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Diffraction of Mössbauer radiation under ultrasonic excitation of crystals

A A Opalenko and A A Kornilova

Faculty of Physics, Moscow State University, 119899 Moscow, Russia

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Abstract. The influence of high-frequency ultrasonic (US) waves on the diffraction of Mössbauer γ quanta in perfect and mosaic Si crystals is studied. Depending on the US amplitude, a strong decrease and oscillation of the elastic component of the Bragg intensity have been observed in the perfect Si crystal. In the mosaic crystal the variation in the Bragg intensity is essentially weaker. The oscillations of elastic scattered intensity are similar for transverse and longitudinal US waves and for surface acoustic waves as well.

1. Introduction

When crystals are subjected to excitation by an ultrasound (US) wave at the frequency ν_s, the Mössbauer spectrum has a central line at the frequency ν_0 corresponding to the elastic scattering of γ quanta and satellites at the frequencies $\nu_0 \pm \nu_s$, $\nu_0 \pm 2\nu_s$, (figure 1) corresponding to the inelastic scattering with absorption and emission of phonons. The intensity of each line is described by

$$J_0^2(Hu) + 2\sum_{n=1}^{\infty} J_n^2(Hu) = 1 \tag{1}$$

where J_n are Bessel functions of the first kind, H is magnitude of the diffraction vector of the γ quanta, u is the ultrasonic wave amplitude, and the total intensity is conserved. This formula is known from classical work on the Mössbauer effect.

However, this is correct only for the kinematic diffraction approximation. For the dynamic diffraction of x-rays in perfect Si crystals an enhancement of the total intensity of the Bragg reflections under US excitation has been observed in both the Bragg and the Laue geometry at all US frequencies [1, 2]. This effect can be ascribed to the extension of a very small reflection region under US distortion of the crystal. This change corresponds to a first-order Bessel function:

$$I \sim J_0(Hu) + 2\sum_{n=1}^{\infty} J_n(Hu). \tag{2}$$

For x-ray diffraction there is no possibility of performing a direct energy analysis of the diffraction beam owing to the large x-ray linewidth. Moreover, only the diffraction of Mössbauer radiation, with a natural linewidth equal to 10^{-8} eV (for ^{57}Fe), allows one to define the change in γ quanta energy under US excitation of a crystal within the frequency range 50–200 MHz.

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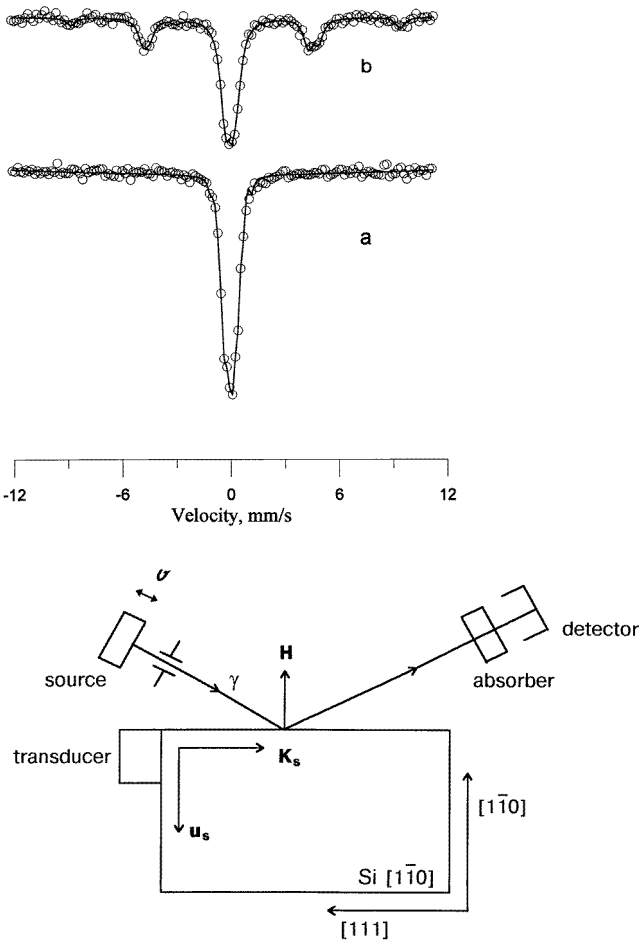

Figure 1. Mössbauer diffraction spectra for the Si(111) reflection: curve a, without any US excitation; curve b, with an excited longitudinal US wave at 50 MHz. The lower figure shows a sketch of the experimental arrangement.

The dynamic diffraction of Mössbauer radiation in a Si single crystal was studied for the first time by Gavrilov *et al* [3]. It might be obvious that, because of the sufficiently large divergence of the incident beam, central and angular satellite reflections are unresolved in the Mössbauer experiment. An enhancement of the total intensity of the Bragg peaks was observed with increasing US amplitude in a wide range of US frequencies, while the elastic component of the diffracted beam decreases in the presence of US waves at low frequencies and increases when the excitation US frequency exceeds the so-called threshold frequency ν_{th}. In [3] ν_{th} was determined as the frequency corresponding to the wavelength equal to the extinction length and the observed increase in the elastic part of the diffracted beam was attributed to the appearance of an anomalous Debye–Waller factor. Oscillation of the elastic intensity was not reported in this paper.

Gavrilov *et al* [4] also studied the dynamic diffraction of Mössbauer γ quanta in Si crystals in the Laue geometry. In this case, the intensity of the elastic scattering in the presence of US waves decreases both above and below the threshold frequency. However, Gavrilov *et al* observed an oscillation in the elastic intensity corresponding to equation (2).

Our detailed Mössbauer diffraction measurements were carried out for both a perfect a and mosaic Si crystal and are in strong contradiction to the results of previous work [3, 4].

2. Experimental part

The measurements were carried out on dislocation-free single Si [111] and Si [110] crystals. The samples were chemically polished and their degrees of perfection were checked by measuring the double-crystal rocking curves for the (111) reflection using Cu Kα x-rays. The half-width of the rocking curves appears to be equal to the theoretical value (about 10.2″) with an accuracy of 0.2″. The travelling transverse US waves were excited by a $LiNbO_3$ piezocrystal; the displacement vector $\boldsymbol{u}$ of the US wave was parallel to the diffraction vector $\boldsymbol{H}$ with frequencies ν_s in the range 140–170 MHz for sample 1 and 155–180 MHz for sample 2 (see sketch in figure 1).

This value of US frequency exceeds the so-called threshold frequency ν_{th} ($\nu_{th} = 90$ MHz for Si(333), $\nu_{th} = 98$ MHz for Si(444) and $\nu_{th} = 127$ MHz for Si(440) [3]).

Separation of the elastic and inelastic scattered γ-rays was achieved by using the Mössbauer technique. A 130 mCi ^{57}Co source diffused into a matrix of Rh was used together with an Fe–Al alloy absorber.

Typical Mössbauer diffractional spectra are presented in figure 1; the central line of the spectrum corresponds to the elastic scattering of γ quanta while the satellites correspond to non-elastic scattering with the absorption and emission of the one or two phonons.

Further, measurements of the two intensities transmitted through the absorber, namely I_∞ (source and absorber are far from resonance) and I_R (in resonance), were performed as functions of voltage V on the piezocrystal (the US amplitude is proportional to V). As the source had a negligible isomer shift relative to the absorber, then the measurement of I_R took place at zero velocity. To obtain the 'out-of-resonance' condition correctly, I_∞ was measured without the absorber, but there was a correction for the intensity for non-resonant attenuation in this absorber. The difference $\Delta I = I_\infty - I_R$ is proportional to pure elastic scattering. The normalized intensities of the elastic scattering for Si under US excitation are shown in figure 2, curves A.

Then, the samples were mechanically distorted by polishing with a 20 μm abrasive. The rocking curves for these samples were broadened by more than a hundred times; so the crystals can be regarded as mosaic type. The results of measurements for these crystals are shown in figure 2, curves B.

Additionally, for perfect and mosaic Si crystals the integral intensities of the reflection under US excitation were obtained as follows:

$$\Delta I_\infty(V) = [I_\infty(V) - I_\infty(0)]/[I_\infty(0) - I_{bg}]$$

where I_{bg} is the intensity of the background, measured near the Bragg angle.

The total intensity I_∞ increased monotonically for perfect crystals (figure 2, curves C) while, for mosaic crystals, I_∞ does not depend on the US amplitude.

3. Discussion

The behaviour of the total intensity in our experiment is in full agreement with x-ray [2] and Mössbauer [3] experiments. In a perfect Si crystal the total intensity increased in accordance with equation (2), which means a linear dependence on u at small values of Hu.

The elastic part of a diffracted beam decreases in proportion to the US amplitude ($u \sim V$) and the magnitude H of the diffraction vector.

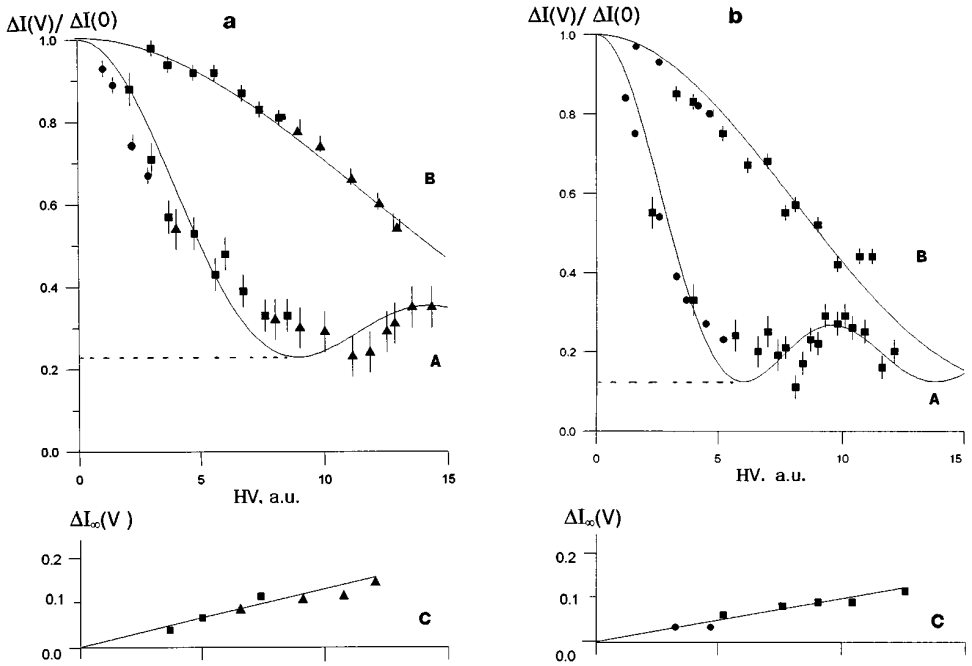

Figure 2. The intensity of elastic scattering of γ quanta in perfect (curves A) and mosaic (curves B) crystals of silicon under excitation by a transverse US wave. (a) Si(111) sample, US frequency of 165 MHz, (111) (●), (333) (■) and (444) (▲) reflections; (b) Si(110) sample, US frequency of 182 MHz, (220) (●) and (440) (■) reflections. The integral intensity of the reflections in a perfect Si crystal is also shown in curves C.

Gavrilov *et al* [3] observed the above-mentioned anomalous Debye–Waller factor for (333) and (444) reflection at $\nu_{th}/\nu_s = 0.68$ and 0.74, respectively ($\nu_s = 132.5$ MHz).

Our experiments were carried out under the following conditions: $\nu_{th}/\nu_s = 0.64$ and 0.69, for the (333) and (444) reflections at $\nu_s = 142$ MHz, and $\nu_{th}/\nu_s = 0.54$ and 0.59 at $\nu_s = 165$ MHz. For the (440) reflection, $\nu_{th}/\nu_s = 0.80$ at $\nu_s = 158$ MHz and $\nu_{th}/\nu_s = 0.70$ at $\nu_s = 180$ MHz. The 'anomalous' Debye–Waller factor was not observed in our experiments. It should be pointed out that there is no qualitative distinction between the elastic scattering under US excitation above the threshold frequency and that below the threshold frequency. For reflections (111) and (220) in figure 2, $\nu_{th}/\nu_s \approx 1 (\nu_{th}(111) = 149$ MHz; $\nu_{th}(220) = 170$ MHz).

The comparison between the results for the perfect and the mosaic crystal for the same US amplitude on the surface suggests that in a perfect crystal the elastic intensity decreases more strongly, highlighting the very important role of multiple coherent inelastic scattering in perfect Si crystals. However, we do not know whether a theory of this phenomenon exists.

In [5] it was found that the intensities of the central line and satellite lines depend upon the US amplitude as $J_n^2(VK)$, where K is the wavevector of the γ quantum. Therefore the intensity of the central line must oscillate with varying US amplitude and at certain values of VK the central line must vanish completely. In figure 2 the solid line corresponds to $J_0^2(HV)$, and the broken line corresponds to the background level, because the area of the

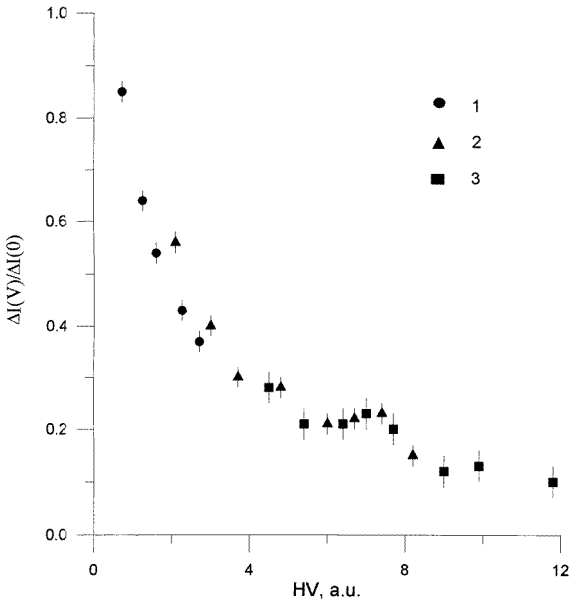

Figure 3. The intensity of elastic scattering of γ quanta in a perfect silicon crystal under excitation by a longitudinal US wave at 50 MHz: ●, (111) reflection; ▲, (333) reflection; ■, (444) reflection.

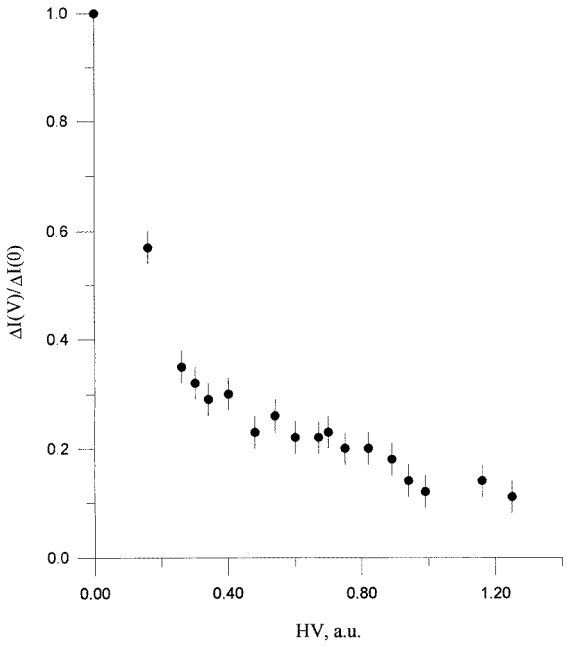

Figure 4. The intensity of elastic scattering of γ quanta in a lithium niobate crystal under the excitation by a surface acoustic wave at 40 MHz for the (030) reflection (a.u., arbitrary units).

US field is not equal to area of the diffractional field on the surface of the sample. $J_0^2(HV)$ somewhat roughly describes our experimental data. However, our experiment cannot be

interpreted in terms of the theoretical approaches of [3]. In that paper the oscillation of the elastic intensity was not observed owing to an insufficiently strong US amplitude because the piezocrystal was excited at frequencies of the third and the fifth harmonic.

Additionally, we have also carried out a diffraction experiment in the Bragg case with longitudinal US waves on a perfect Si crystal (figure 3) and measured the diffraction of γ quanta in a $LiNbO_3$ crystal, under the action of a standing surface acoustic wave (figure 4). One can see that the oscillations of the elastic scattered intensity are similar for transverse and longitudinal US waves and for surface acoustic waves as well.

Acknowledgments

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