

Recoil Effect of Photoelectron in the Fermi-Edge of Simple Metals

In X-ray photoelectron spectroscopy, the emitted electron kicks the atom from which it is ejected in accordance with the conservation of momentum. For an atom with mass M in free space, the recoil energy δE is simply estimated as $\delta E = (m/M)E_{kin}$, where m is the electron mass, and E_{kin} is the electron kinetic energy. This gives rise to a loss in E_{kin} of the emitted photoelectron. This recoil effect is usually negligible because of the enormous mass difference between the atom and the electron, as long as the excitation energy is not very large. Quite recently, however, clear evidence of recoil effects has been found in the hard X-ray photoelectron spectra of graphite [1]. Under the excitation of the core level by X-ray photons with an energy of several keV, the photoelectron spectra show a remarkable shift and broadening as compared to the case of excitation by soft X-rays. Similar spectroscopic features of recoil effects have been observed in other fundamental processes such as elastic electron scattering and neutron scattering [2].

In the present study, we have investigated the recoil effects in the photoelectron spectra for valence levels [3]. In the core level spectra, one may expect that the recoil effect should be observed because core electron is localized around the atom (see Fig. 1). On the other hand, since Bloch electrons in a valence band are delocalized all over the crystal (Fig. 1), it is not clear whether the recoil momentum is shared by all the atoms of the crystal. This would result in no observable recoil effect, just like the recoilless transition in the Mössbauer effect. In contrast to this naïve expectation, we have found a clear evidence of the recoil effect for conduction electrons

in a simple metal from our experiments.

Hard X-ray photoelectron spectra of Au and Al at the excitation energy of 7940 eV and soft X-ray spectra of 880 eV were measured at the undulator beamlines **BL29XU** and **BL17SU**, respectively, using hemispherical electron energy analyzers.

Figure 2 shows the photoelectron spectra around the Fermi edge of Au (diamonds) and Al (circles) measured at 20 K with the excitation energy of 7940 eV. The binding energy scale is calibrated by assuming that the recoil energy in Au ($M=197$) is negligible. It is clear that the Fermi edge of Al ($M=27$) is shifted to higher binding energy. This shift is due to the kinetic energy loss of photoelectron from the Fermi level in Al because of its lighter atomic mass compared to Au, and is an evidence of the recoil effect of Bloch electrons. Furthermore, the edge profile has a slightly larger slope for Al than for Au. Conventional fitting analysis of these Fermi edge profiles using the Fermi-Dirac function (not shown) elucidates an energy shift of 120 meV in Al relative to Au, and Gaussian widths of 160 meV for Al and 124 meV for Au. The broadening of the width in Al is also a sign of the recoil effect because the contribution of the instrumental energy resolution to the width is the same in these spectra.

Soft X-ray spectra of Au (diamonds) and Al (circles) measured at 50 K with the excitation energy of 880 eV are shown in Fig. 3. Although the difference of the spectrum between Al and Au is small in this excitation energy, recoil effect does exist even in soft X-ray spectra.

The observed recoil effect of photoelectrons in the Fermi edge has been theoretically interpreted.

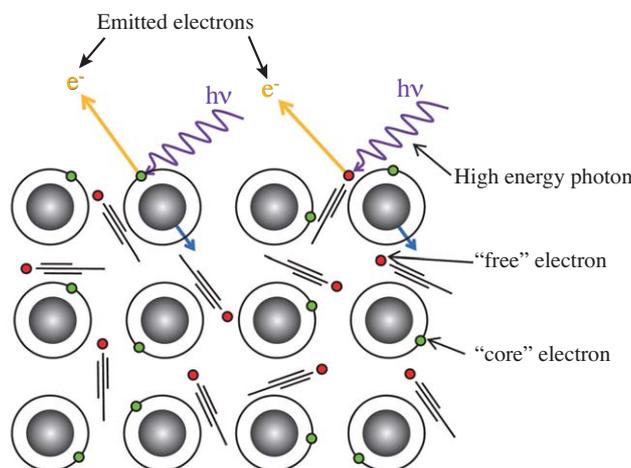


Fig. 1. When a photon ($h\nu$) of sufficient energy hits a sample, it can excite the emission of electrons. For a metal, these electrons can come either from specific atoms (green circles) or from the sea of 'free' valence electrons (red) that extends over the whole sample.

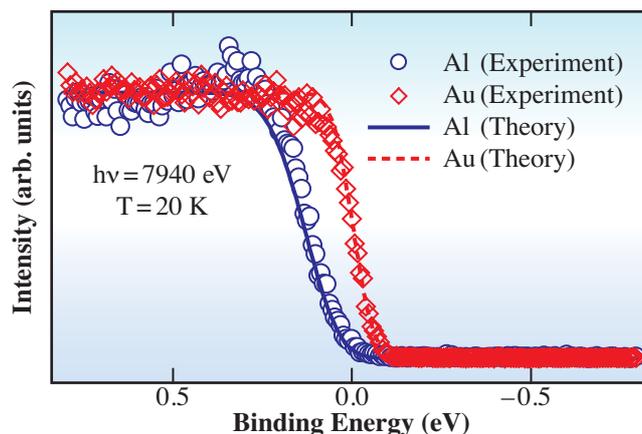


Fig. 2. Photoelectron spectra near Fermi edge of Al (circles) and Au (diamonds) with excitation energy 7940 eV. The zero point of the binding energy is chosen at the observed value of the chemical potential of Au. The solid line and the dotted line indicate the theoretical curves calculated by the Debye model.

A model calculation of Bloch electrons based on the isotropic Debye model (solid and dotted lines in Figs. 2 and 3) reproduces well the experimental spectra. Details of the calculation are described in Ref. [3]. The existence of the photoelectron recoil effect means that the electron is coupled with the crystal lattice, and the wave function of the electron follows adiabatically the atomic motion. Therefore, it is quite surprising that the Bloch electron in the valence band of Al, which is a typical material where the nearly free electron picture works well, shows remarkable recoil effects. The finding of recoil effects in Bloch electrons indicates a new spectroscopic aspect of X-ray

photoelectron spectroscopy, and due care must be taken to interpret changes at and near the Fermi level when using hard X-rays. The magnitude of the recoil effect depends essentially on the mass of the component atoms. Since the photoelectron spectra reflect also the nature of the electronic state in the initial state, the recoil effect in the valence levels of composite materials poses an interesting problem. If the material is composed of atoms with big mass differences, and if the valence levels are made of hybridized orbitals originating in specific atomic species, the modification of the photoelectron spectra will depend on the local density of states of the level.

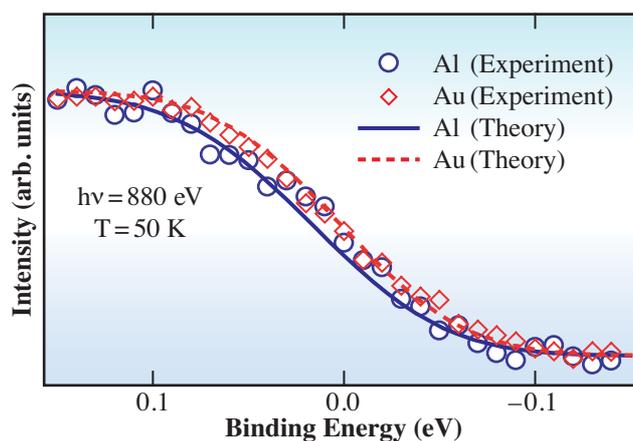


Fig. 3. Photoelectron spectra near Fermi edge of Al (circles) and Au (diamonds) with excitation energy 880 eV. The zero point of the binding energy is chosen at the observed value of the chemical potential of Au. The solid line and the dotted line indicate the theoretical curves calculated by the Debye model.

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