FUNDAMENTAL RESEARCH OF SUBPICOSECOND TIME RESOLVED X-RAY DIFFRACTOMETRY USING ELECTRON LINAC

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Abstract

We have proposed a new pump-and-probe technique, subpicosecond time resolved X-ray diffractometry, which enables the direct observation of the lattice movement in the transient processes of the subpicosecond order. This technique uses a subpicosecond electron pulse from a linac, which produces various radiations as a pump pulse and the characteristic X-ray in a copper target as a probe pulse. Using the EGS4 code, we have found that the subpicosecond X-ray pulse can be generated in a sufficient intensity and purity for this technique. Preliminary experiments have been also performed using the NERL electron linac and several diffraction images were obtained for typical monocrystals such as silicon.

1 INTRODUCTION

Recent remarkable progresses on ultrashort pulse generation technology brought entirely new possibilities to change the current static way of material property research into dynamic one. The electron single pulses of 700 fs[1] and 440 fs[2] are available from the S-band twin linear accelerator (linac) system at the Nuclear Engineering Research Laboratory (NERL). The X-band linac has been planned to be introduced in future, which is evaluated to produce 100 fs pulse[3]. Here, as the new application of these linacs, a new pump-and-probe technique, subpicosecond time resolved X-ray diffractometry is proposed, which enables the direct observation of the lattice movement, namely the temporal change of the three-dimensional atomic arrangement in the transient phenomena of the subpicosecond order. This technique uses a subpicosecond electron pulse from a linac, which produces various radiations like a transition radiation[4] as a pump pulse and the characteristic X-ray in a copper target as a probe pulse.

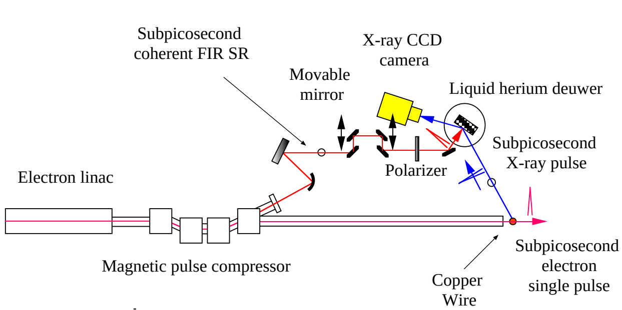

Figure 1: Schematic of linac-based subpicosecond time resolved X-ray diffractometry system.

Fig. 1 shows an example of the system configuration in case of using the far-infrared coherent synchrotron radiation as a pump pulse. The time delay between these two pulses is controlled only through adjustment of the path length of the pump pulse, so this method has an intrinsic advantage that there is no time jitter which may cause serious problems in adding up accumulated results. Further, by choosing the pump pulse properly, various ultrafast phenomena can be treated such as lattice vibration (phonon), thermal expansion, phase transition and so on, which may result in experimental verification for the existing solid state physics and molecular dynamics calculations.

In this study, the converted X-ray probe pulses have been totally characterized numerically using the EGS4 code[5] and their applicability to this technique is investigated. Preliminary experiments have been performed to generate the characteristic X-ray pulses and to obtain their diffraction images for several typical monocrystals at the linac.

2 SIMULATIONS

In this technique, the electron pulses impinge upon the copper wire and converted into KX-ray pulses. The KX-ray generation is mainly occured through the two channels: 1. bremsstrahlung followed by K-shell photoionization (PI) and 2. K-shell electron-impact ionization (EII). The latter process is domimant in the electron energy range of 18-35 MeV available from our linacs but not treated in the default EGS4 code. Here, the modified version of the EGS4 code is used which Y.Namito et al.[6] have developed to include the K-shell EII. In the EGS4 calculation, the incident electron beam with the monoenergy of 35 MeV is pencil-like (zero width) and incident perpendicularly on the central axis of the copper wire.

Strong angular dependencies were found in both the X-ray intensity and spectrum. Fig.2 shows the calculated spectra of forward-emitted and side-emitted X-ray from the $100\mu\mathrm{m}\phi$ copper wire. Each spectrum is normalized to have unity at the KX-ray's peak position($\sim 8.1\mathrm{keV}$). The fraction of the background, mainly due to the bremsstrahlung, of the side-emitted X-ray was smaller than that of the forward-emitted X-ray by a factor of about 100. The characteristic X-ray is isotropically generated, while the bremsstrahlung is forward generated. Most of the X-rays were found to be emitted sharply straight forward. However, we should make use of the side-emitted X-ray in order to realize high S/N measurement.

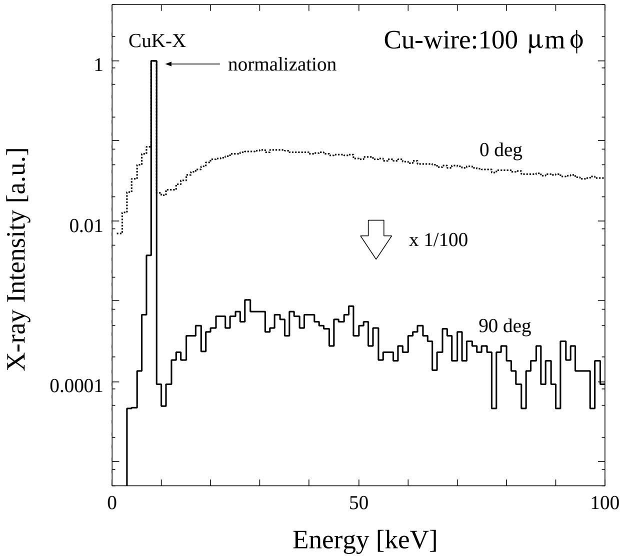

Figure 2: Comparison of (a) forward-emitted and (b)side-emitted X-ray spectra.

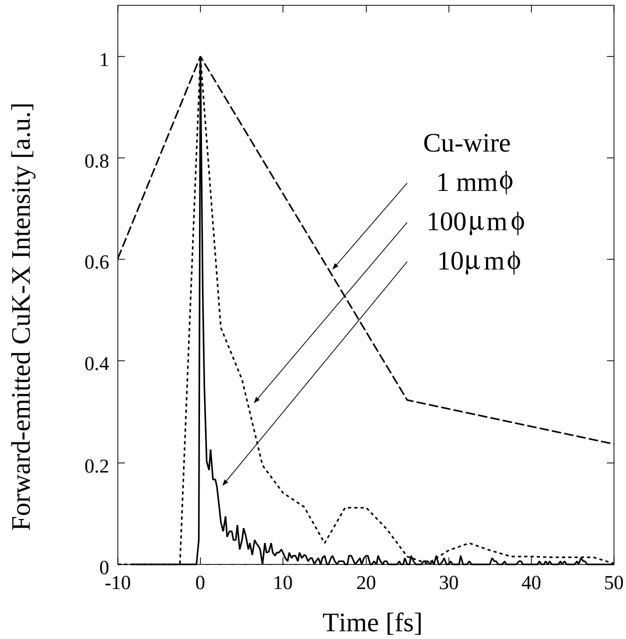

Figure 3: CuK-X pulse shapes for various sizes of copper wire.

As shown in Fig.3, the pulse shapes were also calculated by adding up the transit time of the every trasport step in the EGS4 user code. The hole in the K-shell has an ultrashort lifetime of about 1 fs[7], which was disregarded so that the KX-rays were emitted immediately after the K-shell ionization. The incident electron pulse duration was regarded as zero. The obtained pulse durations of the forward-emitted KX-ray were much shorter than that of the actual electron pulses, that is, subpicosecond. This slight elongation of the KX-ray pulse duration is mainly due to the multiple scattering of the electron before the EII. It should be noted that the side-emitted KX-ray pulse is subject to much more pulse expansion due to the geometrical factor since there is a certain time lag of the KX-ray genaration at upstream and downstream. The light travels 300 μm for 1 ps. Of course, this time lag will be cancelled for the forward-emitted KX-rays if the electron energy is relativistic.

Table 1 summarizes the calculated results for various sizes of the copper wire. The imaging plate (IP) is an excellent imaging device and widely used for not only an X-ray diffractometry but also many purposes. Typically, it has a significant sensitivity to the X-ray energy of less than 150 keV. For the side-emitted X-ray, the S/N ratio is defined as the ratio of the KX-ray to the other IP-sensitive X-ray. The number of the KX-ray reaching the sample crystal surface per pulse is also tabulated. For this derivation, it was assumed that the electron pulse of 1 nC was generated at 10 pps and the sample of several cm^2 located 1 m from the copper wire. Further, the irradiation time necessary to form the Bragg spot composed of the 10000 diffracted KX-rays was roughly estimated assuming that the diffraction efficiency was 1/100. It can been seen from Table 1 that the subpicosecond X-ray pulses are generated in enough intensity and quality for this technique and the copper wire of around 100μmϕ was found to be optimum.

3 EXPERIMENTS

Based on the above information obtained from the EGS4 simulations, preliminary experiments have been done at the NERL linac. The experimental configuraion is shown in Fig.4. The sample monocrystal was mounted on the goniometer, and it was placed inside the lead shielding together with the imaging plate for taking photographs of the X-ray diffraction image. For the photoneutron shielding, the polyethylenes were provided around the lead shielding and the beam dump consisting of the carbon block.

Table 1: Summary of the calculated results.

Diameter of Copper Wire	10 μmϕ	100 μmϕ	1 mmϕ	10 mmϕ
Pulse Duration	0.2-0.3 fs	2-3 fs	20-30 fs	-
S/N Ratio	$\sim$ 7	$\sim$ 6	$\sim$ 0.4	$\sim$ 0.003
KX-rays on Sample	$\sim$ 10	$\sim$ 200	$\sim$ 500	$\sim$ 500
Irradiation Time	$\sim$ 3 hrs	$\sim$ 10 mins	$\sim$ 5 mins	$\sim$ 5 mins

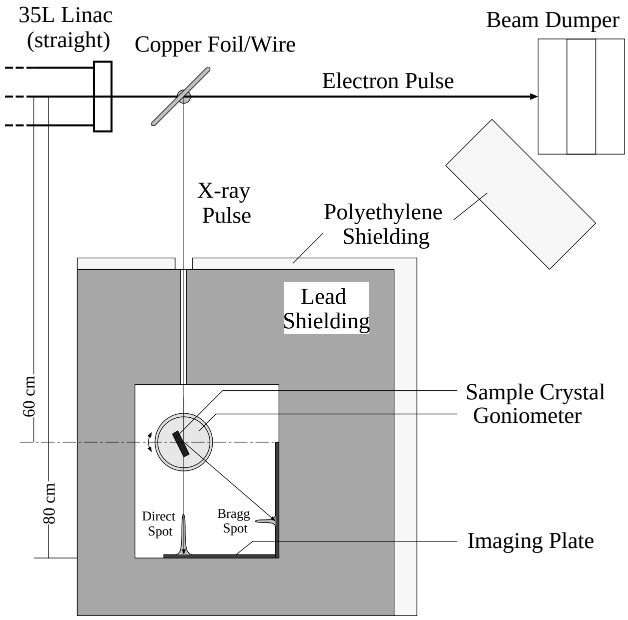

Figure 4: Experimental setup of the X-ray diffractometry at NERL linac.

The X-ray pulses were generated from the copper foil/wire irradiated by the linac electron pulse and lead to the surface of the sample crystal through the slit of 5 mm width in the lead shielding. The linac produced the multi-bunch pulse of 1 ns duration with the electron energy of 28 MeV at the frequency of 50 pps. The electric charge per pulse was a little less than 1 nC and the beam size was about 5 $\mathrm{mm}\phi$.

Fig.5 shows the diffraction image by the Si(111) plane for the silicon wafer. The irradiation time was about 20 minutes and the copper foil of 15 $\mu\mathrm{m}^{\mathrm{t}}$ was employed for the X-ray converter. Three types of spot were seen on the image, the direct X-ray spot from the copper foil, the shadow behind the silicon wafer and the Bragg spot. For Si(111) plane, the Bragg angle of the KX-ray is 14.24 degrees, while the Bragg spot appeared only in the goniometer angle of 14-14.75 degrees. The diffraction images for the GaAs(400) plane and the NaCl(200) plane were obtained as well. Using the copper wire of 100 $\mu\mathrm{m}\phi$, we also succeeded in taking the finer Bragg spot composed of two clearly separated lines due to the $\mathrm{CuK}\alpha_{1,2}$.

4 SUMMARY

The subpicosecond time resolved X-ray diffractometry was newly proposed and its feasibility with the NERL linacs were investigated. By the EGS4 code simulation, the size of the copper wire, as the X-ray converter, was optimized so that the subpicosecond X-ray pulses were generated with the sufficient intensity and quality. Preliminary experiments have been carried out using the NERL linac and the diffraction images of the CuKX-ray were obtaied for several typical monocrystals successfully.

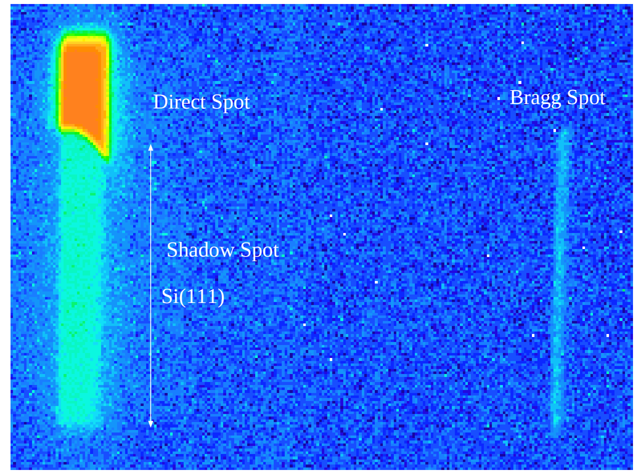

Figure 5: Si(111) diffraction image for 15 $\mu\mathrm{m}^{\mathrm{t}}$ copper foil.

5 AKNOWLEDGEMENT

The authors are thankful for Dr.Namito of High Energy Accelerator Research Organization for his help in using the EGS4 code. We would also thank Dr.Niimura of Japan Atomic Energy Research Institute and Dr.Yoneoka of the University of Tokyo for their help and discussion on the X-ray diffraction experiment.

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