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Photon Energy Range 32 eV to 50 eV

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Photoemission measurements with photon energies ranging from the onset of Na 2p electron absorption to 50 eV have been performed using synchrotron radiation as light source. The results indicate that the rise in the absorption coefficient starting at about 8.5 eV above the onset of 2p electron transitions is due to continuum transitions whereas in the region below most of the structures turn out to be exciton excitations.

Absorption measurements^{1,2} on the Na^+2p core electron excitations in NaCl have shown a rise in the absorption coefficient μ at about 8.5 eV above the onset. 8.5 eV is approximately the fundamental band gap E_g . Most of the other Na halides and as well as also the alkali chlorides near the Cl^-2p onset have shown a similar behaviour. Sagawa *et al.*^{2,3,4} have attributed this rise in μ to two-electron excitations coming from the valence band and the core bands. Theoretical estimations made by Miyakawa⁵ yield a cross section for simultaneous two-electron excitations having a magnitude similar to one-electron excitations. On the other hand Hermanson⁶ has estimated a probability for such a process which is smaller by two orders of magnitude and we have recently been able to prove this experimentally for solid Kr and Xe.⁷

We have made another experimental approach to this problem by measuring the photoemission of NaCl. Our sample was mounted behind the exit slit of a Rowland monochromator⁸ and illuminated at normal incidence. The DESY 7.5 GeV synchrotron served as light source, the resolution was 0.1 eV. The sample ($\sim 500 \text{ \AA}$ NaCl evaporated on gold) was connected to a sensitive DC amplifier (Cary 401) and was surrounded by a gold plated cylindrical cup. A retarding voltage U_R was applied between sample and cup. The spectral distribution of the photocurrent was recorded at different retarding voltages U_R . The measurements were repeated in a plane grating spectrograph with less resolution but with essentially the same results.

First we would like to discuss the different types of processes leading to the emission of photoelectrons in our energy region and also the particulars one has to know for interpreting our data:

1. Though the intensity distribution at the exit slit is not accurately known, it has been verified that the spectrum is continuous and structureless. There is a monotonic increase of intensity to higher photon energies. Normal incidence reflection of NaCl can be neglected.
2. Fig. 1 gives an outline of the relative energy positions of the conduction band CD, the valence band V, the core bands CO, exciton levels EX in NaCl and the Fermi levels E_F , vacuum levels VK, work functions ϕ_N and ϕ_G in NaCl and gold. The external voltage U_R is applied between the sample and the cup. We are now going to give values for the different parameters, some of which are known only with an accuracy of ± 1 eV, which, however, is still good enough for the interpretation of our results.

The bottom of the conduction band is 8.5 eV above the top of the valence band⁹ and 35 eV above the 2p core bands (assuming that the binding energy for the excitons in the absorption measurements is 2 eV). The work function of gold is $\phi_G \approx 4$ eV and as it turned out experimentally

that we obtained saturation of the photocurrent at $U_R \approx 0$ V, then $\phi_N \approx \phi_G \approx 4$ eV follows. The vacuum level of NaCl coincides approximately with the bottom of the conduction band. This can be concluded from a measurement of the highest electron energy occurring at a fixed photonenergy. Therefore eU_R measures the kinetic energy in vacuum and is almost equal to the energy above the bottom of the conduction band. All this implies that the Fermi surface is - as is to be expected for pure samples - in the middle of the band gap of NaCl; this is quite a surprising result for an evaporated film.

3. Electrons excited from a core level into continuum states which suffer no energy loss before they escape from the sample are just rejected at a retarding potential $eU_R = -E_{co} + hv$, E_{co} is the energy separation between the core state and the bottom of the conduction band (35 eV). In the spectra this will give rise to a step at photon energies:

$$hv = E_{co} + eU_R. \quad (1)$$

This step can be distinguished from other structures since by changing U_R the step changes its position in the spectra.

4. Electrons with kinetic energies exceeding the band gap energy E_g (~ 8.5 eV) can suffer energy loss due to electron-electron scattering.

5. Excitons with a core-state hole can decay into high energy photoelectrons (25 eV and more) due to ionization of a valence electron.
6. Metastable excitons can dissociate into those continuum states which are at the same level.
7. Core-state holes can make Auger transitions into the valence band thus emitting energetic electrons (~ 18 eV).

Figure 2 gives the spectral distribution of photoelectrons with retarding potential U_R as parameters. The error in these curves is approximately $\pm 3\%$ for $3 \cdot 10^{-13}$ Amp, $\pm 5\%$ for $3 \cdot 10^{-14}$ Amp and $\pm 15\%$ for $3 \cdot 10^{-15}$ Amp. For comparison we have added our earlier absorption measurement¹ showing the rise in absorption from peak F to G which Sagawa et.al. attributed to two-electron excitations.

a) Peaks A to F are not cut away by a step at a photon energy given by Eq. 1 which would otherwise identify them as continuum transitions according to Point 3. or excitons decaying according to Point 6., but they continuously fade away with increasing retarding potential eU_R . We assume that they are excitons decaying into photoelectrons according to Point 5. (An experimental reason will be given in section b) why Auger decay according to Point 7. is negligible.) The photoelectrons with originally 25 eV and more kinetic energy for peaks A to

F will undergo energy losses according to Point 4. Therefore a whole spectrum of electron energies is emitted. The peaks are preserved up to fairly high retarding potentials, finally merging into the background at values of U_R depending on their intensity and the noise of the detecting system.

b) In the spectrum with retarding voltage $U_R = 6$ V a step of the type discussed in Point 3. is found at photon energy $h\nu \approx 41$ eV. This step reaches peak G (≈ 43 eV) at $U_R \approx 8$ V. For $U_R \geq 9$ V peak G is completely cut away. Therefore we conclude that G is no double excitation but a continuum transition (an exciton excitation decaying according to Point 6. cannot be excluded). As with retarding voltage $U_R \geq 9$ V no structure is left at position G, we conclude that Auger decay of the hole (see Point 7) is a negligible effect.

c) Above peak G electron transitions into one-electron continuum states have kinetic energies high enough to suffer energy loss according to Point 4. therefore many low energy electrons are ejected into this region of the spectrum as may be seen from the different shapes of the spectra at $U_R = 0$ V and $U_R \geq 2$ V. But also electrons with their original energies are left as can be seen from the step at $U_R = 11$ V according to Point 3.

d) At $U_R = 11$ V mainly a background of electrons ejected from the valence band is left for $h\nu < 45$ eV. This reflects the intensity distribution behind the exit slit according to Point 1. The only structure left is the strong exciton B superimposed on this background.

We have given one possible consistent interpretation of our results which appears to us to be the most plausible within the frame of present knowledge on band structure and excitons. Of course we are aware that this has to be tested by extending photoemission experiments to higher retarding voltages and other substances.

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Figure captions:

Fig. 1 Schematic drawing of the energy levels in NaCl sample and gold covered cup, which was connected to the retarding potential U_R . Symbols are explained in the text (Point 2.)

Fig. 2 Absorption coefficient μ in arbitrary units and currents of photoelectrons in logarithmic scale versus photon energy near the Na $L_{II,III}$ edge in NaCl. The parameter of the different curves is the retarding potential U_R .

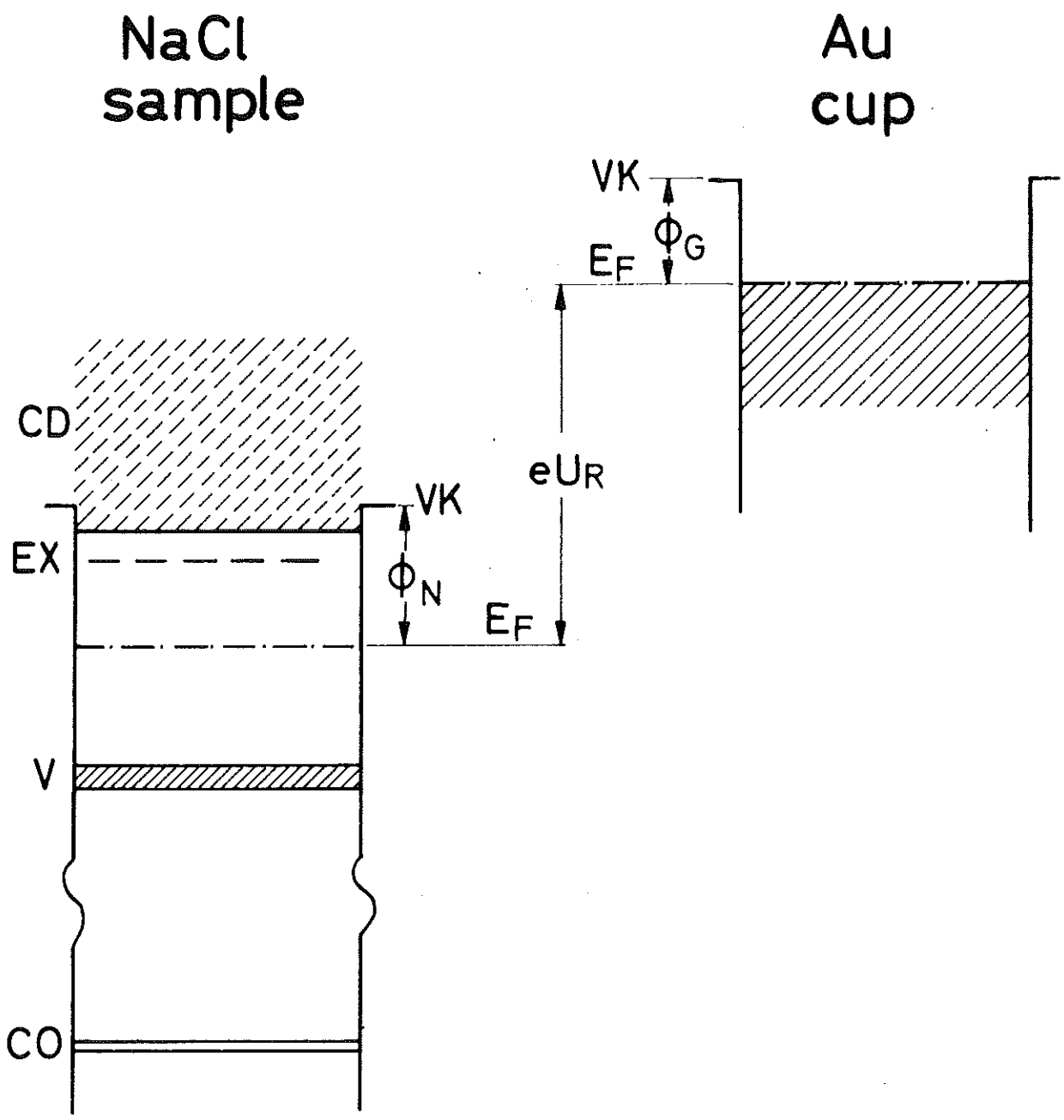


Fig. 1

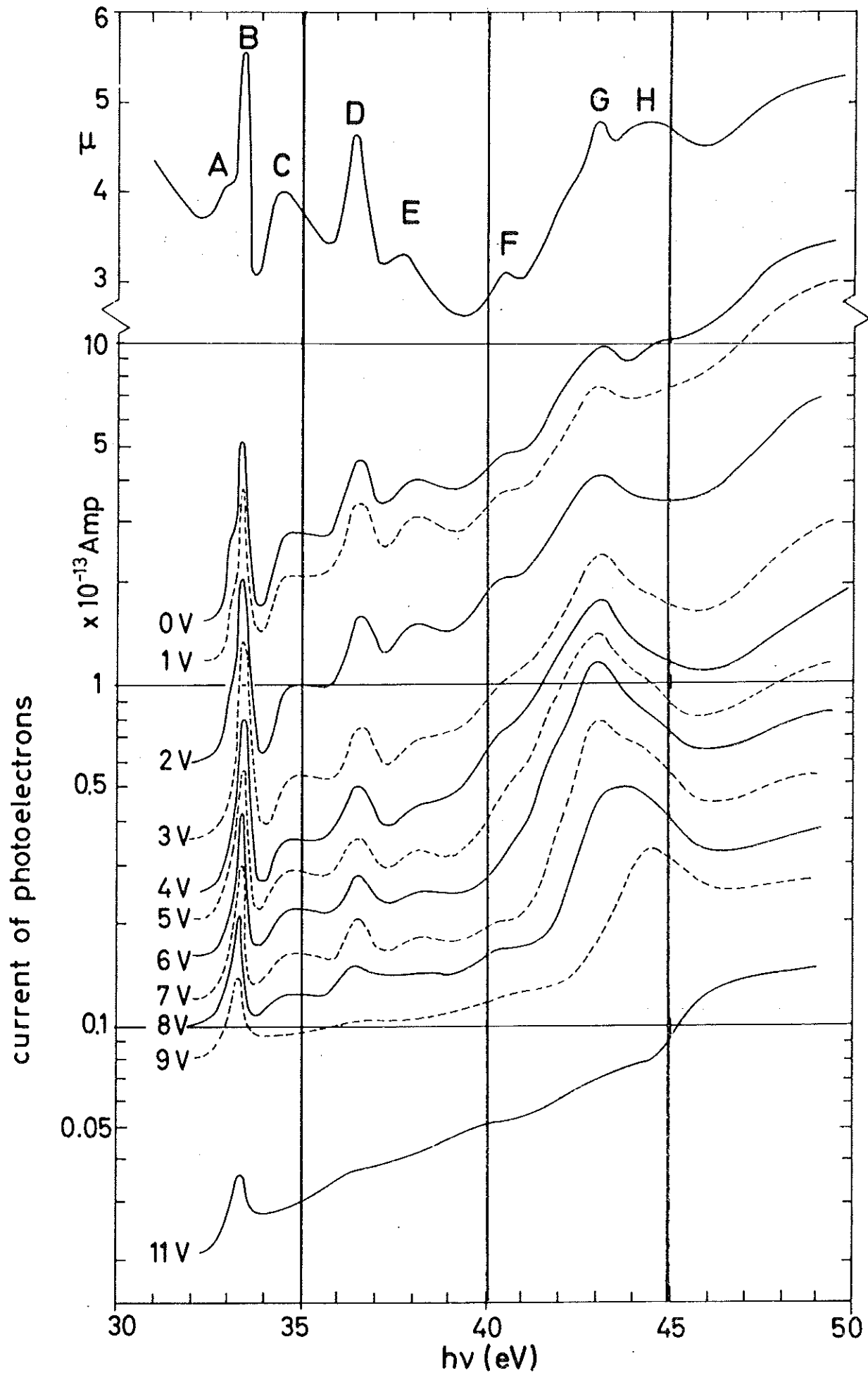


Fig. 2