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To cite this version:

E. Chaînet, J. Derrien, R. Cinti, T. Nguyen, M. De Crescenzi. EXTENDED FINE STRUCTURES IN THE AUGER SPECTRA. Journal de Physique Colloques, 1986, 47 (C8), pp.C8-209-C8-212. <10.1051/jphyscol:1986839>. <jpa-00226161>

HAL Id: jpa-00226161
https://hal.archives-ouvertes.fr/jpa-00226161

Submitted on 1 Jan 1986

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EXTENDED FINE STRUCTURES IN THE AUGER SPECTRA

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Resume - Les structures fines observées dans le spectre Auger de divers matériaux (Fe, Co, Ni, Cu) ont été exploitées avec le formalisme d'EXAFS. Leurs transformées de Fourier permettent de déduire la distance interatomique des premiers voisins qui est en bon accord avec les résultats obtenus par la technique EXAFS effectuée à l'aide du rayonnement synchrotron.

Abstract - We have observed extended fine structures above core-valence-valence Auger transitions on several materials (Fe, Co, Ni, Cu). We analyse these oscillating structures following the same formalism used in EXAFS. The lattice parameters obtained by Fourier transform are in good agreement with those deduced from synchrotron radiation EXAFS data.

INTRODUCTION

Auger electron spectroscopy (AES) is the most currently used technique in order to get information on the chemical composition of the outermost layers of the surface. However the AES technique so far could not provide easily structural information on the investigated samples. The lack of this structural potentiality can now be bridged over thanks to the findings we present in this paper. We demonstrate here that fine structures related to a core-valence-valence (CVV) Auger transition are detectable in Auger spectra/1/. They extended on several hundreds of eV above the investigated CVV Auger lines. We interpret these extended fine Auger structures (EXFAS) as originated from the same interference process which produces extended x ray absorption fine structures (EXAFS) usually observed in the x ray photoabsorption spectra /2/. From the Fourier analysis of our EXFAS spectra, we obtain the radial atomic distribution $F(R)$ of the samples, conferring therefore to the conventional AES technique new capabilities as a surface sensitive structural probe. This is a new approach to local geometry determination to be compared to other fine structures recently observed in energy loss spectra (EELFS) /3/. Fig. 1 schematizes various fine structures found in the electron yield distribution, near the core edge region (as used in EELFS technique) and above Auger transition (as used in EXFAS technique discussed here).

RESULTS AND DISCUSSION

Single crystals, polycrystals and evaporated films were prepared in a ultra-high vacuum chamber. Fig. 2 (a) shows the extended fine Auger structure spectra for a clean Cu(111) surface as a function of the kinetic energy. The upper part corresponds to the conventional $dN(E)/dE$ Auger mode (first derivative $N'(E)$). The lower part corresponds to the second derivative $d^2N(E)/dE^2$ mode ($N''(E)$) in order to improve the fine structure-to-background ratio. The spectra display several extended fine features far from the $M_{23}$ VV Auger lines.
These structures have been observed by other authors /4/ who associated these features to a diffraction effect and no quantitative structural analysis has been achieved. In contrast, we assign these oscillating structures to the same interference process giving rise to EXAFS oscillations. Using therefore the EXAFS analysis procedure, the raw data were background subtracted (dotted line in Fig. 2(a)) and Fourier integrated choosing the $E_0$ threshold energy at the inflection point of the $M_{23}$ VV first derivative Auger line. Figs 2(b) and 2(c) show the magnitude of the Fourier transforms $F'(R)$ and $F''(R)$ spectra of the $N'(E)$ EXAFS and $N''(E)$ EXAFS respectively. The first peak in the two $F(R)$'s is located at $2.20 \pm 0.03$ Å and should correspond to the $2.55$ Å distance between the first nearest-neighbours atoms in the Cu(111) f.c.c. sample. Similar results for Fe and Ni are presented in Fig. 3 and 4 respectively. Fig. 5 shows the Fourier transform of extended fine Auger structures.

**Fig. 1** - Schematic $N(E)$ electron distribution reflected from a surface excited by an electron beam.

**Fig. 2** - (a) EXFAS of Cu(111) detected in the first $N'(E)$ and second $N''(E)$ derivative mode. (b) $F'(R)$ deduced from $N'(E)$. (c) $F''(R)$ deduced from $N''(E)$. The Fourier integral was extended between 3.5 and 9.5 Å⁻¹.

**Fig. 3** - EXFAS of polycrystalline Fe (first derivative and integral) with the corresponding Fourier transforms.
detected for the cobalt film above the MVV Co Auger lines in both the first and second derivative modes. Their corresponding Fourier transforms are sketched in Fig. 5(a) and 5(b) respectively. In order to compare with EXAFS /5/ and EELFS experiments /6/ above the same $M_{23}$ edge, we also draw in Fig. 5(c) and 5(d) the corresponding $F(R)$. The agreement is excellent and the first peak in the $F(R)$ reflecting the first neighbours in the h.c.p. cobalt is located at $2.05 \pm 0.03 \text{Å}$ for all three spectroscopic techniques (EXFAS, EXAFS and EELFS). Corrected for the phase shift /6/, it gives exactly the $2.50 \pm 0.03 \text{Å}$ distance as found from x ray diffraction on the h.c.p. cobalt.

We summarize in Fig. 6 the mechanisms underlying the physical origin of the fine structures observed in Auger spectra. Fig. 6(a) reminds the well-known EXAFS mechanisms. Fig. 6(b) explains the usual Auger process. In this CVV Auger transition, one (valence) electron fills an initial core hole and a second electron (Auger) is emitted from the solid. The kinetic energy of the escaping Auger electron is given by:

$$E_{k}^{(CVV)} = E(C) - E(V_1) - E(V_2)$$

(1)

neglecting the final state hole-hole interaction. The initial core hole in the Auger process was created under primary electron irradiation (energy $E_p$). The ejected core electron could fill either the empty states $\Delta$ above $E_F$ (excited states in the sample) or/and get out into the vacuum. Now if the first electron filling the initial core hole does not come from the valence band as in the CVV Auger process (Fig. 6(b)) but from another state at energy $\Delta$ above $E_F$ (Fig. 6(c)) initially filled by the ejected core electron, then the escaping Auger electron will gain an energy $\Delta + E(V_1)$ according to :

$$E_{k}^{(CAV)} = E(C) + \Delta - E(V_2) = E_k + \Delta + E(V_1)$$

(2)
Moreover if the excited $\Delta$ states of the first electron are EXAFS like modulated in energy, then the measured Auger kinetic energy of the second (valence) electron will contain entirely the EXAFS structures. So we are probing the EXAFS like interference effect experienced by the first electron in its excited state $\Delta$ through the emission of the second (valence) electron. All excited $\Delta$ states above $E_F$ with an energy lower than $\Delta_{\text{max}} = E_p - E_{(C)}$ can be filled by this technique. More detailed calculations will be published elsewhere /7/.

In conclusion, extended fine structures detected in the CVV Auger spectra are interpreted as due to an EXAFS-like effect experienced by one of the two electrons which enter in the Auger process. These EXFAS features confer to the conventional Auger technique new capabilities in the surface structural determination.

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