New horizons in non-linear optics: Helmholtz solitons, spatial fractals & attosecond pulses

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Abstract. We present a brief account of some recent theoretical advances made within the Institute for Materials Research in the field of *broadband non-linear optics*. Particular contexts include: angular aspects of non-linear beam propagation and interactions (Helmholtz soliton theory); the theory of spontaneous spatial optical fractals in non-linear systems; and a profoundly new regime of ultra-broadband multi-frequency Raman generation (that promises spontaneous attosecond pulse trains).

1. Introduction

In this Proceeding, we present an overview of some recent theoretical works that fall collectively under the banner of *broadband non-linear optics*. Firstly, the broad spatial bandwidth associated with obliquely multiplexed or interacting non-linear beams has recently proved to be a goldmine of new analytical developments. These include the derivation of *new families of exact analytical Helmholtz solitons*, new analytical descriptions of spatial Kerr solitons interacting at arbitrary angles, and a novel non-paraxial framework for modelling spatial beams at material interfaces. Secondly, the verification of our proposal of a *generic mechanism for the spontaneous formation of spatial optical fractals* will be outlined. It is useful to describe conventional pattern formation as "single-frequency" in this context, whereby the broadband (or "scale-less") case corresponds to fractal generation. New findings confirm our earlier predictions that the spontaneous fractal-generating mechanism is indeed independent of the details of either the system configuration or the particulars of the host medium non-linearity. Finally, our investigations of broadband Raman effects have recently uncovered a profoundly new regime of multi-frequency generation. It is now well-known that two-colour pumping can lead to the generation of an evolving white light spectrum. A remarkable new self-synchronization effect is reported, in which this bandwidth not only doubles in size but also self-locks to a fixed steady pattern. Some new predictions include a novel means for generating, what appear to be, the *shortest ever attosecond pulses*.

2. Theory of Helmholtz solitons

Paraxial models of optical beam propagation, such as those based upon the Non-Linear Schrödinger (NLS) equation, break down whenever non-paraxial effects can no longer be ignored. The derivation of NLS-type models involves the assumptions that beams are (a) broad compared to their carrier wavelength, (b) of sufficiently low intensity, and (c) propagating at negligibly small angles with respect to the reference direction. Collectively, these criteria constitute the paraxial approximation. When *any one* of these conditions is not met, a beam may be considered as non-paraxial. Over the past three decades, most literature on non-paraxial non-linear optics has been concerned with ultra-narrow beams, i.e. contexts where criterion (a) no longer holds. In this case, the field equation contains higher-order terms derived from order-of-magnitude analyses of Maxwell's equations.

The evolution of moderately intense, broad beams that propagate at non-negligible angles with respect to the reference direction defines a *Helmholtz non-paraxial scenario* [1-5]. Criteria (a) and (b) are then always met rigorously, so that the scalar approximation holds and narrow-beam corrections are unnecessary, but criterion (c) is relaxed. By omitting the slowly-varying envelope approximation [1,2], the electric field is governed by a Non-Linear Helmholtz (NLH) equation, in which the reference longitudinal (z) and single effective transverse (x) coordinates appear symmetrically [1,4]. There is thus no physical distinction between the spatial dimensions, as should be the case in uniform media, and beams may propagate and interact at *any angle* relative to the z-axis. For a Kerr medium, where the refractive index varies linearly with the beam intensity, the NLH equation can be written in normalized variables as:

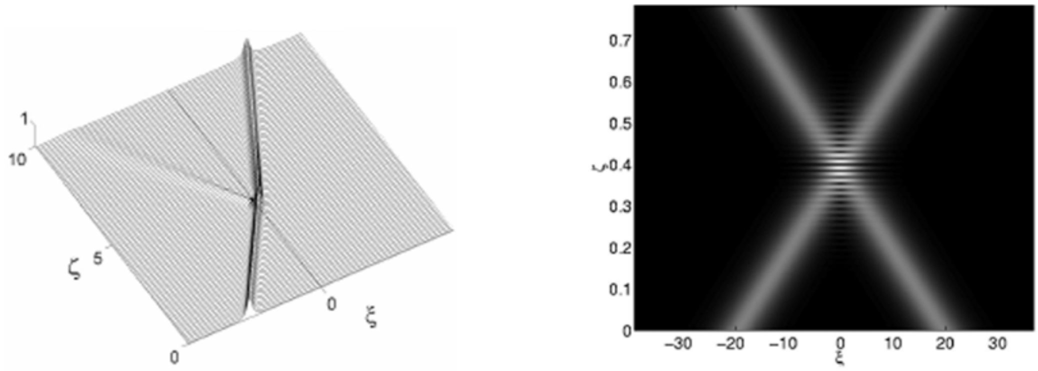
$$\kappa \frac{\partial^2 u}{\partial \zeta^2} + i\frac{\partial u}{\partial \zeta} + \frac{1}{2}\frac{\partial^2 u}{\partial \xi^2} \pm |u|^2 u = 0 . \qquad (1)$$

Here, $\zeta \propto z$ and $\xi \propto x$, and the non-paraxial (inverse-beam-size) parameter is $\kappa \propto (\lambda / w_0)^2$, where λ is the optical wavelength and w_0 the beam waist. u is the electric field envelope, and the $\pm$ sign flags a focusing or defocusing non-linearity, respectively. A key feature of the exact analytical soliton solutions of Eq. (1) [4,5], and other NLH equations, is the connection between the transverse velocity V of a beam in the scaled frame (ξ, ζ), and the propagation angle θ in the unscaled frame (x, z):

$$\tan\theta = \sqrt{2\kappa} V . \qquad (2)$$

The Helmholtz correction term $2\kappa V^2$ is determined solely by the propagation angle θ, and may be *arbitrarily large*. During significant off-axis evolution, one may have that $2\kappa V^2 \sim O(1)$, even though the inverse-beam-size parameter always satisfies $\kappa \ll O(1)$. The generic prevalence of the Helmholtz correction term in soliton solutions confirms our assertion that ultra-narrow-beam (κ-based) order-of-magnitude models are invalid in Helmholtz non-paraxial scenarios.

Our recent work on Helmholtz solitons has involved the derivation of a large number of novel, exact analytical soliton solutions to NLH equations with a range of non-linearities. Numerical simulations have also examined the stability properties of the new solutions. These solutions can be used as non-linear basis functions for modelling angular effects in a vast range of experimental configurations. The problem of 'solitons at interfaces' is a particularly important context where oblique-propagation considerations play a central role. NLS-based models of this fundamental geometry are restricted, by the paraxial approximation, to describing only vanishingly-small incidence angles. Our new Helmholtz formalism captures the *entire range of incidence angles* - see Fig. 1(a). The magnitude of corrections to paraxial-model predictions is discovered to be as large as 100%. Finally, we have considered arbitrary-angle interactions between solitons in both Kerr and non-Kerr media. These configurations are elementary in optics, but again such fundamental considerations lie outside the scope of conventional (paraxial) theory. We have developed new analytical descriptions of soliton interactions for *all non-paraxial angles* – see Fig. 1(b). New physical effects, arising directly from *x-z* symmetry, have also been predicted, and the robustness of Helmholtz solitons has been confirmed rigorously through full numerical simulations.



(a) **(b)**

Figure 1 (a) A spatial soliton beam incident (above the critical angle) on the interface separating two dissimilar Kerr media. (b) Counter-propagating solitons at a non-paraxial interaction angle of roughly 24º.

3. Spontaneous spatial optical fractals

Fractals are self-similar mathematical patterns that *lack* a characteristic scale length. An alternative viewpoint is that such patterns possess structure across *all* scale lengths. Although self-similarity cannot continue indefinitely down to arbitrarily small scales in physical systems (ultimately, due to finite atomic dimensions or diffraction limits), real-world fractals often possess structure across decades of scale, justifying the term *fractal* in their description.

In a recent paper [6], we reported the first spontaneous spatial optical fractals in transverse pattern formation. By considering pattern-formation instability threshold curves of a simple system – the classic diffusive-Kerr slice with a single feedback mirror – we proposed a universal non-linear mechanism for fractal generation in generic systems with feedback. This *spontaneous fractal-generating capacity* is present whenever the spectrum possesses a large number of comparable spatial-frequency instability minima. The Kerr-slice fractals are quite distinct from the transverse fractal eigenmodes of unstable-cavity lasers that we discovered earlier [7-10]. On the one hand, unstable-cavity fractals may be regarded as a *linear* superposition of diffraction patterns with different scale-lengths, which arise from successive round-trip magnifications of an initial diffractive seed. On the other hand, fractals formed in the Kerr slice result entirely from *intrinsic non-linear dynamics* (i.e. light-matter coupling leading to harmonic-generation and/or four-wave-mixing cascades). These processes conspire to generate new spatial frequencies that, in turn, can produce optical structure on smaller and smaller scales, down to the order of the optical wavelength.

We have extended our initial considerations of the Kerr-feedback-mirror system to ring-cavity geometries, where a thin slice of non-linear medium (diffusive Kerr or Maxwell-Bloch saturable absorber) is placed in the free-space path inside the cavity. New linear analyses have shown that the transverse instability spectra of these systems possess the requisite comparable minima that predict the capacity for spontaneous generation of optical fractals. Extensive numerical simulations (including both one and two transverse dimensions) have verified that both the dispersive and absorptive cavity systems do indeed give rise to non-linear optical fractals in the transverse plane (see Fig. 2). Like their feedback-mirror counterparts, the new cavity fractals can be characterized by the properties of the nonlinear material.

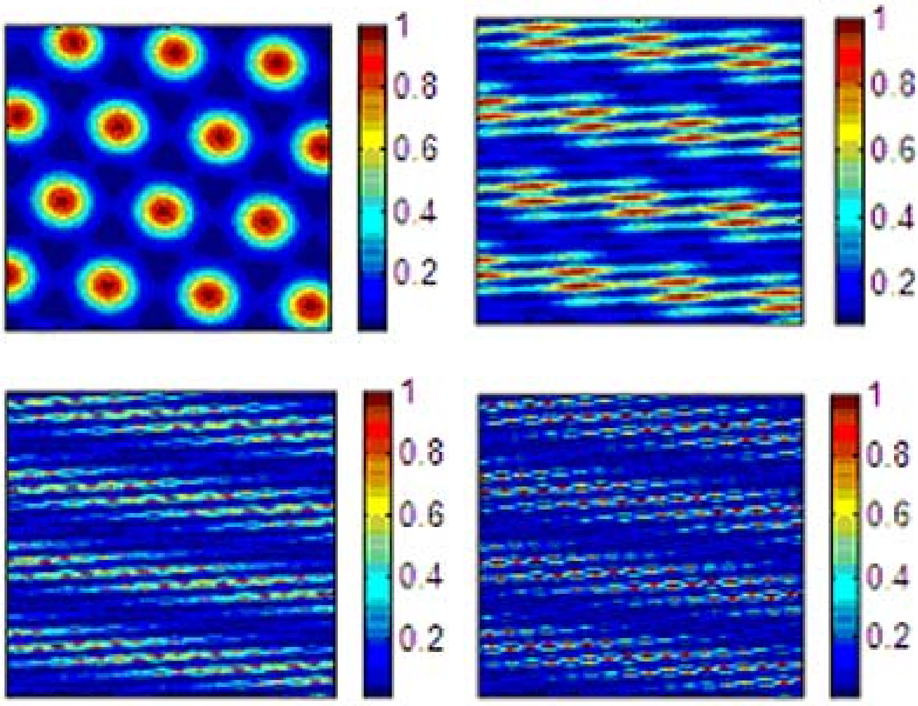


Figure 2 Transverse intensity distribution showing the transition from a conventional (single-*K*) pattern – a hexagonal array– to a fractal mode in a thin-slice Maxwell-Bloch ring cavity. Self-similarity persists down to spatial scales of the order of the optical wavelength.

4. New ultra-broadband Raman effects and attosecond pulse trains

Ultra-broadband Multi-frequency Raman Generation (UMRG) is a known technique for producing temporal bandwidths that can be of the order of the pump frequency. It employs a resonant 2-colour pumping scheme, in which a Raman-active medium is driven by two near-identically-shaped input pulses, whose frequency separation is close to matching the Raman transition with the highest potential gain. This configuration leads to a multi-fold increase in the conversion efficiency over conventional stimulated Raman geometries. The latter tend to involve a single strong pump beam and a much weaker Stokes seed. UMRG optimises the conditions for bandwidth production, with a combination of parametric and non-parametric processes leading to a rapid redistribution of the input energy to higher-order Stokes and anti-Stokes lines.

When transient dynamics and material dispersion are neglected, theory predicts that 2-colour excitation of the S(1) rotational transition of H_2 gas at 1 atm pressure leads to over 30 Raman sidebands of *comparable energy*. By allowing for transiency and dispersive effects, this figure can increase to nearly 50 distinct lines [11-15]. In other Raman media, even denser broadband spectra may be achieved. In air at 1 atm pressure, for example, multi-frequency beams with in excess of 100 comparable-energy components are predicted [16].

Our more recent work has included the derivation of a new exact analytical solution to the plane-wave UMRG equations, in the absence of background dispersion but when a finite amount of detuning is introduced. Full numerical simulations have confirmed our theoretical predictions. We have also considered a new fundamental modification to the UMRG process through inclusion of feedback effects. Our preliminary results suggest that such effects lead to a profoundly new regime of multi-frequency generation (at least in the Raman context, and potentially in a much wider range of non-linear systems). A comparison with the standard broadband Raman effect is shown in Fig. 3. Firstly, modelling predicts that the magnitude of the *bandwidth generated may be enhanced by up to 100%.*

Even more significant is the *discovery of self-synchronization* of the spectrum generated. This latter feature can lead to trains of mode-locked pulses whose individual durations are of the order of attoseconds. By considering the interplay between a wide range of physical processes, supplemented by the novel feedback mechanism, this new regime of feedback-UMRG has the potential to furnish laser sources for probing the dynamical properties of various systems on timescales that are dramatically shorter than existing ultra-fast laser systems.

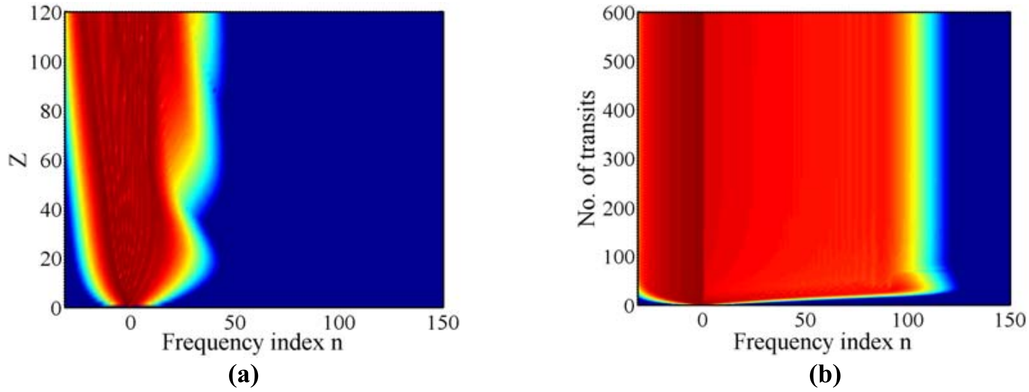


Figure 3 (a) Bandwidth generation in H_2 gas, using the conventional 2-pump arrangement for a Raman cell of normalized length *Z* = 120. (b) Bandwidth enhancement and the self-synchronisation effect arising from our novel feedback-modified system - all other parameters are the equivalent of those used in part (a) of this figure.

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