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THE  $L_{2,3}M_{45}M_{45}$  AUGER SPECTRA OF METALLIC COPPER AND ZINC:  
THEORY AND EXPERIMENT\*

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ABSTRACT

The  $L_{2,3}M_{45}M_{45}$  and  $L_{3,4}M_{45}M_{45}$  Auger spectra of clean metallic copper and zinc were measured. A theory was developed to predict the Auger energies. The theory employs experimental electron binding energies, which were also measured, two-electron integrals, and Hartree-Fock energies. It accounts for multiplet splitting in the  $d^8$  final state, predicting multiplet structure in excellent agreement with experiment in zinc and in very good agreement in copper. It also accounts for "static" atomic relaxation and for static extra-atomic relaxation (screening), which is related to the Friedel theory of alloys. The theory developed here predicts the Auger energies to within 1 eV in zinc and 2-3 eV in copper. Since atomic integrals were used, the success of the theory implies that an atomistic approach to Auger energies is valid for these metals. The magnitude of the extra-atomic relaxation energy ( $\sim 10$  eV) suggests that it may be a crucial factor in Auger energy shifts arising from chemical environment or surface condition.

## I. INTRODUCTION

The Auger effect has both fundamental interest as a relatively simple atomic process and very great practical importance because of its analytical sensitivity. Nevertheless there has been until now relatively little work directed toward a really quantitative understanding of the magnitudes of Auger energies in solids. Recently a previously-neglected relaxation effect<sup>1</sup> was combined with the final-state intermediate-coupling theory of Asaad and Burhop<sup>2</sup> to predict  $KL_1L_1$  Auger energies from empirical binding energies and atomic two-electron integrals.<sup>3</sup> This approach has been successful in predicting the positions of all nine components of the KLL Auger spectrum.<sup>4</sup> In the more precise measurements of Auger energies that are possible in the LMM spectra of transition metals, it should be possible to detect subtle shifts arising from extra-atomic relaxation of the type identified earlier in binding-energy studies.<sup>5</sup> In this paper we report LMM Auger spectra in Cu and Zn, and explain the Auger energies quantitatively in terms of atomic and extra-atomic relaxation.

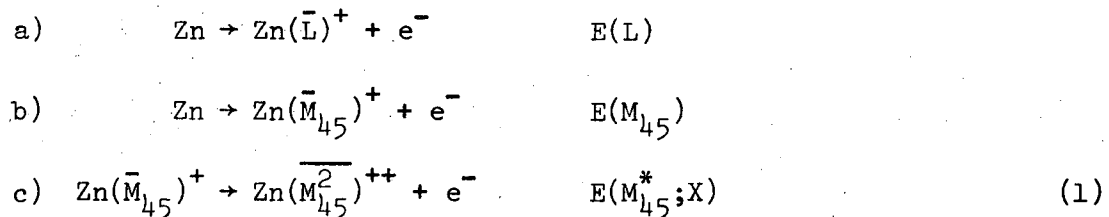
## II. EXPERIMENTAL

The experiments were carried out in a Hewlett-Packard 5950A ESCA Spectrometer modified to operate at a pressure of  $\leq 10^{-9}$  Torr after baking. High-purity single crystals of Cu and Zn were cleaned by argon ion bombardment until the surfaces were free of carbon and oxygen ( $< 0.3$  monolayer). The samples were irradiated with monochromatized Al  $K\alpha$  x rays and the LMM Auger electrons were analyzed. One-electron binding energies were also measured. In each case the LMM Auger spectrum had most of its intensity in three regions of kinetic energy. We shall discuss here only the most energetic of the three regions for both Cu and Zn, because the other two are not yet well-enough resolved to allow unambiguous data analysis. Thus we shall concentrate on the kinetic energy regions 900 - 950 eV for Cu and 970 - 1030 eV for Zn.

In each case the spectral area selected consists of two groups of lines separated by the  $L_2 - L_3$  binding-energy difference. Preliminary analysis shows that these can only be the  $L_3^{M_{45}M_{45}}$  and  $L_2^{M_{45}M_{45}}$  groups, in agreement with earlier analyses of the stronger  $L_3^{M_{45}M_{45}}$  group.<sup>6</sup>

## III. THEORY

The  $LM_{45}M_{45}$  Auger process can be written as the algebraic sum of three simple steps. In Zn, for example, these may be written



where the energy of each process is written on the right, and X denotes the term designation of the two-hole final state. Here steps (a) and (b) are simply the ionizations of an L or  $M_{45}$  electron, the energies of these steps are just the appropriate binding energies, referred to the Fermi energy. Our experimental binding-energy values are given in Table I. Step (c) is more complicated. It is also an  $M_{45}$  binding energy, but in this case with a hole already in the 3d shell (the  $M_{45}$  shell) in the initial state. The extent to which the first 3d hole can be regarded as localized, and the two final-state  $M_{45}$  holes coupled as if they belong to the same atom, is a priori a moot point for open 3d-shell transition metals. In Zn, however, the 3d shell is filled and well below the Fermi energy, so it should be safe to consider the 3d holes as localized. The validity of this approach for Cu would be more difficult to justify a priori, because the 3d "band" in Cu is bound by an average energy of only 3 eV relative to  $E_F$ . The Cu 3d band is also broader than a core level peak would be, and it extends up to within 2 eV of  $E_F$ . The two Cu 3d holes in the final state have nonetheless been treated as localized in the interpretation reported below, and this approach agrees well with experiment.

The energy of step (c) in Eq. (1) can be related to that of step (b), the one-electron binding energy  $E(M_{45})$ . Two additional terms are necessary, however. The first, denoted  $\mathcal{F}(M_{45}M_{45};X)$ , accounts for multiplet coupling in the final state X. The "multiplet" term is easily worked out for the  $d^8$  configuration using multiplet coupling theory<sup>7</sup> and Mann's Slater integrals.<sup>3</sup> The general expressions for  $\mathcal{F}(M_{45}M_{45};X)$  for the five final states that can be formed from  $d^8$  are<sup>7</sup>

$$\begin{aligned}
 ({}^1S) &= F^0 + \frac{2}{7} F^2 + \frac{2}{7} F^4 \\
 ({}^3P) &= F^0 + \frac{1}{7} F^2 - \frac{4}{21} F^4 \\
 ({}^1G) &= F^0 + \frac{4}{49} F^2 + \frac{1}{441} F^4 \\
 ({}^1D) &= F^0 - \frac{3}{49} F^2 + \frac{4}{49} F^4 \\
 ({}^3F) &= F^0 - \frac{8}{49} F^2 - \frac{1}{49} F^4 .
 \end{aligned} \tag{2}$$

Here the orbital notation is suppressed for brevity. The Slater integrals are  $F^k(3d,3d)$  for  $k = 0, 2, 4$ . The calculated values of  $\mathcal{F}(X)$  are given in Table II. The use of  $\mathcal{F}(X)$  alone would be equivalent to making a partial "frozen orbital" approximation, in which multiplet splitting in the two-hole final state is accounted for. The dynamic relaxation energy,  $E_R$ , accompanying photoemission is also taken into account implicitly, by using the empirical 3d binding energies. By dynamic relaxation energy we mean the amount by which the 3d binding energy is lowered from the orbital-energy (Koopmans' theorem<sup>8</sup>) estimate through relaxation of the passive orbitals during electron emission. From the optimized Hartree-Fock-Slater results of Rosén and Lindgren,<sup>9</sup>  $E_R$  has a value of 5.3 eV for a Cu 3d electron in atomic copper.



The second term that must be combined with  $E(M_{45})$  in order to give an accurate estimate of  $E(M_{45}^*;X)$  in Eq. (1) is a correction term accounting for an additional relaxation energy, which we shall denote as  $R(M_{45}M_{45})$ . This is a static relaxation energy. It is the amount by which the binding energy of the second 3d electron (step (c) of Eq. (1)) is reduced because its Hartree-Fock potential is made more repulsive when the passive electrons relax toward the hole left by the first 3d electron, in step (b) of Eq. (1). In the Hartree-Fock formalism  $R(M_{45}M_{45})$  would appear as a decrease in the magnitude of the orbital energy  $\epsilon$  of the second 3d electron after step (b). It is therefore termed a static relaxation energy to distinguish it from the dynamic relaxation energies  $E_R$  that occur during ionization in both step (b) and step (c), as discussed above. Of course this division into static and dynamic relaxation energies is an artifact of reaching the two-hole state by the hypothetical two-step process (steps (b) and (c) in Eq. (1)). Other paths would give different divisions: in a one-step ejection of both electrons, for example, the relaxation energy would be completely of the "dynamic" variety. The total relaxation energy (and the Auger energy) is of course independent of the path chosen for its calculation. In most Auger-energy calculations, including this one, the two-step path is chosen to take advantage of the fact that the one-electron binding energies are known.

Having considered these two contributions to step (c), we can now write  $E(M_{45}^*;X)$  as

$$E(M_{45}^*;X) = E(M_{45}) + \mathcal{F}(M_{45}M_{45};X) - R(M_{45}M_{45}) \quad (3)$$

To calculate  $R(M_{45}M_{45})$  we shall first formally divide it into two parts,

$$R(M_{45}M_{45}) = R(M_{45}M_{45})_a + R(M_{45}M_{45})_e, \quad (4)$$

where the atomic static relaxation term  $R(M_{45}M_{45})_a$ , is the static relaxation energy, discussed previously<sup>1,4</sup> that a free copper or zinc atom would have, and  $R(M_{45}M_{45})_e$  is an additional extra-atomic relaxation energy that arises from electronic relaxation toward the 3d hole from the surrounding lattice. It is relatively straightforward to estimate  $R(M_{45}M_{45})_a$  because, following the work of Hedin and Johansson,<sup>10</sup> this term should be twice the dynamic relaxation contribution  $E_R(M_{45})$ .<sup>1,4,5</sup> Using Rosén and Lindgren's value of 5.3 eV quoted above for Cu, we therefore have

$$R(M_{45}M_{45};Cu)_a \cong 10.6 \text{ eV}.$$

No SCF results for a 3d-hole state in Zn are available, to our knowledge. We could just use the above estimate for Zn as well, with little additional error, but it is also possible to improve on this estimate somewhat. The atomic relaxation energy can be divided into inner-shell, intrashell, and outer-shell contributions. Following Hedin and Johansson,<sup>10</sup> we infer that the first two should be essentially the same for  $Cu(3d^{10}4s)$  and  $Zn(3d^{10}4s^2)$ . The additional outer-shell relaxation energy arising from the additional 4s electron in Zn may be estimated from Mann's integrals,<sup>3</sup> using the equivalent-cores method,<sup>1,5</sup> as

$$\begin{aligned} & [F^0(3d\ 4s) - \frac{1}{10} G^2(3d\ 4s)]_{\text{gallium}} - [F^0(3d\ 4s) - \frac{1}{10} G^2(3d\ 4s)]_{\text{zinc}} \\ & = 1.7 \text{ eV} \end{aligned}$$

We thus estimate

$$R(M_{45}M_{45};Zn)_a \cong 12.3 \text{ eV} .$$

The extra-atomic term is more difficult to estimate with any degree of reliability. This term should be quite sensitive to the environment, and probably the most important consequence of our analysis is to call attention to the magnitude of this environment-sensitive term in the Auger energy. In the cases at hand we are dealing with metals, and it seems safe to assume that the extra unit of charge induced on one atom in the periodic lattice by the appearance of a hole in the filled 3d shell will be completely screened by the conduction electrons. This assumption is the cornerstone of much of the theory of alloys. A formal statement is given by the Friedel sum rule<sup>11,12</sup>

$$z = \frac{2}{\pi} \sum_L (2L + 1) \eta_L(k_F) . \quad (5)$$

Here  $z|e|$  is the excess charge on an impurity atom in an otherwise periodic lattice, and  $\eta_L$  is the resultant phase shift induced in the  $L^{\text{th}}$  conduction-electron partial wave at the Fermi momentum  $k_F$  by the impurity, regarded as a scattering potential. A Zn atom with a 3d hole in a Zn lattice has  $z = 1$ . The phase shifts  $\eta_L(k_F)$  must therefore give a positive sum in Eq. (5), such that a total of one state falls below the Fermi energy and becomes occupied.

The Friedel sum rule is a general self-consistency condition on the lattice potential. As such it must be applicable to this case--a localized 3d hole in the Zn lattice. Because of its generality, however, the sum rule is of limited value in the explicit calculation of  $R(M_{45},M_{45})_e$ . Furthermore, the

potential in this problem is more diffuse than in most alloy problems, because of the radial extension of the 3d hole relative to a "point" charge at the nucleus. We can, however, estimate the screening length of an electron gas for a point charge from the Fermi-Thomas model as<sup>13</sup>

$$\lambda \sim (E_F/6\pi n e^2)^{1/2} \quad (6)$$

This estimate gives  $\lambda \sim 0.5 \text{ \AA}$  for both Cu and Zn. Such a short screening length implies that the localized 3d hole must be almost totally screened by the induced screening charge on the hole-state atom itself. If we assume this to be the case we can estimate an upper limit for  $R(M_{45}, M_{45})_e$ . From the positions of Cu and Zn in the Periodic Table, it is expected that the s and p partial waves should do most of the screening. In the vicinity of the 3d-hole atom the atomic components of these screening states should be approximately described by atomic orbitals having principal quantum number  $n = 4$ . Thus an upper limit for  $R(M_{45}, M_{45})_e$  would be given by the electrostatic energy of interaction between a 3d electron and a 4s or 4p electron,

$$R(M_{45}, M_{45})_e \cong F^0(3d, 4s) - \frac{1}{10} G^2(3d, 4s)$$

or

$$F^0(3d, 4p) - \frac{1}{15} G^1(3d, 4p) - \frac{3}{70} G^3(3d, 4p) \quad (7)$$

Using Mann's tables,<sup>3</sup> we find 9.6 eV and 11.0 eV for the 3d,4s interactions in atomic Cu and Zn, respectively, and 9.6 eV for the 3d,4s interaction in atomic gallium, the first element with a 4p electron in its ground state. Since the nominal configurations of Cu and Zn are  $d^{10}s$  and  $d^{10}s^2$ , we shall use the 3d,4s energy for estimating  $R_e$  in Cu and the (gallium) 3d,4p energy for  $R_e$  in Zn.

These energies are expected to be upper limits, because the "4s" and "4p" conduction electron states are probably more diffuse than these atomic states, but the small screening lengths imply that they are not gross overestimates of  $R_e$ . Combining the two relaxation energies, we have

$$R(M_{45}M_{45}) \lesssim 20.2 \text{ eV} \quad (8)$$

for both Cu and Zn. Combining this with Eqs. (1) - (3), and using the energies in Tables I and II, we have calculated the theoretical Auger energies

$$E(L_{2,3}M_{45}M_{45};X) = E(L_{2,3}) - 2E(M_{45}) - \cancel{F}(M_{45}M_{45};X) + R(M_{45}M_{45}) \quad (9)$$

The results are set out in Table III.

## IV. DISCUSSION

Experimental Auger spectra over the regions of interest are shown in Fig. 1, and experimental values of the Auger energies are also listed in Table III for comparison with the theoretical values. Agreement between experiment and our theoretical values is excellent. It appears that Eq. (9) provides, for the first time, a framework within which it is possible to predict quantitative values of Auger energies in metals in some detail.

Our treatment of the Zn spectrum is on sounder theoretical ground than in the case of Cu, because in Zn the 3d shell is well below  $E_F$ , hence certainly localized. In both Zn and Cu the main lines in the  $L_2M_{45}M_{45}$  and  $L_3M_{45}M_{45}$  groups are separated by exactly the  $L_2 - L_3$  separation to within 0.1 eV. In Zn the positions of the components within each group are also predicted with high accuracy. The main line in such case is interpreted as arising from the three levels  $^3P$ ,  $^1G$ , and  $^1D$ , which are predicted to be nearly degenerate. Since the relative intensities of these three components are unknown, we can only say that the experimental energy of the main  $L_3M_{45}M_{45}$  line,  $991.8 \pm 0.2$  eV, agrees exactly with the predicted energy of the  $^1G$  component (991.9 eV) and is up to 1.3 eV lower than that of the highest-energy component,  $^1D$  (993.2 eV). The two extreme levels of  $d^8$ ,  $^1S$  and  $^3F$ , are also in nearly exact agreement with experiment for the Zn ( $L_3M_{45}M_{45}$ ) group and nearly as good for the Zn ( $L_2M_{45}M_{45}$ ) group, with a maximum discrepancy of only 0.6 eV. The agreement of the multiplet structure within each group is very good evidence for the validity of the multiplet coupling part of this theory. Considering that the relaxation correction of 20.2 eV could hardly be expected to be accurate to better than 10%, the agreement in absolute energy between experiment and our theory is better than we could reasonably expect. We conclude that the Zn spectrum is completely explained.

Copper has more theoretical uncertainties a priori. The width of the d band and its proximity to  $E_F$  weaken the quantitative validity of using localized atomic functions. Thus copper is a test case for studying the extent to which an atomistic approach will give reasonable results in spite of theoretical ambiguities. In this context the agreement between theory and experiment in Table III is very good. The  $L_3M_{45}M_{45}$  and  $L_2M_{45}M_{45}$  groups have exactly the right separation, as mentioned above. The experimental energy of the main line in each group is lower than theory by 2 to 3 eV. This is entirely within the range that we could expect. Delocalization of the d orbitals would tend to decrease electron-electron electrostatic interactions, thus reducing both  $R(M_{45}M_{45})_a$  and  $R(M_{45}M_{45})_e$ . This would decrease the observed Auger energies relative to our atomistic theoretical values, as observed. Better agreement could be obtained if the screening charge in Cu arises mainly from a positive phase shift in the p wave. By extrapolating Mann's  $3d,4p$  integrals to the zinc position, to simulate the interaction for Cu with a 3d hole, we would estimate  $R(M_{45},M_{45})_e = 8.2$  eV, a reduction of 1.4 eV from the estimate used in Table III. This would reduce the discrepancy for copper to only  $\sim 1$  eV. We are disinclined to accept this interpretation, however, because the s wave probably does much of the screening in Cu.

The multiplet structure in Cu is very similar to that expected for  $d^8$ . If the experimental and theoretical spectra are brought into agreement by reducing the latter by 2.5 eV (i.e., by reducing  $R(M_{45},M_{45})$  from 20.2 eV to 17.7 eV), then the main peak in each group can be assigned as the  $^3P$ ,  $^1G$ ,  $^1D$  triad. The  $^3F$  peak falls nicely into place as a shoulder in each case. There is also a peak at the  $^1S$  position in each group, in perfect agreement with the

predicted energy. However, in each group there is also a small additional peak between the  $^1S$  peak and the main peak. The additional peaks fall at different points in the multiplet patterns, and they would be difficult to explain as part of the multiplet structure. Clearly they deserve further study.

In summary, this work demonstrates that it is possible to understand the  $L_{2,3}M_{45}M_{45}$  Auger spectra of Cu and Zn in considerable detail. A theoretical approach was developed that accounts for multiplet splitting, static atomic relaxation, and static extra-atomic relaxation (screening). The theory predicted Auger spectra in excellent agreement with experiment, especially in the case of Zn. Extra-atomic relaxation was found to add about 10 eV to the Auger energies in these metals. Thus future work might profitably be directed toward further exploration of this relaxation energy, by studying Zn and Cu Auger spectra in insulators, for example. It may also be possible to observe separated Auger lines from atoms in the surface and bulk regions of a metal, or to separate the spectra of surface atoms bound to adsorbed species from bulk spectra, because of differences in the extra-atomic static relaxation energy.



FOOTNOTES AND REFERENCES

\* Work performed under the auspices of the U. S. Atomic Energy Commission.

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Table I. Core level binding energies (eV)<sup>a</sup>

Metal	E(L <sub>2</sub> )	E(L <sub>3</sub> )	E(M <sub>45</sub> )
Cu	952.6(2)	932.8(2)	3.0(1)
Zn	1044.9(2)	1021.8(2)	9.9(1)

<sup>a</sup>Error in tenths place appears parenthetically.

Table II. Multiplet energies  $\mathcal{E}(M_{45}M_{45};X)$  for  $3d^8$ , in eV<sup>a</sup>

Element	X = $1S$	$3P$	$1G$	$1D$	$3F$
Cu	31.4	26.3	26.9	25.9	23.9
Zn	35.2	29.4	30.2	28.9	26.7

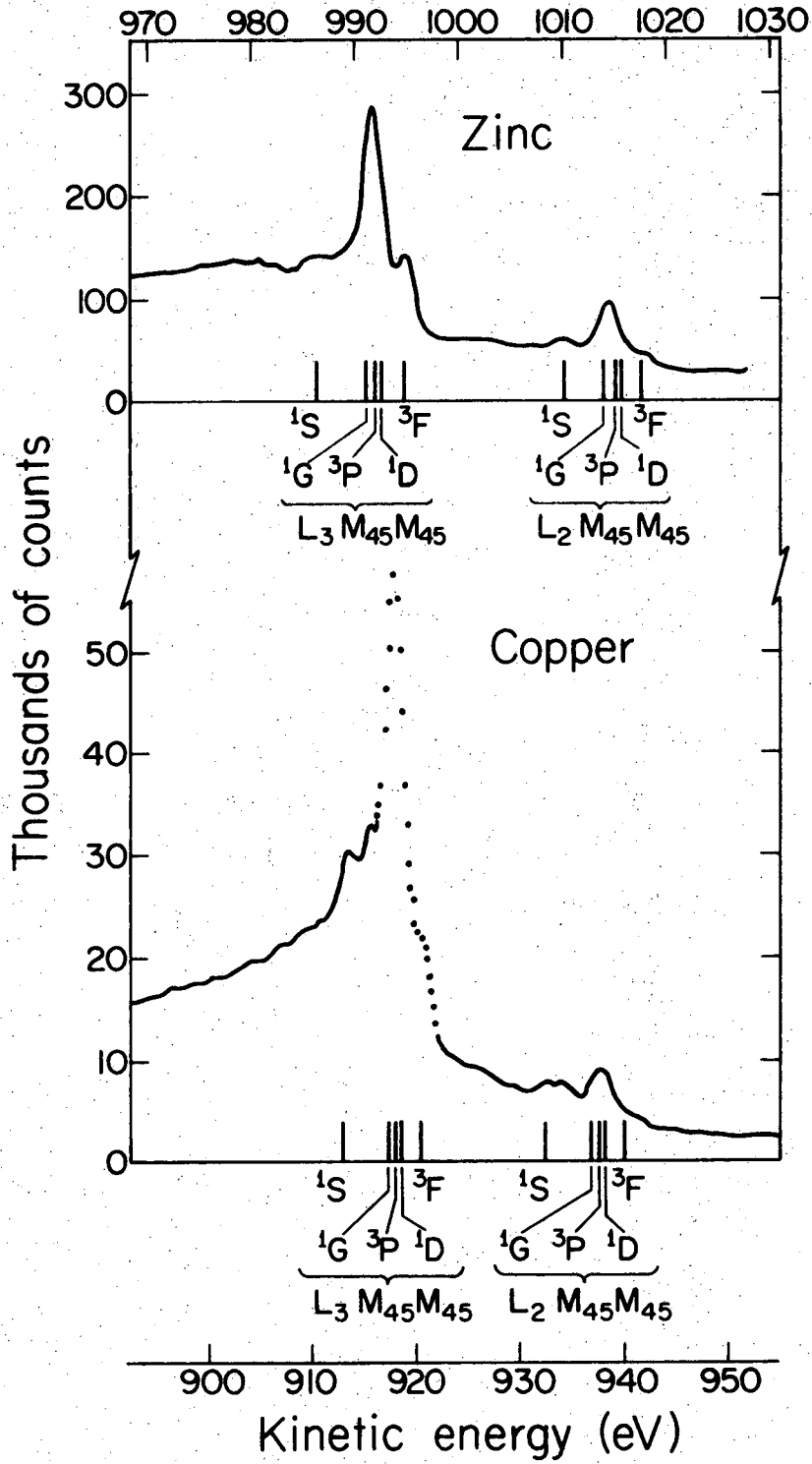
<sup>a</sup>Using  $F^0$ ,  $F^2$ ,  $F^4$  values from Ref. 3.

Table III.  $L_{2,3}M_{45}M_{45}$  Auger energies in Cu and Zn (eV)

Final State, X	Copper		Zinc	
	Expt.	Eq. (9)	Expt.	Eq. (9)
$L_{2}M_{45}M_{45}$	$1S$	(932.8) (934.2)	935.4	1010.4(3) 1010.0
	$3P$		940.5	1015.8
	$1G$	937.8(2)	939.9	1014.9(2) 1015.0
	$1D$		940.9	1016.3
	$3F$	940.6	942.9	1017.9(3) 1018.5
$L_{3}M_{45}M_{45}$	$1S$	(913.6) (915.8)	915.6	986.6(3) 986.9
	$3P$		920.7	992.7
	$1G$	918.0(2)	920.1	991.8(2) 991.9
	$1D$		921.1	993.2
	$3F$	920.6	923.1	995.2(3) 995.4

FIGURE CAPTION

Fig. 1. Experimental  $L_{2,3}$ ,  $M_{45}$ ,  $M_{45}$  Auger spectra from copper and zinc. Kinetic energy scales are given relative to the Fermi energy. Multiplet structures from theory described in text are shown as lines under spectra. They have been moved down in energy relative to values in Table III by 0.5 eV (Zn) and 3 eV (Cu) to facilitate comparison of multiplet structure with experiment.



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Fig. 1

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