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The Instrumental Broadening Function of the Bartels Five-Crystal X-ray Diffractometer

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Abstract

A simple closed-form expression has been derived for the instrumental broadening function of the general Bartels five-crystal diffractometer. The use of this result allows the extraction of sample rocking-curve widths from the measured widths. Such use facilitates the determination of dislocation densities in heteroepitaxic crystals by the measurement of several *hkl* rocking curves using a Bartels five-crystal diffractometer.

1. Introduction

The five-crystal diffractometer described by Bartels (1983*a*,*b*) has become an important tool for the characterization of heteroepitaxic semiconductor materials. The utility of the Bartels five-crystal arrangement results from the small divergence and wavelength spread of the four-crystal monochromator. This monochromator arrangement was first proposed by DuMond (1937) and initial prototypes of the monochromator were demonstrated by Beaumont & Hart (1974) using silicon crystals and synchrotron radiation. Bartels (1983*a*,*b*) subsequently developed a four-crystal monochromator using germanium crystals and a Cu X-ray tube as the source, which has led to the popular use of the instrument.

Slusky & Macrander (1987), van der Sluis (1994) and Möller (1994) have described the instrumental function for the Bartels five-crystal diffractometer. The results of their work can be applied to predict the shapes and widths of diffraction profiles measured with the five-crystal diffractometer if the specimen diffraction profile is known. The published results make possible the assessment of the resolving power of the five-crystal arrangement and also allow predictions of the shapes of diffraction profiles in the tails. On the other hand, the previously published body of theory does not allow direct determination of crystal rocking-curve widths from the measured profiles.

An important application of the Bartels five-crystal X-ray diffractometer is the determination of threading dislocation densities in mismatched heteroepitaxic layers (Ayers, 1994). This application requires the determination of the specimen-crystal rocking-curve widths for a number of different *hkl* reflections. In the present paper, we have derived a simple closed-form expression for the instrumental broadening function of the Bartels five-crystal X-ray diffractometer. The use of this result allows specimen rocking-curve widths to be extracted readily from profile widths measured with the five-crystal instrument.

2. Theory

Here we have derived the instrumental broadening function for the Bartels five-crystal X-ray diffractometer based on a geometrical theory and the following assumptions:

(*a*) the monochromator reflections are from four crystals of identical Bragg angle and occur in a single plane;

(*b*) the natural wavelength spread of the X-ray source used is 'large' compared to the wavelength spread exiting the monochromator (it is shown in Appendix *A* that this is a good assumption as long as the natural wavelength spread of the X-ray source is at least three times larger than the wavelength spread exiting the monochromator);

(*c*) the reflections are specular (*i.e.* the angle of reflection equals the angle of incidence);

(*d*) the diffraction profiles for the crystals used in the monochromator are Gaussian in shape.

The geometry of the Bartels five-crystal diffractometer is shown in Figs. 1 and 2. θ_{Bm} is the Bragg angle for the monochromator reflections, d_m is the spacing of the diffracting planes of the monochromator crystals and λ is the center wavelength of the X-ray line used. The unit vectors normal to the four monochromator crystals are given by

$$\begin{aligned} \mathbf{n}_1 &= (-\sin\theta_{Bm}, \cos\theta_{Bm}, 0), \\ \mathbf{n}_2 &= (\sin\theta_{Bm}, -\cos\theta_{Bm}, 0), \\ \mathbf{n}_3 &= (-\sin\theta_{Bm}, -\cos\theta_{Bm}, 0), \\ \mathbf{n}_4 &= (\sin\theta_{Bm}, \cos\theta_{Bm}, 0). \end{aligned} \tag{1}$$

The unit vectors in the directions of the X-ray beam (center wavelength) incident on the five crystals are given by

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$$\begin{aligned}\mathbf{r}_1 &= \mathbf{r}_3 = \mathbf{r}_5 = (1, 0, 0),\\ \mathbf{r}_2 &= (\cos[2\theta_{Bm}], \sin[2\theta_{Bm}], 0),\\ \mathbf{r}_4 &= (\cos[2\theta_{Bm}], -\sin[2\theta_{Bm}], 0).\end{aligned} \quad (2)$$

The wavelength spread of the four-crystal monochromator may be determined as follows: The diffraction profiles of the monochromator crystals are treated approximately as Gaussian profiles, given by

$$I(\theta) \propto \exp\{[-4\ln 2(\theta - \theta_{Bm})^2]/\beta_m^2\}, \quad (3)$$

where θ is the angle between the incident beam and the diffracting planes of the monochromator crystal and β_m is the full width at half-maximum (FWHM) of the monochromator crystal reflections. A wavelength $\lambda + \Delta\lambda/2$ is attenuated by the four successive reflections so that its intensity relative to that of the center wavelength is given by

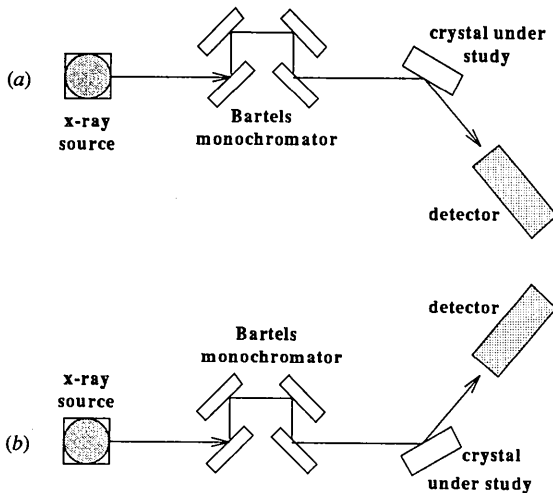

Fig. 1. Schematic of the Bartels five-crystal diffractometer, showing the two experimental configurations. (a) (+, −, −, +, −) configuration. (b) (+, −, −, +, +) configuration.

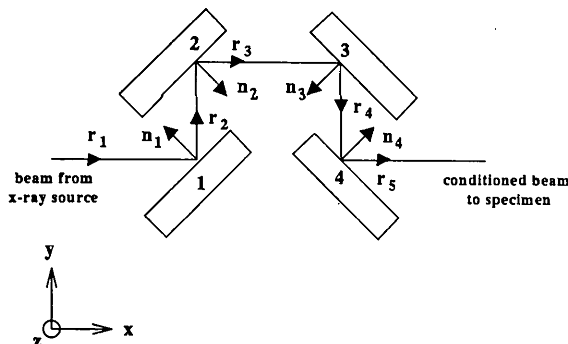

Fig. 2. Geometry of the Bartels four-crystal monochromator.

$$I(\lambda + \Delta\lambda/2)/I(\lambda) = \exp\{[-4\ln 2(\Delta\theta_{Bm})^2]/\beta_m^2\}, \quad (4)$$

where

$$\Delta\theta_{Bm} = \Delta\lambda/2d_m \cos\theta_{Bm}. \quad (5)$$

Hence, the wavelength FWHM exiting the monochromator is given by

$$\Delta\lambda_m = \beta_m d_m \cos\theta_{Bm}. \quad (6)$$

The horizontal divergence of the four-crystal monochromator may be determined as follows: Consider a ray diverging from the central ray by an angle $\Delta\theta/2$. Then, after four successive reflections, the intensity associated with this ray, relative to the central ray, is

$$I(\theta_{Bm} + \Delta\theta/2)/I(\theta_{Bm}) = \exp\{[-4\ln 2(\Delta\theta)^2]/\beta_m^2\}. \quad (7)$$

The horizontal divergence of the four-crystal monochromator is thus given by

$$\Delta\theta = \beta_m/2. \quad (8)$$

The vertical divergence of the four-crystal monochromator is determined as follows. Consider a diverging ray incident on the monochromator, with direction $\mathbf{r}_1 = (\cos[\Delta\varphi/2], 0, \sin[\Delta\varphi/2])$. The angle of incidence for this ray differs from θ_{Bm} by $\Delta\theta$, which may be determined by vector analysis:

$$\mathbf{r}_1 \cdot \mathbf{n}_1 = -\cos(\Delta\varphi/2)\sin\theta_{Bm} = -\sin(\theta_{Bm} + \Delta\theta). \quad (9)$$

Therefore,

$$\Delta\theta = \sin^{-1}[\cos(\Delta\varphi/2)\sin\theta_{Bm}] - \theta_{Bm}. \quad (10)$$

If the monochromator crystals have intrinsic rocking curves that are approximately Gaussian, then after four successive reflections the intensity of the divergent beam relative to the non-divergent beam is given by

$$\begin{aligned}&I(\theta_{Bm}, \Delta\varphi/2)/I(\theta_{Bm}, 0)\\ &\quad = \exp(-16\ln 2\{\sin^{-1}[\cos(\Delta\varphi/2)\sin\theta_{Bm}]\\ &\qquad - \theta_{Bm}\}^2/\beta_m^2).\end{aligned} \quad (11)$$

The vertical divergence of the monochromator (FWHM) may be determined readily from (11) by setting the normalized intensity equal to one half and solving for $\Delta\varphi$. The result is

$$\Delta\varphi = 2\cos^{-1}[\sin(\theta_{Bm} - \beta_m/4)/\sin\theta_{Bm}]. \quad (12)$$

The widths of the diffraction profiles measured using the five-crystal diffractometer are given approximately by

$$\beta^2_{\text{measured}} = \beta^2_{\text{specimen}} + \beta^2_{\text{instrument}}. \quad (13)$$

We have determined the instrumental broadening function $\beta^2_{\text{instrument}}$ with the assumptions that:

(a) as long as the wavelength spread, horizontal divergence and vertical divergence are small, they can be considered to act independently;

(*b*) each of the three effects broadens the measured diffraction profile in the same manner as convolution with a Gaussian profile.

Then,

$$\beta^2_{\rm instrument} = \beta^2_{\Delta\lambda} + \beta^2_{\Delta\theta} + \beta^2_{\Delta\varphi}, \quad (14)$$

where $\beta^2_{\Delta\lambda}$, $\beta^2_{\Delta\theta}$ and $\beta^2_{\Delta\varphi}$ are the instrumental broadening functions associated with the wavelength spread, horizontal divergence and vertical divergence of the monochromator, respectively.

The instrumental broadening function associated with the wavelength spread may be determined from the differential form of the Bragg equation:

$$\beta^2_{\Delta\lambda} = (\Delta\lambda/\lambda)^2 \tan^2\theta_{Bs} = (\beta_m/2\tan\theta_{Bm})^2 \tan^2\theta_{Bs}, \quad (15)$$

where θ_{Bs} is the specimen Bragg angle.

The instrumental broadening function associated with the horizontal divergence is readily determined to be

$$\beta^2_{\Delta\theta} = (\beta_m/2)^2 \quad (16)$$

because the horizontal divergence is in the plane of the specimen diffraction vector.

The instrumental broadening function associated with the vertical divergence may be determined as follows. We will provide the details of the analysis for the $(+,-,-,+,-)$ geometry, but the result is identical for the other configuration. The non-diverging central ray of the X-ray beam incident on the specimen has a direction $\mathbf{r}_5 = (1,0,0)$. Consider a diverging ray with a direction $\mathbf{r}'_5 = (\cos[\Delta\varphi/2], 0, \sin[\Delta\varphi/2])$. With the Bragg condition imposed,

$$\mathbf{r}'_5 \cdot \mathbf{n}'_5 = -\sin\theta_{Bs}. \quad (17)$$

The angle between the diverging ray and the diffracting planes of the specimen, projected into the plane of the diffractometer, is greater than θ_{Bs} by an amount $\Delta\theta$ given by

$$\Delta\theta = \sin^{-1}[\sin\theta_{Bs}/\cos(\Delta\varphi/2)] - \theta_{Bs}. \quad (18)$$

The instrumental broadening function associated with the vertical divergence is

$$\begin{aligned}\beta^2_{\Delta\varphi} &= \{\sin^{-1}[\sin\theta_{Bs}/\cos(\Delta\varphi/2)] - \theta_{Bs}\}^2 \\ &= \{\sin^{-1}[\sin\theta_{Bs}\sin\theta_{Bm}/\sin(\theta_{Bm} - \beta_m/4)] - \theta_{Bs}\}^2.\end{aligned} \quad (19)$$

Combining the above results gives the overall instrumental broadening function:

$$\beta^2_{\rm instrument} \simeq (\beta_m/2\tan\theta_{Bm})^2\tan^2\theta_{Bs} + (\beta_m/2)^2 + \left\{\sin^{-1}\left[\frac{\sin\theta_{Bs}\sin\theta_{Bm}}{\sin(\theta_{Bm}-\beta_m/4)}\right] - \theta_{Bs}\right\}^2. \quad (20)$$

3. Experimental

For comparison with the theoretical results, the instrumental broadening function was experimentally determined for a Bartels five-crystal diffractometer (Blake Industries) employing a Ge (022) monochromator used in the $(+,-,-,+)$ configuration at a temperature of 293 K and using the Cu $K\alpha_1$ X-ray line ($\lambda = 1.540594$ Å). The Philips Cu X-ray source was operated at 30 kV and 10 mA. The line-focused beam was slit-limited to 5 mm length normal to the plane of the diffractometer and 0.5 mm width in the plane of the diffractometer by pairs of slits placed on either side of the four-crystal monochromator. The spacing between the slits was 210 mm. The specimen used for all measurements was a semi-insulating GaAs (001) crystal, 375 µm thick, cut from a 75 mm wafer supplied by Sumitomo. The manufacturer specified an etch-pit density of less than 10^4 cm^{-2} for the wafer. Diffraction profiles were obtained for the 113, 004, 115, 335, 444 and 117 specimen reflections with both the $(+,-,-,+,-)$ and $(+,-,-,+,+)$ geometries. For the asymmetric reflections, the larger angle of incidence was used. A count time of 2 s was used and typical peak intensities measured with a Bicron scintillation counter were 1750 counts s^{-1} for the GaAs 004 reflection and 600 counts s^{-1} for the GaAs 117 reflection The FWHM's for the measured diffraction profiles were determined by least-squares fitting to a Gaussian profile. The instrumental broadening function for the diffractometer was determined by accounting for the natural rocking-curve width for the GaAs specimen:

$$\beta^2_{\rm instrument} = \beta^2_{\rm measured} - \beta^2_i. \quad (21)$$

The GaAs crystal was assumed to behave as a perfect crystal and the values for the intrinsic rocking-curve width β_i for the GaAs specimen were obtained from standard tables for semiconductor X-ray characterization (Ayers, 1990).

4. Results and discussion

Fig. 3 shows four representative diffraction profiles that were obtained experimentally for the 004, 115, 444 and 117 reflections with the $(+,-,-,+,-)$ geometry. In each case, the black squares represent the experimental data and the solid curves represent the Gaussian best fit to the data. For the 004 profile shown, the peak height was 5629 counts and the root mean square error obtained with the best-fit Gaussian curve was only 3.0 counts. For the 117 profile, the peak height was 1239 counts and the root mean square error obtained with the best-fit Gaussian curve was 1.44 counts. All of the diffraction profiles obtained in this study, for both $(+,-,-,+,-)$ and $(+,-,-,+,+)$ geometries, exhibit the Gaussian shape. This is expected after convolution of the intrinsic rocking curve of the sample with the instrumental

distributions. The fact that the diffraction profiles are all Gaussian is key to the validity of the arguments made earlier in this paper. The remaining data are expressed in terms of diffraction-profile widths, which were determined by fitting the measured profiles to Gaussian curves as shown in Fig. 3.

Fig. 4 shows the calculated (solid line) and measured (squares) instrumental broadening functions for the Bartels five-crystal diffractometer used in this work. The theoretical broadening function was calculated for the case of a Ge (022) monochromator used with Cu $K\alpha_1$ radiation at 293 K from (20) using $\lambda = 1.540594$ Å, $d = 2.0002$ Å, $\beta_m = 12''$ and $\theta_{Bm} = 22.65°$. For this case, the calculated values for the divergence are $6''$ (horizontal) and 0.96° (vertical). The calculated wavelength spread is 1.07×10^{-4} Å. This is 23% of the natural width for the Cu $K\alpha_1$ line (Ayers & Ladell, 1988). If an adjustment is made to account for the effect of the natural width of the Cu $K\alpha_1$ line (see Appendix *A*), then it is found that the actual value of the wavelength spread is reduced by 4%.

The instrumental broadening function for the Ge (022) four-crystal monochromator using Cu $K\alpha_1$ radiation calculated from the present theory is

$$\beta^2_{\rm instrument} = 36('')^2 + 223('')^2 \tan^2 \theta_{Bs}. \qquad (22)$$

From Fig. 4, it can be seen that there is good agreement between the calculated and experimentally determined instrumental broadening functions. There was no significant difference between the widths measured in the $(+,-,-,+,-)$ and $(+,-,-,+,+)$ configurations. This shows that the Bartels $(+,-,-,+)$ monochromator

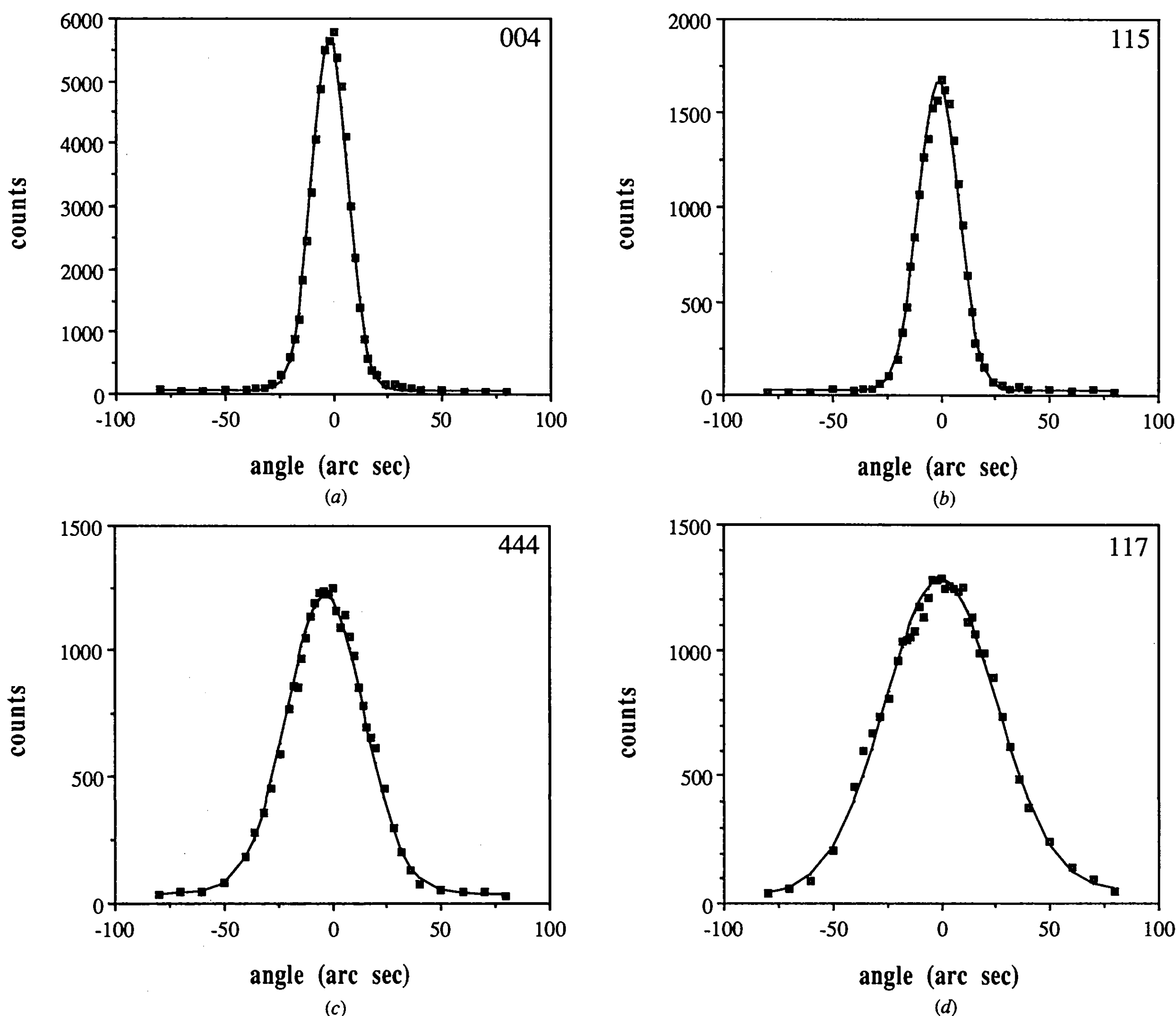

Fig. 3. Measured diffraction profiles for the 004, 115, 444, and 117 reflections from the GaAs (001) specimen using the $(+,-,-,+,-)$ configuration. The black squares indicate experimental data and solid curves indicate the best-fitting Gaussian profile in each case.

configuration effectively eliminates the interaction between the horizontal divergence and the wavelength spread of the monochromator.

5. Conclusions

We have derived the instrumental broadening function for the Bartels five-crystal diffractometer using the $(+, -, -, +, -)$ or $(+, -, -, +, +)$ configurations based on a geometric theory and the simplifying assumption of Gaussian profiles. The present theory contrasts with the previous theoretical work, which predicted the shapes of diffraction profiles measured with the Bartels five-crystal diffractometer for crystals with known rocking curves. Unlike the previously published work, the present theory allows extraction of the rocking-curve widths for crystals of unknown perfection from the widths of profiles measured with the Bartels five-crystal diffractometer. This is of importance for the application of five-crystal measurements to the determination of threading dislocation densities in heteroepitaxic semiconductors.

The theory developed here is general enough that it can be applied to Bartels five-crystal diffractometers with virtually any combination of X-ray source and monochromator reflections. The theory does not apply to the five-crystal diffractometers with the $(+, -, +, -, +)$ or $(+, -, +, -, -)$ geometries because these instruments exhibit important dispersive effects (*i.e.* interactions between wavelength and angle for the conditioned beam).

We have compared the theoretical result with the experimentally determined instrumental broadening function for a Bartels-type five-crystal diffractometer employing Ge(022) monochromator reflections and the Cu $K\alpha_1$ line. There is good agreement between the calculated and experimental results over a wide range of specimen Bragg angle for which $0.5 \leq \tan^2\theta_{Bs} \leq 16.0$.

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APPENDIX A
The influence of the natural width of the X-ray line

The wavelength spread of the four-crystal monochromator was calculated based on the assumption that the natural wavelength spread of the X-ray line is large compared to the wavelength spread exiting the monochromator. In cases where this is not true, the actual wavelength spread exiting the monochromator is reduced below that predicted by (6).

We have analyzed this situation based on the following assumptions:

(*a*) that the natural spectrum of the X-ray line is Lorentzian in shape and may be characterized by its FWHM, $\Delta\lambda_{\rm natural}$ (see Bearden & Shaw, 1935);

(*b*) that the natural spectrum of the X-ray line gives rise to coloring of the spectrum exiting the monochromator, which may be described by the simple multiplication of the (Lorentzian) natural spectrum and the (Gaussian) wavelength spectrum for the monochromator.

Then, the intensity at a wavelength $\lambda + \Delta\lambda/2$ relative to the intensity of the center wavelength λ is

$$I(\lambda + \Delta\lambda/2)/I(\lambda) = [1 + (\Delta\lambda/\Delta\lambda_{\rm natural})^2]^{-1} \times \exp[-4\ln 2(\Delta\lambda)^2/\Delta\lambda_m^2]. \quad (23)$$

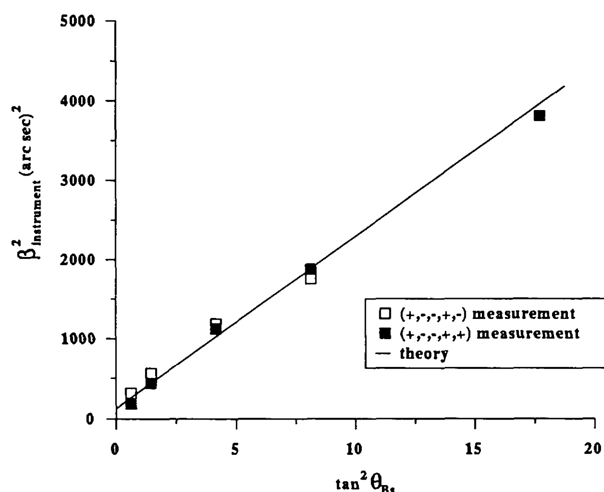

Fig. 4. The instrumental broadening function for the Bartels diffractometer employing a Ge(022) monochromator and Cu $K\alpha_1$ radiation. The calculated result is shown as a solid line; experimental values are shown as squares.

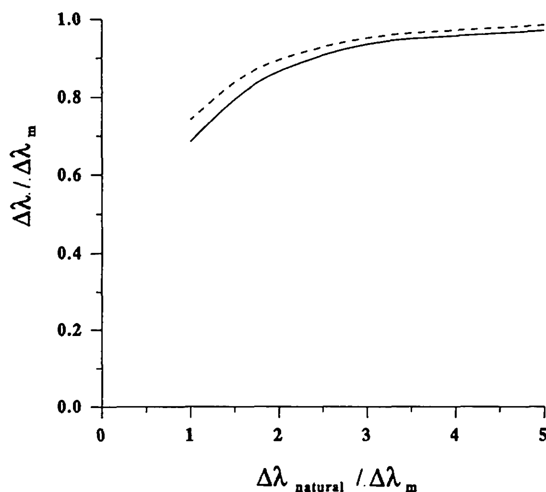

Fig. 5. The normalized wavelength spread $(\Delta\lambda/\Delta\lambda_{\rm monochromator})$ *versus* the normalized natural wavelength spread of the X-ray source $(\Delta\lambda_{\rm natural}/\Delta\lambda_{\rm monochromator})$. The solid curve was determined based on a Lorenzian shape for the natural spectrum and the dashed curve was determined based on a Gaussian shape for the natural spectrum.

The FWHM of the wavelength distribution was determined by setting (23) to 0.5 and solving numerically.

Fig. 5 shows the normalized wavelength spread *versus* the normalized wavelength spread of the X-ray source as a solid line. Also shown for comparison is the result predicted based on a natural spectrum with a Gaussian shape (dashed curve) for which an analytic solution was obtained:

$$\Delta\lambda = [(1/\Delta\lambda_{\text{natural}})^2 + (1/\Delta\lambda_m)^2]^{-1/2}. \quad (24)$$

Several practical points should be made regarding these results:

(*a*) although the actual spectra of natural X-ray lines are asymmetric and cannot be described exactly by a simple Lorentzian or Gaussian profile (Berger, 1986), the results are insensitive to the exact shape of the natural spectrum so that negligible errors result from the use of the Lorentzian approximation;

(*b*) values of $\Delta\lambda/\Delta\lambda_m$ calculated based on (23) are in error by less than 5% as long as $\Delta\lambda_{\text{natural}}/\Delta\lambda_m > 3.0$;

(*c*) values of $\Delta\lambda/\Delta\lambda_m$ calculated based on (24) are in error by less than 5% as long as $\Delta\lambda_{\text{natural}}/\Delta\lambda_m > 3.6$.

Therefore, corrections to (6) are not usually necessary as long as the natural wavelength spread of the X-ray source is greater than three times the wavelength spread predicted by (6).

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