



# Electron pair emission from a Pb surface at room temperature

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## ABSTRACT

We provide a comprehensive study of the electron pair emission from a Pb surface at room temperature. We excited the sample via a primary electron beam or laboratory light source. Besides the excitation of the 6s and 6p valence states the weakly bound 5d core levels are accessible. This allows us to investigate the Auger-photoelectron pairs in coincidence. The electron pair excitation spectra can be largely explained by the underlying electronic structure. Varying the primary energy changes the relative contribution of the 6s and 6p states. The measured double photoemission intensity is dominated by the emission of 5d photoelectrons and the resulting Auger electron. The Auger electron line shape has mainly contributions due the 6p electrons, because the 6s electrons can not lead to the emission of an Auger electron due to energy conservation. From the sum energy spectra we find that the effective Coulomb interaction  $U_{eff}$  is close to zero. The double photoemission intensity without participation of the 5d levels displays rather featureless spectra. Among the materials which display superconductivity is Pb. The explanation of this effect requires the introduction of Cooper pairs. It was theoretically predicted that double photoemission of Cooper pairs is possible. We discuss the experimental feasibility of such measurements.

## 1. Introduction

The properties of matter are decisively determined by the electronic structure. Electrons do not move independently within a solid, but exert a mutual influence via the Coulomb interaction and the Pauli principle. A complete microscopic description of all electrons is impossible. However, it turns out that it is possible to cast the effects of the electron-electron interaction into a mean-field type description. This is also known as the quasi-particle picture. Hence, the access to the underlying electronic structure via photoelectron spectroscopy has been proven very successful. Nevertheless it is desirable to investigate the mutual relations between electrons. This is possible via electron pair emission from surfaces, because the existence of this effect requires a finite electron-electron interaction. It is customary to distinguish between primary electron and photon absorption as (e,2e) and Double photoemission (DPE), respectively.

We have decided to investigate a Pb surface because of particular features in the electronic states. The valence band of Pb has contributions from the 6s and 6p levels which are separated by a gap of 3.1 eV. This means in contrast to transition metals there is no hybridisation between electrons from different orbitals. The spin-orbit split 5d levels possess a binding energy of around 20 eV. This means (e,2e) experiments with typical excitation energies in the range of 30–60 eV can

excite these core levels leading to autoionization states. This aspect is not included in the current theory of (e,2e) and in experiments on other materials core levels were more tightly bound and not excited.

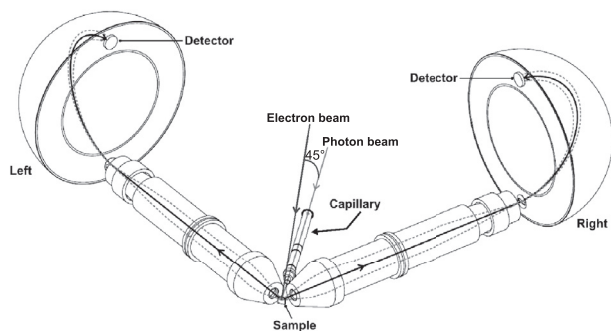
The shallow 5d core levels can also be excited by a laboratory UV light source leading to the emission of 5d photoelectrons and Auger electrons. Although some results have been published using X-ray laboratory sources most studies are conducted using synchrotron radiation [1–5]. We explore the DPE process with and without participation of the 5d core level.

The (e,2e) energy spectra show features which can be largely understood on the basis of the bulk density of states (DOS). The relative contributions of the 6s and 6p valence states and the energy sharing does depend on the primary energy  $E_p$ . We find evidence that the 5d levels play a role via an autoionization pathway. We observe that the DPE intensity is dominated by the emission of Auger-photoelectron pairs. From the Auger lineshape we determine that the effective Coulomb correlation  $U_{eff}$  is close to zero. The DPE intensity directly from the valence band is an order of magnitude smaller than those of the Auger-photoelectron pairs. This would be consistent with a small value of the Coulomb correlation within the valence band [6,7].

Our experiments were performed at room temperature where Pb is not superconducting. The explanation of superconductivity requires the concept of Cooper pairs. The possibility of Cooper pair emission via

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**Fig. 1.** The two transfer lenses of the spectrometer are symmetrically aligned with respect to the surface normal. The photon beam propagates along the surface normal. An electron gun for (e,2e) experiments was available, this primary beam has an angle of 45° with respect to the scattering plane.

single photon absorption has been theoretically predicted [8–10]. We use our data to assess whether this effect can be experimentally observed if the sample is in the superconducting state. We are motivated to address this issue, because of the potential new insights on the pairing mechanism. We find the expected emission rates to be prohibitive. We conclude that only more recent electron spectrometer using the high acceleration voltage of cathode lenses promise success.

## 2. Experimental details

The details of the coincidence spectrometer have been described in more detail elsewhere [11–13]. Therefore we recall only the main aspects. The general layout in Fig. 1 shows two hemispherical electron energy analyzers with a mean radius of 200 mm. They are equipped with wide angle transfer lenses and position sensitive detectors. We label the spectrometers as ‘left’ and ‘right’ respectively. We will quote the kinetic energy with respect to the vacuum level of the sample. The two electron-optical axes of the transfer lenses include an angle of 90° and define the reaction plane, in which the primary photon beam lies. The emitted electrons are detected with energies  $E_{left}$  and  $E_{right}$ . Due to the entrance slits not all electrons which enter the lens will be detected. The orientation of the slits of each spectrometer has a  $\pm 15^\circ$  acceptance within the scattering plane.

All experiments were performed with the photon beam being parallel to the normal which has an angle of 45° with respect to the optical axis of the two transfer lenses. The sample position was maintained during the (e,2e) experiments for which an electron gun was available. The primary electron beam had an angle of 45° with respect to the scattering plane, see Fig. 1. For in-house experiments the system has been upgraded by the attachment of VUV light source which consists of a He lamp and a toroidal monochromator [6,14,15]. We used the He II lines at 40.8, 48.4 and 51.0 eV, respectively. Space constraints do not allow to have the sample at the focus point of the grating. Therefore, we fitted the exit arm of the monochromator with an additional capillary of 200 mm length and 2 mm diameter. This has the added benefit of improved differential pumping. The partial pressure of He during normal operation is  $2-3 \times 10^{-10}$  mbar, while the base pressure was  $5 \times 10^{-11}$  mbar. In order to reduce the primary flux on the sample we placed a set of apertures between the He light source and the monochromator.

We are interested in those events in which a single photon (electron) leads to the emission of an electron pair which we term ‘true’ coincidences. However, it is also possible that two photons (electrons) lead to the individual emission of single electrons which will be recorded by the coincidence electronics. These unwanted events are usually termed ‘random’ coincidences. The ‘true’ coincidences scale linearly with the primary flux, while the random ‘random’ contribution scales quadratically. This allows us to reduce the latter to an acceptable level. This

comes at the expense of a low coincidence count rate. Following standard procedures documented in the literature we are able to remove the aggregate effect of the ‘random’ coincidences [4,16]. The implementation of these procedures for our coincidence spectrometer has been explained previously [13]. At this point we can state that we are able to determine the ‘true’ coincidence rate and energy spectra.

We operated the spectrometer with a pass energy of 150 eV which results in an energy window of 13 eV which can be covered with each spectrometer. The slit size selected was 1 mm, this leads to an energy resolution of 0.35 eV of each spectrometer. The line width of the VUV light (a few meV) can be neglected while the primary electron energy has a width of 0.3 eV. We keep all voltages of the electron-optical components constant for a given coincidence experiment. The sample was a polycrystalline Pb foil which was cleaned via Ar<sup>+</sup> sputtering and annealing up to 150 °C. Auger spectroscopy verified the cleanliness of the surface. The lowest temperature of the manipulator is 120 K. We chose to perform all measurements at room temperature, because the temperature broadening is still smaller than the selected spectrometer resolution. For absolute energy calibration we use the recent work of Iablonsky et. al on polycrystalline and atomic Pb [17].

## 3. Results and discussion

### 3.1. (e,2e) experiments

In our experiments two electrons are emitted from a surface which possess kinetic energies  $E_{left}$  and  $E_{right}$ , respectively. In the following we quote these energies with respect to the vacuum level of the sample. This pair can also be characterized by the energy sum  $E_{sum} = E_{left} + E_{right}$ . In an (e,2e) experiment a valence electron with binding energy  $E_B$  is emitted after a primary electron with kinetic energy  $E_p$  impinges onto the surface. Effectively this process removes one electron from the sample, hence the work function of the surface  $\phi$  has to be considered. Therefore, energy conservation for an (e,2e) process can be formulated as:

$$E_p + E_B = E_{sum} + \phi \quad (1)$$

In the DPE process, a photon with energy  $h\nu$  is absorbed by the sample and two electrons with binding energies  $E_{B1}$  and  $E_{B2}$  are emitted. Therefore, in the energy balance the work function has to be entered twice and we obtain:

$$h\nu + E_{B1} + E_{B2} = E_{left} + E_{right} + 2\phi = E_{sum} + 2\phi \quad (2)$$

A useful reference energy is the maximum sum energy  $E_{sum}^{max}$  which is obtained if the emitted electrons originate from the Fermi level  $E_F$ . From the previous definition it follows that in a (e,2e) process  $E_{sum}^{max} = E_p - \phi$ . For a DPE process it readily follows  $E_{sum}^{max} = h\nu - 2\phi$ . In the last step we have assumed that the energy required to remove an electron pair is equal to twice the energy to remove a single electron. This can only be an approximation, because the electron correlation is ignored at this point. A prominent example may illustrate this. The energy required for single ionization of the He atom is 24.59 eV. For double ionization an energy of 79.01 eV is needed, which is very different from twice the single ionization energy. Similarly, for Auger electron emission it is known that the kinetic energy can be significantly shifted from the position expected by using the binding energies of the involved states. This shift can be identified with an effective electron-electron interaction strength  $U_{eff}$  [18]. In the case of Cu a value of 8 eV is reported. This is the additional energy besides twice the work function which needs to be furnished in order to remove two 3d valence electrons.

The operator leading to single photoelectron emission is the dipole operator, the equivalent for double photoemission is the sum of two dipole operators [19,20]. The evaluation of the transition matrix element with single particle wave functions leads to a vanishing DPE intensity [19,20]. The presence of the electron-electron interaction

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