Localizing Energy Through Nonlinearity and Discreteness

Intrinsic localized modes have been theoretical constructs for more than a decade. Only recently have they been observed in physical systems as distinct as charge-transfer solids, Josephson junctions, photonic structures, and micromechanical oscillator arrays.

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In solid-state physics, the phenomenon of localization is usually perceived as arising from extrinsic disorder that breaks the discrete translational invariance of the perfect crystal lattice. Familiar examples include the localized vibrational phonon modes around impurities or defects (such as atomic vacancies or interstitial atoms) in crystals and Anderson localization of electrons in disordered media. The usual perception among solid-state researchers is that, in perfect lattices—those free of extrinsic defects—phonons and electrons exist only in extended, plane wave states. That notion extends to any periodic structure, such as a photonic crystal or a periodic array of optical waveguides. Such firmly entrenched perceptions were severely jolted in the late 1980s by the discovery that intrinsic localized modes (ILMs), also known as discrete breathers (DBs), are, in fact, typical excitations in perfectly periodic but strongly nonlinear systems.

The past several years have seen this prediction confirmed by a flood of experimental observations of ILMs in physical systems ranging from electronic and magnetic solids, through microengineered structures including Josephson junctions and optical waveguide arrays, to laser-induced photonic crystals. Experimentalists are currently hot on the trail of ILMs in Bose–Einstein condensates (BECs) and biopolymers. Hopes are high that these exotic excitations will be useful in all-optical logic and switching devices and in targeted breaking of chemical bonds, and will prove helpful to the understanding of melting processes in solids and conformational changes in biomolecules.

In this brief overview, we have attempted to capture the excitement and to explain the essence of these remarkable nonlinear excitations. We urge readers to consult some of the several pioneering papers, recent reviews, and Web sites for more details.

Intuition and theory

A good working definition of ILMs (or DBs) is that they are spatially localized, time-periodic, and stable (or at least long-lived) excitations in spatially extended, perfectly periodic, discrete systems. The existence of two distinct names for the same phenomenon is an indication that separate historical paths led to their discovery and provides key insights into the reasons for their existence. A DB is a localized, oscillatory excitation that is stabilized against decay by the discrete nature of the periodic lattice. Box 1 discusses this path to DBs in more detail. An ILM is an excitation that is localized in space by the intrinsic nonlinearity of the medium, rather than by a defect or impurity. Box 2 reviews this path to ILMs.

By the early 1990s, researchers following these two paths had converged on the insight that stable localized periodic modes, whether called ILMs or DBs, were generic excitations in discrete nonlinear systems, and that to study them systematically, one should start with a system of uncoupled nonlinear oscillators—the “anti-continuum limit”—and treat the coupling as a weak perturbation.

To pursue this insight, consider the simple problem of a diatomic molecule, or dimer, modeled initially by a classical system of two coupled anharmonic oscillators. First, imagine that the interoscillator coupling is switched off; that leaves two independent nonlinear oscillators. The nonlinearity of the oscillators means that the frequency of their motion depends on the amplitude or, equivalently, the input energy. In the case of the familiar simple but nonlinear pendulum, for example, the period varies from the discrete nature of the periodic lattice. Box 1 reviews some of the several pioneering papers, recent reviews, and Web sites for more details.

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strength given by $1/(\Delta x)^2$. The notation is a deliberate reminder that our model includes a finite spacing between molecules; the formal continuum limit is obtained by taking $\Delta x \to 0$, in which case the entire second term in the equation becomes $\partial^2 \phi(x,t)/\partial x^2$. As usual, we start by studying the system’s small (linear) oscillations. The local oscillator at each site corresponds to a double-well potential, with degenerate minima at $\phi_0 = \pm 1$. Expanding around the minimum at $\phi_0 = 1$, we obtain the spectrum of the linear waves, $\omega_q^2 = 2 + (2/\Delta x)^2 \sin^2(q/2)$. The linear spectrum consists of a band that is limited by two cutoff frequencies, $2 < \omega_q^2 < 2 + (2/\Delta x)^2$, and is bounded above and below; the upper cutoff arises solely from the effect of discreteness.

As with the plane pendulum, the double-well quartic oscillator has a frequency that, for small oscillations around the minimum, decreases with increasing amplitude of the motion. That means one can create a localized periodic oscillation at a frequency $\omega_q$ lying below the spectrum of linear oscillations, as shown in the bottom panels of figure 1. Setting just one of the nonlinear oscillators in motion at a fairly small amplitude will do the job, so that its frequency is just smaller than the smallest allowed linear frequency. Then if the coupling between the sites is weak enough—$\Delta x$ is large, producing a very narrow band—not only will the fundamental frequency $\omega_q$ of this ILM be below the band of allowed linear excitations, but all harmonics of $\omega_q$ will be above the band. Hence, there will be no possibility of a linear coupling to the extended modes, even in the limit of an infinite system when the spectrum $\omega_q$ becomes dense. This means that the ILM cannot decay by emitting linear waves (that is, phonons) and is hence linearly stable.

To understand the high-energy ILMs that also occur and are shown in the top panels of figure 1, consider again the limit of large $\Delta x$, which creates a weak coupling between the oscillators. If we set a single oscillator in motion, but now with large amplitude, the quartic nature of the potential implies that the frequency of the ILM will increase with increasing amplitude. For large enough amplitude, the frequency will move above the highest frequency in the very narrow—because $\Delta x$ is large—band of linear excitations. For this large-amplitude ILM, all higher harmonics lie above the band because the fundamental frequency does, and so the excitation is linearly stable.

The simple quartic model illustrates how the two critical components—nonlinearity and discreteness—can combine to make ILMs possible. Nonlinearity allows strongly excited local modes to have a fundamental frequency outside the spectrum of small oscillations, and the finite extent of the spectrum in a discrete system allows all higher harmonics of the ILM also to lie outside the linear spectrum. The locations of these frequencies prevent the resonances that, in general, destroy the continuum breathers discussed in box 1. This intuitive understanding of the origin of ILMs/DBs in discrete nonlinear systems was presented in the pioneering paper of Albert Sievers and Shozo Takeno in 1988.

Over the intervening years, both analytic and numeri-
I.M. studies have explored the existence and properties of ILMs in a variety of nonlinear mathematical models of physical systems. Robert MacKay and Serge Aubry, for example, rigorously proved the existence of DBs in networks of weakly coupled anharmonic oscillators. Remarkably, their theorems are insensitive to the lattice dimension: Unlike continuum breathers (see box 1), which occur only in highly constrained 1D systems, DBs are equally common in two and three dimensions.

Additional analytic results established that ILMs occur in one-parameter families, are dynamically stable with respect to perturbations, and are structurally stable with respect to changes of the equations of motion. Combined analytic and numerical studies showed that ILMs act as strong, frequency-dependent scatterers of plane waves and can be quantized. Theoretical studies by Ding Chen (Saclay) and his collaborators gave an explicit algorithm for moving ILMs along the lattice, and calculations of Michel Peyrard (ENS, Lyon) established that ILMs can be generated from thermal fluctuations. The Chen and Peyrard results suggest that ILMs may play critical roles in the transport of energy and other dynamical properties of nonlinear discrete systems, such as melting transitions in solids and folding in polypeptide chains.

**Experimental observations of ILMs**

Much of the present excitement surrounding ILMs comes from numerous recent experimental observations in physical systems. Those systems range from solid state mixed-valence transition metal complexes and quasi-1D antiferromagnetic chains, through arrays of Josephson junctions and micromechanical oscillators, to optical waveguide systems and 2D photonic structures. We look briefly at the nature of ILMs in several of these distinct systems.

**Solids.** The natural periodicity of solid-state materials makes them obvious targets to search for ILMs. But their small length scales and quantum effects make observing or visualizing the excitations experimentally challenging. As discussed in box 3, one quantum manifestation of localization is a redshift in the frequencies of many-phonon excited vibrational states. In recent experiments, Basil Swanson (Los Alamos National Laboratory) and colleagues and K. Kisoda (Wakayama University) found strong evidence for the existence of ILMs in the charge-transfer solid PtCl by measuring resonance Raman spectra. The redshift in the resonances was obtained for up to seven participating optical phonons, indicating the localized excitation of Pt–Cl bonds.

Using a phenomenological model, theorists including Konstantin Kladko (Ingrian Networks) and Nikos Vrangakakis (University of Crete) and their collaborators have reached quantitative agreement with those experiments. But challenges remain for theorists to develop a more microscopic model and for experimentalists to measure explicitly the spatial extent of the ILM. Raman spectroscopy measures only frequency shifts, from which one infers localization.

In magnetic solids, one expects to find localized spin-wave modes that are the direct spin analogs of the ILM phonon modes. Ulrich Schwarz (Cornell University) and his collaborators studied the quasi-1D biaxial antiferromagnet (C6H12NH3)2CuCl4 by driving it with a microwave pulse at the lowest antiferromagnetic resonance frequency of 1.5 GHz. High-intensity microwave pulses drive the spatially uniform antiferromagnetic resonance into a nonlinear region, where it becomes unstable. According to numerical simulations, the resonance decays into a broad spectrum of ILMs, which correspond to localized spin states. Measuring the time-delayed absorption spectra reveals the corresponding redshifts together with observable lifetimes of ILMs up to milliseconds. As in the case of the

**Figure 2. Experimental and schematic images of intrinsic localized modes in an annular Josephson ladder driven by a DC current. In the schematics, Josephson junctions (each about 3 μm wide) lie at the midpoints of each green line segment that signifies superconducting leads connecting the junctions. A pure green ladder in the schematic and a pure green background in the data would signal the superconducting state of all junctions at low DC current—the linear regime. But a large DC current switches some of the junctions into a resistive state that supports a voltage across the junction; in the schematic, red and yellow dots denote resistive junctions having different voltages. These resistive junctions are discrete breather excitations localized at various sites on the ladder, here imaged using a scanning laser microscope. Among the many possible DB states, these data show four: (a) a highly excited, spatially homogeneous resistive state and (b–d) localized states corresponding to several distinct DBs. (Figure adapted from A. Ustinov, ref. 9.)**
Image 29x535 to 353x720

Figure 3. An intrinsic localized mode (ILM) emerges in the output of an optical planar waveguide array (shown on edge) as the power from input laser light increases into the strongly nonlinear regime. Each waveguide of the array extends out of the page, with the laser input (from behind the page) focused near the center. (a) For a laser power of 70 W, linear propagation occurs and the excitation of modes diffracts over the entire array. (b) At intermediate power (320 W), the output pulse narrows, showing increased localization effects. (c) At high power (500 W), the nonlinear nature of the optical medium localizes the output in an ILM located around the input waveguide. (Figure adapted from H. S. Eisenberg et al., ref. 11.)

Box 1. From Continuum to Discrete Breathers

The term “breather” was first applied to a particular solution of the celebrated sine–Gordon equation (SGE)

$$\frac{\partial^2 \theta}{\partial t^2} - \frac{\partial^2 \theta}{\partial x^2} + \sin \theta = 0,$$

which is one of the now famous “soliton” equations (the equation coined by Norman Zabusky of the University of Pittsburgh and Martin Kruskal of Rutgers University). Despite their nonlinearity, such equations are essentially solvable analytically because they correspond to completely integrable, infinite-degree-of-freedom Hamiltonian systems. To appreciate the properties of the nonlinear equation, consider the explicit form of the SGE breather solution:

$$\theta_b(x, t) = 4 \tan^{-1}\left( \frac{\varepsilon \sin(t/\sqrt{1+\varepsilon^2})}{\cosh\left(\varepsilon(x-x_0)/\sqrt{1+\varepsilon^2}\right)} \right).$$

Straightforward but tedious differentiation shows that $\theta_b(x, t)$ solves the SGE exactly for any value of $\varepsilon$. Because the solution $\theta_b$ is clearly periodic in time and exponentially localized in space around $x = x_0$, it satisfies the definition of a breather given in the introduction.

In the limit of small values of $\varepsilon$, for which $\sin \theta \approx \theta$, the SGE reduces to a linear Klein–Gordon equation, the excitations of which are extended plane waves characterized by the dispersion relation $\omega(q) = \sqrt{T + q^2}$. Notice that the spectrum of these plane-wave excitations has a gap and extends to infinite frequency as $q \to \infty$. The fundamental frequency $\omega_b$ of the breather $\theta_b$ always lies in the gap of the linear spectrum: $\omega_b = 1/\sqrt{T + \varepsilon^2} < 1$. However, independent of the value of $\varepsilon$, sufficiently high harmonics of the fundamental frequency must always lie within the spectrum of the linear excitations. And because $\theta_b(x, t)$ contains all odd harmonics of $\omega_b$, that localized mode would appear to couple (albeit in high orders of $\varepsilon$) to the delocalized plane waves. In fact, these couplings all vanish, and the SGE breather remains stable.

Early in the study of solitons, the question arose whether continuum equations other than the SGE could possess exact breather solutions. The most studied case was that of the so-called $\phi^4$ equation, which is just the continuum version of the equation on page 43, a model of coupled nonlinear oscillators. Is there a breather in $\phi^4$? That seemingly simple question launched a multiyear odyssey in computational and mathematical physics. Most of the soliton community argued that there could not be a breather in $\phi^4$ because it was not completely integrable, but most of the computational physicists said there could be, based on evidence from their numerical simulations. An expansion to all orders in $\varepsilon$ suggested the existence of a $\phi^4$ breather, but a “beyond-all-orders” analysis showed that terms proportional to $\exp(-1/\varepsilon)$ produced a coupling to linear plane-wave excitations and caused the putative breather to decay. Subsequently, rigorous results established that the SGE was indeed exceptional and that stable breathers did not exist in other continuum Klein–Gordon theories. But computer simulations in which continuum equations were discretized to be solved numerically provided strong hints that, for a sufficiently large discretization, discrete breathers might well exist. That insight helped motivate the studies described in the main text.
The propagation of photons of certain wavelengths is forbidden—a theoretical study has predicted the existence of ILMs. 12 In the experiment, the individual waveguides are made from aluminum gallium arsenide and are 4 μm wide and a few millimeters long. Arrays contain typically 40–60 waveguides; the strength of the coupling between neighboring waveguides is controlled by their spacing, which varies from 2 to 7 μm. Yaron Silberberg (Weizmann Institute) and his collaborators injected light from a synchronously pumped laser into a single waveguide on the input side of a 6-mm-long sample and recorded the light distribution registered at the output facets (see figure 3). At low light power, the propagation is linear, and the light expands over all waveguides at the output. As the input power increases above a threshold, the width of the output distribution shrinks. And at a power greater than 500 W, the highly localized nonlinear mode confines the light to about three waveguides around the input waveguide; that light output is the signature of an ILM, which, in this context, has been called a discrete soliton.

Photonic crystals—periodic materials in which the propagation of photons of certain wavelengths is forbidden—are another important optical system in which theoretical studies have predicted the existence of ILMs. 12 The optical analog of semiconductors, these artificial crystalline structures provide novel and unique ways of controlling many aspects of electromagnetic radiation, including the possibility of light-induced radiation control—that is, the use of light to switch and channel light. Two years ago, Sergei Mingaleev (now at the University of Karlsruhe) and one of us (Kivshar) predicted the nature of the ILMs that could occur in a composite photonic crystal formed by a regular 2D lattice of rods of two different types of semiconductors. That work awaits experimental confirmation. However, experimental groups led by Mordechai Segev (Technion) and Zhigang Chen (San Francisco State University) have recently used optical induction in a homogeneous nonlinear medium to confirm the existence of this type of ILM in an analog of a 2D nonlinear photonic crystal. Segev’s group used a photorefractive crystal with a strong electro-optic anisotropy to create a lattice with a polarization in the nonelectro-optic direction and orthogonal to that of a probe beam. They observed the light localization in the form of 2D discrete solitons. Similarly, Chen’s group used a nonlinear lattice, created by partially incoherent light, to observe strong localization of one of the waveguides and the continuous variable (time in the equation in box 2) corresponds to the spatial coordinate along the waveguide.

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Box 2. Defect Modes Versus Self-Trapping

Bloch’s theorem guarantees that electrons moving in perfectly periodic, rigid lattices will exist in spatially extended states. But if the lattice is deformable and perfect periodicity lost, the theorem does not apply. In a brief note published in 1933, Lev Landau first described what later became known as the polaron by noting that an electron could be trapped at a strongly distorted location of the lattice. Importantly, Landau did not distinguish between a distortion caused by an extrinsic defect or impurity and one caused by the intrinsic self-trapping of an electron due to the effective nonlinearities that arise from strong coupling to the lattice phonons; either distortion could cause localization. In 1959, Ted Holstein developed an explicit model for this nonlinear self-trapping in one-dimensional lattices and established within certain approximations that the electron’s wavefunction obeyed an equation now known as the discrete nonlinear Schrödinger equation (DNLSE):

$$i\frac{d\psi_n}{dt} + \frac{1}{2}\psi_n - 2\psi_n + \psi_{n-1} + \kappa |\psi_n|^2 \psi_n = 0,$$

where $\psi_n$ is the component of the electron’s wavefunction at lattice site $n$, $f$ represents the electron transfer (hopping) between adjacent lattice sites, and the nonlinear term proportional to $\kappa$ arises from the coupling of the electron to the lattice, which is assumed to be deformable.

The literature on the polaron and the DNLSE is voluminous, so we confine ourselves to just two points that are most relevant to intrinsic localized modes. First, in the final term of this equation, if $|\psi_n|^2$ is replaced by a potential, $V_n$, the equation becomes a discrete version of the standard time-dependent Schrödinger equation. If that potential $V_n$ is localized around some lattice site $n_0$, then it becomes in essence an intrinsic defect that acts just like an extrinsic defect or impurity, able to produce an electronic bound state localized around $n_0$. This self-trapped object is precisely the polaron. Second, the nature of nonlinearity in the DNLSE allows for solutions of the form $e^{i\beta t}$. The absence of higher harmonics means that the spatially localized, time-periodic solutions to the equation are trivial examples of discrete breathers.
Francesco Cataliotti and colleagues strongly suggest that atomic potential is repulsive. Recent experiments by 2). ILMs can therefore be created even if the BEC dynamics are governed by a variant of the DNLSE (see box 48). The recent theoretical, numerical, and experimental results, and the innovative concepts associated with the physics of ILMs have shed considerable light on the complex dynamics, properties, and functions of nonlinear discrete physical systems—from the nanoscale to the macroscale. The study of such systems, and the ILMs they support, underpins applications ranging from smart materials that respond collectively to external stimuli in a coherent, tunable fashion to light-induced, all-optical networks. Only a few years ago, ILMs were almost exclusively the province of theorists. Today, the rapidly expanding list of experimental observations not only establishes the ubiquity of intrinsic localized modes in nonlinear, discrete physical systems but also generates exciting possibilities for future applications both in fundamental science and in technology. Clearly, for ILMs, the best is yet to be.

We have enjoyed many helpful discussions with our colleagues and coauthors who contributed to the field discussed in this article. In particular, we recognize Serge Aubry, Alan Bishop, Oleg Braun, Zhigang Chen, Konstantin Kladko, Arnold Kosevich, Robert MacKay, Sergey Mingaleev, Elena Ostrovskaya, Alexander Ochinnikov, Michel Peyrard, Al Scott, Mordechai Segev, Al Sievers, Yaron Silberberg, Andrey Sukhorukov, Shozo Takeno, George Tsiironis, and Alexey Ustinov.

References


Box 3. Local Modes in Small Molecules

The role that nonlinearity, in the form of anharmonic forces, could play in localization of excitations in small molecules was recognized as early as the 1920s.4 Nonetheless, the idea experienced a checkered history of rejection and rediscovery characteristic of counterintuitive nonlinear phenomena; as late as the 1970s, experimental results that essentially proved the existence of localized modes were still being dismissed as theoretically impossible.

The classic example of nonlinearity in small molecules is the C–H vibrational mode in benzene. This case has been studied primarily in terms of quantum mechanical nonlinear oscillators, and thus establishes a link between our primarily classical analysis of intrinsic localized modes and their existence in quantum systems. From elementary quantum mechanics, one would expect that the wavefunction describing the lowest quantum mode associated with the C–H stretch would have equal amplitude at each of the six possible CH units in the benzene molecule—that is, it would be delocalized around the benzene ring. And this is indeed the case for low-amplitude modes, which, in quantum mechanical terms, refer to excitations containing a small number of phonons. But for large-amplitude motions—those in the nonlinear regime that involve many phonons—the modes are more accurately described by an excitation localized on a single CH unit. The frequencies of the nonlinear modes are shifted to frequencies lower than a simple multiple of the fundamental phonon frequency. And that redshift of the frequencies provides key indirect evidence for the localization. Although these modes are not infinitely stable, they can be very long-lived and are certainly relevant to experiments, not only in small molecules but in molecular crystals and biopolymers.

localization in the regime of large nonlinearities (see figure 4). Both sets of results illustrate the tremendous promise for optically excited and controllable nonlinear localized states as elements of future all-optical logic and switching devices.

The best is yet to be

Armed with the fundamental understanding of the origin of ILMs, experimentalists are becoming increasingly proficient at discovering—or creating—new physical systems in which to study ILMs and their properties. For instance, in a recent experiment designed to explore the important issues of creation, transport, mobility, and interactions of ILMs, a group led by Sievers constructed an array of micromechanical oscillators.10 Nearly identical initial experimental conditions led to ILMs located at widely different sites, an important confirmation that ILMs are in fact localized by intrinsic nonlinear effects rather than by disorder or impurities.

The ability to create periodic optical lattices in which to trap BECs immediately suggests the possibility of creating and observing ILMs in those systems.14 Analytic and computational studies of the dynamical phase diagram of a dilute BEC trapped in a multwell periodic potential reveal that, in the framework of the Gross–Pitaevskii equation (that standard equation in to study BECs), the dynamics are governed by a variant of the DNLSE (see box 2). ILMs can therefore be created even if the BEC’s interatomic potential is repulsive. Recent experiments by Francesco Cataliotti and colleagues strongly suggest that an increase of the BEC density will lead to the generation of ILMs in these systems.

The possible roles of ILMs in biopolymers have also been a focus of concerted theoretical and experimental efforts.15 For instance, the conformational changes and buckling of long biopolymer molecules may occur in response to the excitation of nonlinear localized modes. Conformal flexibility is a fundamental property that differentiates polymers from small molecules and gives rise to many of their remarkable properties. A distinctive feature of biological polymers is the complex structure of their elementary sub-units; that structure can support long-lived nonlinear excitations. ILM excitations have been discussed in connection with the storage and transport of the energy released during adenosine triphosphate hydrolysis, with the local opening of the DNA double-helix, and with the buckling, folding, and collapse of a biopolymer chain to a compact coil, as shown in figure 5. Theoretical results predict that these effects survive in the presence of viscous damping. Thus, although it is speculative, this application of ILMs may prove to be crucial in the kinetics of conformational phase transitions of semiflexible biopolymers in solutions.

The recent theoretical, numerical, and experimental results, and the innovative concepts associated with the physics of ILMs have shed considerable light on the complex dynamics, properties, and functions of nonlinear discrete physical systems—from the nanoscale to the macroscale. The study of such systems, and the ILMs they support, underpins applications ranging from smart materials that respond collectively to external stimuli in a coherent, tunable fashion to light-induced, all-optical networks. Only a few years ago, ILMs were almost exclusively the province of theorists. Today, the rapidly expanding list of experimental observations not only establishes the ubiquity of intrinsic localized modes in nonlinear, discrete physical systems but also generates exciting possibilities for future applications both in fundamental science and in technology. Clearly, for ILMs, the best is yet to be.
t = 200

$\subset$

Figure 5. A biopolymer chain buckles and folds in on itself due to an instability produced by a moving nonlinear localized mode. That mode started as a single-particle excitation at the left end of the 30-particle chain and propagated to the middle. Marked particles indicate the location of the moving ILM excitation as a function of the time $t$ (in dimensionless units). (Figure adapted from S. Mingaleev et al., ref. 15.)