Correlated positron-electron emission from LiF(100)

GA van Riessen, FO Schumann, M Birke, C Winkler and J Kirschner

E-mail: riessen@mpi-halle.mpg.de
Max-Planck Institute of Microstructure Physics, Weinberg 2, 06120 Halle, Germany

Abstract. Using two hemispherical energy analyzers with transfer lenses we have observed positron-electron pair emission from a surface upon low energy positron impact. An 85 eV positron beam from the NEPOMUC positron source was incident upon a LiF(100) surface and emitted electrons were detected in coincidence with the positrons. Energy analysis of the positron-electron pair confirms that electrons are emitted from the valence band upon positron impact. Electron-electron pairs emitted upon electron and positron impact are also discussed. The information obtainable from these experiments has the potential to provide important insights into electron-electron correlation in solids.

1. Introduction
In the past several decades, extensive application of electron-electron coincidence experiments ($e^- e^- e^- e^-$) on atoms and molecules have contributed to a deeper understanding of electron-electron correlation effects. In the condensed phase, electron-electron correlation effects, which underlie phenomena such as magnetism and superconductivity, continue to present a formidable challenge to theory. It follows that electron-electron ($e^- e^-$) coincidence experiments on solids have received burgeoning interest in recent years despite the experimental challenges. It has been demonstrated that information about the correlation between a pair of electrons in the solid can be recovered from the observed momenta of the pair of electrons emitted from the surface upon photon or electron impact [1, 2, 3, 4, 5, 6].

Many theoretical studies have been devoted to the various aspects of the so-called exchange-correlation hole, a depletion of the average electron density around an individual electron in the condensed phase (see, e.g., refs. [7, 8, 9]) that is a direct manifestation of Coulomb and exchange interactions. Its extent in momentum space is a measure of the electron pair-correlation function which is a central quantity in the theory of many-body systems. It has been shown that with $e^- e^-$ pair emission spectroscopy it is possible to directly probe the exchange-correlation hole [1, 2]. Separating the contributions of Coulomb and exchange interactions to the exchange-correlation hole is not straightforward. It is the indistinguishability of two electrons that leads to characteristic exchange effects in $e^- e^-$ interactions. These are absent for positron-electron ($e^+ e^- e^- e^-$) interactions. It follows that by employing a low energy positron beam and observing in time coincidence an electron and positron emitted from a surface after their interaction in the solid may provide a route by which the role of (attractive) Coulomb interactions can be probed without direct influence of exchange interactions.

An understanding of $e^+ e^-$ correlations is also central to understanding positron properties in condensed matter systems. Positron annihilation methods provide a sensitive probe to the electronic structure of solids [10] but interpretation of the information obtained requires a detailed understanding of $e^+ e^-$-correlation. Despite considerable theoretical investigation,
there remains some unexplained discrepancies between theory and experiment, even for simple metals [11, 12].

Recently we reported that correlated $e^+e^-e^-$ pairs emitted from a LiF(100) surface could be detected and that the essential characteristics of their energy distribution could be recognized from simple considerations of the wide band gap of LiF [13]. In this article we present additional experimental detail and present a preliminary analysis of the differences observed between $(e^+,e^+e^-)$, $(e^+,e^-e^-)$ and $(e^-,e^-e^-)$ energy spectra.

2. Experiment

The experiment essentially consisted of the detection of particles (positrons or electrons) emitted upon impact with positrons incident upon a LiF(100) crystal in an arrangement illustrated in Figure 1. The LiF(100) crystal was heated constantly to 150°Celsius to minimize contamination and to mitigate electrostatic charging of the surface. The positron source was a beamline of NEPOMUC located at the research reactor FRM-II in Garching [14]. An 85 eV beam of moderated positrons was magnetically guided in a longitudinal magnetic field of 10 mT to the end of the beamline where it was extracted from the magnetic field through an 8 mm aperture in a shield made from magnetically conductive ARMCO iron and focused to a 1 mm spot by electrostatic optics onto the target. One of seven electrodes was formed by the inner part of the iron shield. A 90% transparent grid was placed at the entrance to the first electrode (at ground potential). The positron beam was accelerated between the grid and the iron shield where the magnetic field lines turn abruptly from the axial to the transverse direction as they are guided outwards through the shield. With sufficient acceleration nonadiabatic transmission through the rapidly varying magnetic field is possible and the beam divergence is small, as was demonstrated by Shi et al [15]. Finite element modeling with the Opera3D (Vector Fields, USA) code confirmed that with appropriately optimized potentials applied the electrodes, positron beam of initial width 7 mm, total kinetic energy of 85 eV and a Gaussian distribution of transverse kinetic energy of FWHM of 2 eV in the solenoid could be completely transmitted and focused to 1mm at the sample as illustrated in Figure b. The residual magnetic field at the crystal was negligible. Transmission was found to be poorer than expected due mostly to the large beam size and energy spread in the solenoid. The primary positron flux was estimated to be $5 \times 10^4$ s$^{-1}$ from the measured rate of positron annihilation at the sample.

Two 200 mm hemispherical analyzer’s ( Scienta R4000) with 2D detectors (multichannel plates (MCP) with resistive anode) were used to detect positrons and electrons and to record energy spectra. The analyzers, sample and positron optics were mounted in a UHV chamber. Either positrons or electrons could be analyzed by reversing the polarity of all the voltages applied to the lens, and analyser components, and by appropriately changing the potential between the front MCP and a grid in front it. The lenses and analyzers were configured to accept electrons or positrons with a mean kinetic energy of 30 eV with an analyser pass energy of 300 eV. Positrons (electrons) with energy $E_{e^+}$ ($E_{e^-}$) between 15 and 45 eV were measured in parallel. The total experimental energy resolution was estimated from the width of the elastic positron peak to be approximately 4 eV. Coincidence timing logic was employed applied to signals originating from the MCPs in order to record the impact parameters at each detector only when they both detected a particle and to measured the time between the detection of the two particles [13]. Data were acquired for $e^+e^-$ and $e^-e^-$pair emission upon positron impact for a total of 62 hours and 11 hours, respectively.

3. Results

Analysis of the time-interval distribution for the detected $e^+e^-$ pairs showed clearly that correlated $e^+e^-$ pairs were emitted from the LiF(100) surface in the energy range observed [13]. The detected intensity of correlated $e^+e^-$ pairs increased rapidly from sum energy $E_{\text{sum}} =$
Figure 1. (a) Simplified diagram of the experiment. (b) Schematic illustration of the magnetic shield and primary positron optics.

Figure 2. (a) Energy distribution of $e^+ - e^-$ pairs emitted from LiF(100) upon 85 eV $e^+$ impact. (b) The $e^+ - e^-$ pair sum energy ($E_{\text{sum}}$) distribution obtained from integrated intensity along a 14 eV wide strip centred along the line $E_{e^+} = E_{e^-}$ shown in (a), together with positron- and high energy resolution electron-excited $e^- - e^-$ pair sum energy distributions. (c) The number of detected pairs as a function of the time interval between the arrival of each particle at the detectors.

$E_{e^+} + E_{e^-}$ of approximately 60 eV toward lower sum energy. We focus our attention to the higher energy region where a weak ridge can be seen that along $E_{\text{sum}} \approx 72$ eV (indicated by a dashed white line in Figure 2a). It is revealed more clearly in Figure 2b which shows the integrated intensity along a 14 eV wide strip centred along the line $E_{e^+} = E_{e^-}$ against the sum energy $E_{\text{sum}} = E_{e^+} + E_{e^-}$. A histogram of the time intervals between the detection of the positrons and electrons in this energy range is shown in Figure 2c. The area of the peak corresponds to the number of correlated pairs, or true coincidences, that arise from single scattering event and the area of the uniform background beneath it gives the number of uncorrelated, or accidental coincidences. Only events corresponding to the area under the peak are included for energy analysis. The contribution from accidental coincidences is clearly very small ($\lesssim 5\%$).

The position of the peak in the ($e^+, e^+ e^-$) energy distribution corresponds to the maximum possible value of $E_{\text{sum}}$ for a correlated $e^+ - e^- e^-$ pair which is given by the difference between the incident positron kinetic energy, $E_i$ and the energy required to transfer an electron from the top of the valence band to the vacuum level, $E_B$. Because the LiF conduction band is close to the vacuum level, $E_B$ is approximately equal to the band gap $E_g = 13.0 \pm 0.4$ eV [3]. The peak can therefore be attributed to exclusively elastic scattering processes involving the incident positron and an electron from the top of the valence band. A corresponding feature is observed in the sum energy distribution of electron pairs obtained with better energy resolution using an electron (Figure 2b). The wide band gap of LiF prohibits an electron emitted from the valence band from losing a continuous range of energy by electronic excitations below $E_g$ (indicated in Figure 2). This leads to a region of low inelastic contributions in the electron-excited $e^- - e^-$ pair spectrum that allows pair emission from the valence band to be clearly distinguished [3]. A corresponding valley is observed in the $e^+ - e^-$ pair energy spectrum below 72 eV. At lower $e^+ - e^-$ pair energy the relative intensity of inelastic contributions increases rapidly. Differences in the electron excited $e^- - e^-$ and $e^+ - e^-$ pair distributions in this region may arise from differences in the inelastic scattering cross-sections for positrons and electrons, but reliable comparison requires similar energy resolution.
Correlated $e^-e^-$ pairs emitted upon 85 eV positron impact were also observed and their energy distribution measured (Figure 2). In contrast to the $(e^+, e^+e^-)$ pair energy distribution, pair intensity increases continuously below about 59 eV. This is consistent with energy being conserved between the $e^-e^-$ pair and the undetected positron, with the maximum energy available to the $e^-e^-$ pair in this case given by $E_i - 2E_b \sim 59$ eV.

Our angle-integrated data do not allow direct pair emission mechanisms to be distinguished from indirect ones, i.e. multiple scattering. However we can speculate that the mechanism of $e^-e^-$ pair emission upon positron impact is an indirect process whereby the incident positron transfers kinetic energy to a valence electron that subsequently interacts with a second electron causing the emission of both. In the case of positron-excited $e^+e^-e^-$ pair emission, we expect that with improved energy resolution and counting statistics, asymmetries in the energy distribution would be revealed, but to first order the mechanism by which the $e^+e^-$ pair are emitted is similar the multi-step scattering processes by which an $e^-e^-$ pair is emitted upon electron impact. We refer to Refs. [16, 17] for discussion of these mechanisms. Angle-resolved experiments will require considerably more time to obtain data with statistics adequate for detailed comparison of the $e^-e^-$ and $e^+e^-$ scattering dynamics but could still be completed in time typical of most $e^-e^-$ coincidence experiments.

4. Summary
We have shown that upon impact with 85 eV positrons, $e^+e^-e^-$ pairs were emitted from a LiF(100) surface. Structure in the pair energy distribution is attributed to the primary positron scattering with an electron at top of the valence band. Comparison of $e^-e^-$ scattering data obtained upon impact with electrons and positrons supports this interpretation. The data indicate that $e^+e^-$ pair emission spectroscopy with energy and angular resolution can be used to probe electronic correlations and mechanisms of electronic excitation in solids by low energy positrons.

Acknowledgments
We are grateful to Dr. C. Hugenschmidt, J. Mayer, P. Pikart and C. Piochacz for their advice and assistance at NEPOMUC.

References
[1] Schumann F O, Kirschner J and Berakdar J 2005 Phys. Rev. Lett. 95 117601
[2] Schumann F O, Winkler C and Kirschner J 2007 Phys. Rev. Lett. 98 257604
[3] Samarin S, Artamonov O M, Suzorova A A, Sergeant A D and Williams J F 2004 Sol. Stat. Commun. 129 389
[4] Feder R, Gollisch H, Meinert D, Scheunemann T, Artamonov O M, Samarin S N and Kirschner J 1998 Phys. Rev. B 58 16418
[5] Berakdar J, Gollisch H and Feder R 1999 Sol. Stat. Commun. 112 587
[6] Fominykh N, Henk J, Berakdar J, Bruno P, Gollisch H and Feder R 2000 Sol. Stat. Commun. 113 665
[7] Ortiz G, Souza I and Martin R M 1994 Phys. Rev. Lett. 80 353
[8] Nekovee M, Foulkes W M C and Needs R J 2001 Phys. Rev. Lett. 87 036401
[9] Perdue J P, Burke K and Wang Y 1996 Phys. Rev. B 54 16533
[10] Puska M J and Nieminen R M 1994 Rev. Mod. Phys. 66 841
[11] Stachowiak H and Borowski E 2005 Phys. Rev. B 71 245107
[12] Boroniski E and Stachowiak H 2005 Physica B: Cond. Matt. 366 168
[13] Van Riessen G, Schumann F O, Birke M, Winkler C and Kirschner J 2008 J. Phys.: Condens. Matt. 20 442001
[14] Hugenschmidt C, Schreckenbach K, Stadlbauer M and Straßer B 2005 Nucl. Inst. Meth. Phys. Res. A 554
[15] Shi M, Gerola D, Waeber W B and Zimmermann U 1995 Appl. Surf. Sci. 85 143
[16] Berakdar J and Das M P 1997 Phys. Rev. A 56 1403
[17] Berakdar J 2000 Nucl. Inst. Meth. Phys. Res. B 171 294