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A UPS STUDY OF THE INTERACTION OF POTASSIUM WITH CARBON MONOXIDE AND BENZENE ON THE Pt(III) SURFACE

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Authors

Kudo, M.
Garfunkel, E.L.
Somorjai, G.A.

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A UPS STUDY OF THE INTERACTION OF POTASSIUM WITH CARBON MONOXIDE
AND BENZENE ON THE Pt(111) SURFACE

M. Kudo*, E.L. Garfunkel** and G.A. Somorjai

Materials and Molecular Research Division
Lawrence Berkeley Laboratory
and
Department of Chemistry,
University of California
Berkeley, CA 94720, U.S.A.

* Institute of Industrial Science, University of Tokyo, 7-22-1
Roppongi, Minato-ku, Tokyo 106, Japan.

**Department of Chemistry, Wright Riemann Laboratory, Rutgers University,
New Brunswick, New Jersey 08903, U.S.A.

A UPS STUDY OF THE INTERACTION OF POTASSIUM WITH CARBON MONOXIDE
AND BENZENE ON THE Pt(111) CRYSTAL SURFACE

M. Kudo, E.L. Garfunkel, and G. A. Somorjai

Materials and Molecular Research Division,
Lawrence Berkeley Laboratory
and
Department of Chemistry,
University of California, Berkeley, CA 94720 USA

ABSTRACT

The interaction of potassium with carbon monoxide and benzene has been studied on the Pt(111) crystal surface by ultra-violet photoelectron spectroscopy (UPS). The adsorptive changes reported in previous studies for carbon monoxide and benzene when potassium is coadsorbed are correlated with the UPS results presented here, and are explained with the aid of a molecular orbital analysis. We find that the valence molecular orbitals increase their binding energy slightly when the potassium is coadsorbed, implying a model in which the adsorbates sense the potassium induced changes in dipole field at the surface.

INTRODUCTION

The physical and chemical properties of alkali metal adatoms on transition metal surfaces are of major interest in surface science because of their promoting effects in catalysis¹. For instance, in the ammonia synthesis potassium increases the rate of reaction by increasing the probability for the dissociative adsorption of nitrogen⁶. For CO hydrogenation reactions, potassium slows the overall rate of reaction, but induces a favorable change in selectivity towards higher molecular weight species⁷⁻¹⁰. Alkali compounds are also used as additives to catalysts for methanol synthesis, and ethylene epoxidation.

Despite almost a century of practical industrial use, the atomic scale behavior of alkali additives has only recently begun to be unraveled²⁻⁵. In recent studies we demonstrated how potassium interacts with various coadsorbed molecules^{2,11}. The molecules which exhibited the most significant changes in chemisorption were carbon monoxide and benzene. The desorption temperature of CO was increased by up to 200 K when potassium was coadsorbed. For benzene, on the other hand, the maximum desorption

temperature decreased by 200 K upon the coadsorption of. A qualitative molecular orbital model was presented explaining these results. In the model we postulated that the positions of the adsorbate molecular orbitals with respect to the Fermi level of the metal substrate are very important in determining to what extent the molecule is affected by potassium. The interaction was assumed to be mediated by the substrate, i.e. direct bonding interactions between the potassium and adsorbate species were less significant. Instead it was proposed that a change in the electrostatic dipole layer at the surface (as evidenced by the change in work function) causes a change in the potential level of the adsorbate orbitals relative to the Fermi energy. This in turn, causes a greater or lesser interaction between the free electron charge density of the metal and the partially occupied adsorbate molecular orbitals lying near the Fermi energy. In this paper we present new ultraviolet photoelectron spectroscopic (UPS) results which further support this model.

EXPERIMENTAL

The experiments were performed in a standard diffusion-pumped ultrahigh vacuum (UHV) chamber (base pressure 1×10^{-10} Torr) equipped with facilities for Auger electron spectroscopy (AES), photoelectron spectroscopy (both UPS and XPS), and mass spectroscopy. A double pass CMA (Physical Electronics 15-155G) was used for electron energy analysis. It also contained a LEED system, a potassium dosing gun, an ion sputtering gun for sample cleaning, and leak valves for gas dosing. The AES and UPS data

were obtained with the aid of a PET-Commodore microcomputer and all the data processing were carried out with this computer. The details of the experimental conditions and the thermal and structural properties of potassium monolayers on Pt(111) are described elsewhere¹².

The valence d-band in transition metals is located within 8 eV of the Fermi level (E_f). Emission below the d-band comes from either the broad s-p band, an adsorbate level, a core level or from "secondary electrons" (those resulting from multiple scattering de-excitation processes). The secondary electron edge is usually 2-3 eV wide and its position is determined by drawing a line tangent to the inflection point and noting where it intersects the baseline. Then the workfunction (ϕ) is determined by subtracting the width of the emission spectra from the photon energy (21.2 eV for He(I)).

RESULTS

K on Pt(111)

In Fig. 1 we show the UPS spectra obtained for potassium on Pt(111) as a function of potassium coverage. The count rate at the secondary electron emission peak in the high coverage potassium surfaces is about five times that of the clean platinum surface, for that reason we have normalized the y-axis for the display. Several spectral features should be noted. First, there is a sharp drop in electron emission at the Fermi level. This

drop is observed in the presence of all adsorbates on Pt(111), as well as on other metals. Two other features observed upon alkali adsorption are an increase in the total secondary electron yield and an increase in the width, E_w , of the spectrum. Figure 2 shows the work function versus potassium coverage. The potassium coverage was determined by using AES peak intensities as calibrated with LEED and TDS elsewhere¹². The coverage value corresponding to room temperature saturation is assigned a value of 1. It corresponds to a $(\sqrt{3} \times \sqrt{3})R30^\circ$ structure with an atomic ratio of 1:3.

As is seen in this figure, the work function decreases with increasing potassium coverage and has a minimum of 1.2 eV at $\Theta_k = 0.5$. Beyond this coverage, the work function begins to increase again with increasing K coverage and reaches a constant value of 1.9 eV. The relation between the surface dipole (μ) and the initial work function change ($\Delta\phi_0$) is expressed by the Helmholtz equation:

$$\Delta\phi_0 = 4\pi/\mu N$$

where N is the coverage of the adsorbed species. Using this equation the value of 2μ for the potassium adsorption system was found to be 13 Debye per K atom at low coverages, somewhat smaller than the value of 18.8 D found by other authors.⁵

CO+K on Pt(111).

In Fig. 3 and Fig. 4 we show the He(I) UPS spectra for CO adsorbed on Pt(111) with and without coadsorbed potassium. On the clean Pt(111) surface, the adsorption of CO is accompanied by several changes in the UPS spectra. As with potassium, CO caused a drop in intensity for the peak lying just below E_f . This is accompanied by the growth of two peaks located at -5.3 and -9.5 eV. For low CO coverages, there is also a peak at -12 eV. This peak is no longer detectable in the difference spectra at higher coverages because a broad peak resulting from increased secondary electron background emission appears near -14.5 eV.

When CO is coadsorbed with potassium, the main features of the spectra seem to be a combination of the results for clean potassium and clean CO overlayers. In Fig. 5 the UPS difference spectra for 0.5 L CO exposures are shown. The peak at -5.3 eV appears not to shift, although by $\Theta_K=0.65$ it has disappeared in the difference spectrum. The peak at -9.4 eV broadens further into a doublet at -8.5 and -9.8 eV. The peak at -12.3 eV shifts to about -13 eV, but becomes undetectable in the more intense secondary electron cascade. The work function upon CO exposure decreases slightly on clean Pt(111), while it increases at medium and higher potassium coverages. The extent of work function increase becomes smaller at the higher potassium coverages, see Fig. 6, corresponding to a decrease in total coverage of CO on the platinum surface.

Benzene + K on Pt(111)

Figures 7-9 show the UPS spectra for benzene coadsorbed with various coverages of potassium on Pt(111). Benzene adsorbed on clean platinum causes a large decrease in emission from the peak just below E_F (Fig. 7), similar to what was observed with carbon monoxide and potassium. Three new overlapping peaks grow up between -3 and -10 eV, labeled #1, #2 and #3 in Figs. 8 and 9. These peaks are also observed for benzene on other metals such as Ni¹³ and Ru¹⁴, and can be associated with the σ and π levels of gas phase benzene, as described in the discussion.

Several interesting changes occur as potassium is added. Peaks #1 and #3 shift to higher binding energy, (see Fig. 9). There are also changes in their relative intensities: for higher potassium coverages, peak #1 and #3 become much larger than peak #2.

Also note the large decrease in work function with increasing benzene exposure on the clean Pt(111) surface, supporting the idea that benzene is a donor on metals¹⁵. The work function on the potassium covered surface decreased upon benzene adsorption, but the extent of the decrease was smaller at higher potassium coverage. This is due in part at least to the fact that the amount of adsorbed benzene becomes less in this circumstance.

DISCUSSION

Blyholder¹⁶ suggested that the bonding of CO to a metal involved not only a 5σ orbital mixing with metal s and p orbitals, but also d-orbitals from the metal backdonating into the 2π (CO) orbital. Since the conjugate d (M)- 2π (CO) orbitals are bonding between the metal atoms and the carbon, and antibonding between the carbon and the oxygen, the backdonation of metal electrons into the 2π orbital leads to a simultaneous strengthening of the M-C bond(s) and weakening of the C=O bond. For many metal-CO systems, the electron orbital levels can be approximated as shown in Fig. 10.

It is reasonable to try to correlate the chemisorption behavior, including the extent of backdonation, with electronic properties of the surface. Nieuwenhuys¹⁷ has shown that by comparing CO adsorption on various metals, the extent of backdonation can be roughly correlated with the work function of the surface. The degree of backdonation has been monitored by several techniques including UPS (the $4\sigma-1\pi$ separation)¹⁸, HREELS and IR (the CO stretching frequency)^{2,19}, surface Penning ionization spectroscopy (SPIES)²⁰ and ELS (the $\pi-2\pi$ and $n-2\pi$) transitions)²¹.

We can explain the enhancement of backdonation upon potassium adsorption by considering the effect that a change in work function has in the energy position of the molecular orbitals relative to the Fermi level. The work function of a metal can be divided into two components: the bulk chemical potential, and a surface dipole potential,²² . The surface dipole potential can itself be divided into two components: one arising from the spillover of the bulk metal wave functions into the vacuum, and a second resulting from the dipole field created by a polarized adsorbate and its image charge²³ . In our coadsorption experiments, we change primarily the surface dipole component of the work function by adsorbing potassium, an electron donor.

Upon CO adsorption a bonding $d-2\pi$ orbital is created. The exact location of the charge within this diffuse orbital will be strongly effected by the surface dipole. If we assume that the surface dipole determines the relative occupation of the metal and 2π parts of this new orbital, i.e. its energy position relative to the Fermi level, then in the potassium free, high work function case, most of the electrons should be localized on the platinum atoms. On the potassium covered, low work function surface, however, the electrons in the $d-2\pi$ orbital would become less localized on platinum, shifting their charge density more onto the carbon and oxygen positions, giving the conjugate orbital more 2π character. Others have analogously explained this by noting that the gas phase 2π reference level should become lowered relative to the surface levels when the dipole field is changing²¹ , lowering the work function.

From molecular orbital theory, we know that the extent of mixing between two overlapping orbitals is inversely proportional to their energy separation. If in our case, by decreasing the surface dipole field we bring the 2π gas phase levels closer to the Fermi level, then the overlap between the 2π level and the metal orbitals should increase. In this model, the molecular orbitals of the adsorbate (at least the 2π level) are not "pinned" to the Fermi level, but move as the electrostatic fields at the surface are changed.

CO adsorption

In the UPS difference spectra we note a peak at around -5.3 eV, see Figs. 3 and 5. This does not correspond to any molecular CO level. This feature has been observed by others for adsorption on platinum²⁴ as well as other metals, and is thought to be due to changes in the d-band. The CO 2π level is known to exist at about -2 eV below E_F , but it cannot be unambiguously observed by UPS. (Its position and occupancy on nickel however, have recently been observed using SPIES^{20,25}). The first (in order of decreasing energy) molecular orbitals of CO that are detected by UPS are the 1π and 5σ levels which appear as overlapping peaks at -9.4 eV. As potassium is added, the peak broadens and a shoulder develops at -8.5 eV. This feature may be considered to result from a shift in the 1π level towards lower binding energy¹⁸. The 4σ level, on the other hand, appears to move from

-12 to almost -13 eV, although its position is just obscured by the large change in the secondary electron edge. We believe that this shift is real since it has also been seen in two other similar studies^{18,5}, the second using He(II). This means that the $4\sigma-1\pi$ spacing increases. Since backdonation into the 2π level will increase the distance between the carbon and oxygen atoms, the 1π bonding level should be destabilized, moving it up in energy^{17,18}. The shift in position of the 4σ level to higher binding energy cannot be interpreted similarly, since it is a nonbonding level. This shift does, however, agree with our initial state argument that the binding energy of the CO levels should increase relative to the Fermi level when the dipole field is changed by potassium.

For moderate potassium coverages, the work function was found to increase with increasing CO exposure, while on the potassium free surface, CO caused the Pt(111) work function to decrease. This behavior can be understood from the donor acceptor model of CO. If the main bonding mechanism on the clean Pt(111) surface is a 5σ bonding donation into the metal and electron transfer to the platinum exceeds backdonation into the 2π CO orbital, then a slight drop in work function should occur as observed experimentally. On the lower work function potassium covered surface, electrons can be donated more easily into the 2π level. Since the 2π CO orbital is initially unoccupied, its filling will result in a large dipole being created between the CO molecule and the surface, with the CO behaving as an electron acceptor. It is interesting to note that in the HRBELS

experiment, vibrational frequencies as low as 1400 cm^{-1} were measured, similar to the gas phase value of 1375 cm^{-1} for a negatively charged CO molecule. If the CO vibrational frequency is any indication of the charge in the 2π level, then it is not surprising that the adsorption of CO on a potassium covered surface results in a large increase in the work function.

Benzene adsorption

In a previous study we showed that upon heating a benzene overlayer deposited on Pt(111), some of the benzene would desorb intact, while the remainder would dissociate yielding hydrogen gas and carbon layer¹¹. Much more of the adsorbed benzene was found to desorb intact if the surface had been pre-dosed with potassium. This was demonstrated both from the larger benzene thermal desorption peak area, as well as the smaller fraction of carbon which remains on the surface (as detected by AES) after heating. In addition, we observed lower temperatures for the benzene desorption peaks as potassium was added. Thus, desorption and decomposition should be viewed as competing reaction pathways. The desorption energy (and pathway), however is the one most strongly affected by potassium coadsorption.

Both the decrease in desorption temperature and increased amount of molecular desorption imply that the benzene-platinum bond is weakened when potassium is present. In the previous TDS study we offered a qualitative molecular analysis of this result based on the molecular orbital diagram for benzene-chromium. In Fig. 11 we present a correlation diagram for benzene approaching a threefold site on a cluster or surface. (A more detailed analysis of this system is presented elsewhere²⁶.) The qualitative features of the two systems are similar although the exact bonding interactions are changed somewhat. Of interest here are the molecular orbitals involved near the Fermi level, i.e., the highest occupied and lowest unoccupied molecular orbitals. All of the filled orbitals of the benzene-metal bond are either bonding or non-bonding between the benzene π -ring and metal d-orbitals. The lowest unoccupied level, however, is anti-bonding between the benzene and chromium orbitals. Consequently, if electrons were added to the system, or similarly if the benzene levels were shifted down (by a change in surface dipole) relative to the metal levels, then the E_{1g}^* anti-bonding level would become populated, weakening the metal-benzene interaction.

The qualitative features of the benzene-metal calculations are confirmed by experiments, in particular, by angularly resolved and photon polarized UPS studies²⁷⁻³⁰. In both the gas phase and adsorbed cases, there is a symmetric coordination of benzene to the metal atom(s), and the benzene is considered as an electron donor. Two interesting features appeared in our UPS studies. First, the shift to higher binding energy of the

benzene peaks No. 1 and No. 3 with potassium. This is consistent with the suggestion that the benzene levels, or more precisely the benzene vacuum reference level²¹, is moving down relative to the potassium free case, because the surface dipole changes. potassium should shift the peaks to lower binding energy. That the peaks move to higher binding energy also implies that the E_{1g}^* level should now be closer to the Fermi level. All of the gas phase benzene orbitals become broadened when adsorbed on the surface, so it is reasonable to consider partial occupation. It is not necessary to require that the levels be either fully occupied or empty. Thus, a continuous decrease in the work function could lead to an increasing occupation of the E_{1g}^* level. This would then result in a continuous weakening of the metal-benzene bond as was observed by TDS.

The second observation is that peaks No. 1 and No. 3 show an increase in intensity relative to peak No. 3. We can assign the peaks by comparison with the organometallic complex and adsorbed phase studies²⁷⁻³⁰. Peak No. 1 is the benzene $e(1g)$ orbital and peak No. 3 contains contributions from 4 different levels, one of which is the $A_{2u}(\pi)$ orbital while the other three are σ -type orbitals. Peak no. 2 is thought to represent the $E_{2g}(\sigma)$ level. It is tempting to suggest that the π -derived orbital peaks might be the only ones

whose intensity is enhanced by potassium adsorption, while the σ -level intensities remain constant. Confirmation of this and a more complete orbital analysis will require angularly resolved and photon polarized UPS studies.

ACKNOWLEDGEMENTS

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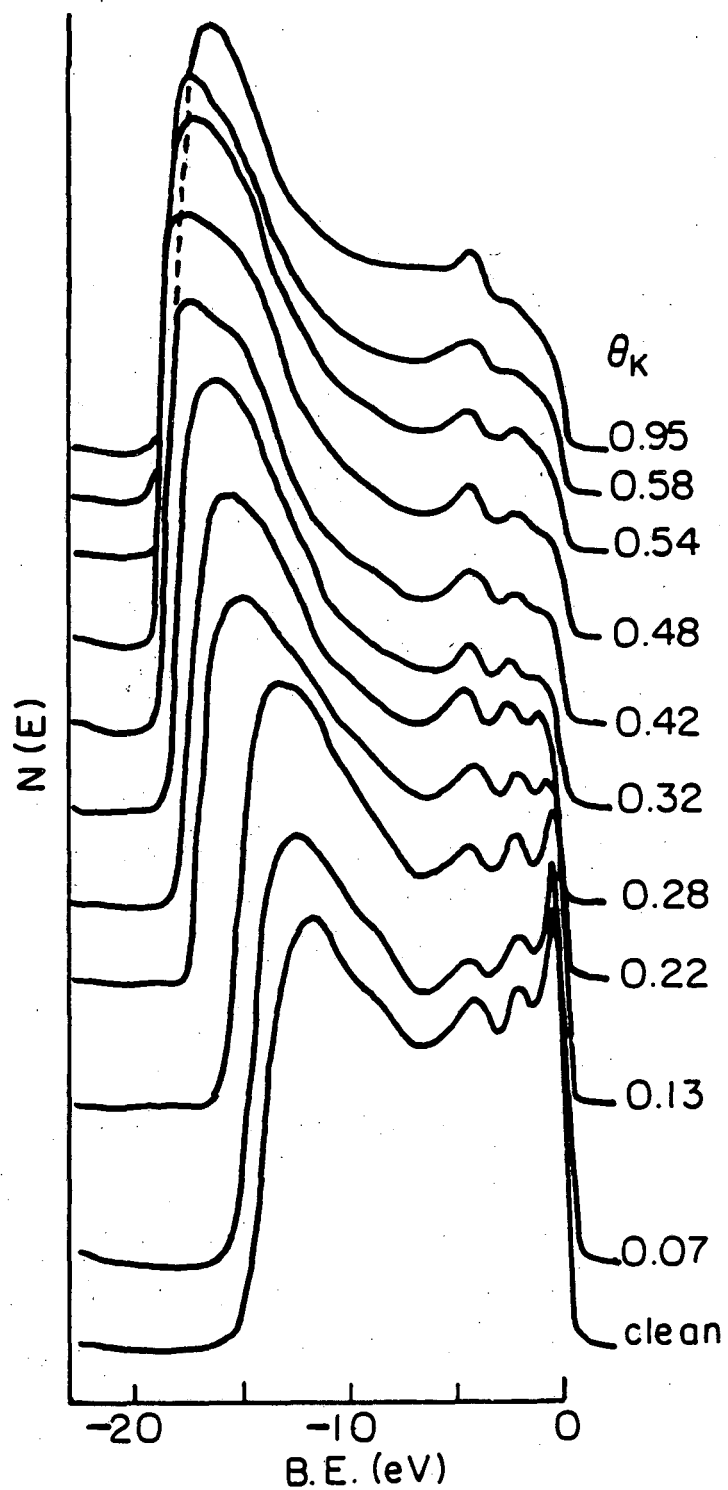
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FIGURE CAPTIONS

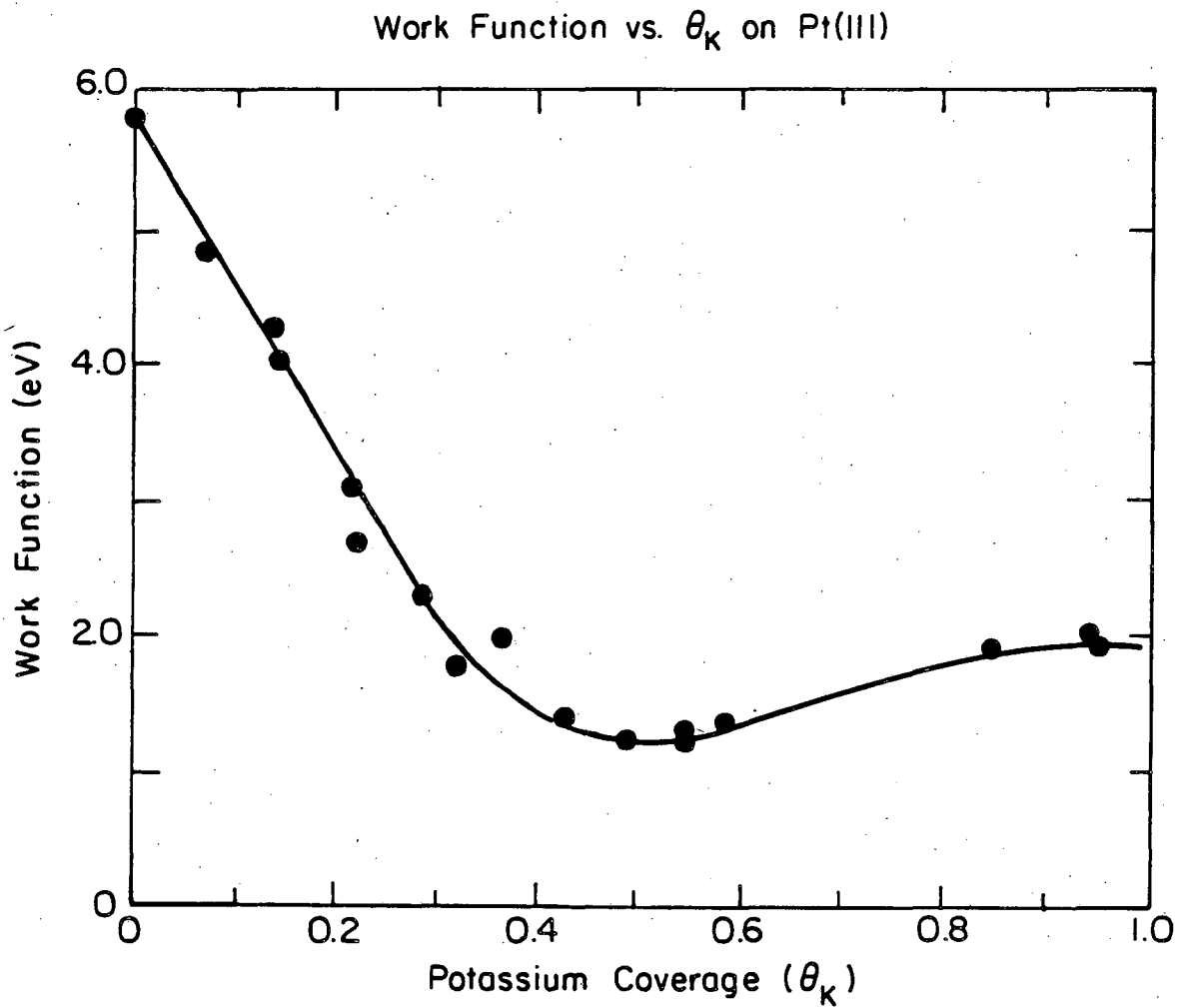
- Fig. 1 UPS spectra from potassium covered Pt(111).
- Fig. 2 Work function change versus potassium coverage on Pt(111).
- Fig. 3 UPS spectra from CO adsorbed on Pt(111). The right half shows difference spectra obtained by using the spectrum from clean Pt(111).
- Fig. 4 UPS spectra from CO and potassium coadsorbed on Pt(111).
- Fig. 5 UPS difference spectra for CO exposure of 0.5 Langmuirs on the potassium covered Pt(111).
- Fig. 6 Work function change on the potassium covered Pt(111) surface versus CO exposure.
- Fig. 7 UPS spectra of benzene adsorbed on Pt(111).
- Fig. 8 Typical UPS spectra of benzene adsorbed on the potassium-covered Pt(111).
- Fig. 9 UPS difference spectra of benzene adsorbed on the Pt(111) surface covered with various amounts of potassium.
- Fig. 10 CO bonding to a transition metal surface (associatively adsorbed state).
- Fig. 11 Sketch of correlation diagram for benzene-chromium complex²⁶. Distance, z , is measured between the benzene and chromium planes.

UPS on Pt(111)



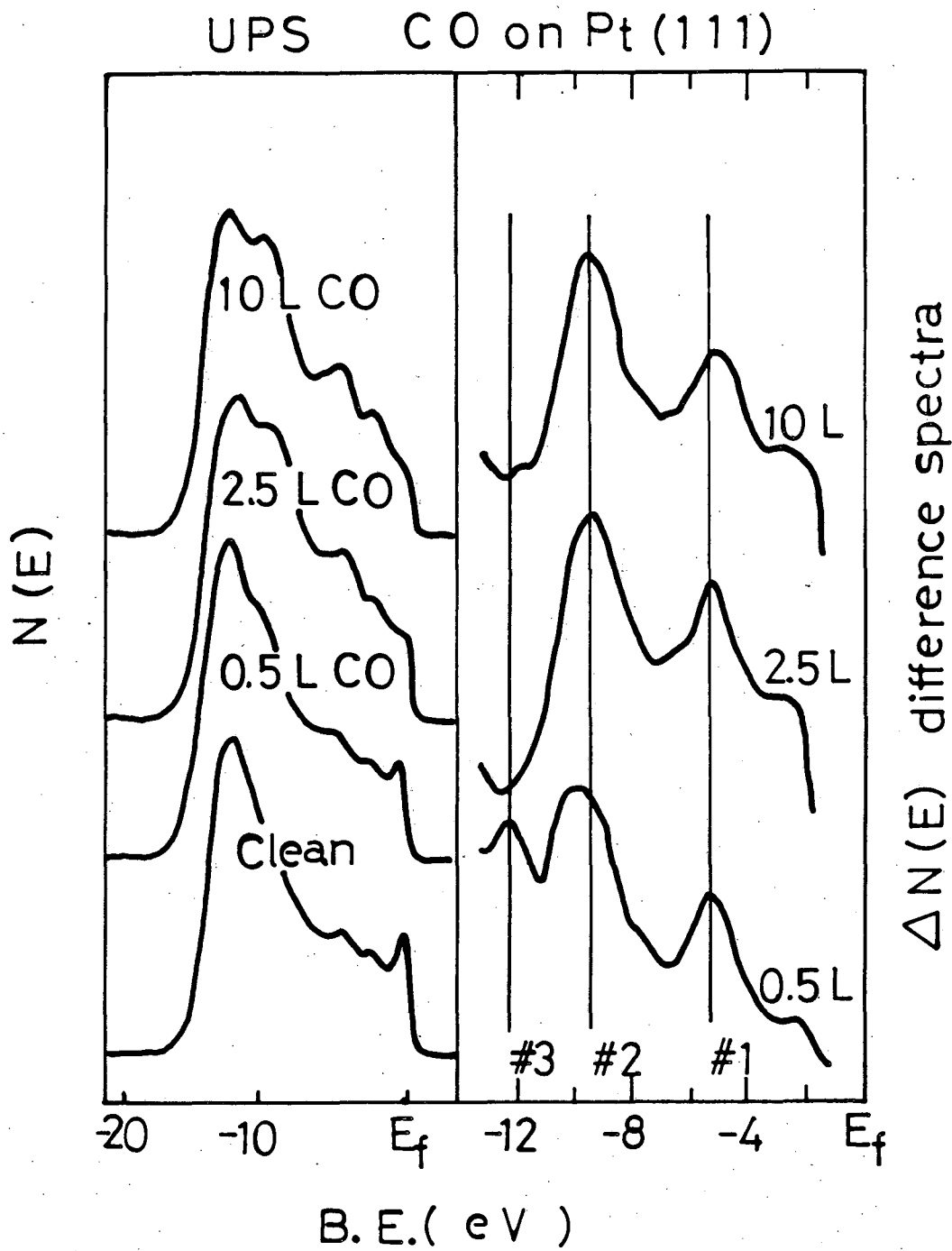
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Fig. 1



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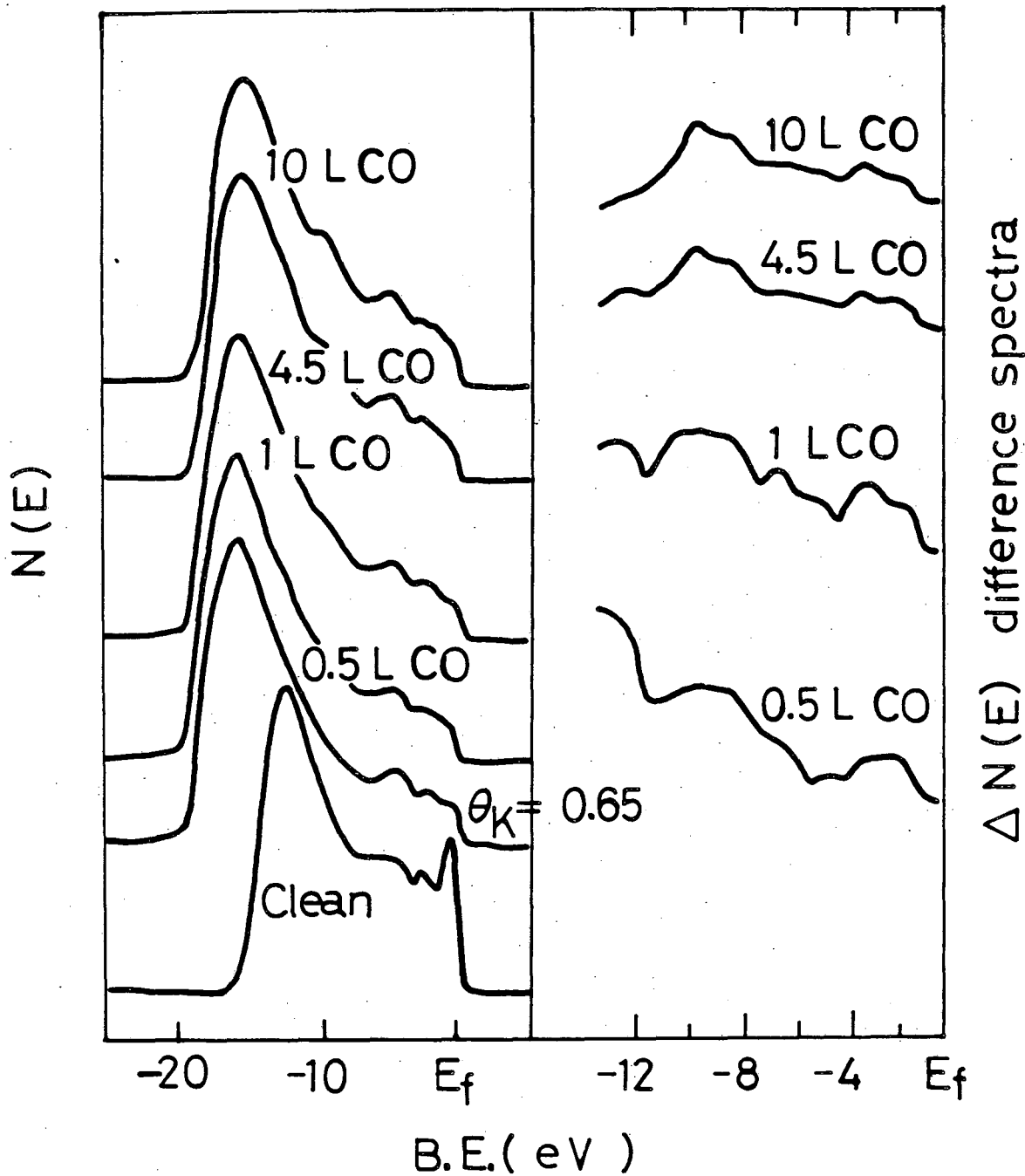
Fig. 2



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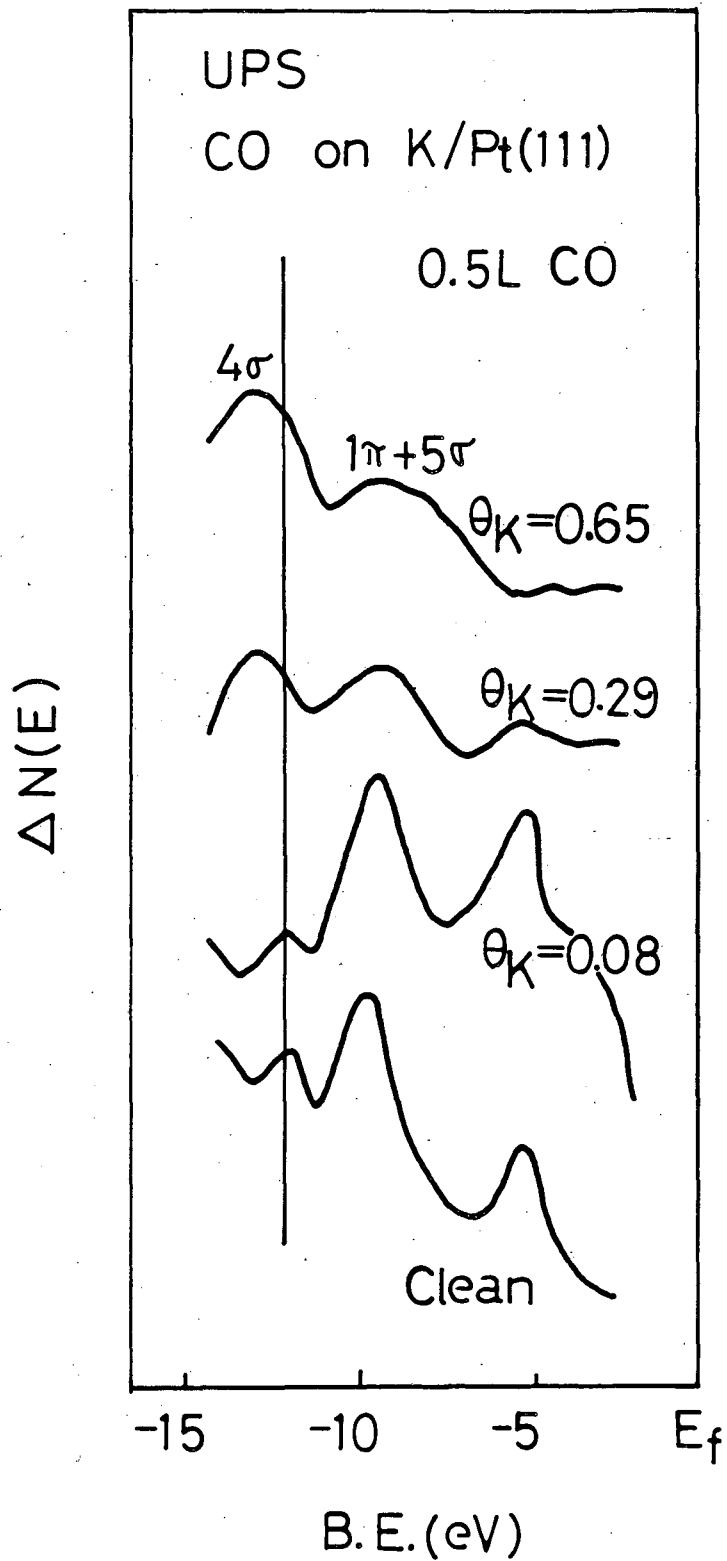
Fig. 3

UPS CO on K/Pt (111)
 $\theta_K = 0.65$



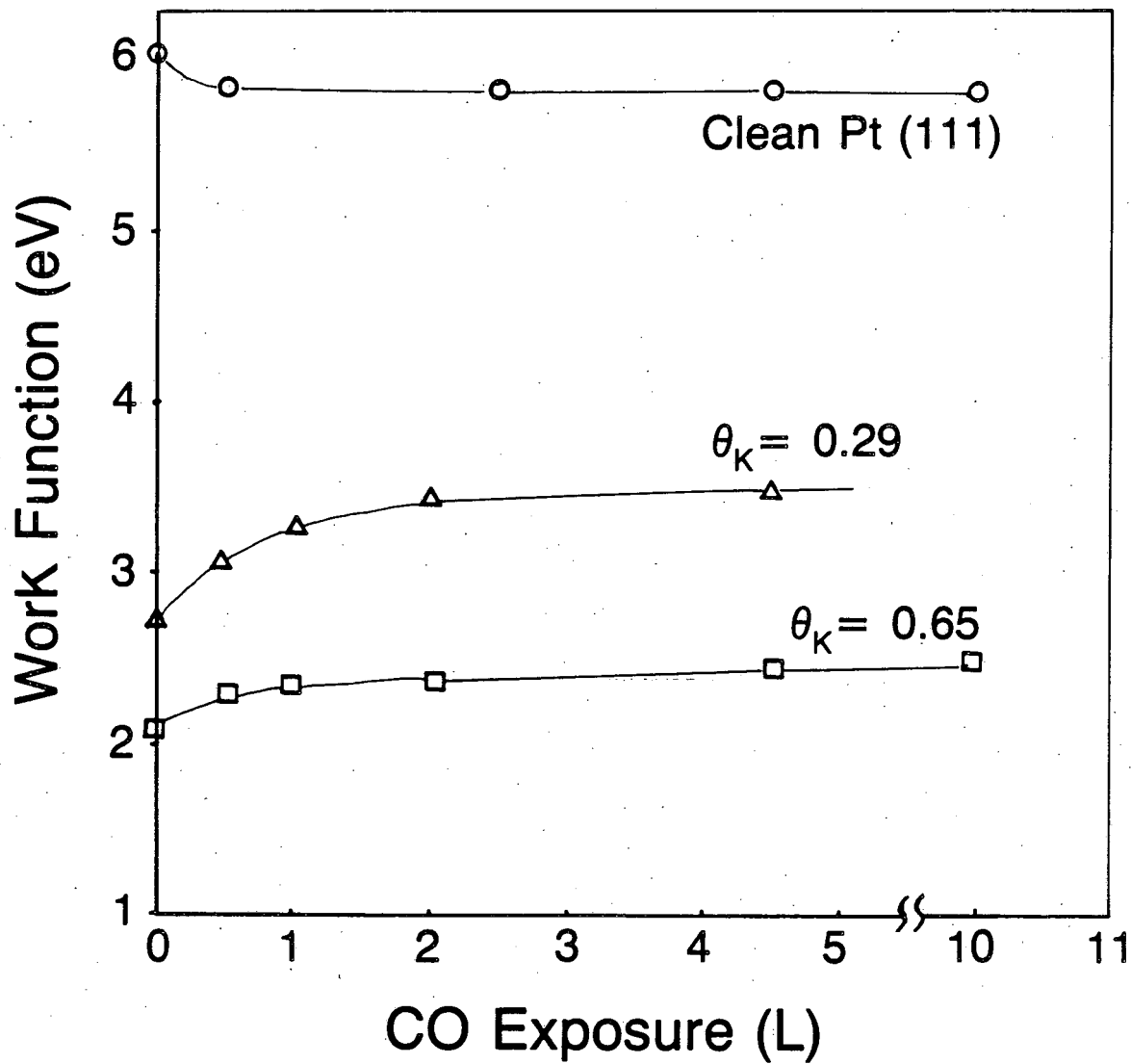
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Fig. 4



XBL 849-3980

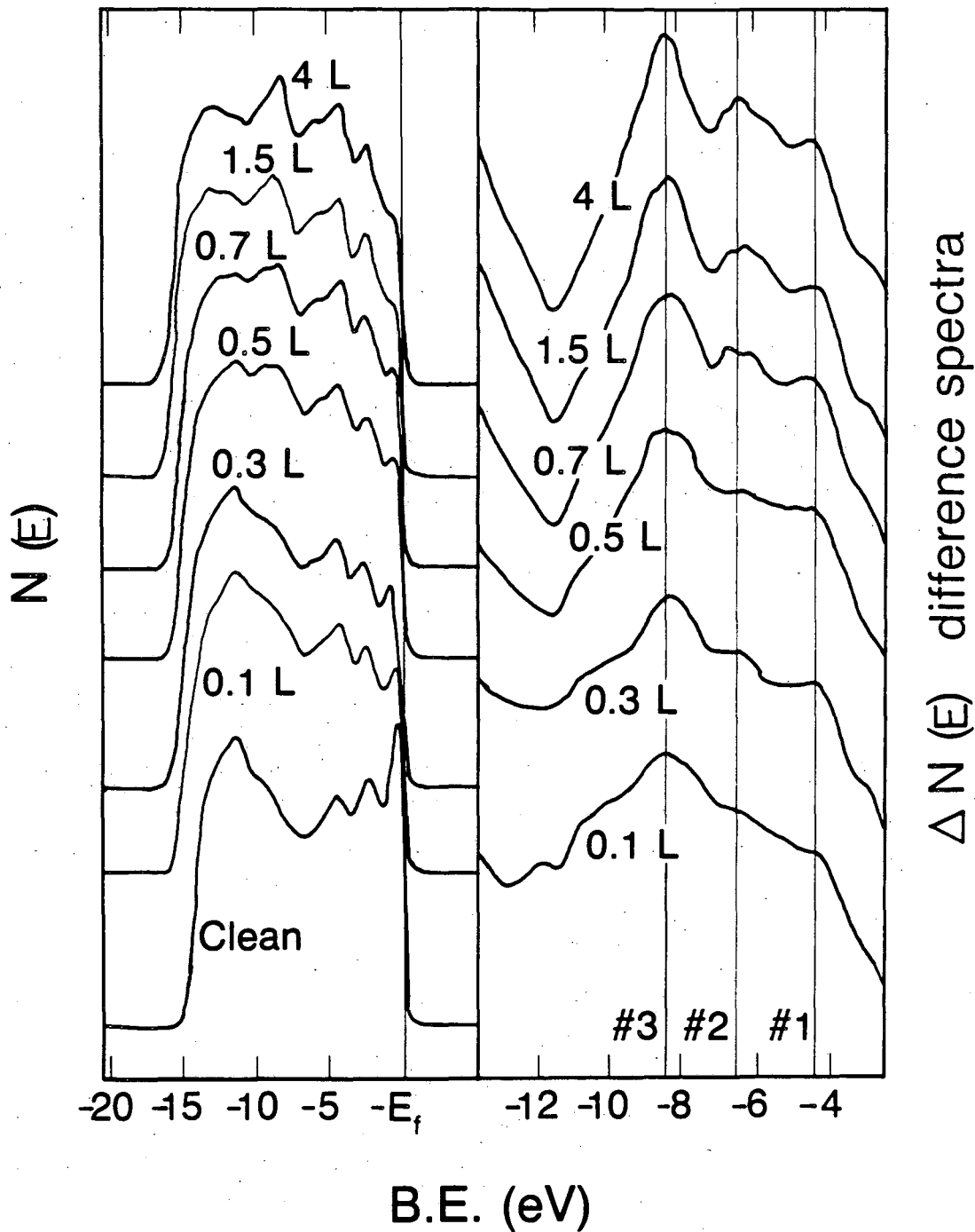
Fig. 5



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Fig. 6

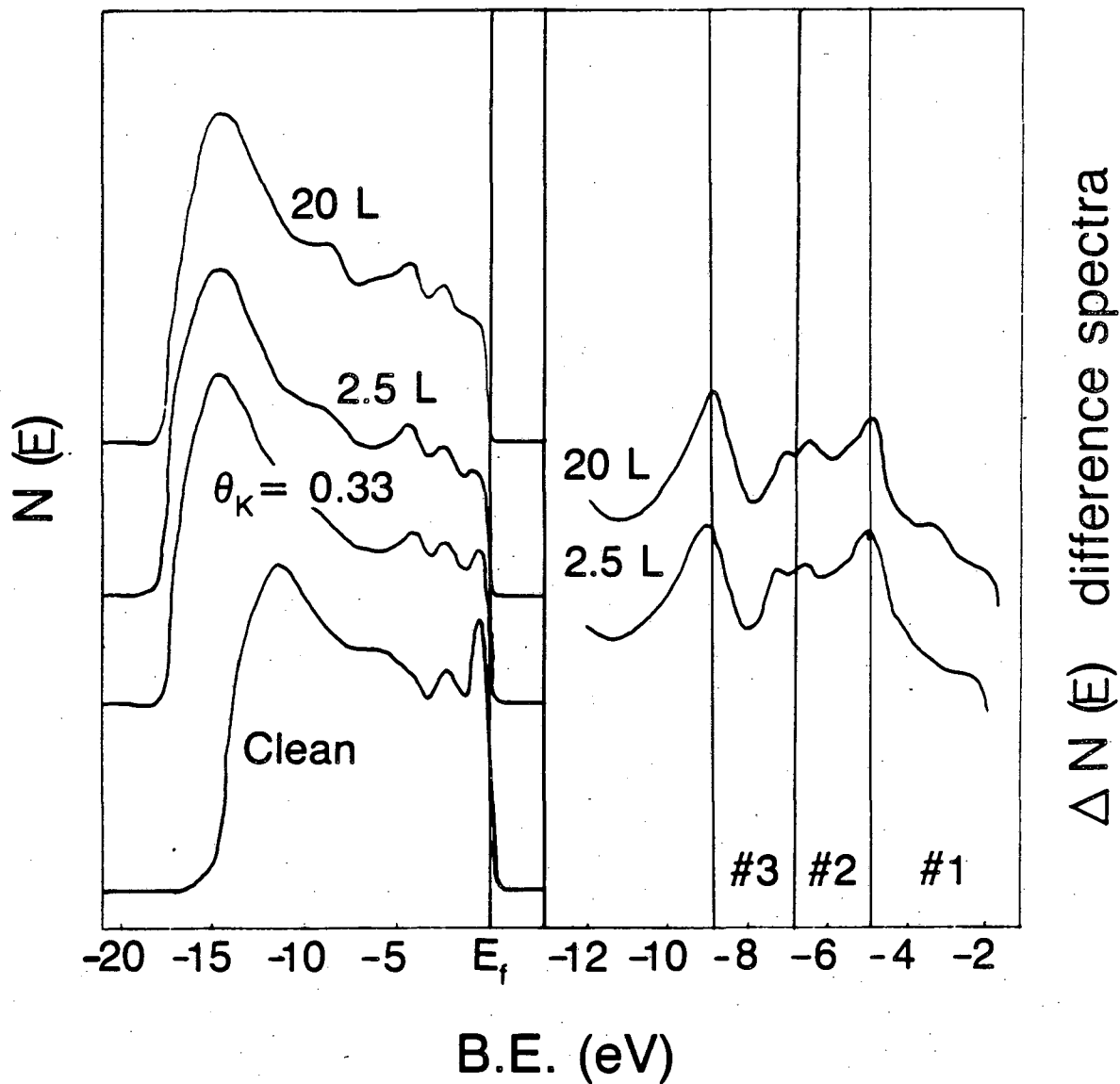
UPS Benzene on Pt (111)



XBL 838-3091

Fig. 7

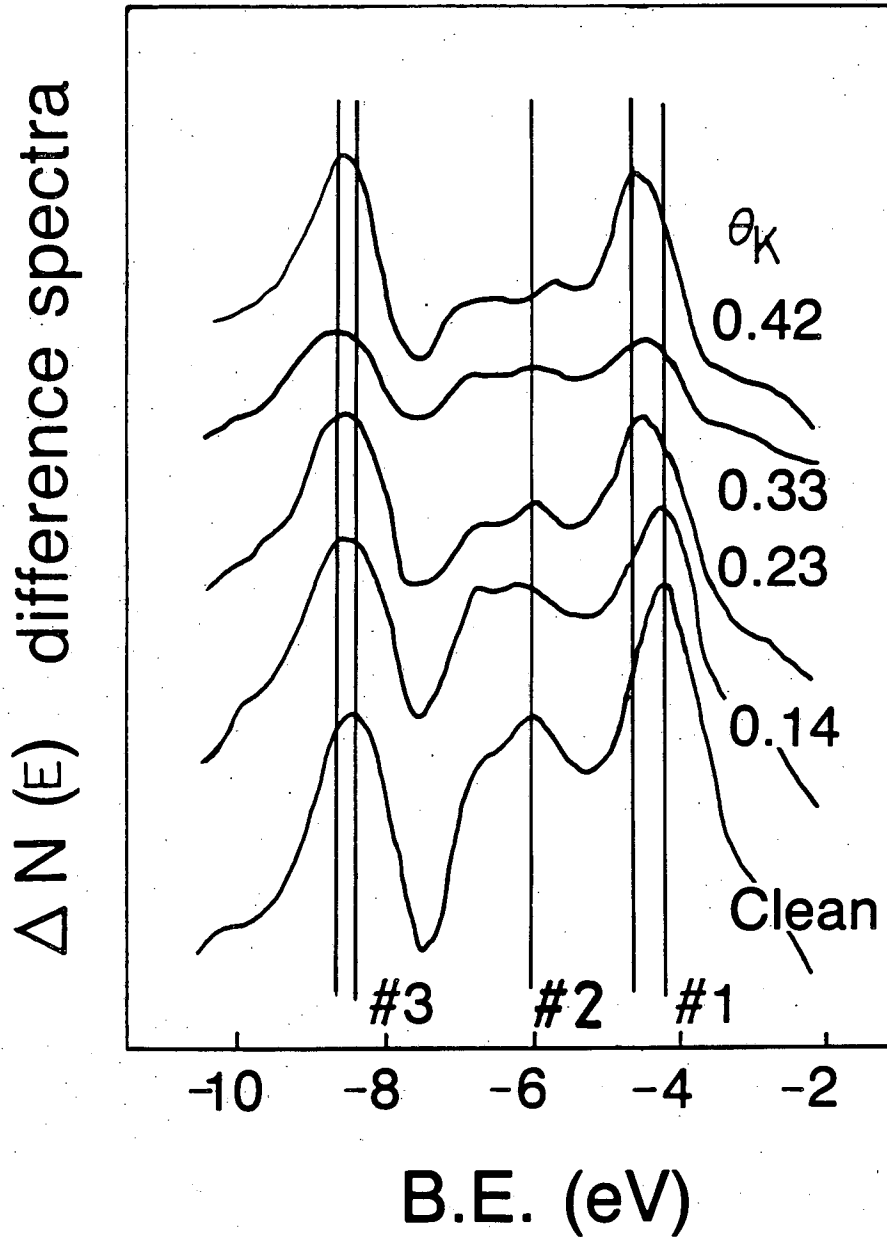
UPS Benzene on K/Pt (111)
 $\theta_K = 0.33$
Benzene exposures listed in
Langmuirs



XBL 838-3073

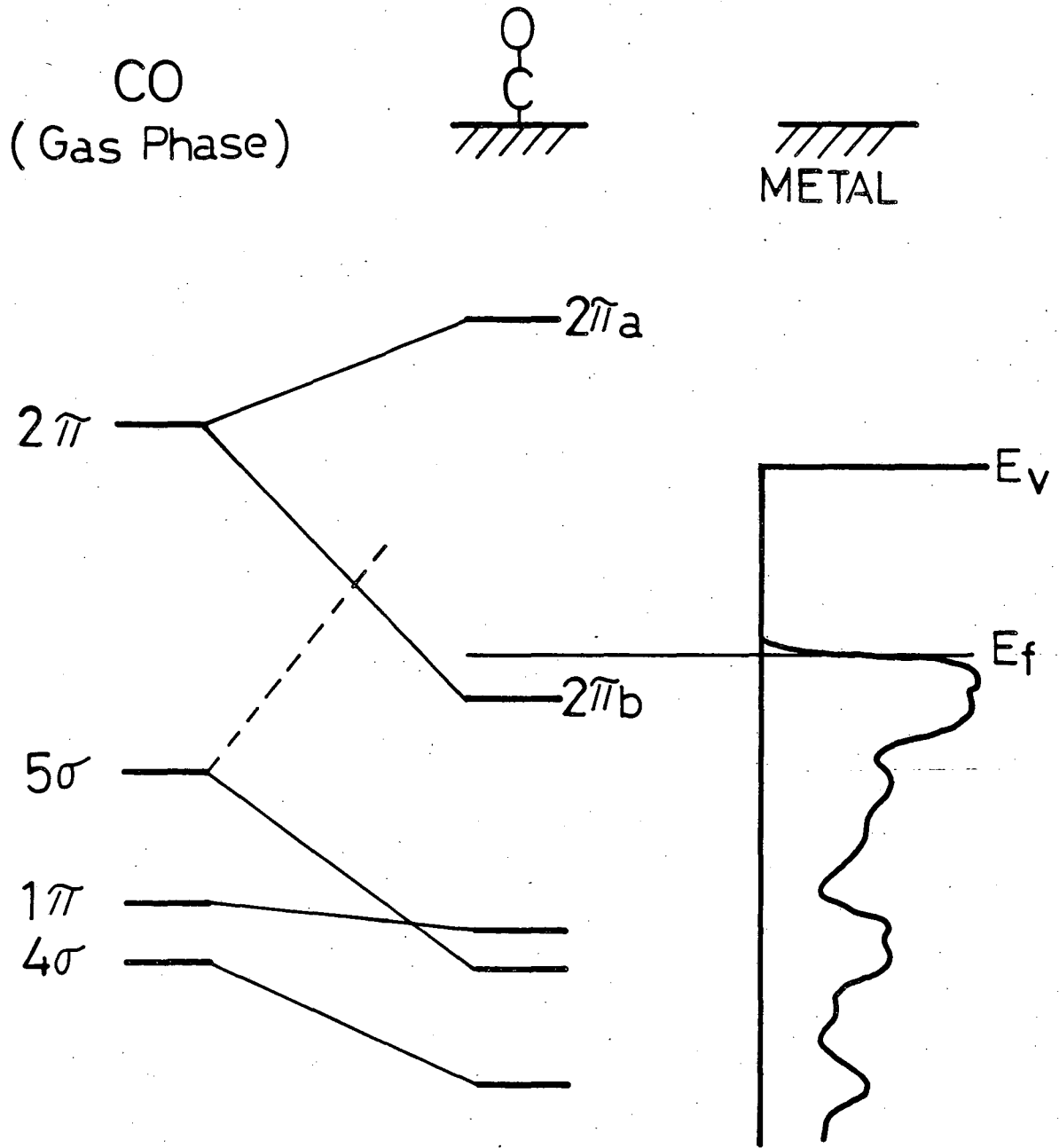
Fig. 8

UPS Benzene on K/Pt (111) difference spectra 2.5 L Benzene exposure



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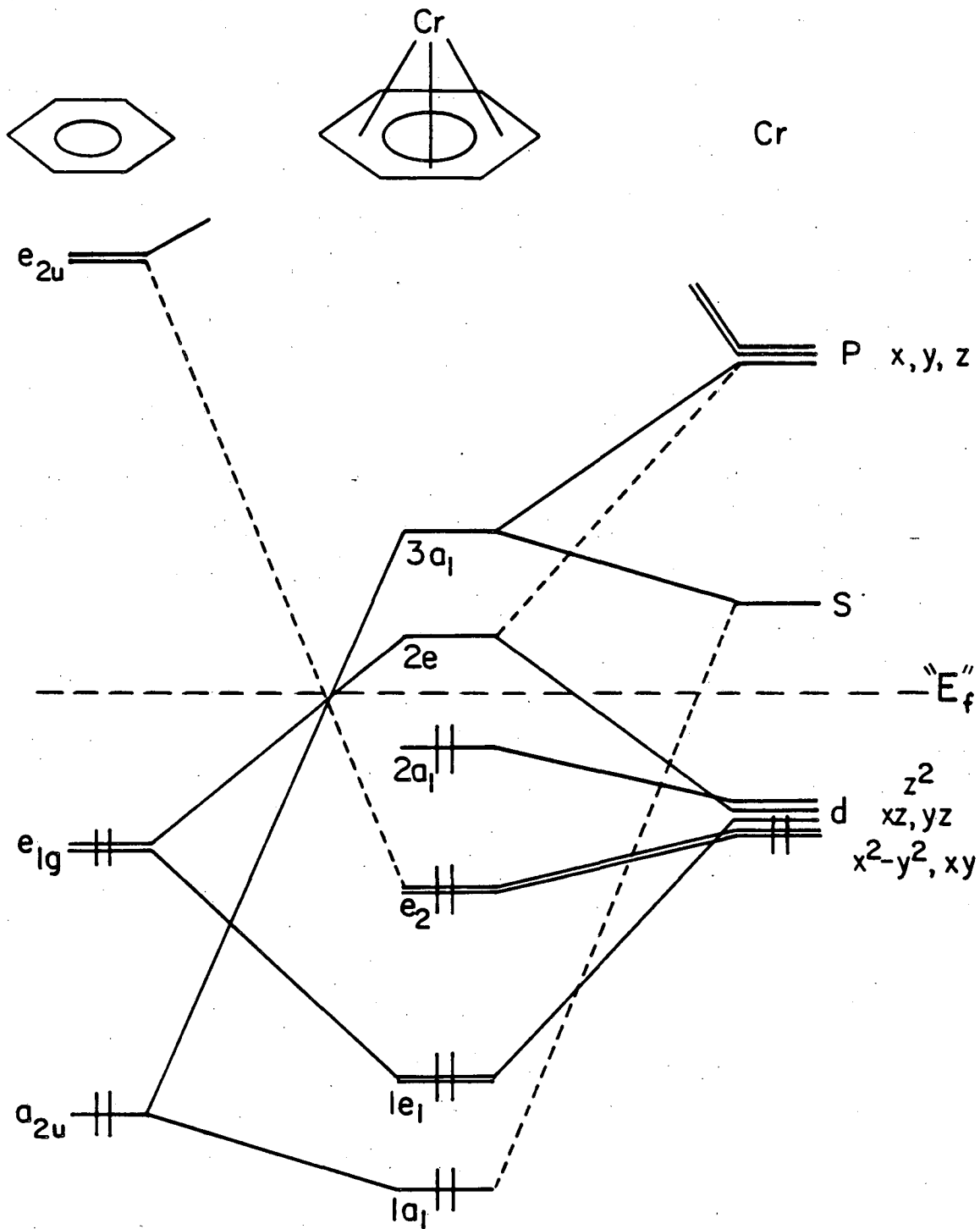
Fig. 9



XBL 849-3979

Fig. 10

Valence Orbitals of Benzene-Cr: Energy levels are approximate



XBL 8211-6884

Fig. 11

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