A compact optical design for Bragg reflections near backscattering

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A very compact in-line X-ray optical design is presented which is optimized for use with Bragg reflections close to backscattering (Bragg angles near 90°). The essential idea is to use a low-order Bragg reflection to couple the X-ray beam into a small channel-cut backscattering crystal. The design is demonstrated in an almost theoretically performing high-resolution monochromator providing 2×10^8 photons s^{-1} in a 0.52 meV bandwidth at 25.65 keV. The monochromator is used to measure inelastic nuclear scattering from phonons in ^{161}Dy-containing samples.

Keywords: X-ray optics; high-resolution monochromators; nuclear resonant scattering; inelastic X-ray scattering.

1. Introduction

Advances in X-ray scattering techniques are often closely linked to improvements in X-ray optics. Particular examples relevant to this work (where we focus on high-energy resolution) include development of backscattering optics (*i.e.* Bragg reflections at angles near 90°) [earlier work includes Kohra & Matsushita (1972), Freund & Schneider (1972), Brümmer *et al.* (1979), Caticha & Caticha-Ellis (1982) and Graeff & Materlik (1982)] which have been crucial for the development of inelastic X-ray scattering (IXS) [see Sette *et al.* (1998) and Burkel (2000), and references therein] and of (non-backscattering) high-resolution monochromators (Faigel *et al.*, 1987; Ishikawa *et al.*, 1992; Toellner *et al.*, 1992; Chumakov *et al.*, 1996; Toellner *et al.*, 1997) which have been important for the development of nuclear resonant scattering (NRS) [see Gerdau & de Waard (2000), and references therein]. Thus new optics provide the potential for the development of new techniques. Furthermore, if a new design provides simplification and improved efficiency for an already available method, then it has an important and immediate applied value, especially if that method is flux-limited (as is the case for both NRS and IXS). Here we present a new optical design that expands the availability of these techniques and provides a performance that is (at least) comparable with presently available designs.

Essentially, two different approaches have been previously used for high-resolution monochromators. On one hand, for experiments where the X-ray energy may be arbitrarily chosen to match an appropriate lattice constant, the use of a single back-reflection is possible [see references above, while a recent relevant example is Verbeni *et al.* (1996)]. However, while being rather efficient, this design is extremely awkward geometrically, since the outgoing beam lies almost on top of the incident beam. On the other hand, for cases where the energy is fixed by other experimental parameters (*e.g.* to match a particular resonant transition), one cannot generally get close enough to backscattering in silicon for reasonable angular acceptance so one is forced to use less efficient but relatively compact high-resolution monochromators [see references above, and also Yabashi & Ishikawa (2000)]. However, these compact designs become increasingly difficult as one approaches closer to 90° Bragg angles, because the separation between the crystals becomes prohibitively large, scaling as $h/2\cos\Theta_B$ (where h is the transverse beam size – 'height' – in the scattering plane).

Here we present a design that provides a bridge between the single back-reflection method and more compact high-resolution monochromator designs. The essential idea of our design is to use a crystal simultaneously in Bragg reflection and in transmission, as is shown in Fig. 1. The incident beam is reflected from a low-order reflection placed *inside* of the backscattering crystal. The beam then reflects from the backscattering crystal, passes through the coupling crystal, reflects off the second face of the backscattering crystal, and is reflected off the 'back' of the coupling crystal and out of the optic. This coupling scheme is feasible for Bragg angles that deviate from exact backscattering by the Darwin width of the coupling crystal (~μrad for most cases of interest).

2. Experimental results

We explored this design in the context of a high-resolution monochromator for nuclear resonant scattering of synchrotron radiation. A detailed drawing of our monochromator is shown in Fig. 2. It differs from Fig. 1 in that the low-order Bragg reflection is cut asymmetrically to help collimate the beam reflected onto the second crystal. It operated at 25.65 keV, using the (6 2 0) and the (18 12 6) reflections in silicon (Bragg angles of 16.3 and 87.4°, respectively). The inner, (6 2 0), coupling crystal was cut with an asymmetry of 15° ($b = -1/22$) corresponding to an angular acceptance of 9.2 μrad and an output divergence of 0.42 μrad, matched to the acceptance of the (18 12 6) high-resolution crystal. The monochromator was designed

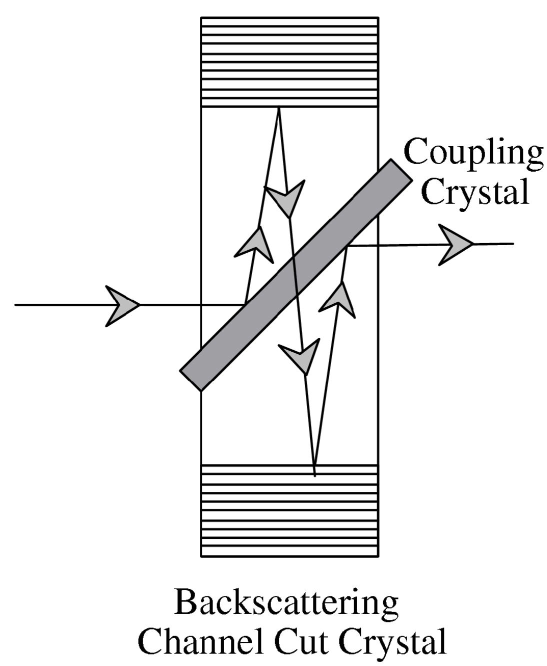

Figure 1
Schematic of the monochromator showing the low-order Bragg reflection, 'coupling crystal', inside of a channel-cut high-resolution crystal operated near backscattering. The arrows show the X-ray beam path.

to have an 8.7 μrad angular acceptance and 1 mm vertical acceptance. The channel in the high-resolution crystal was 60 mm wide while, without the transmission design, it would have needed to be >200 mm for the same vertical acceptance. The blade of the coupling crystal was polished to a thickness of 0.8 mm.

The monochromator was tested at BL35XU of SPring-8 (Baron *et al.*, 2000). X-rays from the third harmonic of a standard in-vacuum undulator (Kitamura, 1998) operating at a 13.22 mm magnet gap were monochromated using Si(111) crystals cooled by a recirculating liquid nitrogen (LN_2) system (Mochizuki *et al.*, 2001). We note that the LN_2 cooling system caused some vibration of the Si(111) crystals (~2–3 μrad amplitude). The beam size was reduced to 0.5 mm × 1.2 mm (vertical × horizontal) by slits at 28 m from the source, as this was the maximum heat load tolerated by the cooling system. A symmetric Si(111) channel-cut crystal was used downstream of the monochromator to help reduce the harmonic content of the beam. The high-resolution monochromator crystals were mounted on a single stage having two independent concentric axes. One crystal was mounted on each axis. These axes each have a resolution of ~25 nrad step^{-1}, and are part of a standard precision optical table set-up at SPring-8 (Ishikawa *et al.*, 1992).

The monochromator performed in near exact accordance with calculations† based on the dynamical theory of diffraction [see, for example, Batterman & Cole (1964), while the specific case of back-scattering is treated in, for example, Caticha & Caticha-Ellis (1982)] after appropriate temperature control was implemented (see below). The bandwidth, determined by measuring the elastic peak of the incoherent nuclear scattering from Dy_2O_3 (95.8% enriched in ^{161}Dy), was 0.52 meV full width at half-maximum (FWHM), as compared with a 0.51 meV calculation (see Fig. 3). The throughput was 2 × 10^8 photons s^{-1} (at 100 mA storage-ring current) as measured with a PIN diode, or 1.5 × 10^{-5} of the incident beam (as measured by both a PIN diode and an ionization chamber). Comparing this with the ~4.5 eV incident bandwidth, one finds an efficiency (the ratio photons s^{-1} meV^{-1} out to photons s^{-1} meV^{-1} in) of 11 ± 1%, while theory (including 50% attenuation on transmission through the coupling crystal) suggests 13 ± 2%.

The temperature control of this design, while not difficult, is an important technical detail that deserves note. The thermal expansion coefficient of silicon near room temperature is about 2.6 p.p.m. K^{-1}, so an 8 mK temperature shift of the high-resolution crystal is sufficient to change the reflected energy by the 0.5 meV bandwidth of the monochromator. Thus both the absolute temperature and temperature uniformity of the high-resolution, (18 12 6), crystal are important issues. We used a pair of thermistors placed just to one side of the X-ray beam to measure both the average temperature and the temperature difference of the two blades of the (18 12 6) crystal (see Fig. 2). This facilitated the use of a small (~25 mW) heater on the second blade to compensate for the unequal thermal load (bandwidth) incident on the two blades. This correction was required for proper function of the monochromator, but was practically rather easy, with manual adjustment every 8 to 12 h, sufficient to preserve the bandwidth of, and flux through, the monochromator. Operation without the heater lead to losses of about a factor of three in output flux.

The monochromator was used to measure inelastic nuclear absorption [see Seto *et al.* (1995), Sturhahn *et al.* (1995), while reviews may be found in Gerdau & deWaard (2000)] from materials containing ^{161}Dy (Baron *et al.*, 2001), and sample spectra are presented in Fig. 4. For Dy_2O_3 (95.8% enriched in ^{161}Dy) at room temperature, both the phonon annihilation (negative energies) and phonon creation (positive energies) are evident (and related by the appropriate temperature – Bose occupation – factor). The second

† Calculations used optical constants based on Cromer & Liberman (1970), McMaster *et al.* (1969), Waasmaier & Kirfel (1994), and code from Brennan & Cowan (1992). All four bounces of the monochromator were treated with proper angle/energy correlations.

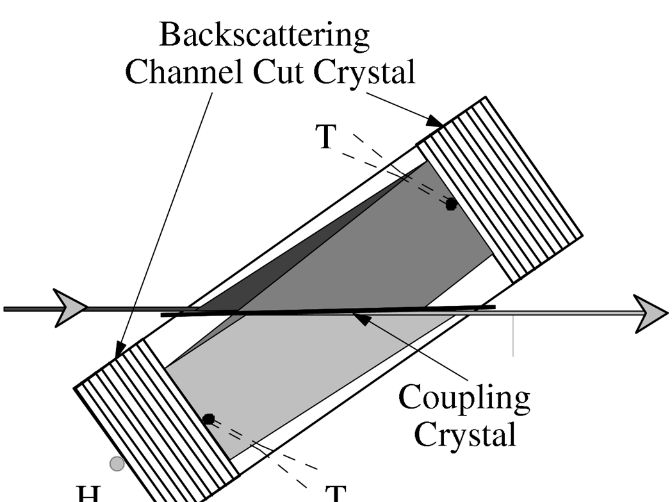

Figure 2
Sketch of the monochromator used in this work. The beam size reflects the effect of the asymmetric reflections. T and H refer to locations of thermistors and a heater, respectively, for temperature measurement and control (see text).

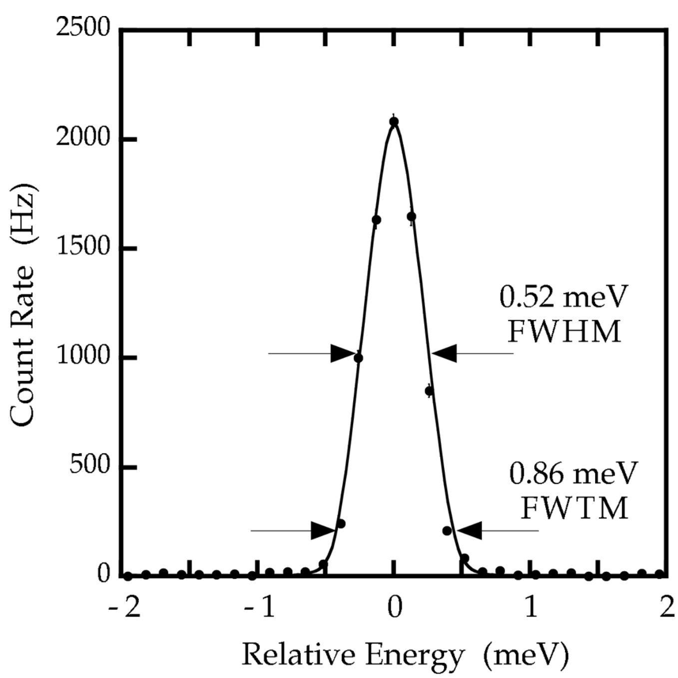

Figure 3
Scan through the elastic peak from nuclear resonant absorption in Dy_2O_3 at room temperature. This gives an upper bound on the resolution, and, in this case (where the sample is not expected to have significant quasi-elastic excitations), should faithfully represent the monochromator resolution, up to a relatively flat background due to absorption with simultaneous creation/annihilation of phonons. See also Fig. 4(*a*). The solid line is the calculation according to the dynamical theory of diffraction.†

sample, DyB_2C_2 (natural abundance, 19%, ^{161}Dy), shows the feasibility of studies of non-enriched materials of current interest [this particular sample shows antiferroquadrupolar ordering (Yamauchi *et al.*, 1999)]. In both cases the disappearance of the tail of the elastic peak at ± 2 meV is evident, showing the good response of the monochromator, as does the resolution of the mode at 18 meV in the DyB_2C_2. In addition, the phonon partial density of states was derived and is provided in the insets, further demonstrating the quality of the data.

3. Discussion

Having demonstrated the feasibility and excellent performance of this monochromator design, we now briefly discuss its application in more generality. One immediate use would be as a replacement for the single-reflection backscattering monochromators used for IXS. Our design provides the convenience of an in-line geometry, but at the cost of reduced flux: the peak reflectivities in silicon, at room temperature, for reflections with $\leq$meV bandwidth are typically 0.5 to 0.75, so the second bounce from the high-resolution crystal reduces the optic efficiency by a factor of two or more (though it also improves the tails of the response). Whether this is an acceptable trade-off depends on a detailed consideration of the experiment. We note that earlier in-line designs [*e.g.* the 'nested' design of Ishikawa *et al.* (1992)] are also feasible for IXS (Schwoerer-Böhning *et al.*, 1998; Sinn *et al.*, 2001) and might provide competition for the design presented here. (Exact comparison is difficult as the monochromators used have slightly different design parameters.)

We note that our design will have some losses dues to transmission through the central crystal. In general, these losses may be reduced by making the central crystal thinner: a similar size crystal has been polished to 0.3 mm, and even thinner should be possible (up to issues of crystal perfection and/or mounting strain). However, the thickness of the coupling crystal affects the offset of the output beam relative to the incident beam, and also the attenuation of the beam passing through the coupling crystal, without traversing the high-resolution bounces. If the coupling crystal is too thin, then the beam traversing all reflections of the monochromator may lie on top of a broad-bandwidth background from that part of the incident beam that is transmitted. Thus, this thickness must be chosen with some care: at higher X-ray energies (*e.g.* >20 keV) a careful choice of asymmetric cut and thickness for the coupling should allow reasonable efficiencies for silicon designs, but at lower energies it may be necessary to use a different material (*e.g.* diamond or possibly even beryllium as the angular acceptances at backscattering can be quite large) for the central crystal.

The use of this design for nuclear resonant scattering is interesting on several levels. On the one hand, this particular monochromator for the ^{161}Dy resonance performs extremely well. On another, there is the possibility of extending this design to other resonances. In the 20–30 keV range, silicon designs should be relatively straightforward. At higher energies, the reflectivity of silicon at backscattering falls off quickly (due both to thermal vibrations and the reduced silicon form factor), but this design still might be feasible in the 30–40 keV range. Finally, an additional possibility is the replacement of the silicon backscattering crystal with a non-cubic crystal having more possible back reflections [*e.g.* Shvyd'ko & Gerdau, 1999]. While significant issues regarding crystal perfection must be addressed, advantages would be that it might be possible to operate at Bragg angles of 89.5° at nearly any energy, and, it might be easier to go to higher energies. However, for the non-silicon case, high-quality crystals would probably be too small to make a full channel-cut crystal, so it might be necessary to make an artificial channel-cut along the lines of Shu *et al.* (2000).

In summary, we have presented a new optical design for use near backscattering, and data from an example showing excellent (near theoretical) performance, including high throughput and extremely good resolution. The optic is immediately useful as a monochromator for nuclear resonant scattering experiments with ^{161}Dy and has potential application both to inelastic X-ray scattering and nuclear resonant scattering with other isotopes.

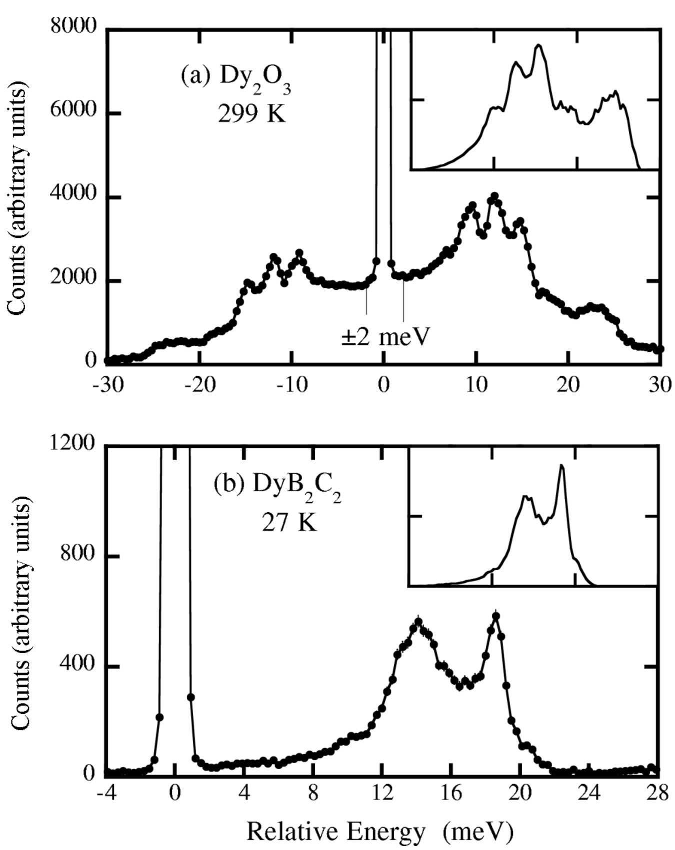

Figure 4
Inelastic absorption spectra from (*a*) Dy_2O_3 powder at room temperature and (*b*) a DyB_2C_2 crystal at 28 K. Collection times were about 8 and 14 h, and elastic peak heights are 1×10^5 and 4×10^4, respectively. See text for details. Lines are to guide the eye. Insets show the derived partial phonon densities of states (30 meV full scale in each case).

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