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Space charge effect and mirror charge effect in photoemission spectroscopy

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Abstract

We report the observation and systematic investigation of the space charge effect and mirror charge effect in photoemission spectroscopy. When pulsed light is incident on a sample, the photo-emitted electrons experience energy redistribution after escaping from the surface because of the Coulomb interaction between them (space charge effect) and between photo-emitted electrons and the distribution of mirror charges in the sample (mirror charge effect). These combined Coulomb interaction effects give rise to an energy shift and a broadening which can be on the order of 10 meV for a typical third-generation synchrotron light source. This value is comparable to many fundamental physical parameters actively studied by photoemission spectroscopy and should be taken seriously in interpreting photoemission data and in designing next generation experiments.

Keywords: Space charge; Mirror charge; Photoemission; Fermi level shift; Fermi level broadening

1. Introduction

Photoemission spectroscopy measures the energy distribution of photo-emitted electrons when materials are irradiated with light (Fig. 1) [1,2]. It is widely used in solid state physics and chemistry for investigating the electronic structure of surface, interface and bulk materials [1,2]. Recently, it has become a prime choice of technique in studying strongly correlated electron systems [3,4], such as high temperature superconductors [5]. The availability of synchrotron light sources and lasers, combined with the latest advancement of electron energy analyzer, has made a dramatic improvement on the energy resolution of photoemission technique in the last decade; an energy resolution of ∼5 meV or better can now be routinely obtained. These achievements have made it possible to probe intrinsic properties of materials and many-body effects [5]. For example, measurements of the superconducting gap on the order of 1 meV, as in conventional superconductors [6] and in some high temperature superconductors [7], have been demonstrated.

On the other hand, the utilization of pulsed light sources, such as synchrotron light or pulsed lasers, has also brought about concerns of the space charge effect [8]. When a large number of electrons are generated from a short pulsed source and leave the sample surface, the electrons will first experience a rapid spatial distribution depending on their kinetic energy. Then, because of the Coulomb interaction, the fast electrons tend to be pushed by the electrons behind them while the slow electrons tend to be retarded by those fast electrons. This energy redistribution will distort the intrinsic information contained in the initial photoelectrons by giving

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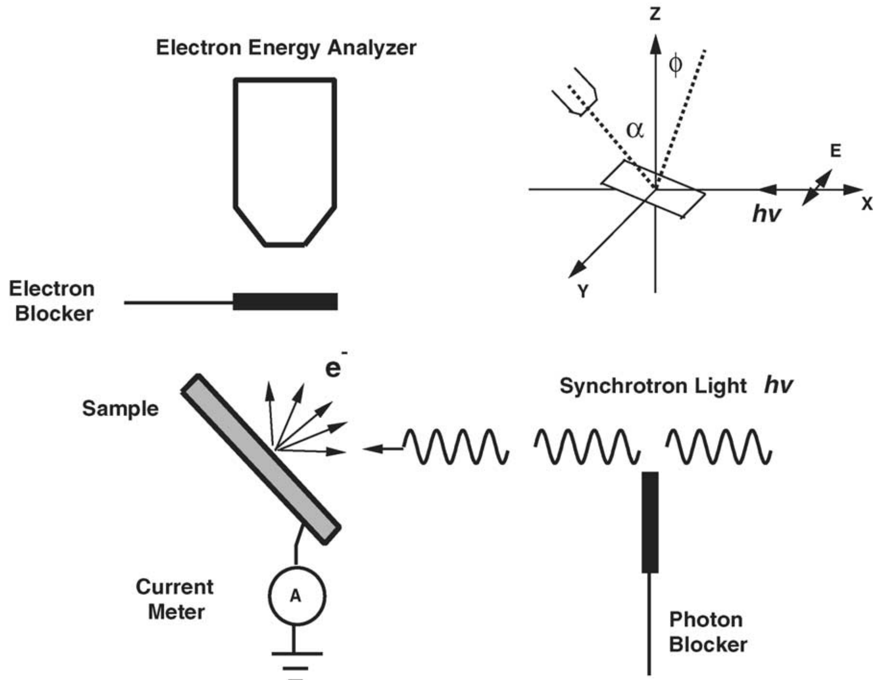

Fig. 1. Schematic of photoemission setup. A pulsed light is incident on the sample, kicking out electrons, and the electrons are collected by the electron energy analyzer. The photon blocker is used to change photon flux while keeping the beamline intact. The electron blocker is used to change the number of electrons collected by the analyzer. The sample current recorded by a picoammeter measures the number of electrons out of the sample which is proportional to the photon flux. In the upright, inset shows the measurement geometry of the light, the sample and the analyzer. The synchrotron light is along the X axis, with its electrical field E in the XY horizontal plane and parallel to Y axis. The sample normal is in the XZ plane and its angle with respect to the Z axis is referred to as φ. The analyzer is rotatable and the lens axis is in the YZ plane. The angle of the lens axis with respect to the Z axis is referred to as α.

rise to two kinds of effects. One is a general broadening of the energy distribution, due to both acceleration and retardation of electrons in their encounters. The other is a systematic shift in the energy. The space charge broadening of the energy distribution has been known for a long time as a limiting factor in electron monochromators and other electron beam devices [9], but it has not been considered in photoemission until very recently [8]. The main concern there was whether such an effect will set an ultimate limit on further improving the energy resolution of the photoemission technique [8].

Here, we report the first experimental observation of the space charge effect in photoemission. In addition, by combining experimental measurement with numerical simulations, we show that the mirror charges (also known as image charges in the literature) in the sample also play an important role in the energy shift and broadening. The combined effect of these Coulomb interactions gives an energy shift and broadening on the order of 10 meV for a typical third-generation synchrotron light source, which is already comparable or larger than the energy resolution set by the light source and the electron analyzer. The value is also comparable to the many-body effect actively pursued by modern photoemission spectroscopy. These effects, therefore, should be taken seriously in interpreting experimental data and in designing next generation experiments.

2. Experiment

The experiment was carried out on beamline 10.0.1 at the Advanced Light Source. This is a third-generation synchrotron source which generates pulsed light with a frequency of 500 MHz and a duration of ∼60 ps. The beamline can generate linearly-polarized bright ultraviolet light with a photon flux on the order of 10^{12} photons/s with a resolving power $E/\Delta E$ of 10,000 (E is the photon energy and ΔE the beamline energy resolution). The endstation is equipped with a high resolution Scienta 2002 analyzer. The analyzer, together with the chamber, is rotatable with respect to the beam while the sample position is fixed. The measurement geometry is illustrated in the upright inset of Fig. 1. There are two angles to define the direction of electrons entering the analyzer with respect to the sample normal: tilt angle φ and analyzer rotation angle α. We measured the sample current to quantitatively measure the number of electrons escaping from the sample which is proportional to the photon flux. With the pulse frequency of 500 MHz at the ALS, 1 nA of the sample current corresponds to 12.5 electrons per pulse.

Fig. 2a shows a typical photoemission spectrum of polycrystalline gold taken with a photon energy of 35 eV. It consists of a Fermi edge drop (E_F) near ∼30 eV, valence band between 20 and 30 eV and a secondary electron tail extending to