Origin of the low-energy tail in the Al $L_{2,3}VV$ Auger spectrum studied with Auger-photoelectron coincidence spectroscopy

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Al $L_{2,3}VV$ auger-photoelectron coincidence (APEC) data are presented and discussed. The well-known ability of APEC to suppress inelastic-scattering contributions to Auger and photoelectron spectra is used to demonstrate that much of the large low-energy tail common to core-valence-valence Auger spectra from many wide-band solids is not due to secondary-electron scattering, but, rather, is an intrinsic part of the core-hole-decay process.

In core-valence-valence (CVV) Auger spectra from wide-band solids a relatively high intensity of emission is observed at low energies. This low-energy tail (LET) typically contains most of the CVV spectral weight. Conventional interpretations attribute the LET to loss processes unrelated to the Auger event, and subtract it off before comparison to theoretical calculations. Examples of this interpretation of data include studies of Al,[1,2] Si,[3–7] Be,[8] Mg,[9,10] Li,[11,12] and C.[13] In this paper Auger-photoelectron-coincidence (APEC) spectra from Al(111) are presented. Although this technique is well known to discriminate against inelastic-scattering processes,[14–16] the LET in the coincidence spectrum is nearly as large as that seen out of coincidence. It is concluded that much of the LET is an intrinsic part of the Auger process.

The conventional picture of the CVV process is illustrated in Fig. 1. A core hole of energy E_c is created by photoionization in a metal of valence bandwidth W. One valence electron fills the core hole, and another is ejected as the Auger electron, leaving two holes in the valence band. The initial state for the Auger process is the core-hole state, and the final state is the two-valence-holes, one-excited-electron state. By conservation of energy, the maximum energy that the Auger electron can have is the core binding energy E_c, corresponding to two holes at the top of the valence band. The minimum energy is E_c minus twice the valence-band width W, corresponding to two holes at the bottom of the valence band. These final-state configurations are illustrated in Fig. 1. The Auger spectrum has width $2W$. Emission outside the range E_c to E_c-2W is forbidden by energy conservation. If it is assumed that all allowed final states are approximately equally well coupled to the initial state, and that surface effects and electron correlation effects can be ignored, then the Auger spectrum can be described by a self-convolution of the valence-band density of states.[17] Note that CVV spectra from narrow-band solids have a much different interpretation, dominated by hole-hole interaction effects in the Auger final state.[18] These materials will not be discussed further.

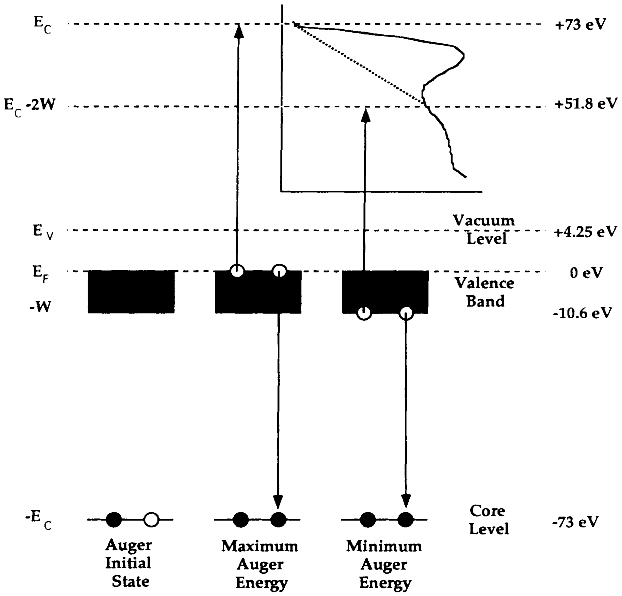

FIG. 1. Schematic illustration of the CVV process. The core-hole energy is E_c, the valence-band width is W, and the Fermi level is the zero of energy. The core hole, or Auger initial state, is shown on the left. Two possible Auger final states, those with maximum and minimum Auger-electron energy, are shown in the center. The Al $L_{2,3}VV$ spectrum is shown on the right. The LET in this spectrum is comprised of all emission at energies below 51.8 eV, plus some continuation into the higher-energy region (Ref. 20). The dashed line represents the approximation to this contribution

Also shown in Fig. 1 is the noncoincidence Al $L_{2,3}VV$ spectrum taken at 200-eV photon energy. It is essentially identical to that observed by Powell.[1] The Al 2*p* core-level binding energy is 73 eV, and the Al valence bandwidth is 10.6 eV.[19] According to the previous discussion, a 21.2-eV-wide spectrum extending from 51.8 to 73 eV, peaked somewhere near 65 to 70 eV, is expected. This is clearly not observed; this spectrum looks more like an edge at 73 eV, with a weak peak at 69 eV, a valley at 61 eV, and slowly increasing emission at lower energies. The extensive spectral weight below 51.8 eV is all part of the LET.[20] As discussed above, energy conservation forbids ordinary *CVV* decay from this region. In order for Auger emission to be seen at these low energies, energy must have gone into some other excitation. These other excitations could occur extrinsic to the core-hole decay, during the transport of the Auger electron to the surface, or they could be an intrinsic part of the Auger process.[21] It is usually assumed that both intrinsic and extrinsic processes play a part in LET generation, but that the dominant cause of the LET is extrinsic inelastic electron scattering. Within this interpretation, the LET may be subtracted from the spectrum without penalty, and the (small) remainder is then searched for surface, matrix element and electron correlation effects. Extensive discussion of the relative importance of each of these effects in different materials can be found in the literature.[1–13,22] A good summary may be found in Ref. 10. Good agreement between theory and the remainder spectrum for Al is generated in Ref. 22. In direct opposition to this picture, our coincidence data suggest that extrinsic inelastic scattering is a minor contributor to the LET, and that much of the LET is intrinsic to the *CVV* decay process.

Data were taken at the National Synchrotron Light Source at Brookhaven National Laboratory, on beam lines U12 and U14. The Al(111) samples were cleaned daily by sputter anneal techniques until photoemission spectra exhibited negligible evidence of oxygen in the sensitive valence-band and 2*p* core-level regions. 100- to 200-eV light was incident normal to the sample. Total energy resolution was typically 1 eV. Two cylindrical mirror electron energy analyzers analyzed the emitted electrons. Coincidence events were accepted only when electrons were received in each analyzer at the same time. The instrumental timing resolution of approximately 10^{-8} sec permits the determination that both electrons came from the same photoexcitation-decay event,[23] and thus the same atom. Data were collected in two distinct modes. In mode 1 (Fig. 3), one analyzer was fixed at a characteristic energy in the photoelectron spectrum, while the second scanned the Auger spectrum. This mode generated coincidence Auger spectra. In mode 2 (Fig. 2), the first analyzer was fixed at a characteristic energy in the Auger spectrum, and the second was scanned through the photoelectron spectrum. This mode generated coincidence photoelectron spectra. In each mode, both ordinary photoelectron or Auger spectra, subsequently called singles spectra, and coincidence spectra were collected. All coincidence spectra presented have had accidental coincidence events removed. The APEC technique was invented by Haak and co-workers for use with x-ray anodes.[14] It was extended to synchrotron sources by us.[15,16] Count rates in singles were typically 10 kHz, while count rates in the coincidence spectra were about 0.5 Hz, with one:one true:accident ratios. Experimental details will be published elsewhere.[23]

Auger-photoelectron-coincidence data strongly suppress the extrinsic secondary-electron contribution to Auger and photoelectron spectra. This has been noted in every previous APEC investigation.[14–16,24] It makes sense that this should be so; both the photoelectron and the Auger electron must escape the solid to be counted in coincidence. This process has a mean free path half as large as the individual Auger electron of photoelectron mean free paths and thus about half as much inelastic scattering is expected.

Figure 2 illustrates the extent to which the detection of extrinsic plasmon loss events is suppressed in the coincidence-photoemission spectrum of Al. In this spectrum, obtained at $h\nu=200$ eV, the primary (i.e., no loss) Al 2*p* photoemission peak occurs at 126 eV while the bulk- and surface-plasmon-loss peaks are seen at 111 and 116 eV, respectively. Superimposed on this curve is the spectrum of the same energy region obtained in coincidence with Al $L_{23}VV$ Auger electrons, scaled so that the primary peaks are of equal intensity. Scaled in this way, we can compare the relative contribution of inelastic secondaries in the singles and coincidence spectra. One can see that the bulk-plasmon peak is severely attenuated

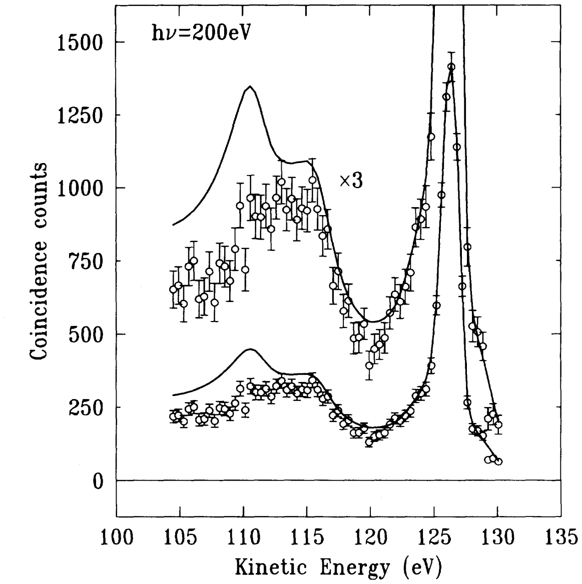

FIG. 2. The Al(111) 2*p* core-level photoemission and plasmon-loss satellite spectra, taken at 200-eV photon energy. The solid lines are the singles spectra, and the circles are the coincidence data (fixed analyzer at 69 eV, the peak in the Auger spectrum). The two spectra are normalized to equal heights at the no-loss photoelectron line at 126 eV. The surface-plasmon-loss feature at 116 eV is attenuated slightly, while the bulk-plasmon-loss feature at 111 eV is attenuated by about 40%, illustrating the ability of APEC to suppress inelastic secondary contributions to photoelectron spectra.

in the coincidence spectrum; the intensity of the bulk-plasmon-loss region is suppressed by about 40% (i.e., the coincidence to singles ratio at 111 eV is ~0.6 when the primary peak heights are equal). Much of the remaining bulk-plasmon intensity in the coincidence spectrum is due to intrinsic plasmon and electron-hole pair losses, so the actual suppression of the extrinsic plasmon contribution is greater than that given by the singles to coincidence ratio.[25] The surface-plasmon peak is not significantly attenuated in the coincidence spectrum because the relative probability of producing a surface loss is not effected by the enhanced surface sensitivity. Only loss processes that can be characterized by a mean free path, or equivalently, a constant loss probability per unit distance, will be suppressed by the coincidence process. The probability of surface-plasmon generation is independent of how far the electron has traveled before reaching the surface, and thus will not be suppressed by coincidence. In principle, the surface-plasmon-loss intensity could even be enhanced. We note that other APEC studies have exhibited even more obvious suppression of inelastic losses.[14-16]

The following simple picture qualitatively accounts for the results presented in Fig. 2. The uv light penetrates about 1000 Å (Ref. 26) into the solid. The electron mean free path is much smaller, about 10 Å at both the Auger and the photoelectron energies.[27] A typical singles event comes from about one electron mean free path below the surface. The probability that this electron escapes without inelastic scattering is e^{-1}, or 0.37. This is the primary part of the spectrum. The probability that it is inelastically scattered is $1-0.37$, or 0.63. This is the extrinsic secondary part of the spectrum. For the singles events, therefore, both primary and extrinsic secondary features occur, but the secondaries have nearly twice as much spectral weight as the primaries and dominate the spectrum. In coincidence spectra, since both electrons must escape the solid, the typical sampling depth is half of an electron mean free path, while the scattering rate per unit distance is the same. Hence, the probability of escape without scattering for coincidence events is $e^{-0.5}$, or 0.61, while the probability of scattering before escape is only 0.39. As a result, the coincidence spectrum has more primaries than secondaries, in particular, the primary to secondary ratio is much larger in coincidence (1.6) than in singles (0.59). While detailed modeling including emission from all depths would be required to obtain quantitative agreement with experiment (exact numbers are model dependent), the qualitative features of the comparison in Fig. 2 are understood within this picture.

In Fig. 3, the Al $L_{2,3}VV$ APEC spectrum in coincidence with the primary $2p$ photoelectron spectrum taken at 200-eV photon energy is shown. The dotted curve is the singles spectrum referenced to the same zero as the coincidence spectrum. The solid curve superimposed on the coincidence spectrum was produced by subtracting a constant background from the singles spectrum (to bring its leading edge to zero) and then scaling the remainder to have the same peak height (at 68 eV) as the coincidence spectrum. Only minor differences exist between the coincidence and the scaled singles spectra. At high kinetic energies, there is a slight enhancement of the threshold emission near 69 eV. This is interpreted as enhanced emission from decay events involving shallow valence-band surface states in the more surface-sensitive coincidence spectrum. In addition, a small shift of the weak plasmon-loss peak to higher kinetic energy is interpreted as a shift in weight from bulk to surface-plasmon loss in the coincidence spectrum. Below 55 eV, the coincidence spectrum is slightly suppressed compared to the singles spectrum, but at 51.8 eV, the lowest possible energy for two-hole final states, it is still 85% of the singles intensity. The point of Fig. 3, and of this paper, is how little difference there is between the singles (solid curve) and coincidence (open circles) spectra. Most importantly, contrary to the above expectations, the relative intensity in the LET region is only slightly reduced in coincidence. If the arguments of the previous paragraph applied, and if the LET were exclusively due to extrinsic in-

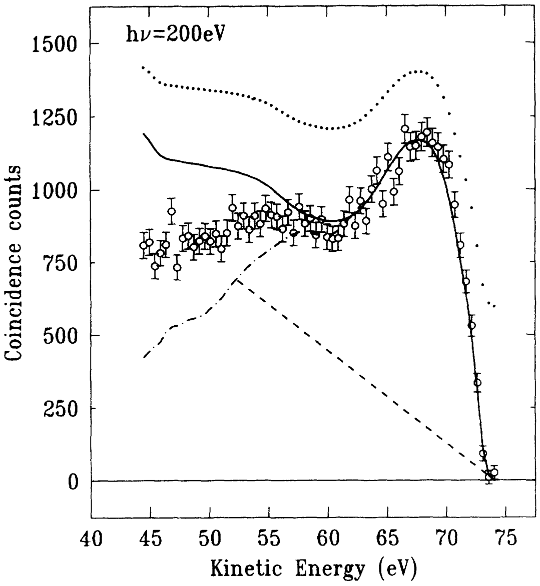

FIG. 3. The Al(111) $L_{2,3}VV$ Auger spectrum taken at 200-eV photon energy. The dotted line is the noncoincidence (singles) spectrum referenced to zero counts. The circles are the coincidence data (fixed analyzer at 127 eV, the peak in the photoelectron spectrum). The solid line is the singles spectrum, with a constant background (from inelastically scattered $2p$ photoelectrons) subtracted. The two spectra are normalized to the same height at 68 eV. Note the very slight difference between the spectra, especially at 51.8 eV, where ordinary two-hole decay is forbidden by energy conservation. The dotted-dashed curve is the coincidence spectrum minus the difference between the singles and coincidence spectra, and is intended to represent a conservative estimate of the intrinsic spectrum. The dashed line is a smooth curve similar to that shown in Fig. 1, the area above which is intended to approximate the two-hole decay contribution to the estimated intrinsic spectrum (see text). The dashed and dotted-dashed curves are included only to show the importance of many-electron decay events; too many approximations are included in their generation for any belief in details of their line shapes.

elastic scattering, then the coincidence spectrum ought to be attenuated by 50% or more.

The procedure for obtaining the solid curve in Fig. 3 requires some justification. The constant background that was subtracted from the singles spectrum is mainly from inelastically scattered $2p$ photoelectrons. These electrons are not part of the intrinsic or extrinsic Auger spectrum with which we are concerned and, as evidenced by the zero count rate at the high-energy threshold of the coincidence spectrum, will not appear in the coincidence spectrum with primary (i.e., unscattered or elastically scattered) $2p$ photoelectrons. Furthermore, to the small extent that we err by subtracting a constant background, we err conservatively. The true background from the $2p$ line will increase with decreasing kinetic energy, so the singles Auger spectrum without this contribution has an even *lower* LET and exhibits an even *smaller* difference from the coincidence spectrum than what is shown in Fig. 3. The overall legitimacy of this subtraction has been checked by changing the photon energy to 100 eV. At that photon energy, the $2p$ photoemission line has a lower energy than the LVV spectrum, and the $2p$ secondaries do not energetically overlap the LVV spectrum. Although we did not have to do any subtraction to see that the LET is nearly as intense in coincidence as in singles spectrum, the $2p$ line itself overlaps the LET making more detailed analysis of the 100-eV spectrum difficult.

The most straightforward interpretation of the lack of attenuation of the LET by the coincidence technique is that the LET is due to events intrinsic to CVV decay; however, other possible explanations need to be considered. The LET might, for instance, be generated by inelastic scattering within the energy analyzer. We have ruled out this possibility by measuring the scattering within the analyzer: it is much too small to account for the LET. Another possibility is that the LET is due to inelastic scattering at the sample surface. As discussed above, such scattering would not be attenuated by the coincidence process. However, the surface-plasmon-loss peak in Fig. 3 is too small to account for the LET, and we know of no other surface-specific loss processes. A third possibility is that the probabilities of escape of the two electrons are not independent. One could imagine, for example, that the Auger electron follows the photoelectron out of the solid. This is unlikely because of the time scales; the photoelectron can move about 500 Å in an Auger lifetime. Since the electron mean free path is only 10 Å, the photoelectron has either been ejected from the solid or dissolved into the Fermi sea before a typical Auger electron is emitted, and any correlation between the two paths is unlikely.[28] Finally, if the excitation profile is strongly peaked near the sample surface, then one would expect no difference between the coincidence and singles spectra. This has been ruled out by the optical measurements of Ref. 26 that show that the light penetrates about 1000 Å.

In the absence of other viable explanations, we conclude that much of the LET is generated intrinsic to the core-hole decay. We note that a similar LET is observed in positron-excited Cu $M_{2,3}VV$ Auger spectra where core-hole creation is confined to the first atomic layer of the solid.[29] A large intrinsic contribution to the LET implies that the core-hole decay process generates other excitations that share energy with the Auger electron. These other excitations could be either plasmons or electron-hole pairs. Plasmon losses typically have characteristic peaks at integral multiples of the zero-momentum plasmon energy, 15 eV for Al, leading to a plasmon-loss peak at 54 eV in the Al Auger spectra. This peak is only weakly evident in the spectra of Fig. 3; one concludes that plasmons are not the dominant LET excitation. The alternative explanation is electron-hole pairs. The generation of electron-hole pairs as an intrinsic part of CVV decay is simply a complex way of saying that more than two electrons have participated in that decay; the Auger final state is not a two-valence hole, one-electron state, but an n-valence hole, $n-1$-electron state, with $n>2$.

It is difficult to quantify just what fraction of the decay events are of this, hereafter called n hole, type. A lower limit can be set by the following observations. As discussed above, at least 50% of the extrinsic part of the LET should be suppressed by coincidence. Then a rough, but conservative, estimate of the intrinsic part of the spectrum can be generated by subtracting the difference between the singles and the coincidence spectra from the coincidence spectrum of Fig. 3. This procedure generates the dashed-dotted curve in Fig. 3, which we will refer to as the intrinsic spectrum. Now note that two-hole decay events must account for all of the Auger intensity at threshold (73 eV) and must have kinetic energy greater than 51.8 eV, with zero intensity at this lower limit. A curve delineating the two-hole part from the n-hole part of the spectrum must satisfy these criteria and maintain a continuous spectrum. If the probability for creation of n-hole states is significant, then their contribution will rapidly rise from zero as the kinetic energy decreases from 73 eV since the phase space for such decays increases. In the absence of a well-defined theory suggesting the details of this line shape, we use a linearly increasing function which equals zero at 73 eV and is equal to the estimated intrinsic spectrum at 51.8 eV. This curve is given by the dashed line in Fig. 3. Any reasonable deviation from this approximation will not change our main conclusion, only numerical details. The discontinuous nature of the slope at 51.8 eV is unphysical, but also is not important at this level of analysis. The area above this curve represents the two-hole part of the intrinsic spectrum and is at most 50% of the total estimated intrinsic spectrum. Furthermore, significant intrinsic intensity at 45 eV implies that there is additional spectral weight at lower energies. We estimate that the two-hole part of the intrinsic spectrum carries only about 35% of the spectral weight, with a precise figure depending on the details of the background subtraction and on the continuation of the spectrum to lower kinetic energies.

The possibility of multielectron Auger decay is not merely ignored by the conventional picture of CVV decay; it is forbidden. When the initial and final states for Auger decay are described by single determinants in the same one-electron basis—the standard independent-electron approximation—the two-electron nature of the Coulomb operator forbids transitions to final states with more than two holes. To get many-electron *CVV* decay, one must explicitly deal with electron-electron correlations. These manifest themselves in two ways: core-valence (*CV*) and valence-valence (*VV*) correlations. *CV* correlations describe the interaction of the valence electrons with the core hole. They are often described in one-electron terms as the response of the valence electrons to the core-hole potential. *CV* correlations are responsible, e.g., for the famous edge enhancement in soft x-ray emission (SXE) spectra.[30] Subsequently, severe distortions in core-core-valence (*CCV*) Auger spectra were observed, and explained as *CV* correlation effects.[31] In *CVV* Auger spectra, *CV* correlations can affect line shape, transition rates, and LET generation. The distortion of *CVV* spectra by *CV* correlation is typically much weaker than that of *CCV* spectra because the final state contains no core hole.[31] Recently, detailed calculations of the effect of *CV* correlation on *CVV* spectra of wide-band solids have been made.[12,22] According to these calculations, line shape and transition rates can be strongly affected by *CV* correlations, but no significant LET is predicted. We conclude that *CV* correlations alone are not the cause of the LET, and that *VV* correlations or a combination of *CV* and *VV* correlations are responsible. *VV* correlations describe the true many-body properties of the valence band, such as plasmons and quasiparticle lifetimes, *VV* correlations are responsible for peak shifts, line broadening, and plasmon-loss peaks observed in valence-band photoemission spectra, among many other interesting phenomena. For example, a recent x-ray photoelectron diffraction (XPD) study of Al (Ref. 32) observed angular variations of the valence-band photoemission intensity that are similar to the XPD pattern of the Al 2*s* core level and attributed this observation to localization of the valence hole. In principle, *VV* correlations can cause plasmon-loss peaks, a LET, shape and transition rate changes to *CVV* spectra. The effect of *VV* correlations on *CVV* spectra have not been calculated, although there are some calculations of plasmon satellite intensity in the literature.[33] The calculated intensities vary widely.

We have presented data and arguments in support of the conclusion that much of the LET observed in *CVV* Auger decay from wide-band solids is an intrinsic part of the Auger process, and due to electron correlation effects within the valence band. Fist and foremost, this conclusion is important because it changes the conventional interpretation of *CVV* decay in a qualitative way; contrary to the nomenclature itself, most *CVV* decay events leave the valence band in a state that contains more than two holes. The important conclusion is that two-hole decay events are in the minority. Second, since so much of the spectral weight occurs in a region forbidden to two-hole *CVV* decay, one must suspect that the line shape within the conventional primary region (the region energy allowed for two-hole decay, 51.8 to 73 eV for Al $L_{2,3}VV$) may be itself significantly perturbed. However, previous investigations have found that very good agreement between data and theory within the primary region can be achieved by simply subtracting the LET from the data.[1–13,16,22] Further theoretical work to find the effect of valence-electron correlations on the primary region as well as the LET of *CVV* spectra is required. Transition rates may also be affected. Note that similar discussions of the effect of electron-electron correlations in generating a LET in other electron spectroscopies such as SXE,[34] core-level photoemission spectroscopy,[25] and valence-electron photoemission spectroscopy[35] have been published. The LET in SXE is small, and due to both *VV* and *CV* correlations. In core-level photoemission it is large, and due to *CV* correlation, while in valence-electron photoemission it is large, and due to *VV* correlations.

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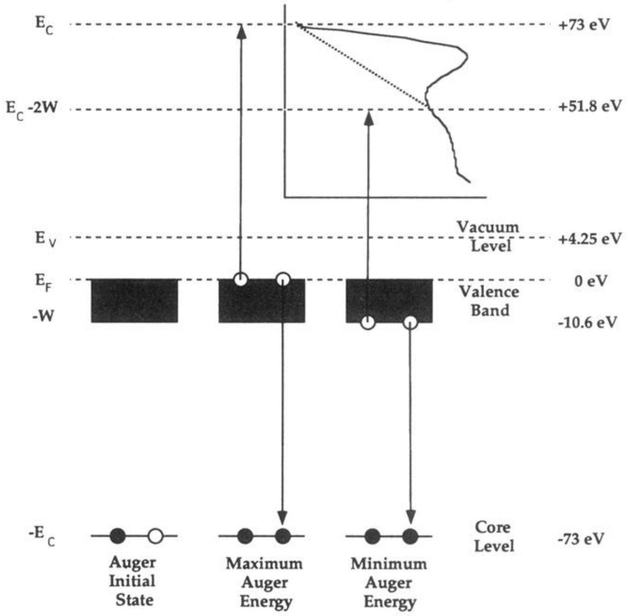

FIG. 1. Schematic illustration of the *CVV* process. The core-hole energy is E_c, the valence-band width is W, and the Fermi level is the zero of energy. The core hole, or Auger initial state, is shown on the left. Two possible Auger final states, those with maximum and minimum Auger-electron energy, are shown in the center. The Al $L_{2,3}VV$ spectrum is shown on the right. The LET in this spectrum is comprised of all emission at energies below 51.8 eV, plus some continuation into the higher-energy region (Ref. 20). The dashed line represents the approximation to this contribution