Spectral widths of the Cu $K\alpha$ lines

John Ayers and Joshua Ladell
Philips Laboratories, North American Philips Corporation, Briarcliff Manor, New York 10510
(Received 26 August 1987)

The spectral widths of the Cu $K\alpha_1$ and Cu $K\alpha_2$ x-ray lines have been determined from a series of x-ray diffraction profiles. All profiles were obtained using a two-crystal Ge (111) monochrocollimator and a single (100) GaAs study crystal. The dispersion of the monochrocollimator was determined from the measured separation of the $K\alpha_1$ and $K\alpha_2$ peaks. The known dispersion was then utilized to calculate the natural widths of the Cu x-ray lines from the observed diffraction profile widths. The Cu $K\alpha_1$ and $K\alpha_2$ lines have full widths at half maximum of 0.000 461(9) and 0.000 61(4) Å, respectively.

I. INTRODUCTION

X-ray diffraction profiles are convolutions of the incident-x-ray spectral distribution, instrumental broadening distribution, and the intrinsic diffraction profile of the study crystal. The spectral and instrumental effects combine to broaden and shift x-ray diffraction peaks.[1] It is therefore important to know the spectral distribution of the incident radiation when interpreting the results of diffraction experiments.

Ladell, Parish, and Taylor[2] derived a model for the Cu $K\alpha$ doublet from the x-ray diffraction data of Bearden and Shaw.[3] The true widths of the $K\alpha$ lines were determined from the observed widths of diffraction peaks using the relationship of Parratt,[4]

$$W_T = W_0 - 2.9(W_c)^{1.7} , \quad (1)$$

where W_T is the true width, W_0 is the observed width in the $(+,+)$ antiparallel position, and W_c is the rocking-curve width of the crystal. The widths thus obtained were 0.000 472 0 and 0.000 635 9 Å for the Cu $K\alpha_1$ and Cu $K\alpha_2$ lines, respectively. These values, however, should be considered upper bounds for the linewidths. If there were any residual instrumental aberrations in the reduced profiles of Bearden and Shaw,[3] or if the calcite crystals used were somewhat imperfect, then the spectral widths could have been overestimated.

Recently, Berger[5] modeled the Cu $K\alpha$ spectrum using four Cauchy functions. The spectral distribution he measured using a single [444] Si reflection resulted in linewidth of 0.000 461 and 0.000 653 Å for the $K\alpha_1$ and $K\alpha_2$, respectively. These results were obtained neglecting the broadening effects caused by the vertical divergence of the incident beam.

Citrin *et al.*[6] determined the Cu $K\alpha$ linewidths from diffraction profiles to be 0.004 574 and 0.000 556 Å for $K\alpha_1$ and $K\alpha_2$, respectively. These linewidths are significantly different from those determined by Bearden and Shaw[3,7] and Berger.[5]

As part of a program to characterize epitaxially prepared structures by x-ray diffractometry, we developed a novel method for x-ray linewidth determination and applied it to the determination of the Cu $K\alpha$ doublet linewidths. In our procedure we use a double crystal germanium (111) monochrocollimator which provides a source of almost parallel incident radiation on the study crystal. This effectively reduces broadening of the observed line profile due to the divergence of x rays of the same wavelength coming from an extended source. The angle-scale representation of the spectral profile is determined by the dispersion effected by the monochrocollimator. Since two monochromating crystals are involved in the dispersion, the usual law of dispersion for a single crystal is inapplicable. A theoretical derivation of the appropriate dispersion law has been determined.[8] For our purposes here, however, the dispersion due to the use of our monochrocollimator has been established empirically from the measurement of line profiles of the $K\alpha_1, K\alpha_2$ doublet for a series of increasing orders of (hkl) reflections. The measured full width at half maximum (FWHM) of each of the various orders of both the $K\alpha_1$ and $K\alpha_2$ were then plotted as a function of $\tan\theta$. The ratio of the slope of the FWHM versus $\tan\theta$ to the slope of doublet separation versus $\tan\theta$ provides a measure of the sought spectral widths.

II. EXPERIMENTAL

X-ray diffraction profiles were obtained for a single (001) GaAs crystal using the biaxial diffractometer described elsewhere.[9] A Rigaku Denke rotating anode generator having a projected focal area of 0.35 mm^2 was operated at 50 kV and 100 mA. The x-ray beam was conditioned by a two-crystal Ge [111] monochrocollimator.[8,9] The first Ge crystal face was tilted 30° down from the vertical and the second Ge crystal face was tilted 30° up from the vertical, to achieve collimation in all directions. The angular divergence was limited to approximately 30 arc sec. Thirteen reflections were observed in both the $(+,+)$ and $(+,-)$ settings.[10] These include the (002), (113), (004), (224), (115), (404), (315), (006), (026), (335), (444), (117), and (155). In the case of asymmetric reflections, where the diffracting planes were inclined to the crystal surface by an angle ϕ, both the $(\theta+\phi)$ and $(\theta-\phi)$ profiles were examined. The

diffraction vector of the study crystal was oriented to be normal to the axis of crystal rotation, to within 0.002° to avoid geometrical broadening.

III. ANALYSIS AND RESULTS

When a single-crystal monochromator is used dispersion of the $K\alpha_1$-$K\alpha_2$ doublet separation can be described approximately by the linear relationship

$$\Delta\theta=\frac{\Delta\lambda}{\lambda}\left[\frac{180}{\pi}\right]\tan\theta\pm\sigma\left[\frac{\Delta\lambda}{\lambda}\right], \tag{2}$$

where $\Delta\theta$ is the observed angular separation between the $K\alpha_1$ and $K\alpha_2$ peaks, $\Delta\lambda$ is the difference in the characteristic wavelengths, and σ is the angular separation of the $K\alpha_1$ and $K\alpha_2$ incident on the study crystal. Expression (2) is valid where $\Delta\lambda$ is small and the characteristic line shapes are symmetric and unaffected by instrumental and crystal diffraction broadening effects. Some departure from (2) should be expected in the real case. The angular separation between the characteristic Cu $K\alpha$ peaks was measured and plotted as a function of $\tan(\theta)$ for the $(+,+)$ and $(+,-)$ configurations (Fig. 1). We found that the dispersion can be described by the linear relationship

$$\Delta\theta=C_1\frac{\Delta\lambda}{\lambda}\frac{180}{\pi}\tan\theta\pm C_2\left[\frac{\Delta\lambda}{\lambda}\right]+C_3\ . \tag{3}$$

The coefficients C_1,C_2,C_3, determined from a least-squares analysis of the line profile data, are reported in Table I.

The two values of the coefficient C_1 are essentially unity as expected in Eq. (2). The coefficient C_2 was determined from the average separation of the experimental $(+,+)$ and $(+,-)$ lines. This experimental value is the projection of σ in the plane of diffraction and agrees with the theoretical value[8] which predicts the effective d spacing of 1.9036 Å for the two crystal germanium (111) monochrocollimator.

TABLE I. Empirical dispersion coefficients (in deg).

	$(+,-)$ Configuration	$(+,+)$ Configuration
X_1	0.997	1.004
C_2	26.3	26.3
C_3	0.0	−0.002

The FWHM of the Cu $K\alpha_1$ and Cu $K\alpha_2$ diffraction profiles were measured and plotted as a function of $\tan(\theta)$ for the $(+,+)$ and $(+,-)$ configurations. The $K\alpha_1$ data are plotted in Fig. 2(a) and the $K\alpha_2$ data are plotted in Fig. 2(b), respectively. Let w_1 and w_2 represent the spectral widths in angstroms for the Cu $K\alpha_1$ and Cu $K\alpha_2$

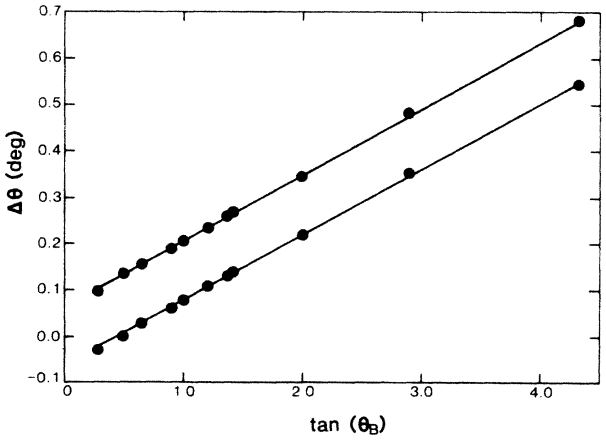

FIG. 1. $K\alpha_1/K\alpha_2$ dispersion relationship for GaAs reflections. $\Delta\theta$ is the angular separation between the $K\alpha_1$ and $K\alpha_2$ peaks. The upper line and lower line were obtained in the $(+,+)$ and $(+,-)$ settings, respectively. The two lines have a slope of 0.1460° and a separation of 0.1289°.

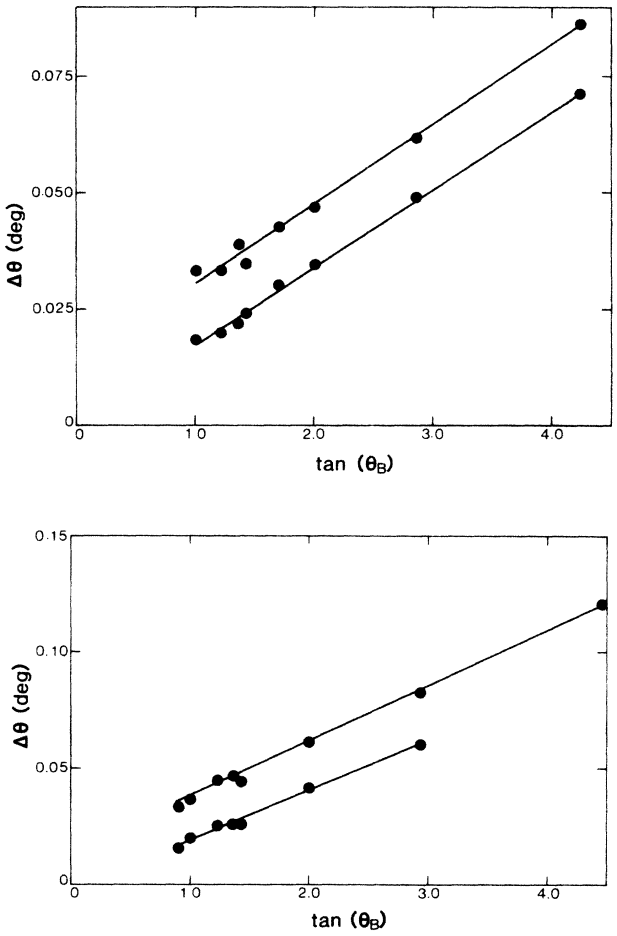

FIG. 2. (a) $K\alpha_1$ dispersion relationship for GaAs reflections. $\Delta\theta$ is the full width at half maximum of the $K\alpha_1$ peak. The upper line and lower line were obtained in the $(+,+)$ and $(+,-)$ settings, respectively. The two lines have a slope of 0.0167° and a separation of 0.0147°. (b) $K\alpha_2$ dispersion relationship for GaAs reflections. $\Delta\theta$ is the full width at half maximum of the $K\alpha_2$ peak. The upper line and lower line were obtained in the $(+,+)$ and $(+,-)$ settings, respectively. The two lines have a slope of 0.0225° and a separation of 0.0199°.

TABLE II. Widths of the Cu $K\alpha$ lines.

Experimental configuration	W_1 (Å)	W_2 (Å)
(+,−)	0.000 453	0.000 584
(+,+)	0.000 470	0.000 638
Average	0.000 461(9)	0.000 61(4)
From C_2	0.000 403	0.000 636

lines, respectively. Similar to expression (3) found for the doublet separation, the linear dependence of linewidth versus $\tan(\theta)$ can be expressed by (4)

$$\Delta\theta = C_1 \frac{180}{\pi} \frac{w_i}{\lambda_j} \tan\theta \pm C_2 \left[\frac{w_j}{\lambda_j} \right] + W(hkl) + C_4 \ , \quad (4)$$

where $j=1$ refers to the $K\alpha_1$ and $j=2$ refers to the $K\alpha_2$ lines, respectively, $W(hkl)$ is the rocking-curve width of the relevant study crystal (hkl) reflection and C_4 the instrumental aberrational widths which add to the angle-scale spectral width of the diffraction profiles.

The dispersion relationships for the individual peak widths consist of two straight lines (Fig. 2), one for the (+,+) case and one for the (+,−) case. Using the known values of C_1 and C_2 for the instrument, the Cu $K\alpha$ linewidths may be calculated either from the slopes or the separation of the two lines. This assumes that the dispersion of the monochrocollimator is linear over the range of wavelengths being considered. The geometrical theory of the skewed crystal monochrocollimator[8] has been derived and verified experimentally. This theory establishes that the dispersion is linear to within 0.1% for the $K\alpha_1$-$K\alpha_2$ separation range.

The Cu $K\alpha$ spectral widths were calculated from the slopes of the dispersion relationships as follows. Each set of data was fit using the least-squares method. For any set of reflections, the slope of the dispersion relationship S is $\Delta(\text{FWHM})/\Delta(\tan\theta)$, where FWHM is the diffraction profile width. The FWHM of the ith spectral line W_i is

$$W_i = \frac{S\lambda\pi}{C_1 180} \ . \quad (5)$$

The spectral widths of the characteristic lines were calculated two ways using the two slopes of the FWHM lines obtained from the (+,+) and (+,−) plots of Figs. 2(a) and 2(b). The Cu $K\alpha$ spectral widths were also calculated from the separation of the (+,−) and (+,+) dispersion lines using the measured value of C_2 and the relationship

$$W_i = \frac{2\sigma\lambda}{C_2} \ , \quad (6)$$

where 2σ is the separation of the dispersion lines.

The spectral widths obtained are given in Table II. The widths obtained from the slopes are considered more accurate.

TABLE III. Reported widths of the Cu $K\alpha$ lines.

	Cu $K\alpha_1$ (Å)	Cu $K\alpha_2$ (Å)
Bearden and Shaw (1935)[a]	0.000 472	0.000 636
Citrin *et al.* (1974)	0.000 457(19)	0.000 556(19)
Berger (1986)	0.000 461(7)	0.000 653(7)
This work	0.000 461(9)	0.000 61(4)

[a]The probable errors of the linewidths were not reported. The authors did report that the four measurements made for each linewidth were consistent to within 1%.

IV. DISCUSSION

Three possible sources of error in the determination of the spectral widths are considered. (1) The width of the intrinsic diffraction profile of the study crystal (rocking curve) $W(hkl)$ is a function of the magnitude of the structure factor $F(hkl)$, as well the glancing angle of incidence $(\theta+\phi)$, and thus varies nonmonotonically with $\tan\theta$. (2) The widths of the nondispersive broadening distributions, that of the rocking curve $W(hkl)$ and C_4, the instrumental and geometrical aberrations, may not be additive with the angle-scale spectral distribution. (3) The accuracy of the determination of observed linewidths is limited by the angular resolution of the diffractometer.

The intrinsic diffraction profile widths $W(hkl)$ for the study crystal GaAs vary from 0.000 08° for $W(024)$ to 0.011° for $W(113)$. The average value of $W(hkl)$ for the reflections measured[11] is 0.0017°. The measured linewidths vary from 0.0085° to 0.087° for the $K\alpha_1$ study and from 0.014° to 0.121° for the $K\alpha_2$ study. The slopes determined after correction [by subtracting the theoretical $W(hkl)$ from the observed widths] were virtually the same (within 2%) as the slopes obtained from the raw data. The variation of the intrinsic diffraction profile widths $W(hkl)$ does not appear to significantly affect the lines of regression at our level of angular resolution.

The error due to the failure of the widths of distributions in convolution to sum to the width of the convoluted result can be estimated. Regarding the observed profile as a convolution of a nondispersive "aberrational" distribution, modeled by a Gaussian function, with the angle-scale spectral distribution, modeled by a Cauchy function, the amounts by which widths are not additive (the "deficits") are calculated. The spectral distribution was extracted by deconvoluting a Gaussian distribution of width 0.01° from the observed profiles. The deficits were obtained by subtracting the widths of the observed profiles from the sum of the widths of the Gaussian and extracted spectral profiles. Then half of each deficit was added to the observed widths. New lines of regression were determined by least-squares analysis of the scatter of the corrected data. Half deficits were used since the models used in the determination were judged to represent the "worst case" situation.

The angular resolution of the diffractometer is 0.0014°. The resulting uncertainty in measuring the separation on the (+,+) and (+,−) dispersion lines is 10% for the $K\alpha_1$ and 7% for the $K\alpha_2$. For this reason there is a large error in determining the spectral widths from C_2 and the measured line separations. Since measuring the slopes of these lines involves much larger angular differences, the

uncertainty can be reduced to less than 3% in any one measurement. A least-squares analysis of many points increases the accuracy of the data obtained from the slope.

V. CONCLUSIONS

A comparison of determined Cu $K\alpha$ spectra linewidths is given in Table III. Our determination for the $K\alpha_1$ is in reasonable agreement with Citrin *et al.*[6] and Berger[5] while our determination of the $K\alpha_2$ linewidth is in reasonable agreement with Bearden and Shaw.[3,7] The major difficulties in arriving at accurate spectral linewidths include improper estimation of the effect of the aberrational distributions and the limited angular resolution of diffractometers. If it is assumed that the width of the observed distribution is equal to the sum of the widths of aberrational and spectral distribution, the spectral widths will be underestimated. On the other hand, overidealized models will lead to overestimates. By involving many reflections in both the $(+,+)$ and $(+,-)$ configurations the reliability of our measurements was enhanced.

---

[1]J. Ladell and N. Spielberg, Acta Crystallogr. **21**, 103 (1966).
[2]J. Ladell, W. Parrish, and J. Taylor, Acta Crystallogr. **12**, 561 (1959).
[3]J. A. Bearden and C. H. Shaw, Phys. Rev. **48**, 18 (1935).
[4]L. G. Parratt, Rev. Sci. Instrum. **6**, 387 (1935).
[5]H. Berger, X-Ray Spectrom. **15**, 241 (1986).
[6]P. H. Citrin, P. M. Eisenberger, W. C. Marra, T. Aberg, J. Utriainen, and E. Kallne, Phys. Rev. B **10**, 1762 (1974).
[7]J. A. Bearden, Rev. Mod. Phys. **39**, 78 (1967).
[8]J. Ladell, J. Ayers, and J. Zola (unpublished).
[9]B. Post, J. Nicolosi, and J. Ladell, Acta Crystallogr., Sec. A **42**, 178 (1984).
[10]Throughout this paper $(+,+)$ refers to the setting in which the dispersive effects of the monochrocollimator and sample add. $(+,-)$ refers to the setting in which the observed broadening is the difference of the monochrocollimator and sample dispersion.
[11]The (001) GaAs crystal was examined, using an identical GaAs crystal cut from the same boule, with a double crystal diffractometer. The rocking-curve FWHM's measured were 0.0029° and 0.0020° for the [004] and [115] reflections, respectively.