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1990 J. Phys.: Condens. Matter 2 3989

(http://iopscience.iop.org/0953-8984/2/17/009)

J. Phys.: Condens. Matter 2 (1990) 3989–3995. Printed in the UK

Pure nuclear broad-band reflection of Mössbauer radiation

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Received 22 November 1989

Abstract. Strong enhancement of the γ,γ channel in nuclear resonant scattering was observed in a pure nuclear Bragg reflection of an almost perfect single crystal of $^{57}FeBO_3$. A broadening of the lines in the Mössbauer reflection spectrum up to 30 natural linewidths and reflectivities $\geqslant 40\%$ were measured at the exact Bragg position. Furthermore interference of the hyperfine transitions gave rise to high reflectivity between the lines. In total, the region of high reflectivity extended over more than a hundred natural linewidths. Such a pure nuclear Bragg reflection can be effectively used for broad-band Mössbauer filtering of synchrotron radiation. In addition, rocking curves were measured with high angular resolution. Special line-shapes, characteristic of Bragg scattering at resonance, were observed.

1. Introduction

In resonant scattering of Mössbauer radiation by single crystals all nuclei scatter in phase when the Bragg condition is fulfilled. In small single crystals the reflected wave arises from a coherent superposition of singly scattered wavelets (kinematical diffraction). In this case the resonant scattering already proceeds via a delocalised excited state, which has an enhanced probability of decay into the radiative channels (Trammell 1961, Afanasev and Kagan 1965a). In large perfect single crystals multiple scattering occurs and leads to a strong collective response of the nuclear system (dynamical diffraction). Important features are the enhancement of the γ, γ channel and the suppression of the γ, e channel (Afanasev and Kagan 1965b, Kagan *et al* 1968, Hannon and Trammell 1969). In stationary conditions, the enhancement reveals itself as a broadening of the lines in a Mössbauer reflection spectrum (van Bürck *et al* 1980, van Bürck 1989, Shvydko and Smirnov 1989). In time dependent experiments, the enhancement appears as a speeding-up of the response to a resonant excitation (Smirnov and Shvydko 1986, van Bürck *et al* 1987).

These effects are of fundamental interest in the context of collective coherent phenomena. They are also of great practical interest in the resonant filtering of synchrotron radiation. In order to obtain Mössbauer radiation out of the white synchrotron beam, nuclear Bragg reflections are employed. Hence the collective nuclear scattering in a Bragg reflection determines the properties of the extracted beam of Mössbauer radiation. Because of this important application, precise and quantitative studies of collective nuclear Bragg scattering are needed.

Collective effects are most pronounced at the exact Bragg position, when the number of nuclei responding in phase reaches a maximum. It is therefore important to study the effects as near as possible to the Bragg position. However, several factors usually contribute to the smearing of the angular resolution, such as slight crystal lattice deformations, finite horizontal and vertical divergencies of the primary beam, and instabilities of the angular setting. The present work was intended to minimise all these influences by utilising the combination of a highly perfect single crystal, a small divergence of the primary beam and a well stabilised γ-diffractometer. Under these conditions the enhancement of the γ, γ channel in nuclear resonant scattering at the Bragg position was studied.

2. Instrumentation and material

Measurements were performed on a two-axis γ-diffractometer with a special temperature control (van Bürck *et al* 1980) and with laser interferometric angular regulation (Maurus *et al* 1984). The γ-source, ^{57}Co diffused into a 12 μm Rh foil of 1×3.5 mm^2 (delivered by Amersham), had an initial activity of 140 mCi, a linewidth of 0.16 mm s^{-1} and a recoilless fraction of 0.74. The beam from the γ-source was collimated using a symmetrical Bragg reflection off a perfect Si crystal. The reflections (1 1 1) or (3 3 3) were used to obtain angular divergencies of 4″ or 1″, respectively. The beam was horizontally limited by a vertical slit. The vertical collimation was determined by the heights of the source (3.5 mm) and the Si(Li) detector (4 mm). The background counting rate of this detector with the source on the diffractometer was 0.05 counts min^{-1}.

The same $^{57}FeBO_3$ crystal platelet was chosen for the experiment as had previously been used by Smirnov *et al* (1986) and Shvydko and Smirnov (1990). Its linear dimensions were about 6×10 mm^2, its thickness about 50 μm, and the (1 1 1) planes were parallel to the surface. A transmission Borrmann topography of this crystal proved its high quality (figure 5(*a*) in Kotrbova *et al* 1985). However, in Bragg reflection geometry a slight bending was revealed. Half of the surface area showed a bending of about 2″, whereas the remaining part was bent by about 5″. Adjustments and preliminary studies of the crystal were performed with Mo Kα x-irradiation in the setting Si(333) $\times$ $^{57}FeBO_3$(444). In this geometry, the x-ray rocking curves of the better area showed half-widths of 5.5–6.2″, which were hardly broader than the folding of the intrinsic half-width (about 2″) and dispersive broadening (about 4″). The surface area finally selected was of about 4×5 mm^2. In the measurements with Mössbauer radiation, the primary beam was well collimated and carefully centred to illuminate only this crystal area.

3. Experiments and results

At first the nearly pure nuclear reflection $^{57}FeBO_3$(222) was measured. In contrast to the procedure in the previous study of this reflection (Shvydko and Smirnov 1989), the crystal was now placed in an external magnetic field perpendicular to the scattering plane. In this field orientation, it is mainly the $\Delta m = \pm 1$ transitions that are excited (van Bürck *et al* 1980), so the (222) spectra could be directly compared with the pure nuclear reflection spectra. The Mössbauer spectra of this reflection were measured for incident beams of about 10′ divergence (figure 1(*a*)) and of 4″ divergence (figure 1(*b*)), both centred at the Bragg position.

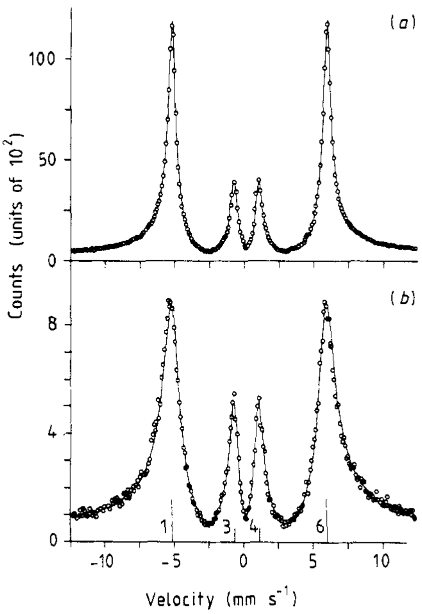

Figure 1. Mössbauer spectra of the nearly pure nuclear reflection $^{57}FeBO_3(222)$ measured for an incident beam of about 10′ divergence (*a*) and of 4″ divergence centred at the Bragg position (*b*). Spectrum (*b*) was fitted via the dynamical theory for resonant diffraction with EPD = 6.0″.

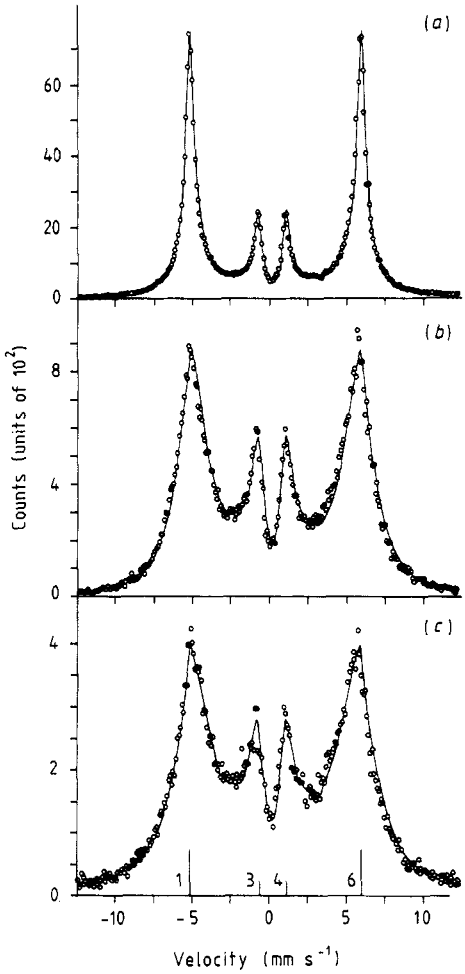

Figure 2. Mössbauer spectra of the pure nuclear reflection $^{57}FeBO_3(111)$ measured for an incident beam of about 10′ divergence (*a*), of 4″ divergence (*b*) and of 1″ divergence (*c*), each centred at the Bragg position. The fit of the spectra (*b*) and (*c*) yielded EPD = 6.5″ and 4.0″, respectively.

The main interest in the present study was the pure nuclear reflection $^{57}FeBO_3(111)$. Because of strong nuclear scattering in an external magnetic field perpendicular to the scattering plane (van Bürck *et al* 1980) and because of the large Lorentz factor of 5.6 connected with the low Bragg angle, this reflection has a very large intrinsic reflection width. This is especially favourable for the observation of a strong enhancement effect. The Mössbauer spectra of this reflection were measured for incident beams of about 10′ divergence (figure 2(*a*)), of 4″ divergence (figure 2(*b*)) and of 1″ divergence (figure 2(*c*)), each centred at the Bragg position.

In addition, the rocking curves of the reflections $^{57}FeBO_3(222)$ and $^{57}FeBO_3(111)$ were measured at resonance. These measurements were performed in order to reveal the characteristic shapes of the intrinsic Bragg reflection curves in the case of a pure imaginary scattering amplitude (Kagan *et al* 1968, Hannon and Trammell 1969). Rocking

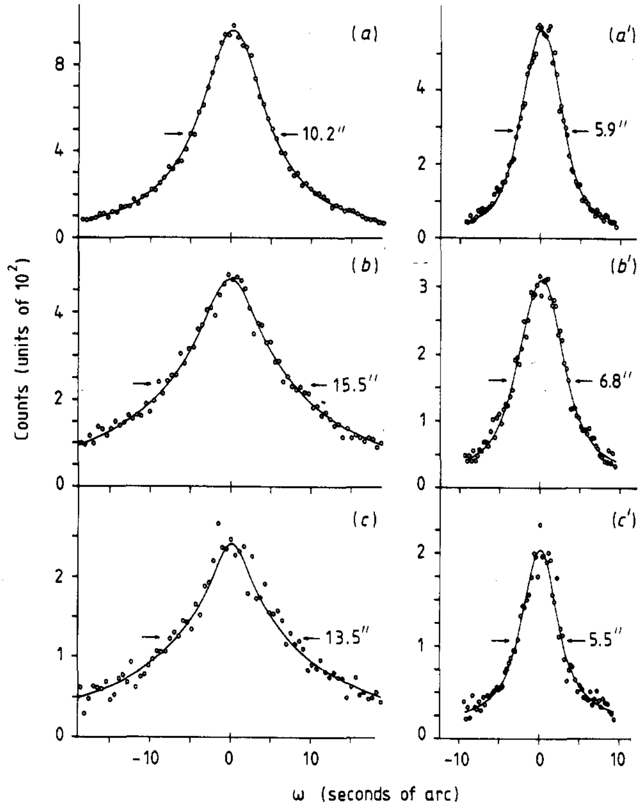

Figure 3. Rocking curves of the $^{57}FeBO_3$ single crystal measured with 14.4 keV Mössbauer radiation at constant velocities in resonance with lines 1 and 6 (*a–c*) and at constant acceleration in the energy range of the Mössbauer spectra (*a′–c′*). The curves were measured in the combinations Si(111) × $^{57}FeBO_3$(222) (*a*, *a′*), Si(111) × $^{57}FeBO_3$(111) (*b*, *b′*) and Si(333) × $^{57}FeBO_3$(111) (*c*, *c′*). From the fit of the curves via the dynamical theory for resonant diffraction the following values of EPD were obtained: 5.0″ for curves (*a*, *a′*), 4.5″ for curves (*b*, *b′*) and 3.0″ for curves (*c*, *c′*).

curves at constant velocities in resonance with lines 1 and 6 of the Mössbauer reflection spectrum were measured in each of the settings Si(111) × $^{57}FeBO_3$(222), Si(111) × $^{57}FeBO_3$(111) and Si(333) × $^{57}FeBO_3$(111). The rocking curves are shown in figures 3(*a–c*). They are rather broad and have specific shapes. In the Bragg position the reflectivity for the recoil-free radiation from the source was measured and was in all cases found to be larger than 40%. For comparison rocking curves were also measured when the source was moved at constant acceleration in the velocity range of the Mössbauer reflection spectra. These rocking curves are also shown, in figures 3(*a′–c′*). They are much narrower and of a different shape, because the nuclear scattering amplitudes decrease off resonance and their real part becomes dominant.

4. Fitting of Mössbauer spectra and rocking curves

Mössbauer spectra and rocking curves were fitted using the dynamical theory for nuclear resonant diffraction in Bragg geometry (Kagan *et al* 1968). In order to compare the results, a common parameter was used in the fit of the spectra, namely the effective divergence of the primary beam (EPD). Here, the crystal was supposed to be absolutely perfect and to be fixed at a precise angular position in the case of the Mössbauer measurements. The actual smearing of the angular dependence of the reflected intensity, which is caused by the various factors mentioned in the introduction, was then taken into account via a Gaussian distribution of the primary beam with an effective full halfwidth equal to the EPD.

The fits are shown as full curves in the figures. In the case of the measurements using a primary beam of 4″ divergence, the EPD values are 6–6.5″ for the Mössbauer spectra and 4.5–5″ for the rocking curves. In the case of the measurements using a primary beam of 1″ divergence, the EPD values were 4″ for the Mössbauer spectrum and 3″ for the rocking curves. The EPD values are larger for the Mössbauer spectra than for the rocking curves because angular instabilities contributed more in the long-time Mössbauer measurements.

5. Discussion

Consistent sets of Mössbauer reflection spectra and of rocking curves were obtained for the same almost perfect single crystal. The results for three different double-crystal combinations (Si(111) × $^{57}FeBO_3$(222), Si(111) × $^{57}FeBO_3$(111) and Si(333) × $^{57}FeBO_3$(111)), each measured by Mössbauer reflection spectra, rocking curves at resonance and rocking curves at constant acceleration, could be satisfactorily fitted by the dynamical theory for resonant diffraction. The effective primary divergence gave values that were about 2–3″ larger than the actual primary beam divergences. This difference is in agreement with the previous findings from x-ray rocking curves: it reflects mainly the slight bending of about 2″ of the selected crystal area.

The rocking curves measured at resonance show the characteristic shapes predicted by the dynamical theory for resonant Bragg diffraction (Kagan *et al* 1968, Hannon and Trammell 1969). They are sharply peaked at the Bragg position and have far-reaching wings. They strongly contrast with the usual Darwin cylinder hat curves connected with real scattering amplitudes (compare the inset of figure 5 in van Bürck *et al* 1980). In reality, however, the sharp peak will always be smeared a little because of angular averaging (finite effective primary beam divergence) and because of energy averaging (Lorentzian energy distribution of the incident beam). Nevertheless, reflectivities larger than 40% were reached, and the characteristic shape is well recognised especially in the case where the primary beam divergence is smallest in comparison with the intrinsic width of the reflection (figure 3(*c*)). These results demonstrate the dynamic character of the scattering.

The Mössbauer reflection spectra measured with 4″ resolution at the Bragg position show strongly broadened lines. The linewidths are 17 or 23 natural linewidths for the (222) or (111) spectra, respectively, as compared with the 7 natural linewidths of the angular integrated spectra. Besides the observed characteristic shapes of the rocking curves, this is another strong manifestation of the enhancement effect. This measurement and the preceding one (Shvydko and Smirnov 1989) demonstrate that line broadenings of the order of 20–25 natural linewidths can already be obtained with a radiation of 4″ divergence, where the intensity of the primary beam is still relatively high. The interference in between the hyperfine transitions is beautifully demonstrated by a comparison between the spectra of the nearly pure nuclear (x-ray-allowed) reflection(222) (figures 1(*a*) and (*b*)) and the pure nuclear (x-ray-forbidden) reflection (111) (figures 2(*a*) and (*b*)). The interference is much more pronounced in the spectra at the Bragg position than in the angular integrated spectra.

In the Mössbauer reflection spectrum measured with 1″ resolution, a broadening of the lines up to 30 natural linewidths was observed. This reveals an enhancement of the coherent channel by a factor of about 200 (van Bürck 1989, Trammell and Hannon 1988). In parallel with the broadening of the lines, strong interference between the lines leads to a region of high reflectivity, which extends over 120 natural linewidths (0.6 μeV). Broadening and interference arise equally for both of the two orthogonal polarisation components of the incident beam. A pure nuclear Bragg reflection with such a broad energy window can effectively be employed for broad-band Mössbauer filtering of synchrotron radiation. Broad-band filtering is of importance for a Mössbauer station when, for example, samples of different compounds of ^{57}Fe are to be excited by the radiation reflected from a universal ^{57}Fe filter.

The enhancement of the γ, γ channel at the Bragg position is also of importance in providing fast Mössbauer filtering of synchrotron radiation. An essential new feature of Mössbauer measurements using synchrotron radiation will be the measurement of the time evolution of the decay, which reveals the nuclear hyperfine splitting (Trammell and Hannon 1978, Gerdau *et al* 1986). For this purpose a filter is needed, that yields a fast pulse of about 1 μeV bandwidth. Strong collimation of the incident radiation to the exact Bragg position will essentially speed up the response of a nuclear Bragg reflection filter. The present experiment demonstrated that with high angular collimation and with $^{57}FeBO_3$ crystals of very high perfection, diffraction conditions can be reached that can be described by an effective primary beam divergence of 3–4″. On this basis fast Mössbauer filter systems (van Bürck *et al* 1990) can be designed.

6. Conclusion

The enhancement of the coherent channel in pure nuclear Bragg reflection was studied at the Bragg position. Consistent sets of Mössbauer spectra and of rocking curves were recorded. The results were satisfactorily fitted by the dynamical theory for resonant diffraction. Special line-shapes in the rocking curves and strongly broadened lines in the Mössbauer spectra were the most important results of the study. A line broadening up to 30 natural linewidths was observed at the Bragg position, corresponding to an enhancement of the coherent channel by a factor of about 200. Line broadening and strong interference of the hyperfine transitions yielded a large energy region of high reflectivity which extended over 120 natural linewidths. Such a pure nuclear Bragg reflection can be effectively used for broad-band Mössbauer filtering of synchrotron radiation. In addition, it can be concluded from the present experiment that fast Mössbauer filtering may be possible.

Acknowledgments

This work has been funded by the Bundesministerium für Forschung und Technologie under contract No KA1TUM4/7 and by the I V Kurchatov Institute.

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