XAFS Beamline

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1. Introduction

On the construction of a new Synchrotron Radiation facility, SPring-8, we intend to arrange a characteristic XAFS experiments including standard XAFS measurements, and aim at a goal of a center radiating the overall XAFS information. For this purpose, we have both short- and long-term programs in the SPring-8 facility. The short-term ones include the development and operation of Broad Energy Band XAFS (BEB-XAFS) experimental station with wide-ranging applications in the fields of material science, chemical science, condensed matter physics, biological science and so on. The long-term program includes the development and implementation of frontier XAFS measurement programs that will open up new methods of the structure determination.

This beam line enable us to start the initial program, and it will be acceptable frontier works opening up to many researchers.

2. Outline of the Experimental Station

The optics of the beam line will basically be arranged for standard XAFS measurements, covering the energy range from 3.5 keV to 90 keV. In the experimental station, an experimental hatch is designed to allow the various XAFS observations. The cryostat (6 ~ 300 K) and furnaces (300 ~ ca.2000 K) are set up. To carry out the one (modulation XAFS) of the main scientific purposes of the beam line, two large optical benches are prepared in the large experimental hatch. The control system and the data-correction systems are independently subjected to two computers.

3. Research Subjects

We have four main research programs for this beam line. Three of these are introduced as follows.

a) XAFS observation in relaxed excited states by using photo-modulation technique

The x-ray absorption fine structure nowadays is regarded as an excellent source information to investigate short-range order around a specific type of an atom in *its steady (ground) state.* In physics or photo chemistry, information about the precise short-range order in either photo-excited states or photo-reaction intermediate states makes it more exciting to investigate various physical phenomena, chemical reactions and so on. A modulation technique [1] has been successfully put forward to observe outer-perturbation responses such as those due to an electric or a magnetic field [2], stress, and irradiation of light. Thus, we intend to perform the XAFS observations of the short-range order in *its relaxed excited state (RES)* by using the photo-modulation technique. A model simulation to inspect its possibility is given as follows [3].

In order to visualize the possibility of the XAFS analysis of local structures in RES, a simple model molecule with a non-degenerate ground state (GS) electronic energy levels denoted by subscript **g** and a photo-excited state denoted by **e** is taken. On exciting the electron once, the molecular coordination will relax to an appropriate stable structure different from that of the ground state, for example, through the Jahn-Teller effect (first-order electron-nuclei interaction). We tentatively assume the structure in RES to be of the same symmetry as GS but with longer bond distances. Corresponding to each electronic state, the bond distances are given by r and $r' = r + dr$, respectively. The XAFS oscillation in GS is $c_g(k) = N_g F \sin 2kr$, where N_g is the occupancy in GS, and F the amplitude of the XAFS oscillation. The XAFS oscillation on photo-exciting is modulated as $c(k) = N_g F \sin 2kr + N_e F \sin 2kr'$, where Ne is the occupancy in RES, and $N_g + N_e = 1$. Therefore, the XAFS oscillation detected by the photo-modulation technique is $dc(k) = 2N_e F \sin k(r-r') \cos k(r+r')$. Figure 1 shows the simulated XAFS oscillations in GS (c), in RES with N_e= 0.01 and dr = 0.02 Å (b), and $dc(k)$ of Photo-Modulation-XAFS (a) as an extreme case.

Thus, we can obtain the XAFS spectra in RES accurately. Its method has a possibility to open a new physical and chemical research field.

b) XAFS in the high energy region

The radiation spectrum from a bending magnet at the SPring-8 Storage Ring with a critical energy of 29 keV shows the high photon density even at 100 keV. This is advantageous for K absorption edges of almost all elements because in XAFS observation at L absorption edge, L_{II} absorption edges perturbs L_{III} XAFS oscillations, resulting in restriction to several hundred eV in energy. On the other hand, it was shown that the lifetime broadening of the core hole becomes more serious for K absorption edges of heavy elements [2]. The short lifetime of the core hole will smear out the XAFS signal. We have examined to detect XAFS signal just above the Pt-K edge (~78 keV) of a Pt foil (0.1 mm thick). Here, a preliminary result is presented together with some problems of XAFS in the high energy region as shown in Figs. 2(a), (b), (c) and (d).

In Fig. 2 (a), it seems that the edge jump of the Pt-K edge is not so sharp in comparison with that of the Pt-L_{III} edge, and that the XAFS signal above the Pt-K edge smears out. The XAFS function $c(k)$ was extracted from the absorption spectrum by following the standard procedure. However, as shown in Fig. 2 (b), a well-defined XAFS signal is found up to 18 $Å^{-1}$.

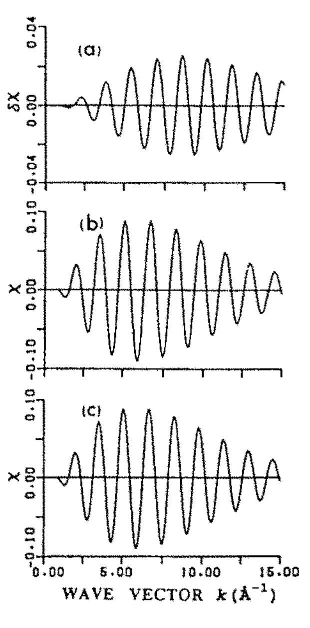

Fig. 1. XAFS oscillations: photo-modulated spectrum (a), in the relaxed excited state by irradiation (b), and in the ground state (c).

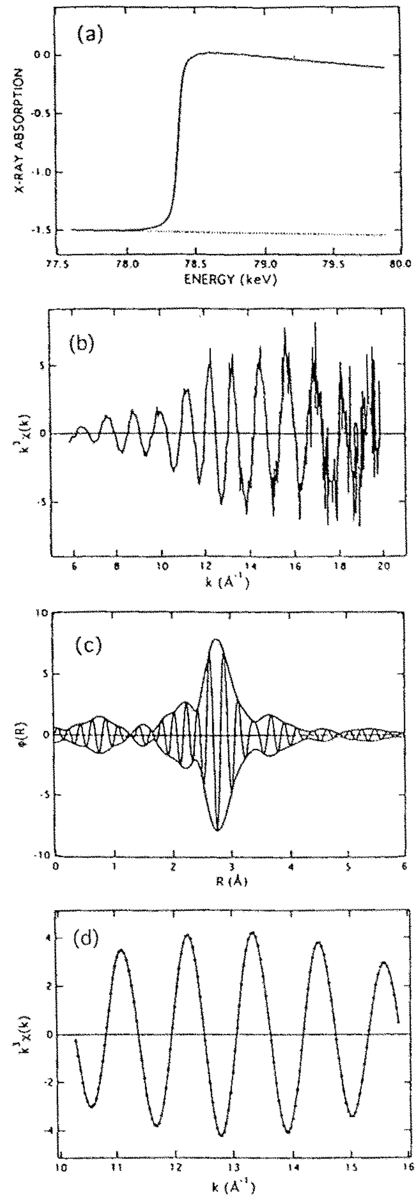

Fig. 2. (a) XAFS spectrum near Pt-K edge of Pt foil at RT, (b) XAFS function $k^3 \chi(k)$, (c) Radial structure function f(R) (magnitude and imaginary part of Fourier transform of the XAFS oscillation), (d) Comparison of an experimental XAFS function $k^3 \chi(k)$ of the first-nearest-neighbor Pt atom (closed circle) and a theoretical function (solid line).

(c) XAFS of dilute system and other applications in high energy region

The XAFS study of dilute systems is a central matter of concern in the various of material science. The experiment is ordinarily performed in a fluorescent mode. However, the conventional SR facilities restrict the kind of elements to be characterized because of poor photon fluxes at high energy region. Recently, the significance of the role of heavy metal elements like 4d-series transition metal and lanthanide metal elements in material sciences has increased. The energy of K-edge absorption by such heavy metal elements lies in a high energy region of more than 18 keV where the fluorescent yield is about 100 %. This is advantageous for the characterization of dilute systems. Using a bending magnet of SPring-8, a photon flux over the actual size of 10 mm $\times$ 1 mm is evaluated to be 2×10^{11} photons/sec at 20 keV in the focused beam. Therefore, at the present beam line, spectra of high quality of S/N can be obtained with short time enough to measurements using a detector covering 10 % of solid angle.

4. Schedule

The beam-line optics and the experimental hatch are scheduled to be arranged at autumn in 1996. The test of the control systems will be started at beginning of winter. The alignment of the optics is expected to be early in 1997.

References

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[2] Stearns, D.G., Phil. Mag. B **49**, 541 (1984).

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