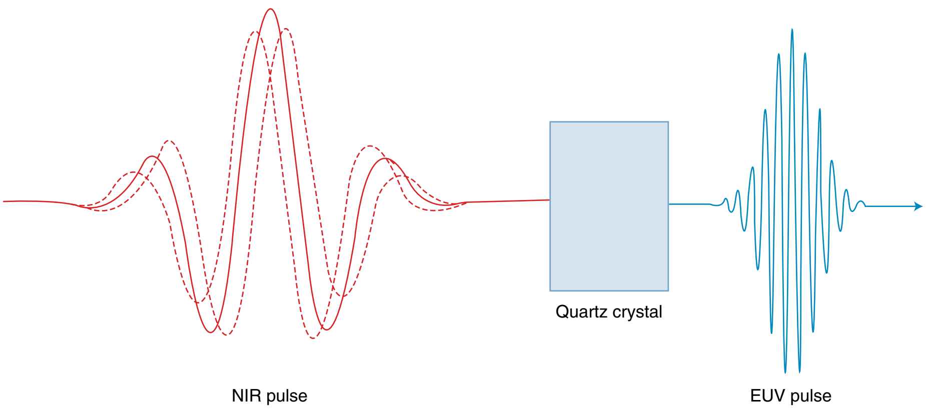

Fig. 1 | Intense near-infrared (NIR) laser pulse goes through thin crystalline quartz to produce a co-propagating coherent extreme-ultraviolet (EUV) pulse. The nonlinear optical response of the crystal produces high-order harmonics, whose spectrum extends to the EUV wavelength range. In the time domain, the duration of the EUV pulse is on the attosecond timescale and its waveform remains locked to the phase of the pump laser field despite fluctuations in the pump laser's amplitude (shown by dashed lines).

conduction bands, the transition dipole matrix elements between the valence and conduction bands, the bandgap and photon energy, for example. Therefore, the dominant microscopic mechanism could easily depend on the material.

Why quartz? A quartz crystal was used to produce the optical second harmonic of a ruby laser 57 years ago, which marked the birth of nonlinear optics[5]. In that experiment, the strength of the applied field was about 10^5 V cm^{-1}, which is a small perturbation to the field-free Hamiltonian. With the advent of ultrafast laser technology, it is now possible to apply ~1,000 times stronger pulses to quartz crystals without causing permanent damage. As a result, high-order harmonics are produced, whose spectra extend to the EUV range[6–9]. A typical experimental set-up is shown schematically in Fig. 1. In Garg and colleagues' work, because of the high applied field, the interaction term of the Hamiltonian is comparable to the field-free Hamiltonian; therefore, the usual perturbation theory of nonlinear optics breaks down[5]. Clearly, the underlying electron dynamics involve attosecond timescales, but the outstanding question was just how the EUV waveforms are synchronized with the electric field of the driving laser pulse. Does that synchronization depend on the laser parameters, as in the gas phase?

To shed light onto the problem, Garg and colleagues consider an interferometry method, which has been successful in similar gas-phase experiments. In their work, photoelectron interferometry is employed to measure the waveform of the EUV pulse produced by solid-state HHG. The EUV beam produced via high-harmonic generation from a thin quartz crystal is focused together with a reference NIR laser beam onto an argon gas target. EUV photons produce photoelectrons from the atoms via photoionization processes. The reference laser's photon energy is too small for direct photoionization but its peak intensity is strong enough to produce electrons via the nonlinear absorption process above-threshold ionization (ATI). The ATI electron spectrum extends to the energy range of the EUV photoelectrons. Then, the ATI and EUV processes interfere, producing fringes that are observed in the electron detector.

The important finding of Garg and colleagues' work is that these fringe positions do not depend on the peak intensity of the pump laser. The fringe positions also remained effectively fixed to the variation in the CEP. Additionally, the entire comb of measured high harmonics from quartz does not show any measurable delay among harmonics, which means there is no intrinsic atto-chirp[2]. These remarkable experimental observations are in contrast to the gas-phase HHG, but are exactly what the Bloch oscillation model has predicted[1,10]. The implications of solid-state HHG are in materials science, in attosecond pulse generation in a compact set-up[8,11] and in high-harmonic spectroscopy[6]. ❐

Shambhu Ghimire
Stanford PULSE Institute, SLAC National Accelerator Laboratory and Stanford University, Stanford, CA, USA.
e-mail: shambhu@slac.stanford.edu

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NONLINEAR OPTICS

Shining light on an old problem

The evolution of modulational instability sidebands in an optical fibre are shown to provide new insights into Fermi–Pasta–Ulam–Tsingou recurrences.

Daniele Faccio and Fabio Biancalana

In this issue of *Nature Photonics*, Mussot et al.[1] demonstrate a technique that exploits nonlinear optics in a glass fibre to provide new insights into an old and famous problem in physics that was first encountered by Enrico Fermi, John Pasta, Stanislaw Ulam and Mary Tsingou in the 1950s. The so-called Fermi–Pasta–Ulam–Tsingou problem relates to the counter-intuitive situation where highly complex physical systems are seen to exhibit unexpected periodic behaviour with a flow

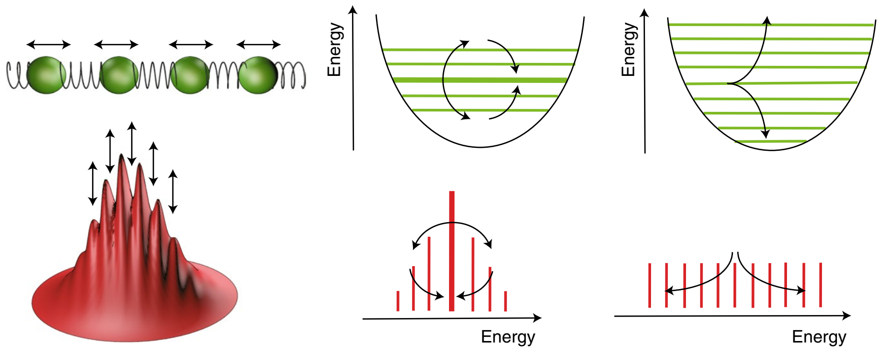

Fig. 1 | Schematic illustrating the original FPU problem. Shown are the chain of coupled oscillators (top left) and the laser pulse analogue of this system (bottom left), together with the energy levels (middle; top and bottom) and recurrent coupling out from the input energy mode and then back in from the outer energy levels (circulating arrows). These modes correspond to different oscillation frequencies in the chain of coupled masses. The energy-level diagrams on the right (top and bottom) show the equipartition (thermalization) of energy across all levels that Fermi, Pasta, Ulam and Tsingou were expecting to observe but didn't.

of energy that oscillates between states on a regular basis. Interestingly, the initial study that revealed this strange behaviour played a pivotal role in the birth of computational physics, exploiting the nascent computing power developed by the Manhattan project (the US initiative to develop a nuclear weapon during the Second World War).

In 1953, Fermi, Pasta, Ulam and Tsingou used the Los Alamos MANIAC computer (previously used for performing calculations on thermonuclear processes) to simulate a one-dimensional chain of coupled masses connected by springs (Fig. 1) with a linear restoring force plus a weak nonlinear correction — a problem that could not be solved analytically. Their expectation was to verify Fermi's statistical prediction that the presence of the nonlinear correction would ultimately lead to an equipartition of the energy across all of the oscillating modes of the system — a phenomenon known as thermalization.

In hindsight, it is hard to decide whether this numerical experiment was more important for the very puzzling result that emerged from it or for the fact that it effectively ushered in the age of computational physics (or experimental mathematics, depending on your viewpoint). Today, physicists and engineers take for granted the ability to be able to perform numerical simulations and often expect or demand to see a numerical comparison with experimental results. But before 1953, this was an unheard-of practice and the attempt to numerically observe thermalization behaviour was truly pioneering.

The result from this simulation, however, showed an unexpected behaviour, which, in turn, has been the founding pillar for other discoveries and studies that continue to date. Instead of thermalizing, the system seemed to spread energy out and then periodically return to its initial state with energy flowing back to the more ordered configuration. These oscillations now come under the name of FPU recurrences (Fermi–Pasta–Ulam) after the trio of names on the 1955 report[2,3] that did not acknowledge Tsingou's contribution in the programming of MANIAC.

The stage was then set for one of the greatest puzzles in modern physics: why was the system not thermalizing as it should do, and what was the physics behind the recurrences?

The quest to answer these questions has led to many discoveries that have branched off in other directions and have become entire research fields of their own. In 1965, Kruskal and Zabusky[4] recognized the connection to the Korteweg–de Vries (KdV) equation and the existence of localized particle-like states called solitons that propagate invariantly though the system and thus break the ergodicity of the problem. It was later discovered that increasing the interaction strength of the system leads to chaotic behaviour and then eventually to the long-sought after thermalization.

More recently, attention has reverted back to experiments with the aim of studying FPU recurrences and physics in real systems. One of the goals is to verify the universality of the FPU problem and flesh out the details of any deviations and connections to solitons and chaos.

One of the main obstacles so far has been the presence of losses in experimental systems: dissipation dampens oscillations and can very rapidly quench any evidence of FPU recurrences. Alongside this difficulty with loss is that real experiments are typically hindered by technical restraints that prevent easy access to all of the system parameters and details, thus providing only a partial picture of the dynamics. A range of systems has been analysed in the past such as water waves (where the major issue is indeed losses)[5] and optical fibre systems[6]. Optical fibres have actually been one of the preferred systems for studying a whole range of physical concepts that are driven by weak nonlinearities and have provided the first ever experimental evidence for example of rogue waves[7] and soliton solutions such as the Peregrine soliton[8].

Now, Mussot et al. have devised a fibre-based system that provides an effectively lossless setting to study FPU dynamics while keeping track of the full details of the evolution with remarkable precision[1]. Losses are overcome by pumping the fibre with a second, counter-propagating laser beam that provides gain that is largest towards the end of the fibre where losses would otherwise be the highest. By carefully balancing the amount of power in the counter-propagating laser beam, they can effectively eliminate losses. At the same time, they keep track of the evolution of the system by monitoring the very small back-reflected (due to Rayleigh scattering) signal along the fibre length. This weak signal is interfered using a reference laser beam (that acts as a local oscillator for a homodyne detection measurement), thus making it possible to monitor the full amplitude and phase of the propagating beam in the fibre with a precision of 20 m along a total fibre length of 7.7 km.

Mussot et al. also inject an intense laser beam into their optical fibre: this beam plays the role of the initial oscillation mode in the FPU problem. They then inject a much weaker set of sidebands, which correspond to adding a small amount of oscillation in other modes in the FPU chain of oscillators (Fig. 1). The final part of their ingenious technical solution is a clever pulse-shaping approach that allows them to precisely control the amplitude and phase of the sidebands, thus allowing the sidebands to act as either an amplitude or as a frequency modulation of the main beam. These modulations, in turn, stimulate a process that in fibre optics is known as modulation instability (MI), whereby weak optical nonlinearity transfers energy from the main beam to the sidebands that gradually increase in number and amplitude. Sure enough, rather than continuously spreading out and 'thermalizing', the energy is seen to periodically oscillate back and forth, refocusing back into the main beam and then out again into the sidebands.

Controlling the nature of the initial state allows Mussot et al. to experimentally observe a feature that had never been observed before in optics, namely a symmetry breaking analogous to the famous Anderson–Higgs mechanism in particle physics and in condensed-matter physics. In fact, the dynamics of the MI can be simplified to a three-wave mixing process treating the MI sidebands as single peaks that exchange energy with the pump. The dynamics of this model show a transition between a parabolic potential to a Mexican-hat potential, depending on whether the frequency considered is outside or inside the MI sideband range, respectively.

Qualitatively different types of FPU recurrence are predicted, depending on the trajectories of the evolution of the system in the phase space. The experimental techniques used by Mussot et al. make it possible to effectively map out the phase space during the evolution of MI along the fibre and thus demonstrate that indeed the amplitude and frequency modulation input conditions lead to very different phase-space trajectories that are separated by a so-called homoclinic crossing point.

The system proposed by Mussot et al. represents a strong step forward in the study of FPU physics, providing a robust system that allows access to the full phase space and details of the recurrences. A key point that remains to be seen is the total distance over which this technique can be extended. In the present work, only two recurrences are observed rather than a long-scale evolution. Expanding this scale will be key for future studies aimed, for example, at investigating the transition to chaos around the homoclinic structure as well as the link to rogue wave formation and the evolution towards final thermalization. ❐

Daniele Faccio[1]* **and Fabio Biancalana**[2]*
[1]School of Physics and Astronomy, University of Glasgow, Glasgow, Scotland, UK. [2]School of Engineering and Physical Sciences, David Brewster Building, Heriot-Watt University, Edinburgh, Scotland, UK.
**e-mail: daniele.faccio@glasgow.ac.uk; f.biancalana@hw.ac.uk*

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VIEW FROM… ICON EUROPE 2018

The super-resolution debate

In the quest for nanoscopy with super-resolution, consensus from the imaging community is that super-resolution is not always needed and that scientists should choose an imaging technique based on their specific application.

Rachel Won

Despite the cold and wintry weather in Bielefeld, Germany, the second International Conference on Nanoscopy (ICON Europe), held 27 February–2 March, attracted more than 200 attendees, a significantly higher number than the last conference, from 21 countries. There were three keynote talks, 18 invited talks, 21 oral presentations and 40 poster presentations. The conference also received tremendous support from industry, with five company presentations and a marked increase in the number of exhibiting companies, proving how hot the research field of nanoscopy is, scientifically and commercially.

Nanoscopy, also known as super-resolution microscopy, has progressed rapidly in the past two decades, transforming biological research. Biological structures can now be viewed well below the diffraction limit, approaching virtually molecular resolution, and their dynamics can be examined on different timescales.

“Nanoscopy is certainly going to reveal processes and features in live-cell biology that have been obscured by blurry images due to the diffraction limit. Over the next 5–10 years, it will become an indispensable tool in cell biology research and give us a crisper picture of what actually is going on in the cell,” said Gregor Drummen from Bio and Nano-solutions, Germany, who was also one of the organizers.

Several interesting super-resolution imaging techniques were presented during the conference. One of them was DNA points accumulation for imaging in nanoscale topography (DNA-PAINT) by Ralf Jungmann from the Max Planck Institute of Biochemistry, Germany. The DNA-PAINT method involves a programmable DNA-based labelling probe for easy-to-implement super-resolution microscopy based on the localization of single molecules. In ‘classical’ single-molecule switching and localization approaches, such as stochastic optical reconstruction microscopy (STORM) or photoactivated localization microscopy (PALM), target-bound dye molecules are switched between so-called dark and bright states to temporally separate their fluorescence emission to enable stochastic super-resolution microscopy.

“However, this switching behaviour is highly dependent on the photophysical properties of the employed dyes, and is hard to predict and control in a programmable fashion. We use the transient hybridization of dye-labelled oligonucleotides to their complementary target-bound DNA strands. The binding is detected using single-molecule fluorescence and can be used to reconstruct super-resolution images. This not only allows the use of virtually any single-molecule-compatible dyes, as the dyes themselves aren’t photoswitched, but furthermore enables easy and spectrally unlimited multiplexing by using the exquisite programmability and specificity of DNA-binding interactions, a technique called Exchange-PAINT,” said Jungmann.

Jungmann further explained that one of the main drawbacks with using DNA-mediated ‘switching’ is that the so-called imager strands, which are dye-labelled oligos, are not fluorogenic, meaning that the dye-labelled oligos also emit fluorescence when they are not bound to their target strands. This limits DNA-PAINT effectively to selective plane illumination approaches, such as total-internal-reflection fluorescence microscopy or light-sheet microscopy. However, Jungmann also said that his group recently realized whole-cell DNA-PAINT using spinning disk confocal microscopes.

Another super-resolution imaging technique was presented by Jasmin Pape from the Max Planck Institute for