Correlation spectroscopy of condensed matter systems

F.O. Schumann, J. Kirschner, K. A. Kouzakov, and J. Berakdar

Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle, Germany

Abstract. We present experimental and theoretical evidence for the potential of the two-particle spectroscopy to explore the electron-electron interaction in condensed matter. The experiment consists of a single electron impinging onto a clean surface. Two electrons are then emitted simultaneously and their momentum vectors are resolved. The measured energy and angular pair correlations within the pair carry direct information akin to the electron-electron interaction in the sample. We also point out that the presence of the Fermi sea leads to a damping, and a suppression of the range of the electron-electron interaction.

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INTRODUCTION

Electronic correlations result in a variety of physical phenomena such as superconductivity and magnetism. Yet, the overwhelming majority of methods for the study of the nature of the electron-electron interaction in matter are based on an effective single-particle picture where electronic correlations are manifested as subsidiary structures in the measured quantities [1, 2]. Here we explore the potential of the two-particle spectroscopy for the study of the electron-electron interaction in matter, specifically a primary single electron incident on a surface results in the emission of (time) correlated electron pairs. We employ a novel time-of-flight coincidence set-up consisting of a small central collector surrounded by a resistive anode.

Here we report two key observations. If the electrons' energies E_1 and E_2 are tuned such that the pair emission from the top of the valence band is possible, a zone of reduced intensity with a diameter of 1.6 Å^{-1} is visible in the coincidence signal whenever the escaping electrons momenta are comparable in magnitudes and directions. This correlation and exchange induced hole disappears if the sample electron originates from below the top of the valence band which indicates the sensitivity of the xc-hole to inelastic, phase-breaking scattering processes. We comment on these findings from a theoretical point of view.

THEORETICAL FORMULATION

Electronic correlation in an N particle system is described conventionally by the reduced two-particle density matrix $\gamma_2(x_1, x_2, x_1', x_2')$. γ_2 is expressible in terms of the N-particle

wave function Ψ as

$$\gamma_2\left(x_1,x_2,x_1',x_2'\right) = N(N-1)\int \Psi\left(x_1,x_2,x_3,\cdots,x_N\right)\Psi^*\left(x_1',x_2',x_3,\cdots,x_N\right)\,dx_3\cdots dx_N. \quad (1)$$

Here $x_j,\ j = 1\cdots N$ label the spin and the position coordinates. For fermions eq.(1) shows that $\gamma_2\left(x_1,x_2,x_1',x_2'\right) = -\gamma_2\left(x_2,x_1,x_1',x_2'\right)$. The two-particle density derives from γ_2 as $\rho_2\left(x_1,x_2\right) = \gamma_2\left(x_1,x_2,x_1,x_2\right)$. For fermions ρ_2 vanishes if $x_2 = x_1 = x$, i. e. $\rho_2(x,x) = 0$. For independent particles $\rho_2\left(x_1,x_2\right)$ is related to the single particle density $\rho(x)$ via $\rho_2\left(x_1,x_2\right) = \rho(x_1)\frac{N-1}{N}\rho(x_2)$. Thus, for overlapping fermions the antisymmetry of Ψ implies a correlation among the particles that results in the existence of the (Fermi) hole in the two-particle density for $x_1 = x_2$ and which we will expose in some details below. The electrostatic Coulomb repulsion between the electrons also contribute to the hole which is quantified conventionally by the xc hole [1, 3] $h_{xc}(x_1,x_2) = \frac{\rho_2(x_1,x_2)}{\rho(x_1)} - \rho(x_2)$. The pair-correlation function g, as defined in the literature, is given by $g(x_1,x_2) = h_{xc}(x_1,x_2)/\rho(x_2) + 1 = \rho_2(x_1,x_2)/[\rho(x_1)\rho(x_2)]$.

Role of exchange

To illustrate the role of exchange let us consider the two-particle probability density $g(\mathbf{r}_1,\mathbf{r}_2)$ (also called two-particle correlation function) associated with a two-particle plane wave $\Psi_{\mathbf{k}_1,\mathbf{k}_2} = \exp[i(\mathbf{k}_1\cdot\mathbf{r}_1 + \mathbf{k}_2\cdot\mathbf{r}_2)]$. The electrons' wave vectors are denoted by $\mathbf{k}_1,\mathbf{k}_2$. Assuming the spin and spatial degrees of freedom to be decoupled we find that g is uniform for the singlet channel. For the triplet channel however it reads

$$g(\mathbf{r}_1,\mathbf{r}_2) = \Psi^*_{\mathbf{k}_1,\mathbf{k}_2}\Psi_{\mathbf{k}_1,\mathbf{k}_2} = 1 - \cos[(\mathbf{k}_1 - \mathbf{k}_2)\cdot(\mathbf{r}_1 - \mathbf{r}_2)]. \quad (2)$$

Assuming that the two electrons are immersed in a Fermi sea and if there are N distinct $\mathbf{k}$ states occupied, we find for the ground-state average of g that

$$\begin{aligned}\langle g(\mathbf{r}_1,\mathbf{r}_2)\rangle &= \frac{1}{N^2}\sum_{ij}(1 - e^{i(\mathbf{k}_i - \mathbf{k}_j)\cdot\mathbf{R}}), & (3)\\ &= 1 - F^2(k_F R), & (4)\end{aligned}$$

where $\mathbf{R} = \mathbf{r}_2 - \mathbf{r}_1$, k_F is the Fermi wave vector, and the function F is given by

$$F(k_F R) = 3\frac{\sin(k_F R) - (k_F R)\cos(k_F R)}{(k_F R)^3}. \quad (5)$$

From these equations we see that the result of exchange is the appearance in *the triplet channel* of a hole (the Fermi hole) at $R = 0$ with an undamped oscillations away from $R = 0$. Averaging over the ground-state Fermi sea leads to a damping of the oscillations and hence to a finite range of the hole. The period of the oscillations are directly determined by the Fermi wave vector, as in the case of Friedel oscillations. In the singlet channel there is no trace of exchange observable in g, however in a more realistic description the Coulomb interaction will contribute additionally to the hole (correlation hole). In a spin-resolved experiment these two contribution can be investigated (but not disentangled).

Pair emission and the two-particle density

The measured coincidence cross sections in the present experiment can be related to ρ_2. To show this we remark that the probability P_{if} for the reaction under consideration is given as $P_{if} = S_{if}S_{if}^*$. Here the S matrix elements are given by $S_{if} = \langle \Psi_{E_f} | \Psi_{E_i} \rangle$ and Ψ_{E_i} (Ψ_{E_f}) is the normalized wave function describing the system in the initial (final) state with the appropriate boundary conditions. The initial state with energy E_i consists of the incident electron interacting with an electron in the valence band in the presences of all other particles in the system (over which we will average eventually). The two vacuum electrons have the energy $E_f = E_1 + E_2$.

Assuming the surrounding medium is not affected while the incident and the valence band electron are interacting and during the emission of the two electrons (i.e. within a frozen-core picture) we find that $\Psi_{E_i} \approx \psi_{E_i}(x_1, x_2)\chi(x_3, \cdots, x_N)$. ψ_{E_i} is the electron pair wave function in the initial state. The function χ describes the surrounding medium. The reduced density matrix (1) takes on the form $\gamma_2(x_1, x_2, x_1', x_2') \approx 2\psi(x_1, x_2)\,\psi^*(x_1', x_2')$. For a further progress we assume the emitted electron pair state ψ_{E_f} to be described by plane waves. Under these conditions the measured, spin (σ_j) unresolved probability reads $P_{if} \propto \sum_{\sigma_1,\sigma_2,\sigma_1',\sigma_2'} \tilde{\psi}_{E_i}(\sigma_1\mathbf{k}_1, \sigma_2\mathbf{k}_2)\tilde{\psi}_{E_i}^*(\sigma_1'\mathbf{k}_1, \sigma_2'\mathbf{k}_2)$, where $\tilde{\psi}_{E_i}$ is the double Fourier transform of ψ_{E_i}.

From the above relations we conclude that the present experiment measures the spin-averaged diagonal elements of the reduced density matrix in momentum space which is the spin-averaged, momentum-space two-particle probability density ρ_2, a quantity that we discussed above.

EXPERIMENTAL DETAILS

The experiments were conducted under UHV conditions featuring a novel time-of-flight spectrometer depicted in fig.1. The sample was a LiF(100) single crystal which was cleaned and annealed. During the measurements the sample was kept at a temperature of $\sim$ 400 K, this avoids the charging up of the sample. Primary electrons delivered from a pulsed electron gun hit the sample with an angle of $\sim$80 degree with respect to the surface normal. Ejected electrons can move towards the spectrometer, where a pair of hemispherical grids ensure a field free region between the sample and a multi-channel plate. The resulting electron avalanches hit two detectors. A central collector accepts electrons only within a solid angle of $\sim$0.1 sr, this detected electrons we refer to as "e_1". A resistive anode is the second detector which allows for a spatial resolution of the impact position. Electrons registered within a solid angle of $\sim$1 sr are termed in what follows as "e_2".

Using fast timing signals and an electronic coincidence set-up allow for the determination of the flight times, which then can be converted into the energies E_1, E_2 when considering the flight path of $\sim$58 mm. The impact position on the resistive anode can now be converted into momentum space. In this way we map out the energy and momentum dependence of the electron pair correlation. The time resolution of the set-up is better than 1 ns as determined by the width of the elastic peak in the time-of-flight spectrum. Thus, the energy resolution depends on the kinetic energy, which for the results

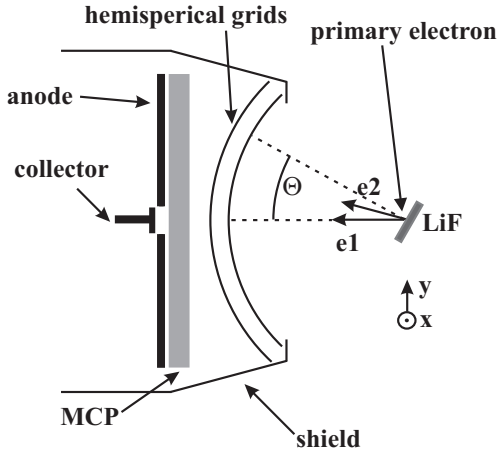

FIGURE 1. Electron pair detection technique. Two electrons with momenta $\mathbf{k}_1$, $\mathbf{k}_2$ and energies E_1 and E_2 are detected in coincidence by a resistive anode and central collector. The polar angle Θ denotes the angle between the surface normal and the central axis of the spectrometer.

presented here is 0.5 eV. The accuracy of the momentum resolution of the coincidence events is primarily determined by the acceptance angle of the central collector and the specific momenta under consideration, a typical value is 0.1 $Å^{-1}$, respectively.

EXPERIMENTAL RESULTS

In fig.2 we plot the 2D energy distribution of the coincidence electron pairs upon excitation with 32.7 eV electrons. We observe the onset of pair emission when the sum energy E_1+E_2 equals $\sim$19 eV, which is indicated by the dashed diagonal line. This position can be easily understood when considering the bandstructure of LiF. The energy required to excite an electron from the top of the valence band to the vacuum level is $\sim$ 14 eV, hence the maximum sum energy of the scattered electron and the valence band electron is expected to be 32.7-14=18.7 eV in agreement with the experiment.

More insight can be obtained if we take advantage of the lateral resolution of the set-up. In a first step we select only those coincidences for which the energies E_1 and E_2 are fixed. In other words we focus on a small region in the 2D energy distribution, as indicated in fig.2. In order to obtain sufficient statistics we actually select an energy window of $\sim$ 1.6 eV around the respective energies. This has been indicated by the square boxes in fig.2 labelled a)-c). We can now proceed and plot the coincidence intensity as a function of the in-plane momentum $k_{\|}$ of the electron "e_2". We have selected three different regimes within the 2D energy distribution highlighted in fig.2 by the black squares a)-c). In the case a) and b) we are right at the onset of pair emission, in other words we move along the dashed diagonal line. Case c) describes the situation if emission below the highest occupied level is possible. In fig.3 we display the resulting momentum distributions. We would like to point out that all momentum plots display a zero intensity at a position where the central collector is positioned. The position and size of this "blind spot" depends on the polar angle Θ and the momentum of the

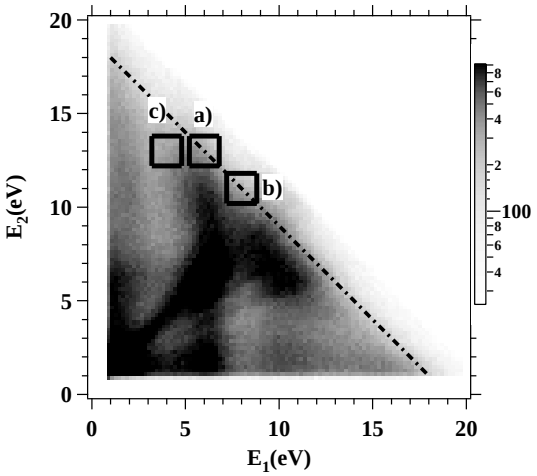

FIGURE 2. The 2D energy distribution for coincident pairs is shown for which the primary energy was 32.7 eV and the polar angle Θ=0°. The energies E_1 and E_2 refer to the electron e_1 and e_2 electron, respectively, as displayed ion fig.1. The dashed diagonal line indicates the onset of the pair emission. The square boxes (width of 1.6 eV) indicate narrow regions which were chosen to map the coincidence intensity in momentum space, see fig.3

electron "e_2". For the plots shown in fig.3 this "blind spot" is centered at $k_{||}$=0 and has diameter of $\sim$0.3 $Å^{-1}$. In fig.3 a) the energies are E_1=6 eV and E_2=13 eV (box a) in fig.2), respectively. We clearly observe that the region $k_{||}$=0 (outside the "blind spot") is surrounded by a region of diminished intensity. The intensity increases for larger $k_{||}$ values and reaches a maximum for $k_{||}$= 0.8$Å^{-1}$. It should be stressed however that these pictures do not conclusively provide information on the size of the exchange and correlation hole because the fall-off of the intensity at large momenta is experimentally inevitable due to the finite size of the detectors.

Fig. (3c) (in which E_1=4 eV and E_1=13 eV) demonstrates a new aspect of the correlated electron-pair emission: Here we observe that the ring of enhanced intensity is essentially filled up. Energetically the sum energy E_1+E_2 has been reduced from 19 eV to 17 eV. This energy difference allows for the excitation of other modes of the sample and opens thus the channel of inelastic scattering processes that lead to a decoherence of the escaping electrons' wave [4]. The influence of these phase-breaking processes on the correlation within the electron pair is illustrated in fig.3 a) and b).

We have also performed experiments where the polar angle was varied. The results are displayed in fig.3 for excitation with 30.7 eV electrons. The energies are selected to be at the onset of pair emission (E_1=6 eV, E_2=11 eV). For a polar angle of 0° the momentum distribution is equivalent to fig.3 a) and b). Changing the polar angle to 10° and then to 20° moves the ring of enhanced intensity with it such that it surrounds the "fixed" electron. This shows that the intensity enhancement is associated with the fixed electron.

We may summarize our observations obtained also with different primary energies as follows: (i) if we select the energies E_1 and E_2 such that the sum energy E_1+E_2 has the largest possible value for pair emission, the 2D momentum plots display an ring of

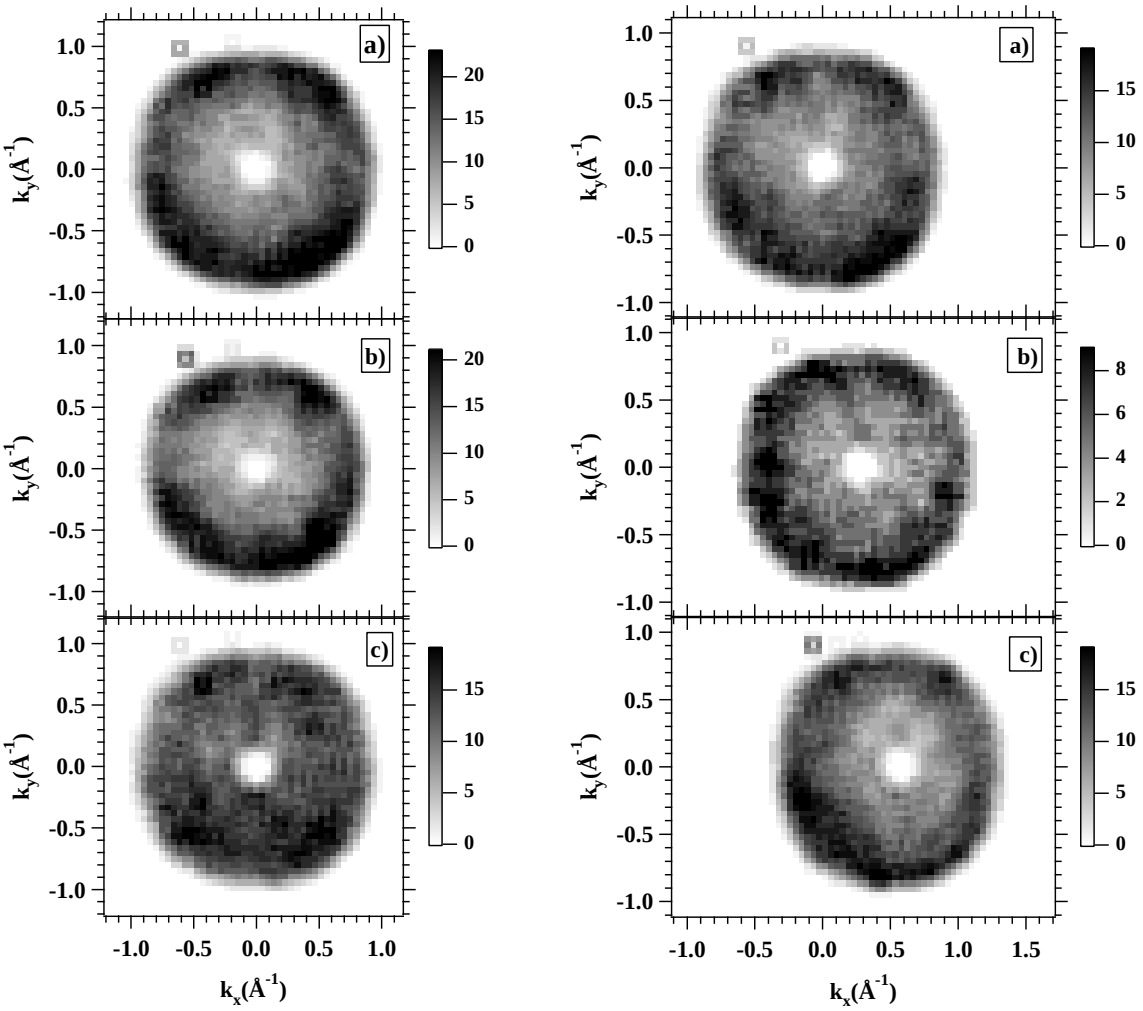

FIGURE 3. **Left panel** shows the 2D $k_{\|}$ distribution of the coincidence intensity (Θ=0°)for different regions in the 2D energy plane, which refer to the boxes a)-c) of fig.2. **Right panel** shows 2D $k_{\|}$ plots obtained upon excitation with 30.7 eV primary electrons. The energies are E_1=6 eV and E_2= 11 eV, respectively. Different polar angles Θ (see fig.1) have been used which are a) 0°, b) 10° , c) 20°.

enhanced intensity which is centered around the "fixed" electron. (ii) if the sum energy is below the maximum value a more or less uniform momentum distribution is the result.

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