spacing or samples that appeared out of order.

Free-electron lasers allow the early stages of ultrafast chemical reactions to be studied with molecular precision[6,7]. Fung *et al.* used X-ray free-electron lasers to investigate a fundamental chemical process: the photoionization of nitrogen molecules, whereby the nitrogen ions N_2^+ and N_2^{2+} are created after photon absorption by molecular nitrogen (N_2). The ionization plays out incredibly quickly, over a period of several hundred femtoseconds (1 fs is 10^{-15} seconds).

The authors used an infrared pulse to trigger photoionization and an X-ray pulse to probe the process, capturing snapshots using a spectrometer with a 'shutter speed' of less than 100 fs. Unfortunately, thermal noise in the instrument introduces timing uncertainty that can exceed 280 fs (ref. 7). This temporal uncertainty, or jitter, makes it difficult to measure quantities such as the vibrating frequencies of the molecular system.

Fung *et al.* used a mathematical technique known as nonlinear Laplacian spectral analysis (NLSA) to recover the true dynamics underlying these snapshots (Fig. 1). In this technique, the data are embedded in a multi-dimensional space characterized by Laplacian functions, which can represent turbulent or intermittent structure in the data. The technique is nonlinear because the data are mapped to a curved surface. The term 'spectral' is used because, just as a prism separates white light into a spectrum of long to short wavelengths, NLSA decomposes the snapshots into their constituent time and space components (known as chronograms and topograms, respectively), arranged from strongest (the largest contributors to the signal) to weakest (the smallest contributors; Fig. 1b,c). The authors' key observation is that the chronograms are robust to jitter. In other words, the data points in the chronograms are uniformly spaced in time, which means that it is, in principle, possible to reconstruct the original data without jitter by recombining the modes.

However, NLSA does not allow such a reconstruction except in simple examples, because it requires a huge number of unknown parameters to be estimated. But reconstruction is not the goal. Instead, the quantities of interest can be observed directly by looking at the strongest 'modes' obtained from NLSA — these modes are derived mathematically from the chronograms and topograms, and describe the essential properties of the nitrogen system. One of the modes reveals an especially interesting result: molecular nitrogen vibrates with three previously unobserved frequencies. These are in the 16–24-fs range — much shorter than those observed in any preceding experiment, but consistent with predictions from quantum mechanical theory.

Aside from this important application to quantum molecular physics, the theory proposed by Fung *et al.* has profound implications for signal processing and spatiotemporal statistics. Techniques for the spectral analysis of time series or spatial data involving uneven sampling or missing data, both with known and unknown timestamps, have been sought since at least the early 1980s (refs. 2,3,8,9). It is remarkable that, in the molecular-nitrogen experiment, the authors can resolve the vibrational modes to about 1 fs spacing. However, an expression for the reduction in temporal uncertainty achieved after application of this technique is desirable. Another question is whether NLSA can return a uniform result if signals are systematically bunched in time. And finally, if NLSA is applicable to unevenly timed samples, it ought to be applicable to unevenly spaced samples in one spatial dimension — but are there nonlinear embeddings that can extend this result into higher dimensions?

The authors verified their observations by numerical simulation, but mathematical rigour and theoretical formalism for the temporal uniformity of the chronogram components are still lacking. Moreover, the presented formulation may still be overly complex. In particular, the authors show that a single cosine curve, sampled with jitter, can be recovered without the need for a nonlinear approach. Given that this simple cosine example underlies all conventional spectrum analysis[10], is there a simpler formulation that does not require the complex machinery of NLSA but still possesses the property of time uniformity in the result? What is the simplest, most basic working example of such a process? This work opens the door to many interesting questions. ■

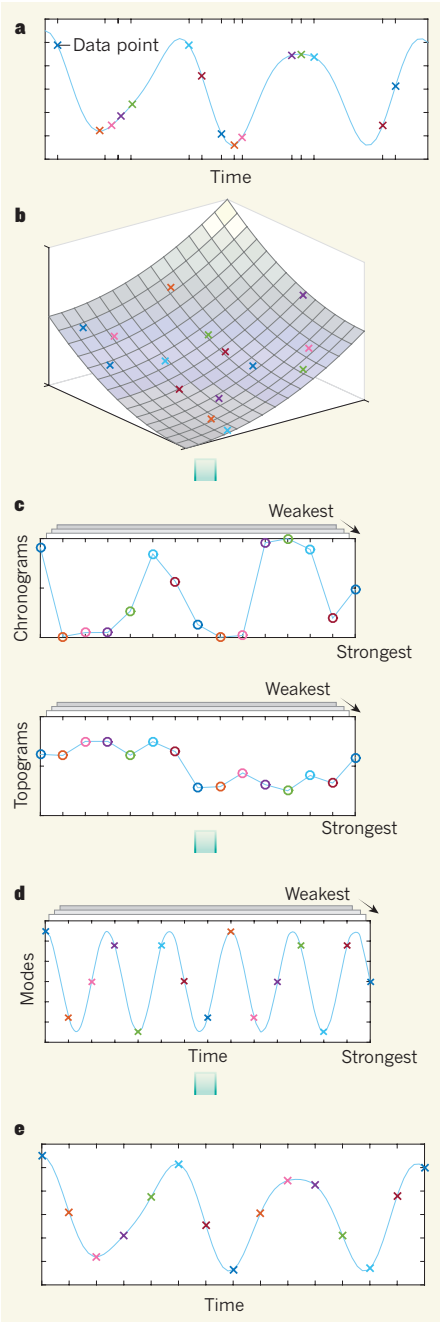

Figure 1 | A mathematical process for analysing space-time data. Fung *et al.*[5] studied the vibrations and break-up of nitrogen molecules in response to a single infrared trigger pulse and a probing X-ray pulse. **a**, The collected space-time data were unevenly distributed in time and sometimes in the wrong order (data from a single spatial coordinate are shown). **b**, **c**, To extract the underlying temporal dynamics, the authors embedded the data in a five-dimensional nonlinear space (**b**, three dimensions are shown, for simplicity), then decomposed the data into a series of time and space components (**c**, chronograms and topograms, respectively), organized from strongest to weakest. **d**, These components can be used to construct modes that describe the essential properties of the nitrogen system and have uniform time spacing. **e**, In principle, the original data without timing errors can be reconstructed from a combination of these modes.

Charlotte A. L. Haley *is in the Mathematics and Computer Science Division, Argonne National Laboratory, Lemont, Illinois 60439, USA.*
e-mail: haley@anl.gov

1. Scargle, J. D. *Astrophys. J.* **263,** 835–853 (1982).
2. Kuhn, J. R. *Astron. J.* **87,** 196–202 (1982).
3. Petit, J. R. *et al. Nature* **399,** 429–436 (1999).
4. Wittman, D. M., Tyson, J. A., Kirkman, D., Dell'Antonio, I. & Bernstein, G. *Nature* **405,** 143–148 (2000).
5. Fung, R. *et al. Nature* **532,** 471–475 (2016).
6. Pellegrini, C. & Stöhr, J. *Nucl. Instrum. Methods Phys. Res. A* **500,** 33–40 (2003).
7. Glownia, J. M. *et al. Opt. Express* **18,** 17620–17630 (2010).
8. Kondrashov, D. & Ghil, M. *Nonlin. Process. Geophys.* **13,** 151–159 (2006).
9. Fuentes, M. *J. Am. Stat. Assoc.* **102,** 321–331 (2007).
10. Ghil, M. *et al. Rev. Geophys.* **40,** 3-1–3-41 (2002).

CORRECTION

In the News & Views article 'Physics: Quantum problems solved through games' by Sabrina Maniscalco (*Nature* **532,** 184–185; 2016), reference 7 was incorrect. The correct reference is https://www.scienceathome.org/games/quantum-moves/game