Two-Electron Coincidence Spectroscopy of Scattering Events at Surfaces

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We report on the first successful two-electron coincidence experiment at solid surfaces in a back-reflection geometry. We found that two-electron coincident electron emission events exist, and that they require a certain threshold energy of the primary electron (relative to the Fermi level) which is not equal to twice the work function of the sample surface [W(100)] as one might expect from the energy conservation law. We give evidence that we observe at low primary energy single electron-electron scattering events between the primary electron and a valence-band electron and give a semiquantitative estimate of the relative probability of such events.

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Electron coincidence experiments of the type one electron in–two electrons out $(e,2e)$ have been very successfully applied in atomic and molecular physics to study the dynamics of electron scattering and the momentum density in the target [1,2]. With solids this concept is much harder to realize, since most of the primary electrons and those excited by them are lost in the bulk. Only transmission experiments with high-energy electrons ($\sim$10 keV) through thin foils have been successfully carried out today [3–5]. They showed convincingly that the momentum density in, e.g., bulk graphite can be measured as a function of crystal momentum by a coincidence technique. Corresponding experiments with low-energy electrons in a back-reflection geometry, to the best of our knowledge, have not been successfully carried out previously [6–8]. On the other hand, a number of early theoretical calculations [9–11] have demonstrated how desirable such a technique would be to study the electronic structure at surfaces from a different point of view than usual. It is now almost routine to determine momentum-resolved energy eigenvalues and densities of electron states in the valence band by photoemission. It would be of similar importance to map out the momentum density in quasimomentum space, i.e., the number of momentum states in an interval dk around a given momentum k. This information is not directly available from photoemission but would be from an $(e,2e)$ experiment performed in coincidence [1–3,9,10]. Our results may be a first step towards realizing this goal. They show that two-electron scattering events at surfaces can be observed in coincidence and demonstrate the important role of the work function.

Our experiment is described schematically in Fig. 1 (left-hand panel). A W(100) crystal is irradiated by electrons from a low-energy gun with primary energy E_p from 5 to 100 eV relative to the W Fermi level. Two

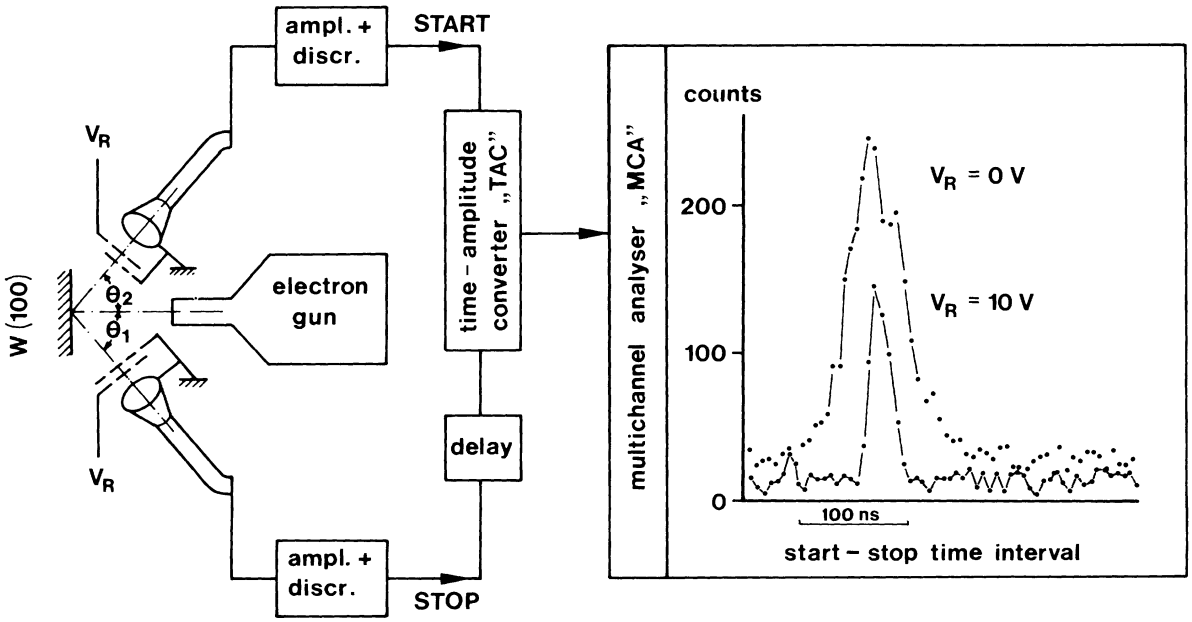

FIG. 1. Schematic view of the two-electron coincidence experiment (left-hand panel) and typical time-of-flight spectra at 40 eV primary with two different retarding potentials V_R at the same primary current and the same measurement time (right-hand panel).

channeltrons, each with a retarding grid energy analyzer, detect electrons from the target in the pulse-counting mode. One of the channeltrons starts a time-to-amplitude converter (TAC), while the other, with an adjustable but fixed delay, stops the TAC. The TAC output is analyzed by a multichannel analyzer (MCA). A typical result is shown in Fig. 1 (right-hand panel), giving the number of start/stop events over the start/stop time interval ($E_p = 40$ eV). Correlated electron emission events appear on the time scale of the MCA at a time given by the delay time in the stop channel plus the flight time difference of the two electrons. Uncorrelated start/stop events may have any time interval between start and stop and thus appear as a "white noise" background in Fig. 1. The electronic time resolution is of order of 5 nsec, i.e., much less than the width of the peaks. The broadening is due to the differences in flight time between fast and slow electrons. This is demonstrated in Fig. 1 by the different widths of the distribution obtained for two different retarding potentials on the grids. Increasing the retarding voltage suppresses the slow electrons, hence decreasing the range of possible transit times between fast and slow correlated electrons. The maximum of the peak corresponds to electrons with equal time of flight (TOF) in the two channels, hence equal energy. The tail on the right-hand side corresponds to fast electrons in the start channel and slow electrons in the stop channel, and vice versa for the tail on the left-hand side.

The very existence of the peaks in Fig. 1 proves the existence of correlated two-electron emission events. The substantial decrease of the coincidence rate at high retarding potential points out the important role of low-energy electrons in two-electron emission events. The fact that the true coincidence distribution curves are located at the same point on the time scale and are single peaked indicates that most of the electrons producing coincidences have similar energy. It is easy to see that the distribution would be, e.g., twin peaked if a two-electron emission process involved always one slow and one fast electron.

Measurements are carried out in vacuum of order of 10^{-11} Torr. The W(100) single crystal (10×5 mm) undergoes a routine final treatment in vacuum [12,13]. The cleaning is checked by LEED monitoring. During the measurements the sample was occasionally cleaned by high-temperature flashes up to 1300°C to desorb CO. The (010) plane of the sample is parallel to the detection plane. The entrance apertures of both detectors are circular, defining a solid angle of about 0.02 sr. The angular positions of the detectors, the sample, and the electron gun were checked optically and also by diffracted electron beams.

The number of true coincidence events is given by the sum of the number of counts in a certain number of the MCA channels, chosen to lie near the maximum of the time-of-flight spectrum, after subtraction of the background. Statistical analysis of the spectra has shown that the distribution of the accidental coincidence events is in good agreement with the Poisson distribution. This fact allows us to extrapolate the background measured outside the maximum of the TOF spectra over a time interval of about 2 μsec, into the region of the true coincidence events to determine their net intensity.

The number of true coincidence events (TCE) was studied as a function of the primary electron energy at fixed geometry and zero retarding potential. The detector positions are symmetrical to the surface normal: $\theta_1 = \theta_2 = 60°$. The primary electron incidence angle is $\phi = 20°$ off the emission plane. The result is shown in Fig. 2. Each point has been obtained by normalizing the TCE number to 10^6 start pulses. In order to determine the TCE probability per incident electron we measured the number of primary electrons necessary to excite 10^6 start pulses and took into account the dependence of the W secondary emission coefficient on the primary electron energy. The number of true coincidence events per primary electron obtained this way as a function of primary energy is shown in Fig. 2. The curve shows a roughly

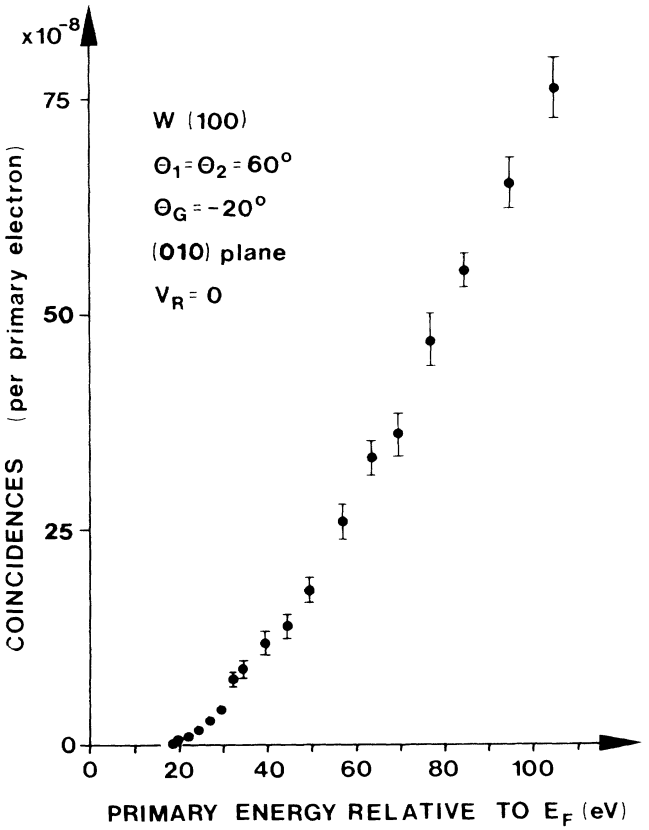

FIG. 2. True coincidence events per primary electron as a function of the primary electron energy referred to the Fermi level of the sample. Both detectors are in the same plane and have an angle of 60° relative to the surface normal. Note the threshold near $E_p = 18$ eV, below which no true coincidences have been observed.

quadratic energy dependence with a clear threshold energy near 18 eV above E_F. This threshold energy is much higher than one would expect for the simultaneous emission of two secondary electrons excited in a "collision cascade," which is the standard mechanism of secondary electron creation [14]. Since secondaries can be observed at an energy just above the vacuum level of the sample, the minimum primary energy for the emission of two secondaries is twice the work function. In the case of W(100) this amounts to 9.2 eV [15]. Such a threshold was not observed in our experiments.

The existence of a true coincidence electron emission threshold and a rough estimate of the minimum primary electron energy can be derived from the following simple kinematic considerations. Since we detect electrons in the back-reflection geometry, it is evident from the kinematics of particles with equal masses that the scattered electrons can only move in the forward direction because of the total momentum conservation. Therefore, it is necessary to include the event of elastic scattering on the lattice or on some other scattering center with a large mass into the kinematics of scattering into the back hemisphere. Let us assume for simplicity that the primary electron after the elastic scattering moves along the normal to the surface from the solid into vacuum (Fig. 3). In this case, as a result of scattering with a valence electron, two secondary electrons appear nearly at the threshold energy, both having energy equal to $\frac{1}{2}E_p$. In the case of scattering with an electron at rest, the scattering angle is 90°; i.e., each of the secondary electrons moves at an angle of 45° relative to the normal to the surface. Emission of such electrons into vacuum is possible if the normal component of the electron kinetic energy exceeds the magnitude of the potential barrier, which is equal to the work function. In this case the emitted electrons move parallel to the surface. It is easy to see that the appearance of correlated secondary electron pairs in vacuum is possible when the primary electron energy is 4 times as large as the work function of the sample, this energy in the case of W(100) being 18.6 eV relative to the Fermi energy. This value agrees with the threshold energy shown in Fig. 2. Since the detectors are positioned 60° from the normal to the surface, the threshold based on the preceding arguments should be 24.8 eV. We have no simple explanation yet for this discrepancy.

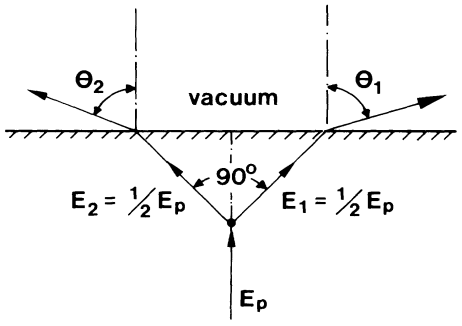

FIG. 3. Kinematics of a two-electron coincidence emission event with primary energy near threshold. After the collision two electrons travel nearly parallel to the surface with energy $E = \frac{1}{2}E_p$.

Indeed, the considered scattering geometry corresponds to the minimum primary electron energy, which can be seen more clearly from the following. The angle between two secondary electrons is strictly defined by the energy and momentum conservation laws and is equal to 90°, except in a trivial case of "knock-on" collisions when the primary electron stops and the secondary continues moving in the same direction. Therefore any deviation from symmetry relative to the normal to the surface appearing in the scattering geometry will lead to a decrease of the normal component of the velocity of one of the electrons and to an increase of that of the other. This means that one or the other of the electrons will not be able to cross the surface barrier and that a coincidence event will not be observed. (The inclusion of exchange does not alter this argument.) Of course, a more realistic kinematic scattering diagram should take into account, at least, the refraction of the electron trajectory at the interface for given detection angles, as well as the finite energies of valence electrons involved in the scattering event. Nevertheless, the diagram shown in Fig. 3 gives a reasonable idea of what should be taken into account in a more detailed consideration. We emphasize that the energy threshold for the detection of true coincidences appears because of the requirement of energy *and* momentum conservation in an elementary event of electron-electron scattering. In the case of conventional secondary electron spectroscopy, when the coincidence technique is not used, another secondary electron may have any momentum direction since in the cascade process of secondary electron generation any momentum information is lost [14].

To prove that the work function indeed plays a dominant role for the threshold energy of the electron coincidence events we manipulated the work function of the sample. Its effect on the position of the TCE threshold is shown in Fig. 4. On the W(100) surface, either a Cs layer has been deposited, which results in decreasing the surface work function down to 1.8 eV, or oxygen has been absorbed, which results in increasing the work function up to 5.5 eV. The work function changes have been checked by the shift of the I-V curve of the target current. The number of true coincidence events is normalized to 10^6 start pulses. Figure 4 shows that the threshold of the TCE appearance shifts in the same direction as the work function. The magnitude of this shift of the threshold energy agrees with the work function change multiplied by a factor of 4, as expected from the above arguments. This result proves directly the dominant role of the surface in these experiments.

We note that the coincidence electron yield near the threshold in Fig. 4 can be well approximated by a linear

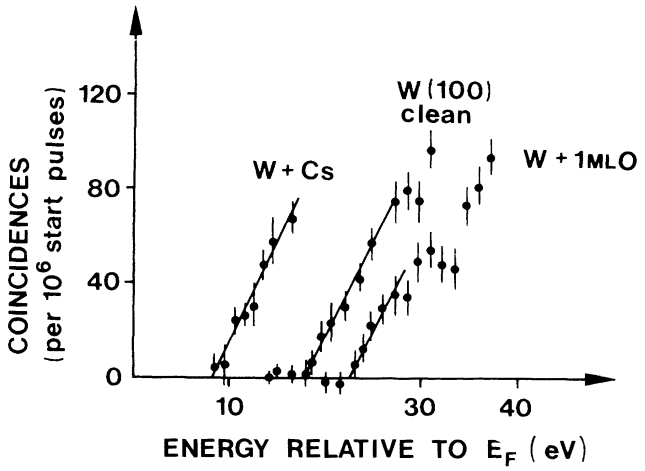

FIG. 4. Enlarged view of the threshold region for three different values of the work function of the W(100) crystal created by adsorption of Cs or O. Note the shift of the threshold to either side of that for clean W(100) corresponding to a decrease or an increase of the work function. The geometry of the experiment is similar to that of Fig. 2, but the number of coincidence events is normalized to 10^6 counts in the "start" channel.

energy dependence. This proves, rather than contradicts, the result found in Fig. 2, where an approximately quadratic dependence was observed. The reason is that here we normalized to the "start" pulses of the TAC. The normalization to the "stop" events leads to the same result. Thus, the coincidence rate, when normalized per *electron detected* in either of the channeltrons depends linearly on primary energy., When normalized per *primary electron* (such as in Fig. 2) the coincidence probability will be proportional to the product of the phase spaces accepted by each of the detectors. Since the latter are equal, a quadratic yield function is to be expected, in agreement with Fig. 2.

As seen in Fig. 2 the number of coincidence events per primary electron is of order of 10^{-7} (around 40 eV primary energy). This might lead to the conclusion that the effect observed here is of very minor importance. However, the low coincidence yield is primarily due to the back-reflection geometry of the experiment, and the small combined phase space of the two detectors, rather than to low intrinsic two-electron scattering probability. Let us assume we had a (hypothetical) device capable of detecting each two correlated electrons over the full half sphere in front of the sample. The (measured) angular distribution of the electrons (cosine to good approximation), and the (calculated) solid angle of the detectors imply that we would then obtain a coincidence yield of a few percent per incident electron. Further, the number of back-reflected electrons with no or small energy loss in this energy range is of order of 10^{-1}. Thus we arrive at the conclusion that for a back-reflected electron the probability of undergoing a scattering event leading to the emission of two electrons in coincidence is of order of 10% or more. Thus, the study of electron scattering events by coincidence techniques may lead to a new venue towards understanding elementary electron-electron interaction in the near-surface region of solids. For example, an obvious extension of the present experiments would include a polarized electron source and a magnetic sample to study explicitly the role of exchange in electron-electron scattering. This in turn is of great importance for, e.g., the understanding of the giant magnetoresistance effects discovered recently.

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