

Nonequilibrium Precondensation of Classical Waves in Two Dimensions Propagating through Atomic Vapors

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The nonlinear Schrödinger equation, used to describe the dynamics of quantum fluids, is known to be valid not only for massive particles but also for the propagation of light in a nonlinear medium, predicting condensation of classical waves. Here we report on the initial evolution of random waves with Gaussian statistics using atomic vapors as an efficient two dimensional nonlinear medium. Experimental and theoretical analysis of near field images reveal a phenomenon of nonequilibrium precondensation, characterized by a fast relaxation towards a precondensate fraction of up to 75%. Such precondensation is in contrast to complete thermalization to the Rayleigh-Jeans equilibrium distribution, requiring prohibitive long interaction lengths.

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Bose-Einstein condensation (BEC) has been reported in a variety of quantum systems, such as ultracold atoms and molecules [1], exciton polaritons [2–4], or photons [5], where the bosonic character of the particles is crucial. On the other hand, it is known that an ensemble of classical waves can exhibit a phenomenon of condensation, whose thermodynamic properties are analogous to those of the genuine quantum BEC, despite the classical nature of the system [6–15]. Indeed for waves traveling in random directions in a nonlinear medium, wave thermalization and condensation can occur. Such spontaneous formation of large scale coherent structures is encountered in many fields of physics, such as astrophysics, low-temperature condensed matter, hydrodynamics, plasma physics, and optics. As a remarkable fact, in spite of its formal reversibility, a nonintegrable Hamiltonian system can exhibit self-organization induced by its natural thermalization towards the equilibrium state [6–10,12,14–20]. Wave condensation is a spectacular example of this type of self-organization processes, which results from the divergence of the classical Rayleigh-Jeans equilibrium distribution.

Here we present an experimental system allowing us to study the time evolution of such wave condensation in two dimensions. In contrast to ultracold atom experiments, the wave under consideration is the electromagnetic field of a laser beam, rendered spatially incoherent by passing through a diffuser. At variance with many ultracold atom experiments, we also consider a situation here without an external potential.

An important aspect of this experimental work is the study of fast relaxation to out of equilibrium states in the initial process of two-dimensional thermalization. Indeed,

achieving complete thermalization and condensation of random nonlinear waves through nonlinear optical propagation is known to require prohibitive large interaction lengths [17–19]. The existence of fast relaxation to out of equilibrium states is an open problem that is attracting a growing interest in different research communities [21–24], including long range interacting systems, where fast relaxation occurs towards quasistationary states [25,26] or one-dimensional, nearly integrable quantum systems, where experimental signatures of prethermalization have been observed [27–29]. At variance with the usual approach characterizing wave condensation in the far field spectrum, here we identify a fast initial relaxation through the analysis of the optical near field, which reveals the existence of a phenomenon of precondensation that occurs far from thermal equilibrium for short propagation lengths. Our work thus contributes an experimental observation (supported by numerical simulations) of fast relaxation to out of equilibrium states.

The nonlinear Schrödinger equation (NLS) describing the experiment can be rewritten as [30]:

$$i \frac{\partial \psi}{\partial z} = -\frac{1}{2k_0} \nabla^2 \psi + \gamma |\psi|^2 \psi, \quad (1)$$

where ∇ is the gradient in the transverse surface section of the beam $\mathbf{r} = (x, y)$, while the longitudinal variable z plays the role of the time evolution. The wavelength of the optical wave is $\lambda = 2\pi/k_0$, and γ describes the strength of the nonlinearity.

The incident speckle field is characterized by a transverse correlation length σ_c , which also determines the

initial transverse momentum distribution. The healing length denotes the relevant transverse length scale for which linear and nonlinear effects are of the same order $\Lambda = \sqrt{z_{NL}/(2k_0)}$, where $z_{NL} = 1/(\gamma I_0)$ is the nonlinear length scale, and $I_0 = \langle |\psi|^2 \rangle$ is the intensity averaged over the relevant transverse surface of the beam (see below). In this Letter, we consider defocusing nonlinearities ($\gamma > 0$) corresponding to repulsive interactions. We recall that, in addition to the intensity I_0 (‘particle number’), the NLS Eq. (1) also conserves the total energy (Hamiltonian) $H = E + U$, which has a kinetic (linear) contribution $E(z) = (1/2k_0) \int |\nabla\psi|^2 d\mathbf{r}$, and a nonlinear contribution $U(z) = (\gamma/2) \int |\psi|^4 d\mathbf{r}$.

We stress that this type of classical wave condensation occurs in the spatial frequency domain and at the same wavelength as that of the incident laser. This is in contrast to the condensation of photons reported in [5], where the effect of condensation also occurs for temporal frequencies and is accompanied with inelastic light scattering via interactions with a thermal bath. In the situation considered here, no thermal bath is present; we deal with a microcanonical statistical description, where the total energy H plays a role analogous to the temperature (note that, in analogy with kinetic gas theory, the kinetic energy $E(z)$ provides a natural measure of the amount of randomness in the incoherent wave). This is a key difference with respect to the broader notion of condensation used to characterize different phenomena in optical cavities, which are inherently forced-dissipative systems [4,43–47]. The possibility to engineer the initial conditions for the nonlinear propagation illustrates the potential of this experiment to explore novel regimes of the universal two dimensional NLS equations.

The experimental setup is depicted in Fig. 1. The output of a 1 W fibre laser, tuned below the D2 line of Rb at 780 nm (allowing for defocusing nonlinearities [30]), is used to realize a speckle field by passing through a diffuser providing a Gaussian distribution of incident wave vectors in the transverse plane. The correlation length of the speckle field can be adjusted by changing the size of the illumination area on the diffuser (see Fig. 1). This allows us to tune the kinetic contribution $E(z=0)$. This speckle field is then sent onto the atomic vapor with a Gaussian envelope

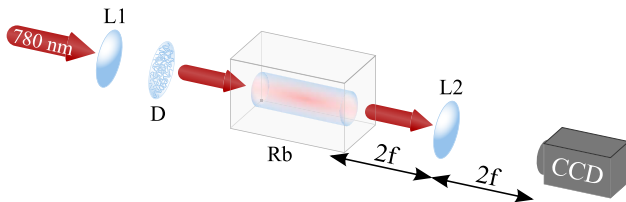


FIG. 1. *Experimental setup.* After passing a first lens ($L1$) a 1 W laser is impinging on a diffuser (D) creating a speckle field at the input face of a heated atomic vapor cell containing rubidium atoms. The output field is imaged on a CCD camera using a second lens ($L2$) with focal distance f .

on the order of $w_{sp} \approx 5$ mm and the average intensity I_0 around the center of the beam is thus proportional to the total power $I_0 \propto P/w_{sp}^2$. The nonlinear medium consists of a $L = 7$ cm long heated vapor cell containing a natural mixture of rubidium atoms. By adjusting the temperature of the cell, we can vary the atomic density ρ_{at} of the atoms by several orders of magnitude. The narrow atomic resonances allow efficient control of the linear index of refraction and its nonlinearity is due to the excited state saturation, which can be tuned by changing the incident laser frequency ω_L away from the atomic resonance ω_{at} by $\Delta = \omega_L - \omega_{at}$ or by adapting the power of the incident laser beam. We are thus able to realize a nonlinear phase shift up to $\Phi_{NL} = k_0 L \Delta n = 20\pi$, with a nonlinear index of refraction of $\Delta n = 20\pi/(k_0 L) \approx 10^{-4}$, and a transmitted power larger than 70%, so that $z_{NL} = L/20\pi \approx 1$ mm and $\Lambda \approx 10$ μm [30].

The light transmitted after nonlinear propagation can be analyzed either in real space (near field) through imaging onto a CCD camera (see Fig. 1) or by investigating the (far field) momentum distribution. Experimental measurements of the far field spectrum are delicate and extremely sensitive to details of the optical setup and detection scheme [48]. At variance with [14], our analysis is based on near field measurements, which will be shown to provide the appropriate framework to define the notion of ‘non-equilibrium precondensation.’

In Fig. 2, we illustrate the experimental results of the near field data for increasing values of L/z_{NL} , obtained by changing the nonlinear distance z_{NL} at constant L . As the laser intensity in the wings of the Gaussian envelope vanishes, the nonlinear interaction is prominent only in the central part of the speckle field. We therefore analyzed the near field intensity by performing spatial averaging ($\langle \cdot \rangle$) within a sufficiently small region of interest of the beam, where the statistics of the random wave are almost homogeneous.

For a very far detuned laser, inducing a vanishing nonlinearity, the transmitted intensity distribution $P(I)$

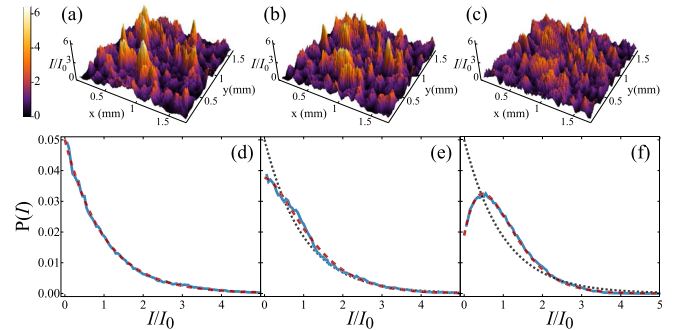


FIG. 2. *Near Field Speckle:* (a)–(c) near field images for $L/z_{NL} = 0, 3.5\pi$ and 14.4π ; (d)–(f): corresponding intensity histograms, showing the emergence of a nonzero value for the maximum of $P(I)$ with corresponding precondensation fractions n_0 : (d) $n_0 = 0$, (e) $n_0/I_0 = 0.5$, (f) $n_0/I_0 = 0.7$. The dotted black line refers to the exponential (Gaussian statistics), the dashed red line is a fit to the probability density (2).

exhibits an exponential decrease [see Fig. 2(d)], as expected for random waves with Gaussian statistics. By increasing the nonlinearity, the probability distribution strongly deviates from Gaussianity, and the maximum of $P(I)$ gradually shifts away from $I = 0$ [see Fig. 2(f)]. Such a deformation of the distribution is a robust phenomenon that also occurs for non-Gaussian heavy tailed statistics. Also, note that similar deformations of the probability density have apparently been reported in 1D optical systems [49–51]. However, the 1D NLS equation is integrable and does not exhibit thermalization or precondensation, so that the 1D probability density $P(I)$ is of a different nature than in 2D [30].

Analogous with BEC, the deformation of the probability distribution in Fig. 2 reflects a reduction of intensity fluctuations that precedes the establishment of long-range phase coherence [52,53]. We analyze the transmitted near field distribution by decomposing the field into a homogeneous (plane wave) condensate component and an incoherent component with statistical Gaussian fluctuations, $\psi(\mathbf{r}, z) = \sqrt{n_0} + \phi(\mathbf{r}, z)$. Although the coherent component does not refer to a purely homogeneous plane wave, such a decomposition proves robust and relevant to our analysis [30]. The intensity distribution $P(I)$ is given then by:

$$P(I) = \frac{\exp(-\frac{I+n_0}{I_0-n_0})}{I_0-n_0} \mathcal{I}_0\left(\frac{2\sqrt{n_0 I}}{I_0-n_0}\right), \quad (2)$$

where $\mathcal{I}_0(x)$ is the modified Bessel function of zeroth order and $I_0 = \langle I \rangle$. In the limit $n_0 \rightarrow 0$, the distribution (2) reduces to a pure exponential that characterizes a Gaussian field, $P(I) = \exp(-I/I_0)/I_0$. In the opposite limit, $n_0/I_0 \rightarrow 1$, one obtains $P(I) = \delta(I - I_0)$ as expected for a pure condensate plane wave solution. According to (2), the precondensate is simply related to the variance of the intensity fluctuations, $n_0/I_0 = \sqrt{2 - \langle I^2 \rangle / I_0^2}$, which, by energy conservation, are related to the prethermalized kinetic energy. Note in Fig. 2 that the intensity distributions observed in our experiment remain broad. This invalidates the standard Bogoliubov approach [1,23,54], which requires a sharp peaked intensity histogram around I_0 .

We stress the fact that Eq. (2) does not require the field to be in a thermal equilibrium state, so that the probability density (2) is valid even far from equilibrium. This is in contrast with the equilibrium probability density that is derived on the basis of equilibrium statistical mechanics, see Ref. [50]. Actually, the precondensation effect occurs very far from thermal equilibrium and does not require the establishment of an equilibrium state, i.e., the Rayleigh-Jeans spectrum. This is a striking difference with the usual equilibrium condensation arising from the divergence of the Rayleigh-Jeans distribution, featured by a marked peak at $k = 0$. We recall that complete thermalization to the Rayleigh-Jeans equilibrium spectrum requires extremely long propagation lengths that are not accessible

experimentally, as revealed by numerical simulations (see Fig. 4) or through the analysis of nonequilibrium kinetic equations [17]. At variance with this fully developed equilibrium condensation, here we identify a non-equilibrium precondensation effect that is characterized by a fast relaxation of n_0/I_0 for small propagation lengths available experimentally. Surprisingly, this initial stage of precondensation, featured by an accumulation of relatively long-wavelength modes around $k \approx 0$, provides a good indication for the final condensate fraction at Rayleigh-Jeans thermal equilibrium.

We report in Fig. 3(a) the precondensate fraction, extracted by fitting the probability distribution (2) to the experimental intensity histogram, as a function of the effective propagation distance L/z_{NL} . One clearly observes a continuous increase of n_0/I_0 for different values of the initial speckle correlation length. The Hamiltonian evolution of the random wave can be understood in a microcanonical statistical description, where the total energy H is the relevant parameter in the absence of an external heat bath governing the temperature for a canonical description. Accordingly, we have studied the transition to precondensation by varying the energy H , while keeping constant the “number of particles” (I_0 fixed). Such a transition is reported in Fig. 3(b) as a function of a dimensionless Hamiltonian, which can be conveniently expressed in terms of the ratio of the healing length Λ and the initial correlation length σ_c : $\tilde{H} = 1 + (\Lambda/\sigma_c)^2$ [30]. This representation shows that the nonlinear evolution consists of an exchange between the initial linear contribution $\langle E \rangle_0 \propto 1/\sigma_c^2$, and the initial nonlinear contribution $\langle U \rangle_0 \propto \gamma I_0^2$ of the Hamiltonian. This total energy can be tuned in our experiment by either changing the correlation length σ_c , or the nonlinear index of refraction (by changing the laser detuning or the laser intensity). As a remarkable result,

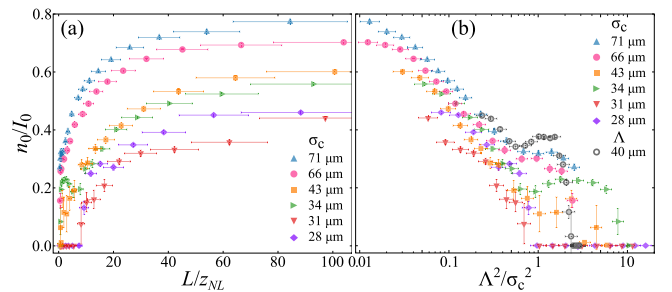


FIG. 3. Precondensate fraction at the constant laser power ($P = 1$ W) and sample length ($L = 7$ cm): (a) as a function of propagation distance L/z_{NL} for different values of the speckle correlation length σ_c . The nonlinear length z_{NL} is varied via the laser frequency Δ . (b) As a function of Λ/σ_c , obtained by changing the healing length Λ via the laser detuning for various correlation lengths σ_c (full symbols) or by changing the correlation length σ_c at constant detuning and corresponding healing length $\Lambda = 40 \mu\text{m}$ (gray circles). Error bars are derived from uncertainties of the nonlinear phase calibration and from fitting, respectively.

the precondensate fraction seems to only depend on the ratio between the linear and nonlinear contributions, $\langle E \rangle_0 / \langle U \rangle_0 = (\Lambda / \sigma_c)^2$. We report in Fig. 3(b) n_0 / I_0 as a function of $(\Lambda / \sigma_c)^2$ for various different initial correlation lengths and different values of the nonlinear interaction. The collapse of the data to an almost unique universal curve is a good indicator of the relevance of $\tilde{H} = 1 + (\Lambda / \sigma_c)^2$ to describe precondensation. Note that the dispersion of the different curves may be ascribed to the impact of a nonlocal nonlinearity [55,56].

Numerical simulations of the NLS Eq. (1) show that the precondensate fraction increases in a significant way in the initial stage [see Fig. 4(a)], a feature that can be described analytically: $n_0 / I_0 \approx 2\sqrt{2}(\Lambda / \sigma_c)(z / z_{NL})$ [30]. Note that, as compared to the theory and simulations, the experimental results show a delay for the initial growth of n_0 / I_0 , a feature that can be associated to a first correction of a nonlocal nonlinearity [30]. The growth of n_0 / I_0 then rapidly saturates to a quasistationary value after few nonlinear lengths z_{NL} . This fast process is characterized by a transfer and subsequent equilibration of the kinetic (E) and nonlinear (U) energies to their prethermalized quasisteady values, which in turn determine the amount of precondensate fraction n_0 / I_0 [30]. In marked contrast to such a short-time relaxation of E and n_0 , the spectrum of the random wave exhibits a very slow thermalization to equilibrium. This is revealed by the far-field, zero-momentum condensate $n_0^{\text{FF}} = n(k=0)$, whose relaxation to equilibrium requires several thousands of nonlinear propagation lengths, see Fig. 4(b) (blue line).

This slow relaxation is set by the photon-photon collision rate scaling as $1/\gamma^2$ [17], whereas the nonlinear length z_{NL} scales as $1/\gamma$ (reminiscent of the chemical potential in BEC). The far-from equilibrium nature of the precondensation process is also evidenced by the fact that the system does not exhibit a long-range phase order, as it would be expected for the thermalized 2D NLS equation below the Berezinskii-Kosterlitz-Thouless transition [57,58]. Indeed, precondensation is characterized by a fast decay of the correlation function [30], in contrast with the power-law behavior found at equilibrium. This means that precondensation does not refer to a “quasi-condensate” in the sense of the Berezinskii-Kosterlitz-Thouless theory [58].

Precondensation is characterized by an accumulation of particles (power) toward $k \sim 0$, as revealed by the far-field spectrum (momentum distribution) in Fig. 4(c). Observe that this strongly nonlinear effect ($\langle E \rangle_0 < \langle U \rangle_0$) cannot be described by a weak turbulence kinetic approach [8,16,20]. This shows an important property, namely the multimode nature of the effect of precondensation. At variance with n_0^{FF} , that refers to the pure zero-momentum occupation, here the precondensate refers to a slowly varying coherent field $\psi_c(\mathbf{r}, z)$, characterized by low-frequency components $k \lesssim 1/\Lambda$. The conventional decomposition of the field discussed above through Eq. (2) can then be refined by the substitution $\sqrt{n_0} \rightarrow \psi_c(\mathbf{r}, z)$, i.e., $\psi(\mathbf{r}, z) = \psi_c(\mathbf{r}, z) +$

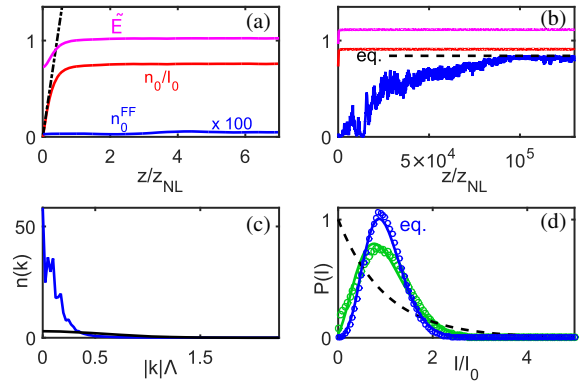


FIG. 4. *Numerical simulations of precondensation:* Precondensate fraction n_0 / I_0 (red lines), zero momentum population n_0^{FF} / I_0 (blue lines) and normalized kinetic energy \tilde{E} (magenta lines) in early stage (a) and in the long-term evolution (b). The dashed dark lines in (a) denote the analytical prediction for small z , in (b) the thermal equilibrium state. (c) Momentum spectrum of the initial wave at $z = 0$ (black line) and at $z = 100z_{NL}$ (blue line), showing an accumulation of particles “near” $k \sim 0$. (d) Near field intensity histogram: the initial field (dark dashed line), after $z = 100z_{NL}$ (green circles) and the fit of Eq. (2) (green line), and corresponding intensity histograms after $z = 1.5 \times 10^5 z_{NL}$ (in blue).

$\phi(\mathbf{r}, z)$, where $\phi(\mathbf{r}, z)$ denotes the rapidly varying incoherent component and $n_0 = \langle |\psi_c|^2 \rangle$. Importantly, the multimode analysis reveals that the bare intensity distribution (2) is well corroborated by a refined multimode intensity distribution, as revealed by the remarkable agreement between the simulations and the bare intensity distribution (2), see Fig. 4(d) at $z = 100z_{NL}$ and at full equilibrium ($z = 150 \times 10^3 z_{NL}$). In addition, precondensation proves robust with respect to the intensity moments ($\langle I^p \rangle$) used to compute n_0 / I_0 , or the frequency cutoff that is known to regularize the ultraviolet catastrophe inherent to classical waves. These aspects validate the simple model (2) used to analyze precondensation [30].

In conclusion, we have reported the observation of a phenomenon of non-equilibrium precondensation of classical waves in two dimensions. This experiment can be extended to study a classical analogue of BEC after a sudden quench below the critical temperature (in 3D) [59–62], the Berezinskii-Kosterlitz-Thouless transition (in 2D) [57], or the formation of nonthermal fixed points [63,64]. The possibility of shaping the initial conditions further allows the study of the growth of long range coherence using, e.g., a Gaussian beam with small fluctuations corresponding to nonlinear filtering of high frequency components [65], in relation with spatial beam self-cleaning in multimode fibers [66,67]. This experimental platform also paves the way to the study of a variety of phenomena in the key area of quantum fluids of light, such as superfluid behaviors [3,68–71], strongly nonlinear shocks [72,73], nonlocal effects [55,56], the development of turbulence cascades [8,9,74], or quench dynamics in the framework of the Kibble-Zurek mechanism [75].

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