

A method to determine the absolute harmonic content of an X-ray beam using attenuation measurements

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We present a new method for determining the absolute harmonic content of an X-ray beam. The technique is applied to determine the harmonic content of a synchrotron beam to high-accuracy by measuring the X-ray attenuation of a large number of aluminium foils with thicknesses varying over several orders of magnitude. Earlier methods always determined relative quantities such as the *effective harmonic content*, which are dependant on experimental geometry and not transferable between detectors. We use a more fundamental and useful parameter: the *harmonic-photon percentage*. Copyright © 2009 John Wiley & Sons, Ltd.

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Introduction

A synchrotron beam contains a spectrum of energies. In most experiments, the beam is passed through a monochromator in order to select the desired energy from the spectrum. However, the monochromatation process is never perfect and the resultant beam will have a finite bandwidth and may contain X-ray energies of higher-order harmonics.^[1,2] The harmonic photons that are present in the X-ray beam after it passes through the monochromator have energies that are integer multiples of the fundamental.^[3] For example, a double-crystal monochromator utilising a monolithic silicon 111 crystal allows through odd multiples of the fundamental energy E_f (e.g. $3E_f$, $5E_f$...).

Most X-ray experiments require a monochromatic beam and the presence of higher-order contributions to the spectral distribution cause a systematic error. This systematic error can be quantified and corrected for if an accurate determination of the harmonic content of the X-ray beam is made.

The method of determining the harmonic content of an X-ray beam using attenuation measurements has been known for some time^[4–6] and has been applied to synchrotron radiation.^[1,7] Previous work was only able to determine an effective harmonic parameter that depended on the properties of the beam as well as the particular detectors and experimental geometry being used (the effective harmonic parameter is defined in Ref. [1] and also in Eqn 2 of this paper). While these measurements were useful for the particular detectors upon which the measurements were made, the results did not reflect the fundamental properties of the beam and were not transferable to other detectors and/or experimental configurations.

We present a method for measuring the harmonic content of an X-ray beam which is parameterised in terms of the percentage of photons that are harmonic. We apply this method to determine the harmonic-photon percentage of a synchrotron beam at 31 energies between 5 and 8 keV.

Background

In the ideal situation of a perfectly monochromatic beam, the X-ray mass-attenuation coefficient of a sample is measured in transmission geometry using the Beer-Lambert equation:

$$\left[\frac{\mu}{\rho}\right]_{[\rho t]} = -\log\left(\frac{I}{I_0}\right) \quad (1)$$

where I and I_0 are the beam intensity exiting and entering the sample, respectively. $\left[\frac{\mu}{\rho}\right]$ is the mass-attenuation coefficient. $[\rho t]$ is the integrated column density which is the integral of the density of the foil along the path of the X-ray beam.

When the X-ray beam contains more than one energy, Eqn 1 does not have a unique solution for the mass-attenuation coefficient $\left[\frac{\mu}{\rho}\right]$.^[8] If harmonic energies are present then the measured value of the mass-attenuation coefficient depends on the thickness of sample being investigated. While this effect causes a systematic error in X-ray attenuation measurements, it provides useful information about the harmonic content of the beam.

If we assume that the beam contains X-rays at the fundamental energy E_f and from a higher-order harmonic energy E_h we can derive the equation describing the effect of harmonics on X-ray attenuation:

$$\frac{D}{D_0} = (1 - \alpha)e^{-\left[\frac{\mu}{\rho}\right]_f[\rho t]} + \alpha e^{-\left[\frac{\mu}{\rho}\right]_h[\rho t]} \quad (2)$$

where α is the effective harmonic parameter and $\left[\frac{\mu}{\rho}\right]_f$ and $\left[\frac{\mu}{\rho}\right]_h$ are the attenuation coefficients at the fundamental and harmonic energies. D and D_0 are readings of the downstream and upstream detector, respectively.

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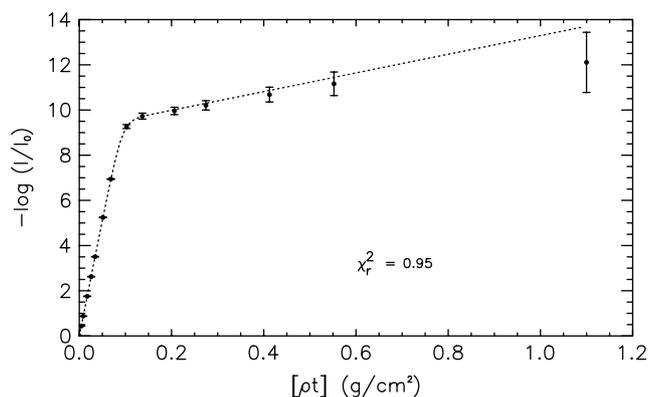


Figure 1. The measured attenuation at 6.104 keV plotted against the integrated column density $[\rho t]$ of the aluminium foils. The graph can be partitioned into two straight regions, indicating the presence of two energies in the X-ray beam. The presence of these harmonic energies was modelled and the result is plotted as a dotted line.

The presence of multiple energies in the X-ray beam can be diagnosed by plotting attenuation against $[\rho t]$. The dotted line in Fig. 1 was modelled using Eqn 2. The beam was assumed to contain two energies and so has two distinct regions of differing gradient. If the beam contained three X-ray energies then the graph would have three distinct gradients and so on. This type of plot can be used as a general tool to identify the presence of multiple energies in an X-ray beam.

Experimental Details

The experiment was performed at beamline 20B of the Photon Factory synchrotron in Japan. We present results from that experiment for X-ray energies between 5 and 8 keV. Measurements were made of the attenuation of 15 aluminium foils with thicknesses ranging between 10 and 4000 μm . The foils were mounted on the perimeter of a circular daisy wheel; the daisy wheel rotation was controlled using a stepper motor so that measuring the attenuation of all 15 foils was relatively quick.

Figure 1 plots the measured attenuation against integrated column density $[\rho t]$ of the aluminium foils at 6.104 keV. The data plotted in Fig. 1 was fitted to Eqn 2 to determine the effective harmonic parameter. The values of α and $[\mu/\rho]_f$ were determined simultaneously during the fit. The mass-attenuation coefficient of aluminium for the third harmonic X-rays was derived from theory (the FFAST tabulation.^[9]) More precisely, the ratio of the attenuation coefficients of the third and first harmonics was taken from the theory. The mass-attenuation coefficient of the third harmonic was then calculated by multiplying that ratio by $[\mu/\rho]_f$.

The results of the best-fit model are plotted in Fig. 1 as a dotted line and the agreement with experiment is excellent ($\chi_r^2 = 0.95$). The effective harmonic parameter at this energy was determined to be $(1.33 \pm 0.09) \times 10^{-4}$. This process was repeated and the effective harmonic parameter was determined for 31 energies between 5 and 8 keV.

We tested for the presence of fifth harmonic radiation. The determined levels did not differ significantly from zero during the experiment. This is consistent with the expected spectral distribution of the beamline.

Aluminium-foil samples contain impurities, with iron and silicon typically occurring at the highest levels. The effect of impurities was

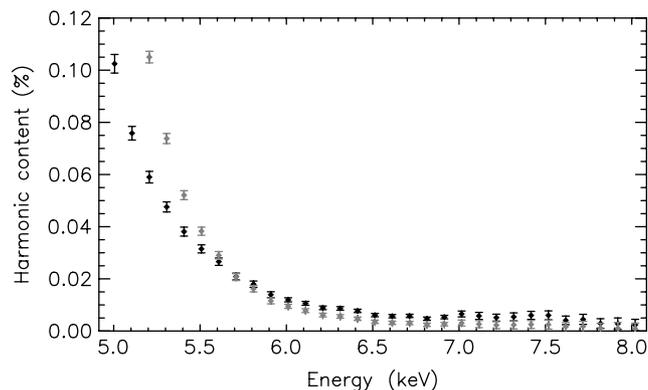


Figure 2. The harmonic-photon percentage is plotted in black along with error bars at 31 energies between 5 and 8 keV. The effective harmonic content (multiplied by 100 to convert from a fraction to a percentage) is plotted in grey. At 5 keV the effective harmonic content is much larger than the harmonic-photon percentage (0.25 against 0.10 respectively).

assessed by analysing the energy-dependence of our results. Their effect was not significant compared with the other experimental uncertainties.

Determining the Harmonic-Photon Percentage

The harmonic-photon percentage can be determined from the effective harmonic parameter by correcting for a number of factors specific to the detectors and experimental geometry. The fundamental and harmonic X-rays have different detector efficiencies, detector-current yields and are absorbed and scattered to differing degrees by the air. Once these effects are taken into account, one is left with the fundamental measure of the harmonic component of the beam, the harmonic-photon percentage.

The function to convert between these two parameters can be derived for the case of a transmission X-ray absorption measurement using matched ionisation chambers. The formula for the ratio of harmonic to fundamental photons $\frac{\Phi_3}{\Phi_1}$ is:

$$\frac{\Phi_3}{\Phi_1} = \left(\frac{\alpha}{1 + \alpha} \right) \frac{E_1 P_{IC1+air}^{E_1}}{E_3 P_{IC1+air}^{E_3}} \left(\frac{1 - P_{IC1}^{E_1}}{1 - P_{IC1}^{E_3}} \right), \quad (3)$$

where α is the effective harmonic parameter, E_1 is the fundamental energy and E_3 is the harmonic energy. $P_{IC1+air}^E$ is the probability that of an X-ray of energy E will be transmitted through the upstream ion chamber (IC1) and air path. The probability of transmission through a material was calculated using the formula $P = 1 - \exp\left(-\left[\frac{\mu}{\rho}\right][\rho t]\right)$. If an X-ray passes through two materials then $P_{1+2} = P_1 \times P_2$. The mass-attenuation coefficient of the nitrogen gas and window material of the ion-chambers and the various gasses in air were taken from the FFAST tabulation.^[9] The value of the integrated column densities was calculated on the basis of the design drawings and geometry of the experiment.

The value of the harmonic-photon percentage was determined using formula 3 and is plotted in Fig. 2. The maximum harmonic content occurred at a nominal beam energy of 5 keV, where approximately 1 out of every 970 X-rays was harmonic with energy 15 keV.

Discussion

The harmonic-photon percentage is a more transferable parameterisation of the harmonic content of an X-ray beam. There are many experiments in which this would be useful, such as for X-ray absorption fine structure (XAFS) measurement made simultaneously in transmission and fluorescence geometry.^[10–12] Harmonic X-rays cause a systematic error in XAFS measurements,^[1] but this effect can be corrected for determining the harmonic-photon percentage. The appropriate correction could be applied both to the fluorescence and the absorption data. This demonstrates a clear advantage over the effective harmonic parameter which is not transferable between detectors.

The method outlined in this paper could be extended to measure more general spectral distributions. The transmission spectrum of an X-ray beam $T([\rho t])$ is governed by Eqn (4):

$$T([\rho t]) = \int_0^\infty \phi(E) e^{[\frac{\mu}{\rho}] [\rho t]} dE = \mathcal{L} \left[\phi(E([\mu/\rho])) \frac{dE}{d[\frac{\mu}{\rho}]} \right] \quad (4)$$

where \mathcal{L} refers to the Laplace transform. One can then invert this equation to obtain $\phi(E)$ the spectral distribution.^[5,13] If the results are obtained using detectors with an energy dependent response (e.g. ionisation chambers) then $\phi(E)$ is the *effective* spectral distribution. The method outlined in this paper can be used to transform $\phi(E)$ into the true spectral distribution and this would be a useful extension of our technique.

Conclusion

We demonstrated a method for measuring the absolute harmonic content of an X-ray beam, parameterised using the harmonic-photon percentage. The technique used X-ray attenuation measurements of a large number of aluminium foils and was applied to determine the harmonic content of a synchrotron beam at 31 energies between and 8 keV. Our technique is an improvement over earlier methods because its results are more fundamental and are transferable between detectors and experimental geometries. A general method was also presented for obtaining the spectral distribution of an X-ray beam by measuring its transmission spectrum.

References

- [1] C. Q. Tran, Z. Barnea, M. D. de Jonge, B. B. Dhal, D. Paterson, D. J. Cookson, C. T. Chantler, *X-Ray Spectrom.* **2003**, 32, 69.
- [2] M. D. de Jonge, Z. Barnea, C. Q. Tran, C. T. Chantler, *Phys. Rev. A* **2004**, 69(2), 022717.
- [3] W. L. Bragg, *Proc. Cambridge Phil. Soc.* **1912**, 17, 43.
- [4] L. Silberstein, *J. Opt. Soc. Am.* **1932**, 22(5), 265.
- [5] B. R. Archer, L. K. Wagner, *Med. Phys.* **1982**, 9(844), 844.
- [6] M. Rubio, R. T. Mainardi, *Phys. Med* **1984**, 29(11), 1371.
- [7] J. L. Glover, C. T. Chantler, Z. Barnea, N. A. Rae, C. Q. Tran, D. C. Creagh, D. Paterson, B. B. Dhal, *Phys. Rev. A* **2008**, 78(5), 052902-1.
- [8] D. C. Creagh, J. H. Hubbell, *Acta Crystallogr.* **1987**, 43(1), 102.
- [9] C. T. Chantler, *J. Phys. Chem. Ref. Data* **2000**, 29, 597.
- [10] J. D. Grunwaldt, P. Kappen, L. Basini, B. S. Clausen, *Catal. Lett.* **2002**, 78(1), 13.
- [11] H. Huwe, M. Froba, *J. Synchrotron Radiat.* **2004**, 11, 363.
- [12] G. Chen, H. Jain, S. Khalid, J. Li, D. A. Drabold, S. R. Elliott, *Solid State Commun.* **2001**, 120(4), 149.
- [13] P. H. Huang, K. R. Kase, B. E. Bjärngard, *Med. Phys.* **1983**, 10, 778.