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Correction of the deterministic part of space–charge interaction in momentum microscopy of charged particles

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ABSTRACT

Ultrahigh spectral brightness femtosecond XUV and X-ray sources like free electron lasers (FEL) and table-top high harmonics sources (HHG) offer fascinating experimental possibilities for analysis of transient states and ultrafast electron dynamics. For electron spectroscopy experiments using illumination from such sources, the ultrashort high-charge electron bunches experience strong space–charge interactions. The Coulomb interactions between emitted electrons results in large energy shifts and severe broadening of photoemission signals. We propose a method for a substantial reduction of the effect by exploiting the deterministic nature of space–charge interaction. The interaction of a given electron with the average charge density of all surrounding electrons leads to a rotation of the electron distribution in *6D* phase space. Momentum microscopy gives direct access to the three momentum coordinates, opening a path for a correction of an essential part of space–charge interaction. In a first experiment with a time-of-flight momentum microscope using synchrotron radiation at BESSY, the rotation in phase space became directly visible. In a separate experiment conducted at FLASH (DESY), the energy shift and broadening of the photoemission signals were quantified. Finally, simulations of a realistic photoemission experiment including space–charge interaction reveals that a gain of an order of magnitude in resolution is possible using the correction technique presented here.

1. Introduction

The advent of intense pulsed photon and electron sources with pulse lengths in the femtosecond range opened the door to a new generation of experiments targeting ultrafast dynamics and short-lived transient states [1]. On the laboratory scale, numerous table-top devices for the production of ultrashort EUV or X-ray pulses [2] or photocathodes for electron guns [3] have been developed and are currently under development. Free electron lasers (FELs) [4] in particular have emerged as fascinating research tool to study atoms, molecules and solids with a time resolution of a few femtoseconds and a spatial resolution down to the Å range. One key question in condensed matter physics is how complex and often unexpected phenomena emerge from the competing interactions in a quantum many-body system, leading to remarkable macroscopic phenomena such as high-Tc superconductivity, colossal magnetoresistance and metal–insulator transitions. Time-resolved studies after controlled excitation with light pulses out of equilibrium offer the possibility to identify the most relevant relaxation channels and to potentially gain insight into the driving forces for unexpected ground state properties; for a recent perspective article, see [5].

Photoelectron spectroscopy (PES) is a well-established technique to study the electronic properties; however the information depth is limited to the topmost surface layers. New possibilities for increased depth information come from high-brilliance Synchrotron sources and FELs in the hard X-ray regime. The large mean free path of electrons with kinetic energies above ~5 keV opens access to bulk properties in hard X-ray PES (HAXPES) experiments. Due to the high pulse intensity of such sources, the Coulomb interaction of the particles released within a single short pulse becomes substantial and can result in prohibitively large energy broadenings [6] and a loss of angular- and even of spin

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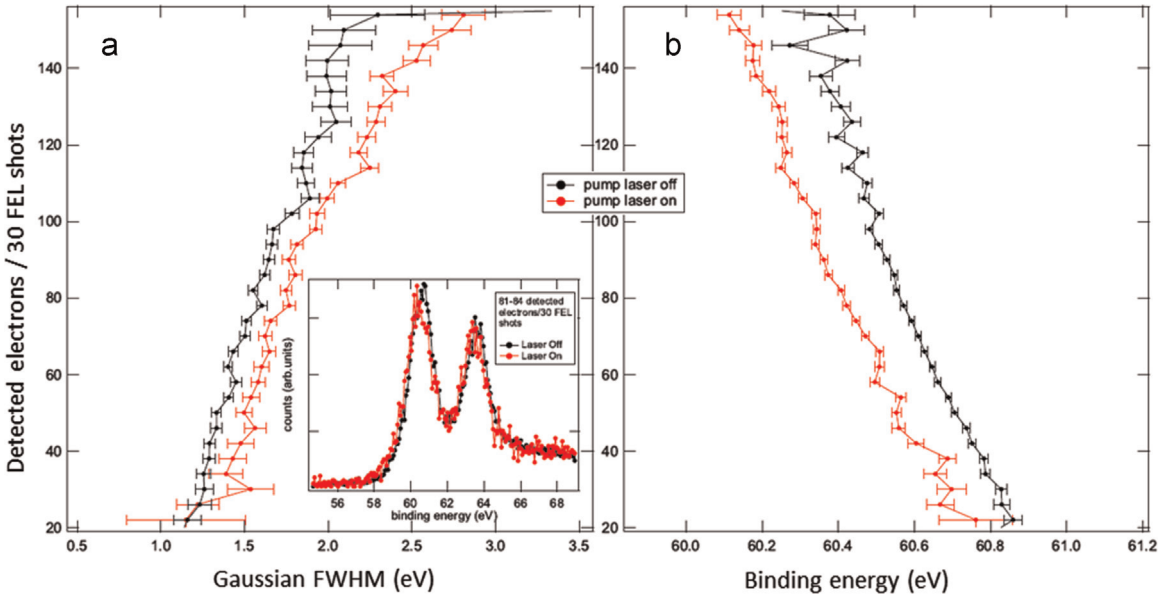

Fig. 1. Space–charge induced broadening (a) and apparent binding energy shift (b) of the Ir $4f_{7/2}$ peak in respect of the number of detected electrons per 30 FEL shots. The measurements where performed at the free-electron laser FLASH at DESY at an excitation energy of 190.7 eV, with (red trace) and without (black trace) a 800 nm optical pump pulse (2.1 mJ/cm^2, ^{2}50 kHz). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

information [7]. This is commonly referred to as the *space–charge problem*. As a consequence, electron spectroscopic and imaging methods like angle-resolved photoelectron spectroscopy (ARPES) and photoemission electron microscopy (PEEM) with pulsed sources are facing a dramatic loss in performance.

In common PES, e.g. using hemispherical analyzers or time-of-flight spectrometers being based on the entrance lens optics of such analyzers, the only way a sufficient energy resolution can be achieved is by reducing the photon intensity necessitating an increased acquisition time. The large retardation ratio between the initial kinetic energy, typically >5 keV, and pass energy through the analyzer (of the order of a few 100 eV) increases the beam size. In order to cope with the high lateral magnification of the entrance lens (necessary to fulfill Liouville's Theorem) the photon spot on the sample must be of the order of 50 μm or less. Repulsive Coulomb forces act during the expansion of the electron cloud in the vicinity of the small probing spot on the sample surface. Once the initial energy and angular distribution is washed out, it cannot be reconstructed and part of the information is lost.

This effect is clearly evident in Fig. 1, which shows data extracted from typical photoemission spectra taken with a hemispherical electron analyzer (Scienta SES 2002) at the free electron laser FLASH at DESY (Hamburg). The $4f_{7/2}$ core-level of an Ir(111) single crystal was studied at an excitation energy of 190.7 eV. The spectra were sorted according to the number of detected electrons in a bunch train of 30 bunches with a separation of 4 μs (black error bars, denoting the statistical error). Additionally, we excited the sample with a 2.1 mJ/cm^2 pump laser at 800 nm (red error bars). In this case, we create slow electrons in front of the sample surface by multi-photon excitation. The effect of space–charge induced broadening and apparent binding-energy shift with increasing electron number is clearly visible. The ordinate shows the number of detected electrons, which is proportional to the number of emitted electrons. The space–charge effect (sum of deterministic and stochastic contribution) is enhanced in the presence of the pump laser and hence the presence of a slow electron cloud at the sample surface.

In this paper we describe a method to minimize space–charge induced effects in photoelectron spectroscopy experiments by manipulating the time where Coulomb repulsion between photo-emitted fast electrons and slow secondary electrons is significant. The influence of the Coulomb interaction on the electron trajectories is minimized by the following means: (i) Accelerating the electrons away from the source region with a high electrostatic field that acts differently on electrons with different energies, (ii) rapid separation of the momentum discs of fast and slow electrons and (iii) exploiting the fact that the deterministic part of the Coulomb interaction originates from the interaction of a given electron with the average charge density of all other electrons.

3D particle tracking techniques have been performed for a monochromatic electron ensemble and for a model distribution of two different electron species (fast electrons model the valence range and slow electrons the secondary cascade). The electric-field distribution in the source region was assumed to have realistic parameters common to electrostatics cathode lenses. Details of the space–charge effect and its spatiotemporal evolution were determined for randomly generated initial bunch charges between 0.1 and 100 fC with spatial and temporal Gauss profiles of 10 μm and 30 fs FWHM, respectively (note that Fig. 1 shows only the intensity of the detected electrons being much smaller than the total number or released electrons). The results indicate that the deterministic part can be corrected with a momentum microscope: a cathode-lens type instrument optimized for best resolution in reciprocal space. The simulations reveal a gain of one order of magnitude in spectroscopic performance. The first experiments using synchrotron radiation qualitatively confirm the expected behavior for photon fluxes where the space–charge interaction becomes significant.

2. Momentum microscopy

Momentum microscopy is a novel technique that was developed for detecting the *k*-distribution of an ensemble of charged particles in a parallel-imaging device. Fig. 2 illustrates the basic principle of such an instrument. Electrons emitted from the sample are accelerated by a strong electrostatic immersion field forming part of the cathode lens (the planar sample surface acting as cathode). The backfocal plane of the objective lens is the image plane for the reciprocal (or Fourier) image. The radial coordinate in

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