Exact Bragg backscattering of x rays

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Exact 180° Bragg scattering of pulsed synchrotron radiation was observed by using a semitransparent detector and a time-of-flight technique. The angular dependences of Bragg scattering of monochromatic 14.413 keV x rays with only 0.5 μeV bandwidth were studied by utilizing the $(1\ 3\ \bar{4}\ 28)$ reflection of an Al_2O_3 crystal at different temperatures. By heating the crystal first the exact backscattering shows up achieving maximum intensity and 1.7 mrad angular width at 372.40 K. By increasing the temperature it develops into the usual Bragg reflection with much narrower angular profile. The measured dependences are in agreement with the theory. The fit is sensitive to a 5×10^{-9} Å variation of interplanar distance. [S0163-1829(98)08609-3]

Bragg scattering of x rays with angles close to 180° is a well established experimental technique. It is successfully applied for monochromatization of hard x rays to meV and sub-meV bandwidths.[1–8] Despite this fact the properties of backscattering remain experimentally not fully studied. So far, to our knowledge, in no experiment were x rays observed which were reflected from a crystal exactly opposite to the direction of the incident beam.[9] It is due to this fact that up to now applications involving exact backscattering, e.g., Fabry-Perot[10,11] and other types of x-ray backscattering interferometers are not realized. Our purpose was to observe exact Bragg backscattering of x rays, to study its angular and energy dependences, to observe its transition to the usual Bragg scattering and to test the validity of the dynamical theory of x-ray diffraction under these extreme conditions.

Observation of scattering at exactly 180° has an obvious experimental difficulty. The x-ray source or the detector is blocking the reflected or the incident x rays, respectively. To overcome this problem we have applied a semitransparent detector with a good time resolution and made use of the pulsed structure of synchrotron radiation. The time-of-flight $\tau=2L/c$ to the crystal and back to the detector separated by a distance L was used to distinguish between the incident and reflected radiation pulses, see Fig. 1.

Our experiment ($L=9.9$ m and $\tau=66$ ns) was performed at the wiggler beam line BW4 at HASYLAB (Hamburg). The DORIS-III positron storage ring was operated in the five-bunch mode producing radiation pulses from the wiggler every 192 ns ($\sim$ 200 ps duration). A double-crystal Si(111) monochromator provided radiation with 3 eV bandwidth at the energy of the nuclear resonance in ^{57}Fe (14.413 keV). The ^{57}Fe nuclei in an iron foil of 10 μm thickness enriched in ^{57}Fe to 95% were excited resonantly by the incident beam. They emit 14.413 keV photons predominantly in the forward direction[12,5] within a narrow energy band of less than 0.5 μeV width and with an average delay of $\sim$ 40 ns. Only these delayed resonant quanta are counted to make use of the radiation with 0.5 μeV bandwidth. As a semitransparent detector served an avalanche photodiode with a 100 μm thick sensitive Si wafer.[13] Its time resolution was 1 ns. This time resolution was thus used twice in the experiment. Firstly, to distinguish between the incident and reflected radiation pulses and secondly to count only resonant photons in the 0.5 μeV band. The beam divergence was 20 μrad in the vertical (z,x) and 80 μrad in the horizontal (z,y) plane.

Sapphire single crystals, instead of Si or Ge which are standard in x-ray crystal optics, were used as backscattering mirrors. Several reasons motivated this choice. Bragg's law $2d_{hkl}\sin\theta_B=hc/E$ for x-rays with energy E scattered from crystal planes (hkl) with interplanar distance d_{hkl} reduces in the case of backscattering ($\theta_B=\pi/2$) to $E_B=hc/2d_{hkl}$. Every reflection (hkl) has its Bragg energy E_B. However, because of the cubic symmetry of Si and Ge many reflections have the same d_{hkl} and thus the same Bragg energy. Therefore the number of matching energies is low: once per $\simeq$ 500–250 eV in the range 10–25 keV. Furthermore, pure exact backscattering in crystals with such a high symmetry is not possible due to multiple beam Bragg diffraction,[10] and the reflectivity for hard x rays with $E>25$ keV decreases rapidly because of the relatively low Debye temperatures.

By contrast crystals with lower symmetry like hexagonal Al_2O_3 with high Debye temperature and low photoabsorption suit much better for backscattering. Sapphire single crystals allow exact backscattering at least once per $\simeq$ 15 eV in the range 10–25 keV and even more often for harder x rays. By heating or cooling Al_2O_3 for not more than 100 K from room temperature one can fulfill the backscattering condition for any x-ray energy above 10 keV. E.g., for the

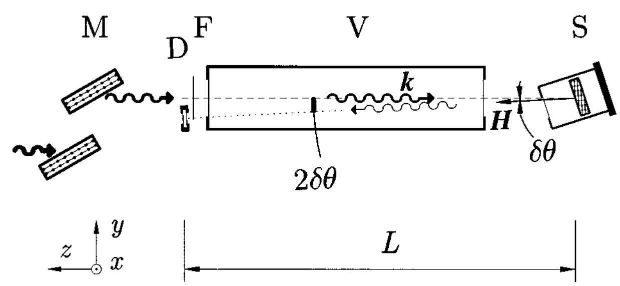

FIG. 1. Scheme of the backscattering experiment; M is the Si(111) monochromator (shown rotated by 90° about ***z*** axis); D is the semitransparent x-ray detector; F is the 10 μm ^{57}Fe foil; V is the 8 m vacuum tube; S is the Al_2O_3 single crystal in the oven; $\delta\theta$ is the angle between the wave vector ***k*** of the incident radiation and the scattering vector ***H***.

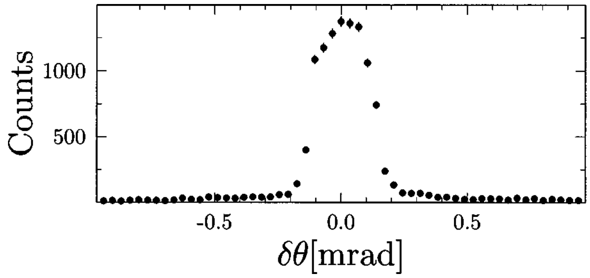

FIG. 2. Angular dependence of the exact Bragg scattering of the 14.413 keV x rays with 3 eV bandwidth from an Al_2O_3 crystal measured at different angular deviations $\delta\theta$ from normal incidence to the $(1\ 3\ \bar{4}\ 28)$ reflecting planes. The detector was not moved. Therefore the width of the rocking curve is given by the angular acceptance of the detector.

14.413 keV ^{57}Fe energy the $(1\ 3\ \bar{4}\ 28)$ back reflection in sapphire can be predicted at 380 K with angular acceptance of 1.3 mrad, energy bandwidth of 6.2 meV, and 88% reflectivity (the crystal data of Refs. 14,15 were used).

In our experiments sapphire crystals in form of a disk 15 mm in diameter and 1 mm thick cut perpendicular to the c axis were employed.[16] The crystal was installed in an oven on a four-circle diffractometer at the end of the vacuum tube, see Fig. 1. At first the crystal c axis was oriented parallel to the incident beam by detecting the exact back reflection $(0\ 0\ 0\ 30)$ of x rays with an energy $E=14.315$ keV. Then the $(1\ 3\ \bar{4}\ 28)$ planes were set perpendicular to the incident beam by detecting the exact back reflection $(1\ 3\ \bar{4}\ 28)$ of the incident 14.413 keV x rays with the broad 3 eV band. The angular position of the crystal at which it reflects these x rays back into the detector with maximum intensity was taken as the reference point $\delta\theta=0$ for the exact backscattering, see Fig. 2. As a next step the temperature of the crystal was scanned to find the temperature region at which the 14.413 keV resonant quanta in the 0.5 μeV band are reflected.

Figure 3 shows such temperature scans recorded at different angular deviations $\delta\theta$ of x rays from normal incidence to the $(1\ 3\ \bar{4}\ 28)$ planes. The temperature in the oven was controlled with a relative accuracy of 1 mK. At $\delta\theta=0.2$ mrad the maximum of reflectivity is achieved at $T_R=372.40 \pm 0.01$ K. The width of the reflection curve is 100 mK corresponding to 10.0 meV energy width. This is more than the expected 6.2 meV. With increasing $\delta\theta$ the temperature where maximum reflectivity is reached increases proportional to $\delta\theta^2$. This square dependence which is observed only for backscattering has a remarkable consequence, namely an extraordinarily large angular acceptance as demonstrated below.

Figure 4 shows the angular dependences of Bragg scattering of 14.413 keV resonant radiation measured at different temperatures T of the same Al_2O_3 crystal. Below the reference temperature at $\Delta T=T-T_R<0$ the Bragg scattering scarcely takes place. Approaching T_R the Bragg scattering builds up with the maximum at $\delta\theta=0$ and reaches the maximum angular width (full width at half maximum) of 1.7 mrad at $\Delta T=0$. With increase of the temperature above T_R the maximum reflectivity deviates from $\delta\theta=0$, the rocking curve narrows and gradually develops into the conventional

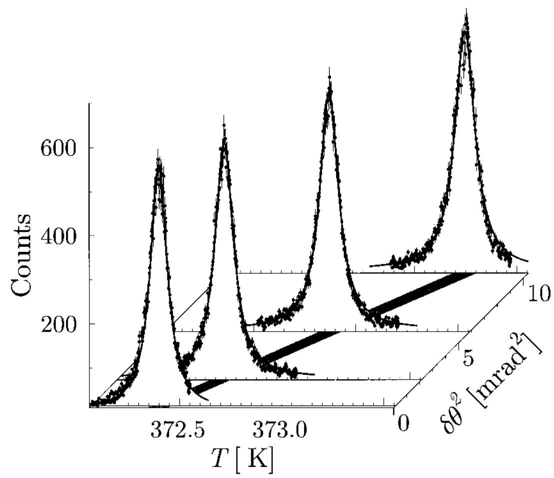

FIG. 3. The temperature dependences of Bragg scattering of resonant 14.413 keV photons with 0.5 μeV bandwidth from an Al_2O_3 crystal measured at different deivations $\delta\theta$ from normal incidence to the $(1\ 3\ \bar{4}\ 28)$ reflecting planes. Solid lines are fits with Lorentzians. The width of the curve at $\delta\theta=0.2$ mrad is 100 mK (equivalent to 10.0 meV x-ray bandwidth).

Bragg reflection. Nevertheless the width of the Bragg reflection at $\delta\theta=2.78$ mrad still has the large value of 0.24 mrad.

The reflectivity of 14.413 keV nuclear resonant radiation was measured to be 64%, close to the theoretical value for a perfect crystal, at the exact 180° Bragg scattering condition.

The angular dependences were fitted by using the dynamical theory of x-ray diffraction. The principles underlying the theory (see, e.g., Refs. 17,18) are valid for backscattering too.[1,19,20] The solution for the angular and energy dependences of Bragg scattering is expressed through the scattering amplitudes χ_0, and χ_h, crystal thickness ℓ, asymmetry parameter b and the parameter $a=(\boldsymbol{H}^2+2\boldsymbol{Hk})/\boldsymbol{k}^2$ which is a function of the energy E and the angle of incidence $\theta=\pi/2-\delta\theta$ of the x rays. The total reflectivity in a nonabsorbing $(\chi''_{0,h}=0)$ and semi-infinite crystal occurs within the interval (a_+,a_-) where

$$a_{\pm}=\pm 2|\chi_h|/\sqrt{|b|}-\chi_0(1/b-1). \tag{1}$$

We have found that the following presentation for a:

$$a=4\frac{E_B}{E}\left(\frac{E_B}{E}-\sin\theta\right) \tag{2}$$

can be used which is valid for any value of the angle $0\leqslant\theta\leqslant\pi$. However, the theoretical approaches of Refs. 17 and 18 use the approximation $a=2(\theta_B-\theta)\sin 2\theta_B$ which fails in the backscattering region. As it was shown in Ref. 20 another approximation is applicable for backscattering $(|\delta\theta|\ll 1)$:

$$a=2[\delta\theta^2-2(E-E_B)/E_B]. \tag{3}$$

Figure 5 is a kind of DuMond diagram modified for backscattering (note the $\delta\theta^2$ dependence), i.e., the spectral-angular region of total reflection which is depicted according to Eqs. (1), (3) for the particular case $b=-1$. The diagram helps to see that our experimental results (Figs. 3,4) qualitatively agree with the main theoretical predictions of Refs.

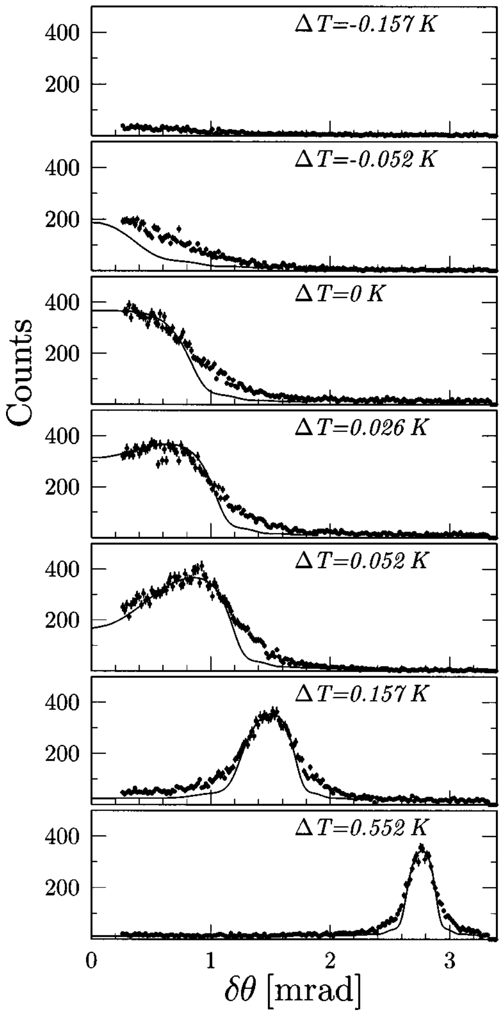

FIG. 4. The angular dependences of Bragg scattering of monochromatic 14.413 keV x rays with 0.5 μeV bandwidth measured at different temperatures T of an Al_2O_3 crystal utilizing the ($1\ 3\ \bar{4}\ 28$) reflection. $\delta\theta$ is the angular deviation from normal incidence of the x rays to the ($1\ 3\ \bar{4}\ 28$) reflecting planes. $\Delta T = T - T_R$ where T_R=372.40 K. Solid lines are the fits using the dynamical theory of Bragg scattering.

1,19,20. It also shows that the largest angular acceptance of $\sqrt{2|\chi_h|}$ and simultaneously the highest relative energy resolution $|\chi_h|$ are achievable in the exact backscattering geometry. The fits of Fig. 4 were performed by using the results of Refs. 17 and 18 but with the nonapproximated parameter a of Eq. (2).

We have made certain assumptions to take into account the observed broadening of the temperature profiles and angular curves compared with the theoretical widths for an ideal crystal. It is supposed that the broadening is caused by a variation of the interplanar distance d_{hkl} with crystal depth. As the real distribution is not known, it is simply assumed that at some depth the atomic planes are shifted by half of the radiation wavelength relative to the ideal undisturbed position. Accordingly the coherent response of the rest of the crystal does not contribute significantly to the scattering. The crystal thus has a smaller effective thickness. We have ascertained that the angular and energy dependences calculated for the crystal with a thickness $\ell = 70$ μm may describe satisfactorily our experimental results as it is shown by solid

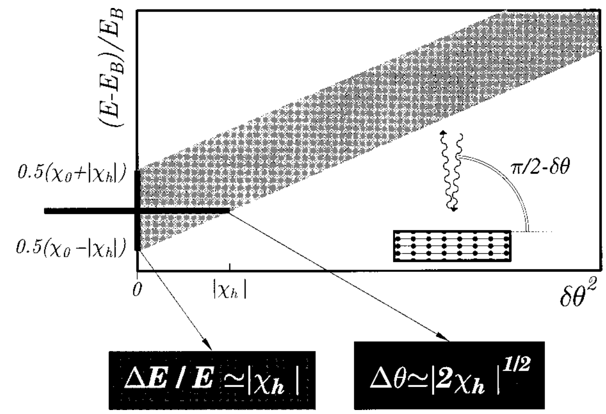

FIG. 5. Spectral-angular region of total reflection of the Bragg backscattering. Note that the abscissa scales with $\delta\theta^2$.

lines in Fig. 4. By taking crystal defects more precisely into account one could certainly describe better the wings of the angular distributions. It is remarkable that the fit turned out to be sensitive to as small as a 5×10^{-9} Å variation of the average interplanar distance. The temperature dependence obtained from this fit is $d_{hkl} = d_R(1 + 6.944\times10^{-6}\ \Delta T/\mathrm{K})$ where $d_R = 0.430\,108$ Å. By using this dependence one obtains for the variation of the energy of backscattered x rays with crystal temperature a value of $dE/dT = 0.100$ eV/K. The following data were used in the calculations: $E = 14.4132$ keV, $\chi_0 = (-78.395 + i0.289)\times10^{-7}$, $\chi_h = (-4.036 + i0.129)\times10^{-7}$.

In summary, exact 180° Bragg scattering of synchrotron radiation was observed by using a semitransparent detector and the time-of-flight technique. The angular dependences of Bragg scattering of resonant 14.413 keV (0.5 μeV bandwidth) photons were studied at different temperatures of Al_2O_3 crystals. It was observed that by heating the crystal first the exact backscattering shows up with an extremely broad angular width. By increasing the temperature it develops into the usual Bragg reflection with much narrower angular profile. The results are in a good agreement with the dynamical theory of Bragg scattering. The fit is sensitive to a 5×10^{-9} Å variation of the interplanar distance.

We have found by extended simulations that for x rays of any energy in the range 10–70 keV noncubic and hard single crystals like sapphire allow backscattering with significant reflectivity at a temperature in the range of 200–450 K. This may open a broader field of applications of Bragg backscattering in x ray optics with enhanced luminosity and high energy resolution. Backscattering mirrors for x ray interferometers and resonators would be possible. Especially attractive is the fact that the longitudinal coherence length of nuclear resonant photons is in the order of meters. Also for any Mössbauer transition in the mentioned energy range not only one reflection can be found to be used for a meV or sub-meV energy-resolution backscattering monochromator. This will allow observation of new nuclear excitations and promote further the Mössbauer spectroscopy in time domain[21,5] as well as the phonon spectroscopy by means of inelastic nuclear scattering.[22–24] Backscattering experiments with sapphire crystals of better quality are in progress.

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