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Quantitative Analysis Using Asymmetric Adaptive Pulse Processing

Richard B. Mott[1], Owen E. Healy[1], Nicholas W. M. Ritchie[2], and Abigail P. Lindstrom[2]

[1] PulseTor LLC, 1580 Reed Road, Suite C2, Pennington NJ 08534
[2] National Institute of Standards and Technology, Gaithersburg, MD 20899-8371

Previous work [1] demonstrated that pulse-by-pulse adaptive digital filtering improves the precision of X-ray quantitative analysis for a given sample electron dose, with no loss of accuracy compared to conventional pulse processing. The improvement stems from better energy resolution compared to short fixed digital filtering for the same throughput. The gain in precision is greatest for small peaks below about 5 keV near to or overlapping with larger peaks, such as Al in NIST K412 glass, which has favorable implications for analysis at low accelerating voltages.

That work used only a small set of digital filters for ease of generating standards. The time intervals between X-ray arrivals are randomly unequal. Asymmetric processing, as described by Koeman [2], reduces the noise component of resolution by allowing different integration times before and after an X-ray arrives. It promises further gains in analytical precision, but it also requires more sophisticated peak shape modeling to derive appropriate fitting spectra from standards.

Table 1 shows the resolution gained by asymmetrical filtering. The matrix diagonal is conventional symmetric filtering (shaded). Entries in bold are the filters used for last year's results. The axes are leading and trailing integration times in nS. Entries are Al-K resolution in an Al_2O_3 sample at 7 nA, 319 kcps input rate, 196 kcps throughput rate. Figure 1 is a resolution contour plot of these data.The matrix is symmetric within a few tenths of an eV, which is expected since resolution should not depend on arrival order for each pair of intervals. For each diagonal entry, resolution improves as we move up or to the right (increasing the integration time toward the wider interval).

Table 2 gives the probability of occurrence for each of the cells in Table 1 at an input count rate of 320 kcps. The interval pair probability depends only on the input rate, regardless of the composition of the sample. At 320 kcps ICR and 38% dead time, nearly 27% of the X-rays fall in the upper right matrix cell with six times the integration period and 30 eV better resolution relative to the shortest filter, while 77% are measured with 2.4 uS integration on one side (top row or right column).

The table sums to 1, and the lower-left entries are so small because the time bins are of unequal size. The rectangle with each diagonal entry at a lower left corner sums to the fraction of X-rays measured with that diagonal entry's resolution or better. Since the input rates generated from the unknown and the standards will be different in general, the procedure for generating fitting standards uses the noise components derived from Table 1 with the probabilities of Table 2, which can be readily computed for any input count rate.

References
[1] R.B. Mott, O.E. Healy, N.W.M. Ritchie and A.P. Lindstrom. *Microscopy and Microanalysis* 19 (Suppl. 2), (2013) 1252-1253.
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2400 nS	97.6	94.4	91.9	88.9	86.7	85.1	83.9	82.6	82.0	81.8
2000 nS	97.9	94.9	92.3	89.1	87.0	85.7	84.2	83.0	82.4	81.9
1600 nS	98.3	95.5	93.0	89.9	87.7	86.3	84.9	83.8	83.0	82.6
1280 nS	99.3	96.2	93.9	90.8	88.8	87.3	**85.9**	84.9	84.1	83.7
1040 nS	100.2	97.3	94.9	91.8	90.0	88.6	87.4	86.2	85.7	85.4
880 nS	101.4	98.2	96.2	93.2	91.0	89.9	88.4	87.6	86.8	86.7
720 nS	103.0	100.0	97.8	**95.0**	93.1	91.9	90.7	89.8	88.9	88.9
560 nS	105.7	102.7	100.5	97.9	96.3	95.0	93.8	92.9	92.3	92.1
480 nS	107.7	104.9	102.6	100.1	98.2	97.4	96.1	95.0	94.5	94.4
400 nS	**110.3**	107.4	105.6	102.7	101.3	100.2	99.3	98.4	97.7	97.4
Filters	400 nS	480 nS	560 nS	720 nS	880 nS	1040 nS	1280 nS	1600 nS	2000 nS	2400 nS

Table 1. Al-K resolution vs. filter times. Diagonal (shaded) entries are symmetric filtering. Resolution improves above and to the right of each diagonal entry, as leading (row) or trailing (column) integration times increase.

2400 nS	0.0134	0.0130	0.0252	0.0239	0.0227	0.0319	0.0388	0.0428	0.0373	0.2687
2000 nS	0.0019	0.0018	0.0035	0.0033	0.0032	0.0045	0.0055	0.0063	0.0056	0.0373
1600 nS	0.0021	0.0021	0.0040	0.0038	0.0036	0.0051	0.0063	0.0069	0.0063	0.0429
1280 nS	0.0019	0.0019	0.0037	0.0034	0.0033	0.0046	**0.0056**	0.0063	0.0055	0.0388
1040 nS	0.0016	0.0016	0.0030	0.0028	0.0027	0.0038	0.0046	0.0051	0.0045	0.0319
880 nS	0.0011	0.0011	0.0021	0.0020	0.0019	0.0027	0.0033	0.0036	0.0032	0.0227
720 nS	0.0012	0.0012	0.0022	**0.0021**	0.0020	0.0028	0.0035	0.0038	0.0034	0.0239
560 nS	0.0013	0.0012	0.0024	0.0022	0.0021	0.0030	0.0037	0.0040	0.0036	0.0252
480 nS	0.0007	0.0006	0.0012	0.0012	0.0011	0.0016	0.0019	0.0021	0.0018	0.0131
400 nS	**0.0007**	0.0006	0.0013	0.0012	0.0011	0.0016	0.0019	0.0022	0.0019	0.0135
Filters	400 nS	480 nS	560 nS	720 nS	880 nS	1040 nS	1280 nS	1600 nS	2000 nS	2400 nS

Table 2. Probability for measured X-rays falling into each cell in Table 1.

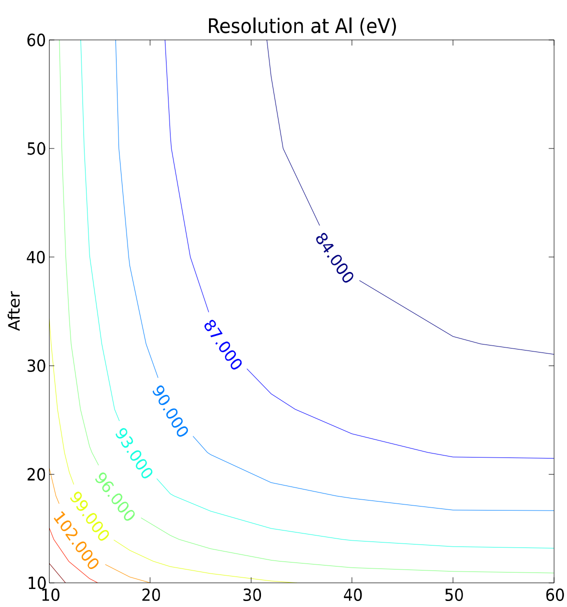

Figure 1. Table 1 as resolution contour plot.

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