24]. Its presence here, and its energy, intensity, and lineshape, characterizes the nature of the localized shake hole in the valence band.

The valence band DOS for polyethylene in the presence of a core hole, obtained as described above, is shown in Fig. 3a. It shows relatively sharp peaks indicative of localized states, for both the s and p DOS. For comparison, the valence band DOS for diamond in the presence of a core hole is shown in Fig. 3b, obtained exactly as for polyethylene [2]. In diamond, U₀ is equal to just 5 eV, compared to 10 eV for polyethylene, reflecting the increased screening in 3-D diamond. Clearly no localized valence band states exist for diamond in the presence of a core hole, and no shake/Auger satellites are found in the Auger lineshape [2].

The theoretical probability for shakeoff upon core ionization of a carbon atom in methane is estimated to be around 24% [25,3]. Furthermore, it has been shown that this probability is relatively independent of its molecular environment [25,26]. The experimental relative intensities of the shake/Auger satellites in ethane, ethylene, cyclohexane and benzene are around
21-24%, compared to 17-21% in polyethylene (the latter depends on whether the
Pepper or Kelker data is used). This indicates that in polyethylene, as well
as in the gas phase molecules, most of the shake holes remain localized
efficiently long to cause a satellite contribution.

We indicated above that the minimum subunit orbital is a methyl group
orbital, and that a shake hole could not be delocalized over the whole chain
or else no satellite would be visible. We can therefore consider two extreme
cases for the nature of the valence shake hole; either 1) it is localized on
the methyl group with the core hole, or 2) it is delocalized on some larger
sub-cluster of the alkane chain. Now in the final state for case 1), the total
repulsion energy is 3U0; it decreases to U0+2U0 when one hole moves off
this methyl group. AU, as defined above, then is U(U1-U0) or twice what it is
for the normal Auger lineshape. For case 2), the total repulsion energy is
U0+2U0, where U0 is the repulsion between a hole created by the Auger
process, and localized on the initial methyl group, and the shake hole
localized on some larger subcluster of the chain. When one Auger hole moves
off the initial methyl group, the net repulsion reduces to U0+2U0, and
thus AU is now equal to that for the normal Auger process. We conclude that
for case 1) the satellite lineshape should be very different from the normal
lineshape because of the different AU; in case 2) it should be the same. Case
1 is clearly appropriate for methane gas, and indeed the experimental Auger
spectrum reveals different lineshapes for the normal and satellite
contributions [3].

The strong similarity between the shake/Auger and normal Auger
lineshapes in Fig. 1b clearly indicates that for polyethylene case 2 is more
appropriate. We conclude that in polyethylene the shake hole delocalizes onto
some larger sub-cluster of the alkane chain. The nature of this delocalized
orbital can be obtained from the size of the shift,

\[ \delta_{s} = U_0 - 2U_0 - U_{mm} \]  

of the shake/Auger satellite relative to that for the normal Auger lineshape.

Eq. 8 is derived exactly as eq. 5 above, except for the change in sign in
front of U0 and U0 (here we have hole-hole repulsion instead of hole-
electron attraction). The required experimental downward shift is 18 eV. A
good estimate of Umm can be obtained from the shift in the primary peak in
the p DOS upon creation of a core hole, which is 6 eV as shown in Fig. 3a. In
the final state, the shake hole is indistinguishable from the Auger valence
holes, thus \( \delta_{s} \approx U_{mm} - 3 U_{mm} \) and \( U_{mm} \) must be 8 eV, assuming pairwise
additivity. This represents the approximate hole-hole repulsion of an ethane
or propane molecule, i.e. the holes are localized on the core hole methyl group
but with significant population on neighboring methyl groups.

Similar interpretations, to be published elsewhere, of the Auger
tlineshapes of the gas phase molecules, methane, ethane, ethylene, cyclohexane,
and benzene, reveal that \( \delta_{s} \) for the initial-state Auger satellite is
relatively constant at -18 eV, inspite of the fact that the two-hole molecular
hole-hole repulsion energy is increasing as the molecule gets smaller. This
strongly indicates, contrary to that indicated previously [27], that the two-
hole kev lineshape primarily reflects molecular delocalized holes (although
hole-hole correlation effects are evident), but the ka-\( \nu \nu \)'s three-hole
lineshape reflects holes largely localized onto a few methyl groups.

5. Summary

In summary, we have identified and interpreted satellites arising from
resonant electron excitation and shakeoff in the Auger lineshape of
polyethylene. This is the first identification of such satellites in extended
covalent solides. Our results provide direct experimental evidence for partial
delocalization of the excitonic electron and shake hole onto nearest
neighboring methyl groups. Further, the valence two-hole final states are

FIGURE CAPTIONS

Fig. 1. a) Comparison of the experimental lineshapes (after background subtraction and loss deconvolution as described in ref. 1) reported by Dayan and Pepper [4] and Kelber et al [5], with the theoretical total lineshape as determined in this work. The Kelber spectrum was energy aligned to that of Peppers. The components in order of increasing energy are the ke-vva, kvv, ke-vve, and ke-v. Their relative intensities were determined by the best fit to the experimental spectra and their lineshapes were determined as described in the text.

b) Comparison of the difference spectra (Pepper's experimental spectrum minus the theoretical kvv component) with the sum of the satellite components as indicated above.

Fig. 2 a) Comparison of the empirically determined DOS as described in the text with the theoretical DOS as reported by McCubbin and Manne [13,14]. The theoretical DOS were shifted upward by 2 eV to provide better agreement with the empirical DOS. The spc, and pse components, determined as described in the text, are also shown.

b) Comparison of the DOS self-fold (NNN) and the Cini distorted self-fold (CNN) obtained by utilizing eq. 1 and a AU of 1.2 eV for the pse+pse contribution and 3 eV for all of the others. The
Contributions in order of increasing energy are the $\sigma^*$, $\pi^*$, $\pi^*$, and $\pi^*$-$\sigma^*$ self folds, and the $\pi^*$-$\pi^*$, $\pi^*$-$\pi^*$, and $\pi^*$-$\pi^*$ cross folds.

Fig. 3 a) Comparison of the polyethylene DOS (N) with the distorted DOS (C(N)) obtained by utilizing eq. 1 applied to N with a core hole polarization potential (i.e. $\Delta U$ in eq. 1) of 10 eV. The optimum $\Delta U$ was determined by the best fit to the EELS lineshape [9] (which mimics the XAS). The occupied and unoccupied DOS of McCubbin and Mann [13] are shown as reported in refs. 14 and 10, respectively, but they were energy shifted for best agreement with the EELS and XPS (Fig. 2) data.

b) Comparison of the diamond DOS (N) with the distorted DOS (C(N)) obtained as above with an optimal $\Delta U$ of 5 eV. The theoretical DOS, reported in ref. 29, and XAS, reported in ref. 28, are shown; the DOS were energy shifted for best agreement with the XAS.
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