Does the Maximum Entropy Method Improve Sensitivity?

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Maximum entropy reconstruction has been applied to inverse problems in a variety of fields *(1–3)*. In NMR spectroscopy *(3)*, for example, maximum entropy reconstruction has been used to obtain dramatic improvements in signal-to-noise ratio over conventional discrete Fourier transform spectrum estimates. Unfortunately, it is not clear just how maximum entropy reconstruction achieves these impressive results. This is largely because in the general case there is no analytical expression for the maximum entropy reconstruction, which must therefore be obtained by numerical methods. In order to gain some insight into the maximum entropy method, we consider maximum entropy reconstruction applied to a special class of reconstructions for which a formal analytical solution *can* be found. We show that the reconstructions take the form of a single non-linear transformation applied point-by-point to the discrete Fourier transform of the data. This result explains many published examples of maximum entropy reconstructions, and shows that the maximum entropy method does not, by itself, improve sensitivity.

Figure 1, reproduced from Sibisi *et al.* *(3)*, is typical of maximum entropy spectrum reconstructions appearing in the literature. The feature of the reconstructions most often emphasized is the suppression of noise near the baseline; however, close examination reveals another characteristic feature: while de-emphasized relative to strong signals, the structure of the noise near the baseline is mostly preserved. A third characteristic feature is shown in the reconstruction in Figure 2: noise near the baseline is suppressed more effectively than noise superimposed on a peak. All of these effects can be explained by the following analysis.

the introduction of a Lagrange multiplier λ to form the objective function

$$Q = -\sum_{\omega=1}^{N_\omega} \hat{f}_\omega \log \hat{f}_\omega - \lambda \sum_{t=1}^{N_t} \left| \frac{1}{\sqrt{N_\omega}} \sum_{\omega=1}^{N_\omega} \hat{f}_\omega e^{2\pi i t\omega/N_\omega} - d_t \right|^2 . \tag{3}$$

The solution we seek corresponds to a critical point of Q, with the value of λ chosen so that $C = C_0$. There is no known formal solution to this problem for the general case $N_\omega > N_t$, and the general problem must be solved numerically by simultaneously optimizing the N_ω elements of $\hat{f}$.

However, when $N_\omega = N_t$, Parseval's theorem provides the equality

$$\sum_{t=1}^{N_t} \left| \hat{d}_t - d_t \right|^2 = \sum_{\omega=1}^{N_\omega} \left| \hat{f}_\omega - f_\omega \right|^2 , \tag{4}$$

where f is the Fourier transform of d. Thus, equation (3) becomes

$$Q = -\sum_{\omega=1}^{N_\omega} \hat{f}_\omega \log \hat{f}_\omega - \lambda \sum_{\omega=1}^{N_\omega} \left| \hat{f}_\omega - f_\omega \right|^2 . \tag{5}$$

This problem is easily solved; at a critical point we must have

$$0 = \frac{\partial Q}{\partial \hat{f}_\omega} = -(\log \hat{f}_\omega + 1) - 2\lambda(\hat{f}_\omega - \mathrm{Re}(f_\omega)) \qquad \text{for all } \omega. \tag{6}$$

Let the function $\delta_\lambda(z)$ be defined as the solution to the equation

$$0 = -(\log \delta + 1) - 2\lambda(\delta - \mathrm{Re}(z)). \tag{7}$$

¿From the definition of δ_λ and equation (6), the solution to the maximum entropy problem is

$$\hat{f}_\omega = \delta_\lambda(f_\omega) \qquad \text{for all } \omega. \tag{8}$$

and is applied to each frequency independently, the reconstructed spectrum possesses exactly the same structure as the original spectrum. (For example, if the intensity is greater at frequency j than at frequency k before reconstruction, then the same must be true after reconstruction.) This means that if 95 per cent of the noise values in the discrete Fourier transform spectrum are expected to be less than some value W, then in the reconstructed spectrum 95 per cent of the values will be less than $\delta_\lambda(W)$. Consequently peak identifications—as signal or noise—based on an expected noise distribution (such as a Gaussian distribution) will always give the same results for the conventional Fourier transform spectrum and for the maximum entropy reconstruction.

Although we have been concerned only with a special case, some of the phenomena described above, such as noise remaining when it is superimposed on a peak, are frequently observed in the more general case where $N_\omega > N_t$ *(9)*. Maximum entropy reconstruction has been and will no doubt continue to be the subject of vigorous debate *(7,11,12)*. The analysis presented here illustrates, for a specific case, just how maximum entropy reconstruction works, and demonstrates in concrete terms that maximum entropy reconstruction can improve signal-to-noise ratio without improving sensitivity.

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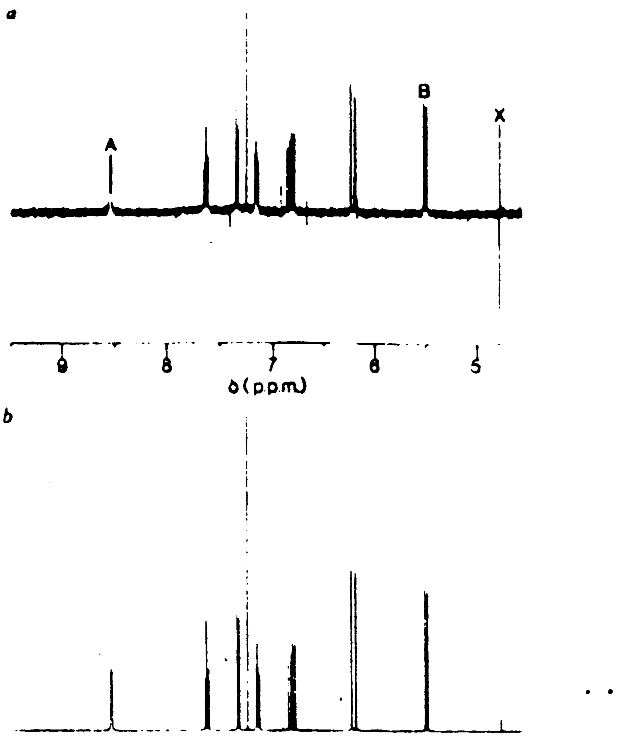

Fig. 3 *a*, ^{1}H spectrum of 2-vinyl pyridine produced by conventional Fourier transform. *b*, Corresponding MEM spectrum.

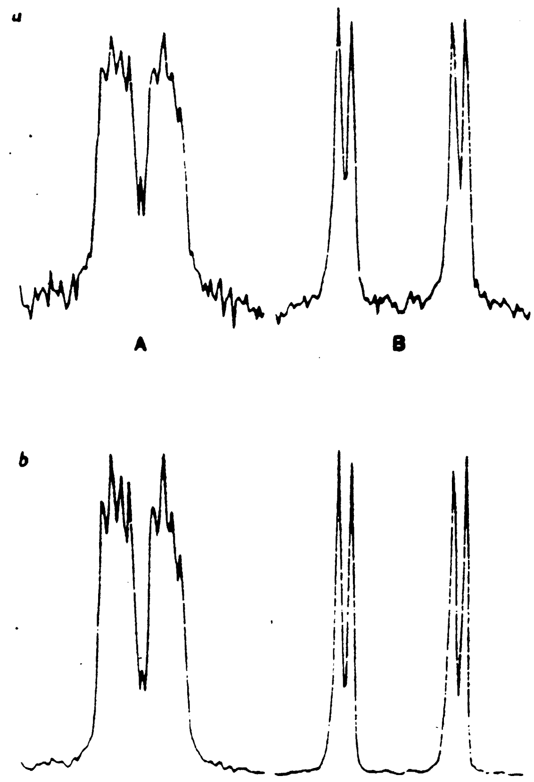

Fig. 4 *a*, Expansions of the multiplets A and B in the conventional spectrum (Fig. 3*a*). *b*, Expansions of the multiplets A and B in the MEM spectrum (Fig. 3*b*).

time (1.6 μs). We corrected the baseline of the FID and converted it from quadrature to cosine form[21] over 8,192 points before zero filling to 16,384 points. The Cambridge MEM algorithm[22] was used to find the two principal phase parameters, by maximizing the entropy over both the spectrum and these parameters simultaneously. The phase parameters were then used for both the conventional FT and MEM spectra shown in Fig. 1.

In the conventional FT spectrum, Fig. 1*a*, the signals assigned to C-3, C-4, C-5, C-6, C-7 and C-8 are reasonably clear of the noise, but the signal from C-2 is only marginally higher than some of the noise peaks (marked with asterisks). In the corresponding MEM spectrum obtained from the same data (Fig. 1*b*), noise is dramatically decreased, so that the peak for C-2 appears much more convincing. The reliability of the lines shown in the MEM spectrum can be assessed by comparison with a control FT spectrum (Fig. 2) produced from the same sample, using a longer pulse time (13 μs) to increase the signal-to-noise ratio. This proves that the C-2 line picked out by maximum entropy is a true signal, as opposed to the noise peaks marked with asterisks in Fig. 1*a*. The ability of MEM to discriminate in favour of real signals, and against noise of similar intensity, was reproduced in three other test spectra determined in the same conditions.

The ^{1}H data were recorded in a similar manner on a Bruker WH-400 instrument. We selected a moderately noisy spectrum to give a clear comparison between MEM and conventional procedures. Figure 3*a* shows the conventional Fourier spectrum, and Fig. 3*b* the corresponding MEM spectrum. In addition to the obvious suppression of noise, MEM has also sharply reduced the instrumental artefact, X. Two representative multiplets, A and B, are plotted on expanded scales in Fig. 4*a* (conventional) [illegible]

[illegible]

These results demonstrate that MEM provides a potentially important addition to the range of techniques currently available for producing NMR spectra. Details of the Cambridge MEM algorithm used to produce these spectra will be published elsewhere[22]. The disadvantage of MEM is the extra computing involved, amounting to about 1 min on a IBM3081 mainframe computer. The algorithm requires eight times the store and about 30 times the central processing time of an ordinary Fourier transform. Further work is in progress on the application of MEM to ^{1}H and ^{2}H NMR spectra, and also on the more sophisticated approach in which a two-dimensional map is reconstructed from one-dimensional data[11].

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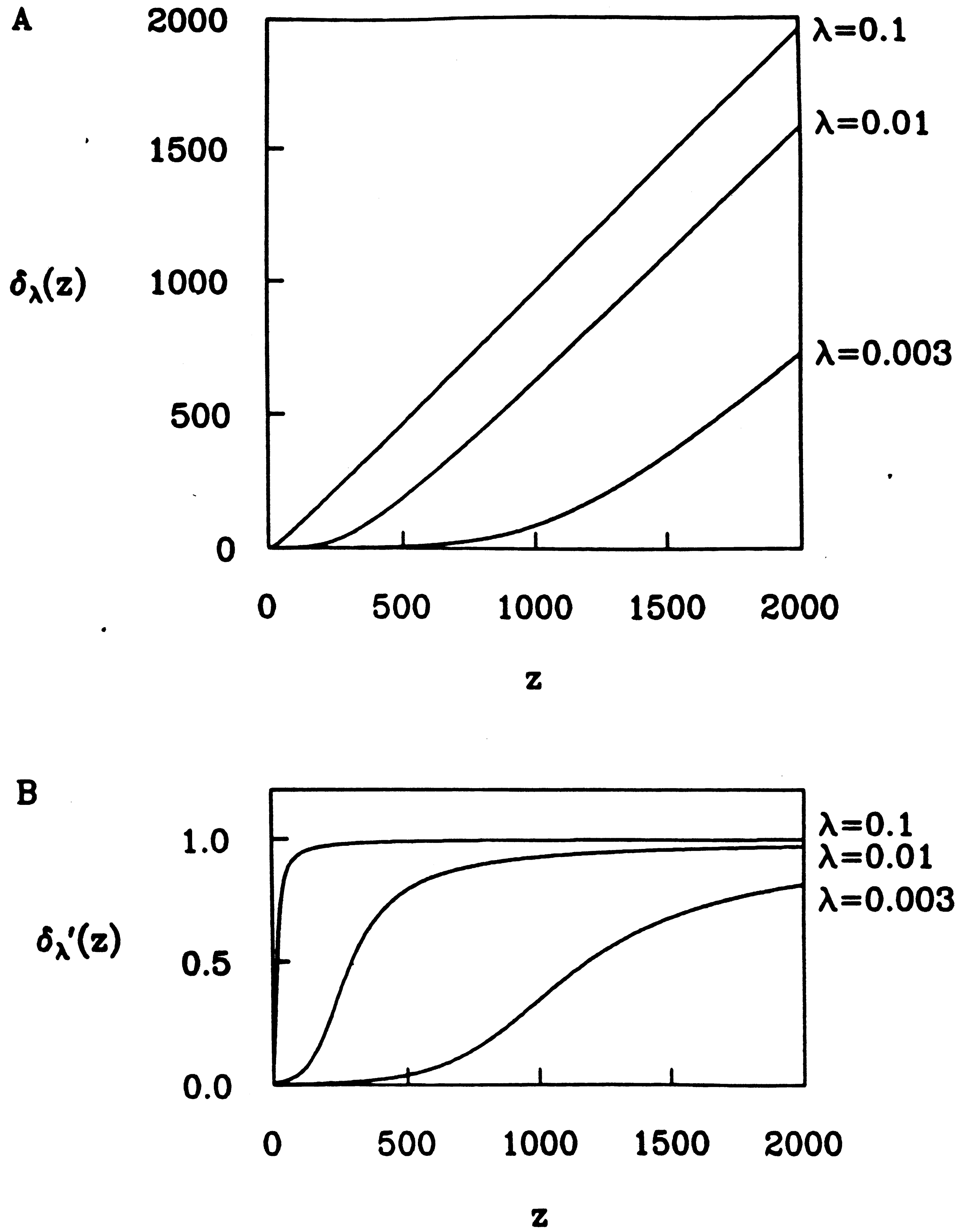
A
2000
1500
1000
500
0
$\delta_\lambda(z)$
0
500
1000
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2000
z
λ=0.1
λ=0.01
λ=0.003
B
1.0
0.5
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$\delta_\lambda'(z)$
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λ=0.1
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λ=0.003

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