X-ray Monochromator with $2 \times 10^{-8}$ Energy Resolution


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An X-ray beam of $3 \times 10^7$ photons s$^{-1}$ with $2 \times 10^{-8}$ relative energy resolution has been obtained at a third-generation synchrotron undulator X-ray source using the (13 13 13) Bragg reflection from a silicon perfect crystal. The production of these 25.70 keV X-rays with 450 ± 50 µeV bandpass opens up new possibilities in X-ray optics and spectroscopies.

Keywords: X-ray optics; monochromators; dynamical theory; energy resolution; inelastic scattering; silicon (13 13 13).

1. Introduction

X-ray beam monochromatization to sub-meV energy resolution with a high degree of intensity and collimation is a challenging task in modern synchrotron-radiation-based research for its application in important branches of science. An interesting use would be the study of the dynamics of density fluctuations in condensed matter by inelastic X-ray scattering. The complementarity to inelastic neutron scattering comes from the different energy-momentum region explored by the two probes. In particular, the small momentum transfer regions of materials with a large speed of sound are so far inaccessible to neutrons, whilst they could be readily explored using 20 keV X-rays with meV energy resolution. This possibility is of extreme importance in liquids and disordered materials, which have characteristic collective excitations only in this energy-momentum region as a consequence of the lack of translational invariance.

High-energy resolution X-ray monochromators are based on Bragg diffraction from perfect crystals. At a given diffraction order $h$, the intrinsic energy resolution, $(\Delta E/E)_h$, is proportional to the lattice spacing and the atomic form factor of the considered reflection. The best resolution is achieved with high reflection orders, where the form factor decreases rapidly. If photoelectric absorption is small, one can obtain values of $(\Delta E/E)_h$ of the order of $10^{-8}$ at photon energies $E \approx 26$ keV. The corresponding energy resolution, $\Delta E$, is then in the meV and sub-meV range, a value characteristic for the study of dispersion relations associated with density fluctuations in solid and liquid materials.

The practical realization of such a monochromator demands perfect crystals with a relative lattice spacing variation, $\Delta d/d$, in the diffracting volume smaller than the desired energy resolution. At present, this is achieved only with silicon crystals (Ando, Bailey & Hart, 1978). Moreover, in order to maintain the energy resolution without reduction of the reflected intensity, the divergence of the incoming photon beam must be comparable to or smaller than the intrinsic angular acceptance of the considered reflection. This angular quantity, defined as the Darwin width, $\omega_D$, in the dynamical theory of X-ray diffraction (Zachariasen, 1944), is given by $\omega_D = (\Delta E/E)_h \tan \theta_B$, where $\theta_B$ is the Bragg angle. At Bragg angles very close to 90°, where $\tan \theta_B$ tends to infinity, the Darwin width of high-order reflections can become comparable to the divergence typical of X-ray beams produced by undulators on third-generation storage rings. This match in phase space allows full utilization of the intensity provided by the synchrotron source. Combined with a high-order Bragg reflection, the production of a photon beam with an extremely high degree of monochromatization and optimal intensity becomes possible.

The use of backscattering geometry to obtain very high energy resolution was first demonstrated by Graeff & Materlik (1982) and further developed by Dorner, Burkel & Peisl (1986) for applications in inelastic X-ray scattering. These authors achieved an energy resolution of 9 meV at the Si(999) reflection in backscattering, i.e. a relative energy resolution of $5 \times 10^{-7}$ at 17.794 keV X-ray energy, with a photon flux of $\sim 10^6$ photons s$^{-1}$ on the sample (Burkel, 1991). The highest energy resolution obtained so far for an X-ray crystal monochromator was reached with a four-crystal device in a dispersive arrangement, first developed by Faigel, Siddons, Hastings, Haustein & Grover (1987) for applications in resonant nuclear scattering with synchrotron radiation. Here, an energy resolution of 5 meV was obtained at the 14.4 keV iron Mössbauer resonance using
the Si(10 6 4) reflection. This corresponds to \( \Delta E/E = 3.5 \times 10^{-7} \), and was reached with an angular acceptance of 2 \( \mu \)rad.

2. Experimental

We report here the results obtained with an instrument based on high-order reflections in a silicon perfect crystal, and a backscattering geometry with \( \theta_B \) very close to 90°. The experiment was performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble. The photon source is an undulator providing a collimated beam with 25 \( \mu \)rad vertical and 40 \( \mu \)rad horizontal divergence. After pre-monochromatization to \( \Delta E/E = 2 \times 10^{-4} \) with an Si(111) double crystal, the photon beam impinges on the backscattering silicon crystal, which is set at a Bragg angle of 89.98° (\( \tan \theta_B = 2865 \)), and diffracts in the vertical plane. A specific Si(h h h) reflection is chosen by tuning the pre-monochromator to the correct energy. In this configuration, the intrinsic energy resolution \( \Delta E/E \) of the Si(h h h) reflection is obtained as long as \( WD = \Delta E/E \tan \theta_B \) is larger than 25 l.trad (the vertical beam divergence). This sets the minimum \( \Delta E/E \) value to less than \( 10^{-8} \) and allows the Si(13 13 13) reflection, which has an intrinsic energy resolution of \( 2 \times 10^{-8} \), to be investigated.

The energy analysis of the backscattered photon beam is achieved utilizing an additional silicon crystal, operating at the same reflection and backscattering angle as the monochromator. The diffracted intensity from this analyser crystal is monitored in a fixed geometry whilst varying the relative temperature, \( \Delta T \), between the monochromator and the analyser crystal. In fact, a change in \( \Delta T \) induces a relative variation in lattice constant \( d/d = \alpha \Delta T \), where \( \alpha \) is the coefficient of thermal expansion \( [\alpha = 2.56 \times 10^{-6} \text{ K}^{-1}] \) in silicon at 294 K (Ibach, 1969). The \( d/d \) scan is analogous to a \( \Delta E/E \) scan; in fact, according to Bragg’s law, \( E = hc/2d \sin \theta_B \), and therefore, at fixed \( \theta_B \), \( \Delta E/E = -\Delta d/d = -\alpha \Delta T \). Under the described experimental conditions, where the geometrical contributions to the energy resolution are constant and small, the temperature scan provides a direct measurement of the Si(13 13 13) intrinsic energy resolution.

3. Results and discussion

Fig. 1 summarizes the experimental set-up, and Fig. 2 shows the temperature scan. The y axis reports both the temperature difference and the corresponding energy difference. The measured width of the curve is 650 \( \pm 50 \) \( \mu \)eV full width at half maximum (FWHM). This corresponds to the convolution of the intrinsic energy resolution of the monochromator and analyser crystal. Assuming the theoretical lineshape for the two crystals in the deconvolution (Zachariasen, 1944), the resulting value for each crystal is 450 \( \pm 50 \) \( \mu \)eV, giving a relative energy resolution \( \Delta E/E = 2 \times 10^{-8} \), in good agreement with the theoretical value. This is the highest energy resolution obtained so far with a tunable X-ray monochromator based on silicon perfect crystals. The corresponding photon flux of \( 3 \times 10^7 \) photons s\(^{-1}\), measured with a calibrated silicon photodiode, is close to the total available intensity in the considered bandwidth of the undulator X-ray source.

An important implication of this result, i.e. the production of a high intensity, very high energy resolution X-ray beam, is to open up applications of X-ray inelastic scattering spectroscopies to a domain so far limited only to neutron techniques (Sette et al., 1995). As stated in the introduction, the complementarity between X-rays and neutrons is particularly relevant in those energy and momentum transfer regions which are very difficult to access with neutrons (energy transfers of a few meV and momentum transfers smaller than 10 nm\(^{-1}\)). In the momentum transfer region corresponding to correlation lengths of 0.1—50 nm, only with X-rays could it be possible, at present, to measure the high-frequency dynamics of disordered systems as liquids, amorphous materials and biological samples (Sette et al., 1995).

Finally, the production of such a high-resolution X-ray beam gives an assessment of the perfection of available silicon crystals. The energy resolution obtained implies that...
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Figure 2
Temperature scan between the monochromator and analyser silicon crystal, at the $\langle 13\,13\,13 \rangle$ reflection with a Bragg angle of $89.98^\circ$. This corresponds to an X-ray energy of $\sim 25.7026$ keV. The dots are the experimental points taken with a temperature step of 1 mK. The solid line is a guide to the eye. The spectrum is normalized to the peak intensity. The energy scale, also shown, is obtained from the temperature difference between the two crystals, $\Delta T$, as $\Delta E = -E \alpha \Delta T$, where $\alpha$ is the coefficient of thermal expansion of silicon. At $T = 294$ K, $\alpha = 2.56 \times 10^{-6}$ K$^{-1}$ (Ibach, 1969). The FWHM is $650 \pm 50$ \text{µeV}$, and corresponds to the convolution of the analyser and monochromator response function. Assuming that each crystal contributes a comparable amount, the quasi-Gaussian convolution would give an energy resolution of $450$ \text{µeV}$, which compares well with the calculated intrinsic value.

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References