of the electrons above the transition, or that some fortuitous cancelation has occurred.

If the additional conductivity in the non-Ohmic region is due to a depinned sliding charge-density wave, the carriers involved are in a condensate. This condensed state of electron-hole pairs results from a situation in which a finite fraction of conduction electrons occupy their ground-state configuration. The condensate has no entropy and can transport no heat and therefore produces no TEP. The situation is similar to that which arises in conventional superconductors where the TEP drops to zero at the transition temperature, the supercurrent arising from a condensate with no entropy. Therefore a sliding charge-density wave *by itself* will not contribute to the measured TEP which means that some other mechanism must exist in order to explain the observed contribution to the TEP.

The most direct interpretation of the TEP data is that the applied field is freeing the single-particle carriers which have been frozen at the CDW transition. This would result in the TEP returning to a value consistent with the pretransition value (which is observed) and also be consistent with the observed saturation of the conductivity. However, this implies that the CDW is being depleted by the electric field which appears incompatible with the interpretation of the x-ray data mentioned earlier.

In conclusion, we have reported the first observation of a nonlinear or electric-field–dependent thermopower. Our results suggest that sliding charge-density waves alone, Zener tunneling, or hot-electron effects are not responsible for the observed thermopower. The simplest interpretation involves the electric-field breakdown of the charge-density–wave state. In the light of recent x-ray studies on $NbSe_3$ it is clear that further work is required to resolve this.

We would like to acknowledge useful discussions with S. Alexander, A. J. Berlinsky, J. W. Brill, T. Holstein, P. Monceau, R. Orbach, and P. Pincus. This research was supported in part by the National Science Foundation, Grant No. DMR 76-83421 and in part by the U. S. Office of Naval Research, under Grant No. N00014-77-C-0473. One of us (P.M.C.) acknowledges receipt of an A. P. Sloan Foundation Fellowship.

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Negative Shake-Up Energy in Core Ionization

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(Received 29 December 1978)

It is shown that the dynamical screening phenomena, which have recently been found to affect the shape of adsorbate core-hole spectra, occur also in the core ionization of certain molecules. For K-shell ionization of paranitroaniline we find shake-up transitions of *negative* energy which cause a multipeak structure of the corresponding line in the ESCA (electron spectroscopy for chemical analysis) spectrum. The present first-principles results lead to a new interpretation of the origin of the dynamical screening processes in adsorbates.

Recently renewed interest has arisen in screening processes affecting the core-level spectra of atoms and molecules adsorbed on metal surfaces.[1-5] Numerical calculations on the self-consistent atom-jellium model by Lang and Williams[1] suggested a physical picture which is quite different from the surface-plasmon screening model discussed previously.[6] Because of the electrostatic potential of the core hole, unoccupied valence levels of the adsorbate are pulled below the metal Fermi level and the screening occurs by partially filling up these levels.

The dynamical aspects of the screening process have been discussed by Schönhammer and Gunnarsson,[2,3] by Hussain and Newns,[4] and by Gadzuk and Doniach[5] with the use of the simple model Hamiltonian

$$H_0 = \epsilon_c n_c + [\epsilon_a - U_{ac}(1-n_c)]n_a + \sum_k \epsilon_k n_k + \sum_k (V_{ak} c_a^\dagger c_k + V_{ak}^* c_k^\dagger c_a), \qquad (1)$$

where ϵ_c denotes the core-level energy, ϵ_a the energy of the unoccupied adsorbate level which couples to the metal states $|k\rangle$ via V_{ak}. $c_a^\dagger$ ($c_k^\dagger$) is the creation operator for an electron in state $|a\rangle$ ($|k\rangle$) and $n_a = c_a^\dagger c_a$, etc. U_{ac} is the electrostatic interaction energy between the charge distributions of orbitals $|a\rangle$ and $|c\rangle$.

The dynamics of the ionization process can be briefly described as follows.[2] If U_{ac} is sufficiently large, the adsorbate level ϵ_a, which lies above the Fermi level in the unperturbed system ($n_c = 1$), is pulled below the Fermi level in the core-ionized state ($n_c = 0$). In the case of weak adsorbate-metal interaction V_{ak}, charge transfer to the adsorbate is hindered and a single core line appears at the "unrelaxed" position ϵ_c. In the case of strong adsorbate-metal coupling, on the other hand, the adsorbate level $|a\rangle$ is quickly filled and a single core line appears at the "fully relaxed" position $\epsilon_c + \epsilon_a - U_{ac}$. In the intermediate case, a well-developed double-peak structure results.[2] It should be noted that a model equivalent to (1) has been introduced already some years ago by Kotani and Toyozawa[7] to describe the splitting of core lines in transition-metal compounds.

The multipeak structure of the core-hole spectral function predicted by the model Hamiltonian (1) for suitable values of the parameters has recently been detected in the ESCA (electron spectroscopy for chemical analysis) spectra of chemisorbed molecules, specifically for CO on Cu, N_2 on W, and N_2 on Ni.[8,9] The C, O, or N K-shell lines, which appear as well-defined lines in the gas phase ESCA spectra of N_2 and CO, exhibit a pronounced multipeak structure in the chemisorbed state. The question arises whether the dynamical screening phenomena causing the multipeak structure of the core-hole spectra of adsorbates can occur also in free molecules. Upon looking through the literature we became aware of the work of Pignataro and others,[10,11] who have discovered clearly developed multipeak structures in the nitrogen K-shell ESCA spectra of nitroaniline and related highly polar aromatic compounds. In this communication we show that the mechanism leading to these multipeak structures is indeed related to the screening mechanism described by the Hamiltonian (1). In particular, we show that the multipeak structure in nitroaniline is the consequence of a *negative* shake-up energy in this molecule. This negative shake-up energy is the molecular counterpart to the pulling down and filling up of the virtual adsorbate level described above. Since in the free molecule the dynamical screening effects can be treated starting from first principles, this offers the possibility to arrive at a more detailed understanding of the complex screening processes on surfaces and to elucidate the meaning of the models used for their description.

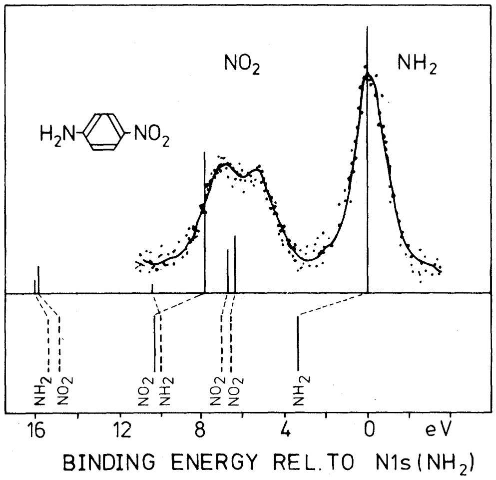

FIG. 1. N 1s energy region of the Al $K\alpha$ ESCA spectrum of paranitroaniline after Tsuchiya and Senō (Ref. 11) (the full line serves as a guide for the eye) together with the calculated ionization spectrum. The energies of the corresponding unperturbed states are shown in the lower panel (see text).

The N 1s energy region of the Al $K\alpha$ ESCA spectrum of paranitroaniline (PNA), $NO_2C_6H_4NH_2$, as recorded by Tsuchiya and Senō,[11] is shown in Fig. 1. The N 1s lines resulting from the nitrogen atoms in the NH_2 and NO_2 groups, respectively, are clearly separated because of the large chemical shift in this highly polar molecule. The N 1s (NO_2) line shows a double-peak structure which is reminiscent of the double-peak structure of the N 1s line of N_2 on W, for example.[8] The N 1s (NH_2) line, on the other hand, appears as a single line.

As a starting point we have performed a standard *ab initio* Hartree-Fock (HF) calculation[12] on PNA in its experimental geometry using the STO-3G basis set.[12] This calculation yields the HF orbital energies ϵ_i and the complete list of Coulomb and exchange matrix elements V_{ijkl}. In what follows we show that these data are already sufficient to understand the multipeak phenomenon in PNA. To describe shake-up lines we have to consider two-hole, one-particle (2h-1p) configurations, i.e., configurations involving a core hole and a valence-electron excitation from an occupied to an unoccupied orbital. Since we are interested in the lowest satellite lines, it suffices to consider the excitation from the highest occupied orbital (π) to the lowest unoccupied orbital (π^*). To first order in the electron-electron interaction, the lowest (triplet) $\pi \to \pi^*$ excitation energy of the neutral molecule is given by

$$E_{\pi^*\pi} = \epsilon_{\pi^*} - \epsilon_\pi - V_{\pi^*\pi\pi^*\pi}. \tag{2}$$

To the same order the $\pi \to \pi^*$ shake-up energy, i.e., the excitation energy *in the presence of the core hole,* is given by the expressions

$$\tilde{E}_{\pi^*\pi}{}^{(1,2)}(c) = \epsilon_{\pi^*} - \epsilon_\pi + F_{\pi^*\pi}{}^{(1,2)}(c), \tag{3}$$

where

$$F_{\pi^*\pi}{}^{(1,2)}(c) = (V_{c\pi c\pi} \pm V_{c\pi\pi c}) - (V_{\pi^*\pi\pi^*\pi} - \alpha^{(1,2)} V_{\pi^*\pi\pi\pi^*}) - (V_{c\pi^*c\pi^*} - \alpha^{(1,2)} V_{c\pi^*\pi^*c}), \tag{4}$$

$\alpha^{(1)} = \frac{1}{2}$ and $\alpha^{(2)} = \frac{3}{2}$; c denotes the core orbital from which ionization takes place. The difference between $\tilde{E}_{\pi^*\pi}{}^{(1)}(c)$ and $\tilde{E}_{\pi^*\pi}{}^{(2)}(c)$ is due to the two possibilities to couple the spins of three electrons.

For PNA we find $\epsilon_{\pi^*} - \epsilon_\pi = 11.79$ eV and $V_{\pi^*\pi\pi^*\pi} = -5.70$ eV. Thus the triplet excitation energy is $E_{\pi^*\pi} = 6.09$ eV. On the other hand, we find that, for $c = \mathrm{N}\ 1s\ (\mathrm{NO_2})$, $F_{\pi^*\pi}{}^{(1)}(c) = -13.54$ eV and $F_{\pi^*\pi}{}^{(2)}(c) = -14.56$ eV, yielding the shake-up energies $\tilde{E}_{\pi^*\pi}{}^{(1)}(c) = -1.75$ eV and $\tilde{E}_{\pi^*\pi}{}^{(2)}(c) = -2.78$ eV. The $\pi \to \pi^*$ excitation energy in the presence of the N 1s (NO_2) core hole is thus negative. The origin of this puzzling phenomenon is that $V_{c\pi^*c\pi^*}$ in Eq. (4) is very large, namely 12.44 eV, while $V_{c\pi c\pi}$, which enters with positive sign, is relatively small, namely 5.11 eV. Note that $V_{c\pi^*c\pi^*}$ corresponds to the electrostatic interaction energy U_{ac} in the Hamiltonian (1), which is responsible for the pulling down of the virtual adsorbate level below the Fermi level. The exchange integrals in Eq. (4) are small compared to the direct Coulomb integrals and cause the energy difference between $\tilde{E}_{\pi^*\pi}{}^{(1)}(c)$ and $\tilde{E}_{\pi^*\pi}{}^{(2)}(c)$. Considering the same $\pi \to \pi^*$ excitation for ionization out of the N 1s (NH_2) orbital, we find that $F_{\pi^*\pi}{}^{(1)}(c) = 1.51$ eV and $F_{\pi^*\pi}{}^{(2)}(c) = 0.23$ eV, leading to shake-up energies $\tilde{E}_{\pi^*\pi}{}^{(1)}(c) = 13.30$ eV and $\tilde{E}_{\pi^*\pi}{}^{(2)}(c) = 12.01$ eV. For N 1s (NH_2) ionization the $\pi \to \pi^*$ shake-up energies are thus *larger* than the corresponding triplet excitation energy of the neutral molecule. The reason is that now $V_{c\pi^*c\pi^*}$ is relatively small, namely 4.16 eV, while $V_{c\pi c\pi}$ is large, namely 11.61 eV.

To obtain the core ionization spectrum a more sophisticated calculation has been performed. The spectrum is given by the imaginary part of the core-hole one-body Green's function.[13] The Green's function has been computed using an approximation scheme described in detail elsewhere.[14] In this scheme the self-energy[13] $\Sigma(\omega)$ is constructed by diagonalizing the Hamiltonian in the space of the 2h-1p and 2p-1h configurations. All configurations which can be constructed considering the two N 1s core orbitals, the eleven occupied orbitals highest in energy and the seven unoccupied orbitals lowest in energy, have been included. The Green's function is then obtained by solving the Dyson equation.[13]

The calculated spectrum is shown in Fig. 1, together with the experimental spectrum. In the lower part of the figure the energies of the unperturbed states are shown. The solid lines represent the N 1s (NO_2) and N 1s (NH_2) single-hole states. The dashed lines represent the poles of the self-energy part which interact with either the N 1s (NO_2) or N 1s (NH_2) single-hole state to give the ionization spectrum shown in the upper part of the figure. It is seen that the splitting of the N 1s (NO_2) line observed in the ESCA spectrum is qualitatively reproduced by the calculation. The two satellite lines appearing on the low-energy side of the N 1s (NO_2) "main" line originate from the $\pi \to \pi^*$ transition with negative excitation energy discussed above. The appearance of two negative-energy satellite lines instead of one is due to a nonnegligible intravalence exchange element $V_{\pi^*\pi\pi\pi^*}$. This type of interaction is not contained in the model Hamiltonian (1). For ionization out of the N 1s (NH_2) orbital a quite different behavior is found: 85% of the intensity associated with the N 1s (NH_2) orbital appear in the main line at lowest binding energy. The satellite lines are weak and situated at considerable higher energy. The qualitative features of the spectrum are easily understood from the foregoing discussion, which has shown that the

$\pi \rightarrow \pi^*$ excitation energy is pushed upwards upon N 1*s* (NH_2) ionization, but strongly pulled downwards upon N 1*s* (NO_2) ionization.

The specific properties of PNA which are responsible for the negative shake-up energy for N 1*s* (NO_2) ionization are easily rationalized considering the charge distribution of the molecular orbitals. The valence charge of the aromatic ring in PNA is highly flexible and resembles in this respect the conduction electrons in a metal. Most of the charge associated with the highest occupied molecular orbital (π) resides on the NH_2 part of the molecule. The charge associated with the π^* orbital, on the other hand, is mainly concentrated on the NO_2 part of the molecule. It is now clear that the presence of a core hole on the N atom of the NO_2 group lowers the $\pi \rightarrow \pi^*$ excitation energy, since the core hole is effectively screened by the valence charge flowing towards this atom in the $\pi \rightarrow \pi^*$ excitation. A core hole on the N atom of the NH_2 group, on the other hand, must increase the $\pi \rightarrow \pi^*$ excitation energy, since valence charge is pulled away from the core hole in the $\pi \rightarrow \pi^*$ excitation. As quantitatively shown above, the energy gain through the screening of the N 1*s* (NO_2) core hole is large enough to make the $\pi \rightarrow \pi^*$ excitation energy negative.

From the present first-principles results we can learn about the interpretation of the model Hamiltonian used for the description of screening processes in adsorbates. The essential features of the present calculation can be understood within the framework of a simple model Hamiltonian. In neglecting spin and exchange, it reads

$$\begin{aligned} H = {} & \epsilon_c n_c + \epsilon_\pi n_\pi + \epsilon_{\pi^*} n_{\pi^*} + (1-n_c)(1-n_\pi) V_{c\pi c\pi} \\ & - (1-n_c) n_{\pi^*} V_{c\pi^* c\pi^*} - (1-n_\pi) n_{\pi^*} V_{\pi\pi^*\pi\pi^*} \\ & - (1-n_c)(c_\pi^\dagger c_{\pi^*} + c_{\pi^*}^\dagger c_\pi) V_{c\pi c\pi^*}. \end{aligned} \tag{5}$$

The last term of the Hamiltonian (5) represents the interaction between the unperturbed states and causes the multipeak structure of the N 1*s* (NO_2) line in PNA. The Hamiltonian (5) is formulated in the basis of the ground-state HF orbitals of the system. In the adsorbate-metal case it is customary to write the Hamiltonian in the basis of the one-particle states of the separated adsorbate and metal. By transforming Eq. (5) to such a basis and adopting the notation of Eq. (1), the Hamiltonian takes the form

$$\begin{aligned} H = {} & H_0 + (1-n_c) \sum_{k,k'} W_{kk'} c_k^\dagger c_{k'} \\ & + (1-n_c) \sum_k (W_{ak} c_a^\dagger c_k + W_{ak}^* c_k^\dagger c_a), \end{aligned} \tag{6}$$

with H_0 given by Eq. (1). For convenience only a single unoccupied adsorbate orbital $|a\rangle$ is considered as in Eq. (1). Two interaction terms are seen to arise in addition to the usually adopted Hamiltonian H_0.[2-5] The first represents the scattering of metal electrons by the suddenly appearing core hole in the adsorbate. It is the analog to the well-known scattering of conduction electrons by deep core holes in metals, which causes the x-ray threshold singularities and the skewness of the core-level x-ray photoemission lines.[15] The second term describes the mixing of the adsorbate and metal one-particle states due to the appearance of the core hole. The interaction matrix element W_{ak} arises from the Coulomb interaction between core and valence electrons and is independent of the magnitude of the adsorbate-metal coupling V_{ak} included in H_0. From the foregoing discussion of PNA we conclude that it is this *core-hole-induced* mixing of adsorbate and metal states which is responsible for the multipeak core-level line shapes. The effect of the last term in Eq. (6) might be partly included by redefining the hopping matrix element V_{ak} in Eq. (1), but this obscures the physics of the process. Clearly the adsorbate-metal coupling strength determined in this way from the analysis of experimental core-level line shapes does not bear any relationship to the strength of the chemical bond between adsorbate and metal.

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Giant Hyperfine Anomaly between Bound Negative Muon and Rh Nucleus in Pd Metal

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(Received 4 January 1979)

The Knight shift of negative muon (μ^- Pd) in Pd metal has been determined to be $-(9.0 \pm 0.7)\%$ at $T = 11$ K, revealing an unusually large hyperfine anomaly between μ^- Pd and its equivalent isotope Rh*Pd*; $H_{\rm hf}(\mu^- {\rm Pd})/H_{\rm hf}({\rm Rh}Pd) = 0.64 \pm 0.05$, or $\epsilon_\mu - \epsilon_N = -(36 \pm 5)\%$. Its implication is discussed in terms of the spatial distribution of the electron spin density in transition metals.

The hyperfine anomaly, the change of the hyperfine field between different nuclear states of the same isotope, reflects spatial distributions of both nuclear magnetization and electron spin density. This problem was first studied theoretically by Bohr and Weisskopf,[1] and then, rigorously by Stroke, Blin-Stoyle, and Jaccarino.[2] They considered the radial decrease of s-electron density that is probed by nuclei of finite size. Another possible cause of hyperfine anomaly is the change of $|\psi_e(0)|^2$ due to the change of the charge distribution of the probe nucleus (so-called Rosenthal-Breit-Crawford-Schawlow correction,[3] as discussed in Ref. 2). A large amount of experimental data have been explained by taking into account the nuclear magnetization distributions consistent with the nuclear wave functions, but neither the nuclear structure nor the mechanism of the hyperfine field become clear from these studies simply because the nucleus is too small to produce a large effect sensitive enough to discriminate between models. It would be dramatic, if we could find a much more extended magnetic probe to detect the electron spin density.

In this context, we paid special attention to the hyperfine field probed by bound muons.[4] Polarized negative muons that have stopped in a material immediately reach the ground state ($1s_{1/2}$) of the muonic atom and stay for a certain length of time τ_μ (τ_μ varies from 2.2 μsec in the lightest atoms to 80 nsec in heavy atoms). The average polarization is decreased to about $\frac{1}{6}$ due to the spin-orbit coupling, but is still large enough to observe spin precession (negative-muon spin rotation). The density of the bound μ^- is given by $|\psi_{1s}{}^\mu(r)|^2$. The muon wave function $\psi_{1s}{}^\mu(r)$ is, for a point nucleus, $\exp(-Zr/a_\mu)$ with $a_\mu = \hbar^2/m_\mu e^2 = 260$ fm, but, for heavy nuclei, where a_μ becomes close to the nuclear radius, it is modified due to the finite extension of nuclear charge. In either case, the bound muon is distributed largely outside the nucleus. However, when viewed from atomic electrons, the bound muon is still concentrated around the nucleus so that it should behave like an impurity nucleus of apparent charge $(Z-1)e$. Compared to its equivalent nucleus of true nuclear charge $(Z-1)e$, the bound muon has