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Rapid computer analysis of X-ray diffraction films

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Abstract

We illustrate the use of a Macintosh-based scanner and computer program to reduce X-ray powder diffraction films, yielding a quantitative profile of intensity vs. scattering angle. Our method is rapid and convenient, yet it produces reliable results even from complex diffraction patterns having a low signal-to-noise ratio. Our software can also be used to reduce image-plate data.

Discussion

The purpose of this note is to illustrate a new approach that we have developed for analyzing X-ray powder diffraction films (Nguyen and Jeanloz, 1993). Our method is simple, inexpensive, and rapid, yet it offers results that are both reliable and quantitative; the software can also be used on data from image plates. We therefore believe that it may have wide-ranging applications in research and education.

Film has commonly been used for recording X-ray powder diffraction patterns. In addition to being inexpensive, it has the advantage of being a sensitive, two-dimensional integrating detector with high spatial resolutions. Thus, it is especially useful for small samples and for specialized applications; examples include the samples of approximately picoliter size studied in the ultra-high-pressure diamond cell, and the powder diffraction films obtained from single crystals in the Gandolfi camera. Indeed, for low diffraction intensities, when the intensities are not saturated, film is in many ways an ideal recording medium. Its major drawbacks are that it is difficult to quantify the intensities (Klug and Alexander, 1974, sections 6–7), the pattern is not amenable to digital enhancement or filtering, uncertainties in the results are not easily determined, and reading the film can be time consuming and tedious.

These drawbacks are largely overcome if the film is digitized. Using modern computers and microdensitometers, a fully quantitative pattern can be obtained (e.g., Meade and Jeanloz, 1990). A similar approach is used with image plates, which are scanned by laser and have the advantage over film of much greater dynamic range (i.e., less readily saturated at high intensities).

To take advantage of the simplicity and economy of film, we have developed a Macintosh-based computer program that can be used with a common scanner to obtain a digital pattern of intensity as a function of diffraction angle (Nguyen and Jeanloz, 1993). For many applications, especially in the classroom, a routine 300 dots per inch (dpi) scanner provides adequate spatial resolution of ~85 μm. Such scanners are widely available and can be driven with a variety of image-analysis programs (e.g., Adobe Photoshop). Higher resolution scanners (600–1200 dpi, corresponding to a resolution of 42–21 μm) may be required for more specialized applications, but even these instruments are relatively accessible. Perhaps the greatest problem with increasing resolution is that the data files rapidly become larger and are therefore more difficult to manipulate and store. In our experience, file sizes of ~0.5–10 Mb are not uncommon.

The computer program we have developed is a refinement of that described by Meade and Jeanloz (1990). It takes the two-dimensional scanned image of the film and collapses or integrates each diffraction arc to a single point at the appropriate diffraction angle. Another way to think of it is that the diffraction pattern is a series of coaxial elliptical arcs having ellipticity that increases systematically with scattering angle (intersection of the diffraction cones with the cylindrical film surface, with ellipticity ranging from 0 at $2\theta = 0°$ to infinite at $2\theta = 90°$). The computer program performs a nonlinear least-squares fit to the diffraction ellipses.

The product of the computer analysis is two one-dimensional profiles of intensity vs. diffraction angle, with one profile for each half of the film on either side of the primary X-ray beam. These profiles can be compared with each other for consistency and can be averaged to obtain the best estimate of the diffraction pattern. Note that any noise that does not follow the appropriate elliptical arc for a given scattering angle (e.g., scratches on the film) tends to be suppressed. Also, the integration or summation along each diffraction arc gives an approximately 20-

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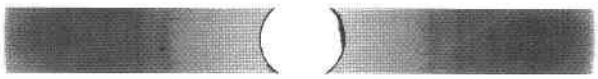

Fig. 1. X-ray powder diffraction pattern of a peridotite-composition sample at 135 GPa, after subsolidus laser heating.

fold enhancement in the signal-to-noise ratio over the original film (Meade and Jeanloz, 1990; Nguyen and Jeanloz, 1993).

The new program has the advantage of speed and convenience (cf. Piltz et al., 1992). The illustrative cases we present were processed on a Macintosh IIfx in <1 min. Adding the time required for scanning (2–3 min), we can realistically estimate that diffraction films can be reduced to a pair of digital profiles of intensity vs. diffraction angle in <5 min. One of the advantages of this speed is that the raw image file can be processed in a variety of ways prior to analysis. For example, the image contrast can be varied, or spurious markings on the film (e.g., Laue spots from diamond anvils) can be removed. The program itself has been combined with Wayne Rasband's Image program, developed at the National Institutes of Health and available in the public domain. Additional details are given in Nguyen and Jeanloz (1993), and the program may be obtained by contacting them.

We illustrate our method with four Debye-Scherrer films from two samples taken to pressures of 54 and 135 GPa in a Mao-Bell type diamond cell. Each sample consists of <10 μg peridotite that has been converted to the perovskite-dominated high-pressure assemblage through subsolidus laser heating at high pressures (O'Neill and Jeanloz, 1990). Two of the films were collected in situ, through the diamond cell, after laser heating but while the sample was still at pressure (monochromatized Mo$K\alpha$ radiation from a rotating anode generator, with a sample-to-film distance of ~50 mm); the other two films were obtained from the samples after decompression to ambient conditions (filtered Cu$K\alpha$ radiation from a tube, with sample-to-film distances of ~29 and 57 mm). Further experimental details are given by O'Neill and Jeanloz (1990, in preparation).

The image of the film acquired at 135 GPa is shown in Figure 1. It was obtained with a Sharp 600-JX scanner at 600 dpi, using transmitted illumination (however, the image has been printed at 300 dpi). Although transmitted illumination is best, incident illumination also provides usable scans (the results are improved if one places a bright, diffuse reflector, such as white paper, behind the film).

The quality of the film pattern is not unusual for powder diffraction patterns obtained at ultrahigh pressures, especially for complex assemblages of phases (O'Neill and Jeanloz, 1990). To analyze the film, there must be at least one matching pair of diffraction rings. In the present approach, the geometry is set by one pair, whereas several pairs of diffraction rings have been used in previous work (cf. Meade and Jeanloz, 1990; Nguyen and Jeanloz, 1993). If necessary, a polycrystalline standard such as Au can be mixed with the sample to provide the necessary diffraction pattern; we have done that in the case of amorphous samples, for example (Kruger and Jeanloz, 1990).

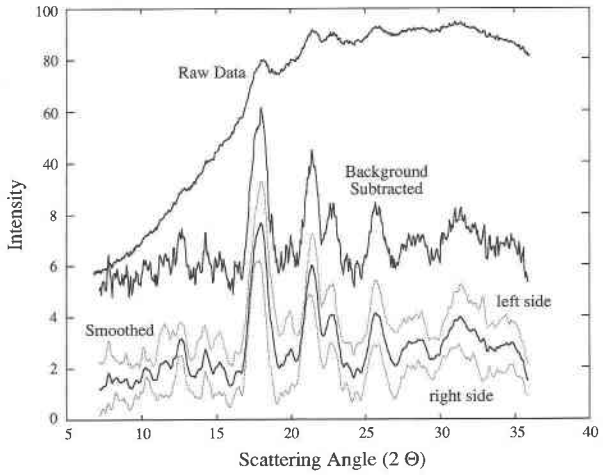

Fig. 2. Collapsed version of the diffraction pattern shown in Fig. 1, comparing the raw, integrated data (top) with the corresponding profiles after background subtraction (middle), using a high-pass filter and subsequent smoothing (bottom) with a low-pass filter (see text). Results from the left and right sides of the film are compared with the average profile of the two sides in the final pattern; only the average profiles are shown for the raw and background-subtracted data.

Upon collapsing, the diffraction pattern emerges more clearly and can now be filtered digitally (Fig. 2). Moreover, the quality of the fit to the diffraction pattern can be quantified, such that errors are reliably estimated. Appropriate background subtraction, intensity calibrations, and corrections for texturing are essential before the intensities can be used quantitatively, however (e.g., for Rietveld refinement). It should be noted that, unlike microdensitometers, desktop scanners bin the optical density of the film into 256 gray levels, which must also be taken into account if quantitative intensity measurements are required.

To remove the background and smooth the diffraction pattern (Fig. 2), we apply high-pass and low-pass Butterworth filters, respectively (Gonzalez and Wintz, 1987, chapter 4). That involves multiplying the Fourier-transformed data by two Butterworth functions,

$$B = [1 + (\sqrt{2} - 1)\, z^{2n}] \qquad (1)$$

with $z = C/x$ for the high-pass filter and $z = x/C$ for the low-pass filter, and then back-transforming the data. Here, x is the spatial wavenumber, C the cutoff frequency, and n the order of the filter. Figure 2 illustrates the result of applying first-order filters ($n = 1$) with cutoff frequencies $C = 10$ and 50 (x in units 2π per pixel).

The final pattern contains at least six to eight resolvable peaks (overlapping diffraction lines in several cases); these are much more evident than in either the original film (Fig. 1) or the raw data after collapsing (Fig. 2). Perhaps

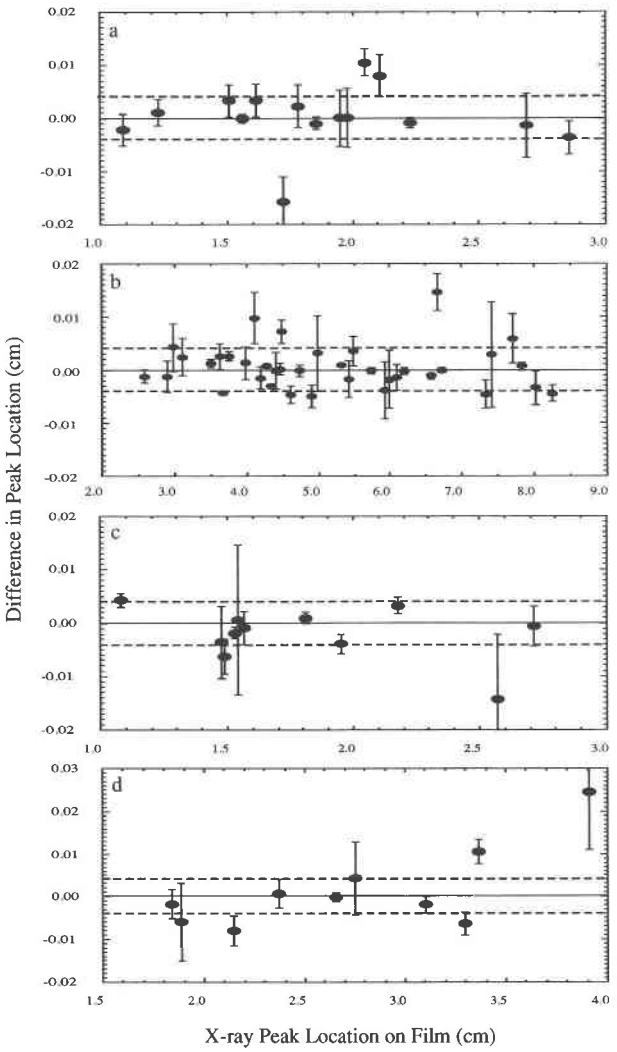

Fig. 3. Comparisons of numerical methods and digitizing hardware in analyzing complex diffraction films having low contrast. Differences in the locations of diffraction lines obtained either using two numerical methods (**a** and **b**) or using two methods of digitizing (**c** and **d**) are shown. Error bars are inversely proportional to line intensities, and ±1 pixel is indicated by the dashed lines.

more significant is the fact that one can compare the diffraction patterns from the two sides of the films. For example, the fact that the peaks on the left side of the film at $2\theta \sim 12$ and 19° do not appear on the right side raises a suspicion that they may be spurious. As the signal-to-noise ratio is close to 1 in both cases, further documentation would be required to consider seriously these peaks as part of the diffraction pattern. This built-in redundancy of digitized film records is especially useful for examining complex diffraction patterns (e.g., O'Neill and Jeanloz, 1990, in preparation).

We have previously documented the reliability of both the digitization and the computer analysis when examining high-quality films with very clear diffraction patterns (Meade and Jeanloz, 1990; Nguyen and Jeanloz, 1993). Here, we illustrate the reproducibility of results obtained from films that are complex and of lower quality. Our intention is to document the bias that can enter into the digitization and the numerical collapsing (least-squares fitting) of poorly resolved diffraction patterns.

Figure 3a and 3b summarize the diffraction-peak locations derived from two separate scans (600 dpi) of films of the 135 GPa sample, one film at pressure and one after decompression. Each film was scanned twice, then one scan was reduced with the original program of Meade and Jeanloz (1990), and the other with the newer program of Nguyen and Jeanloz (1993). Although both programs solve the identical equations, the numerical algorithms used are quite different and, because the diffraction profiles are noisy, could yield different results from the nonlinear least-squares analysis.

Our comparison illustrates the reproducibility of separate scans and of the software employed, with the digitization hardware remaining the same (Fig. 3a, 3b). We find that 12 out of 15 and 27 out of 37 diffraction lines are reproducibly located to within ±1 pixel (42 μm). That is, the standard deviation is less than the nominal resolution of the digitization. One typically obtains enhanced resolution from high-quality films because the summation involved in collapsing the film serves to average the multiply sampled positions of each diffraction line. It is encouraging to see that such enhancement can still occur for poorly resolved patterns, although the results are clearly subject to greater scatter and bias than for high-quality patterns (Meade and Jeanloz, 1990).

Figure 3c and 3d summarize results for the 54 GPa sample, at pressure and after decompression (the diffraction pattern of Fig. 3c is described by O'Neill and Jeanloz, 1990). In this case, we use the same software and compare different digitization methods (microdensitometer with a 25-μm resolution vs. a 600-dpi scanner), with readings by eye supplementing the densitometer results in Figure 3d. The agreement among different digitizing techniques remains good: standard deviations are $\ll 1$ pixel in Figure 3c and approach ~1.5 pixel in Figure 3d. There is also a hint that systematic biases may be present for this last pattern, in contrast with the others. However, given the few data points and their relatively large uncertainties, any biasing is too small to be unambiguously demonstrated. Overall, the comparisons shown in Figure 3 illustrate that complex, poorly resolved X-ray diffraction films can be scanned and reduced to yield diffraction patterns having a resolution approaching, if not exceeding, that of the digitization.

Acknowledgments

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