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Impurities in Bose-Einstein Condensates: From Polaron to Soliton

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We propose that impurities in a Bose-Einstein condensate (BEC) in a multimode cavity transversely pumped by a laser form an experimentally accessible and analytically tractable model system for the study of impurities solvated in correlated liquids and the breakdown of linear response theory. As the strength of the coupling constant between the impurity and the BEC is increased, which is possible through Feshbach resonance methods, the impurity passes from a large to a small polaron state, and then to an impurity-soliton state. This last transition marks the breakdown of linear response theory.

It is well known that ultracold atomic physics provides realizations of interesting quantum many-body systems [1–3]. In particular, the emergence of spatial order and other forms of spontaneous symmetry breaking in quantum systems can be studied in BECs confined to transversely laser-pumped cavities [4, 5]. We focus on the problem of an impurity in a correlated quantum liquid near a continuous (or weakly first-order) symmetry breaking transition. The role of quantum fluctuations on impurity solvation and transport in correlated liquids like water [6] is currently the subject of active debates in the physical chemistry literature. The failure of linear-response theory plays a central role in these debates [7].

The physics of impurities in ultracold quantum gases [8–10] and in *uniform* BECs has already been well explored [11–16]. The interaction between a (neutral) impurity and the Bogoliubov excitations of the BEC maps in the continuum limit onto the so-called Fröhlich Hamiltonian [17–20], a linear-response theory that has been extensively applied to charged impurities in polarizable media, or polarons. Such a “Bose-polaron” can undergo a transition [21] from a *large polaron* state, which is well described by the Fröhlich continuum theory, to a *small polaron* state, in which the impurity is self-trapped on length-scales of the order of the inter-particle spacing of the medium [22, 23].

In a transversely pumped multimode cavity, a BEC undergoes a spontaneous phase transition from the uniform state to a state in which the density of the condensate is periodically modulated. This transition is described by a quantum version of the *Landau-Brazovskii* (QLB) theory for fluctuation-driven first-order phase transitions [5]. In this Letter we will combine the Fröhlich Hamiltonian description for impurities and the QLB theory for symmetry breaking in BECs to investigate the fate of a BEC polaron near the onset of spontaneous positional ordering, and determine whether impurities in a BEC in a multimode laser-pumped cavity can serve as a model system for the study of the effects of quantum fluctuations on solvation and the breakdown of linear response theory.

Model. Our model is defined by a Lagrangian that is the sum of three terms, respectively referring to the impurity particle, the condensate, and the lossy cavity. The Lagrangian of an impurity particle in a BEC condensate is

$$L_1 = \frac{1}{2} M_I |\dot{\mathbf{R}}|^2 - \int d^3r V(\mathbf{R} - \mathbf{r}) \rho(\mathbf{r}, t), \quad (1)$$

with M_I the impurity mass [24], $\mathbf{R}(t)$ the impurity location and $\rho(\mathbf{r})$ the deviation of the local density of the condensate from the mean density $n_0 = N_0/\mathcal{V}$ (N_0 and \mathcal{V} are the number of bosons in the condensate, and the volume of the system, respectively). Next, $V(\mathbf{r})$ is the interaction potential between the impurity particle with the bosons. In the *s*-wave Fermi approximation, $V(\mathbf{r} - \mathbf{r}') = g_{IB} \delta^{(3)}(\mathbf{r} - \mathbf{r}')$ with the pseudopotential $g_{IB} = 2\pi a_{IB} \hbar^2 / M_I$, where a_{IB} is the impurity-boson *s*-wave scattering length and M_I the reduced mass of a impurity-boson binary system. The Lagrangian for the excitations of the BEC reads

$$L_B = \frac{1}{\mathcal{V}} \sum_{\mathbf{q}} \frac{\hbar}{\zeta_q} \left(|\dot{\rho}_{\mathbf{q}}|^2 - \omega(\mathbf{q})^2 |\rho_{\mathbf{q}}|^2 \right) + L_{NL}. \quad (2)$$

Here, $\rho_{\mathbf{q}}(t) = \int d^3r \rho(\mathbf{r}, t) e^{-i\mathbf{q} \cdot \mathbf{r}}$ and $\zeta_q = n_0 \epsilon_0(q) / \hbar$ with $\epsilon_0(q) = (\hbar q)^2 / 2M_B$ the free boson dispersion relation. For a uniform BEC, the dispersion relation is given by the Bogoliubov spectrum $\hbar \omega_0(q) = \sqrt{\epsilon_0(q)(\epsilon_0(q) + 2n_0 g_{BB})}$ with $g_{BB} = 4\pi a_{BB} \hbar^2 / M_B$ the pseudopotential for boson-boson scattering (a_{BB} is the boson-boson scattering length). Non-linear terms are represented by L_{NL} . For BECs in an optical cavity, both the boson-boson and boson-impurity scattering length are experimentally adjustable parameters. The Fröhlich Hamiltonian of BEC polarons in uniform condensates is recovered upon canonical quantization of the linear and quadratic terms of equations (1) and (2).

The modes of a BEC inside a laser-pumped optical cavity, are mixed Bogoliubov excitations and electromagnetic modes [4]. The mode frequencies can be depressed and driven to zero when the frequency of the transverse laser matches that of a low frequency mode of the cavity. Near the instability threshold, the spectrum can be approximated as [5]:

$$\omega(\mathbf{q})^2 \simeq \Delta + \lambda \mathcal{R}^2 (|\mathbf{q}| - q_0)^2, \quad (3)$$

Here, q_0 is the wavenumber of the cavity mode, which is of the order of the inverse of the radius \mathcal{R} of the cavity radius [4, 5]. Δ is the gap in the harmonic mode spectrum, and λ is a phenomenological parameter determining the range of wavevectors over which the depression takes place. It is itself determined by the width of the cavity resonance and other factors [5]. In mean field theory, this Lagrangian describes a continuous ordering transition at $\Delta = 0$ from a uniform phase

to a density-modulated phase with modulation vector q_0 and modulation amplitude proportional to $|\Delta|^{1/2}$ [25–27].

The modes of the BEC are coupled to the electromagnetic modes outside the pumped cavity that act as a reservoir. These are included in the form of a distribution of harmonic oscillators coupled linearly to the BEC modes. Their Lagrangian is $L_E = \sum_j \frac{1}{2} m_j \{\dot{x}_j^2 - \omega_j^2 x_j^2\} - \sum_{j,q} C_{j,q} \rho_q x_j$. The last term represents a linear coupling of the reservoir degrees of freedom x_j to the modes ρ_q with coupling constants $C_{j,q}$. The nature of the dissipation is determined by these coupling constants through the oscillator spectral density $J_q(\omega) = \frac{\pi}{2} \sum_j \left(\frac{C_{j,q}^2}{m_j \omega_j} \right) \delta(\omega - \omega_j)$ [28]. We will restrict ourselves to the simplest case of “Ohmic” dissipation with $J_q(\omega) = \eta_q \omega$, for low frequencies where η_q is an effective friction coefficient [29]. The classical equation of motion for the impurity, obtained by minimizing the total action, is of the Langevin form with a friction coefficient that diverges at the MF critical point as η/Δ^2 , where $\eta \equiv \eta_{q_0}$.

The equilibrium partition function \mathcal{Z} of the impurity is proportional to the functional integral $\int \exp(\mathcal{S}_T) \mathcal{D}[\mathbf{R}(t)] \mathcal{D}[\rho_q(t)] \mathcal{D}[\{x_j(t)\}]$ over all degrees of freedom, which must obey periodic boundary conditions in “imaginary time” $0 \leq s \leq \beta$ ($\beta = 1/k_B T$). Here, \mathcal{S}_T is the Euclidean action. The path integral over the environmental oscillators can be carried out analytically. The remaining path integrals over condensate modes and particle trajectories will be discussed separately for positive and negative Δ .

$\Delta > 0$. For positive Δ , the non-linear terms (L_{NL}) do not play a significant role. The density fluctuations can then be integrated out, leading to an effective action for the particle trajectories:

$$\mathcal{S} \simeq - \int_0^{\tilde{\beta}} \frac{1}{2} \left(\frac{d\tilde{\mathbf{R}}}{d\tilde{s}} \right)^2 d\tilde{s} - \tilde{\mathbf{f}} \cdot \int_0^{\tilde{\beta}} \tilde{\mathbf{R}}(\tilde{s}) d\tilde{s} + \alpha \int d^3 \tilde{q} \int \int_0^{\tilde{\beta}} d\tilde{s} d\tilde{s}' G_{\tilde{q}}^{(2)}(|\tilde{s} - \tilde{s}'|) e^{i\tilde{\mathbf{q}} \cdot [\tilde{\mathbf{R}}(\tilde{s}) - \tilde{\mathbf{R}}(\tilde{s}')]}, \quad (4)$$

where $\alpha = \frac{g_{IB}^2 q_0^3 \zeta \hbar}{4(2\pi)^3} \left(\frac{M_I}{\hbar^2 q_0^2} \right)^3$ plays the role of a dimensionless coupling constant while $\zeta \equiv \zeta_{q_0}$. We shifted to dimensionless quantities, indicated by tildes, setting $M_I = \hbar = q_0 = 1$ [30]. An infinitesimal external force $\tilde{\mathbf{f}}$ is included in the action, in order to compute the effective mass. The kernel, for q around 1, is given by

$$G_q^{(2)}(\tau) = \frac{1}{\beta} \sum_{n=-\infty}^{+\infty} \frac{e^{i\omega_n \tau}}{\chi(q-1)^2 + \Gamma + \gamma|\omega_n| + \omega_n^2}. \quad (5)$$

The summation is over dimensionless Matsubara frequencies $\omega_n = 2\pi n/\beta$. The dimensionless distance to the MF critical point of the QLB is defined here as $\Gamma = \Delta(M_I/\hbar q_0^2)^2$, the dimensionless friction coefficient as $\gamma = \eta(M_I/\hbar q_0^2)$, and the dimensionless field rigidity as $\chi = \lambda(\mathcal{R}q_0)^2(M_I/\hbar q_0^2)^2$. The path integral over the particle trajectories is performed variationally [31] by defining a suitable Gaussian trial action. For

the present case we choose

$$\mathcal{S}_t = - \int_0^{\beta} \frac{1}{2} \left(\frac{d\mathbf{R}}{ds} \right)^2 ds - \mathbf{f} \cdot \int_0^{\beta} \mathbf{R}(s) ds - \frac{1}{2} \int \int_0^{\beta} \mathcal{K}(|s - s'|) |\mathbf{R}(s) - \mathbf{R}(s')|^2 ds ds', \quad (6)$$

where the kernel $\mathcal{K}(\tau) = \frac{1}{\beta} \sum_{n=-\infty}^{+\infty} \mathcal{K}_n e^{i\omega_n \tau}$ with $\mathcal{K}_n = C/(D + \gamma|\omega_n| + \omega_n^2)$, is similar to the actual kernel. The constants C and D play the role of variational parameters that are determined by applying Feynman’s inequality for trial actions: $F(f) \leq F_t + \beta^{-1} \langle \mathcal{S} - \mathcal{S}_t \rangle_t$. Here $F(f) = -k_B T \ln \mathcal{Z}$ is the free energy of the particle. Expectation values are computed using the Gaussian trial action. For a free particle with mass M^* subject to a force f , the second derivative of the free energy with respect to the applied force equals $\partial^2 F / \partial f^2|_{f=0} = \hbar^2 \beta^2 / 12 M^*$ (in actual units). Using this as the definition of the effective mass [32] and applying the trial action gives: $M^* = \frac{\beta^2}{24} [\sum_{n>0} g_n]^{-1}$ where $g_n = [\omega_n^2 + \frac{\gamma}{\beta}(\mathcal{K}_0 - \mathcal{K}_n)]^{-1}$. In the limit of $\gamma = 0$, the kernel $G_{q=1}^{(2)}(\tau)$ decays as $\exp(-\sqrt{\Gamma}\tau)$, as in Feynman’s theory of large polarons [33]. In the opposite limit of $\gamma \rightarrow \infty$, quantum fluctuations of the field are suppressed and only the $n = 0$ term remains, corresponding to MF classical static structure factor $1/[\chi(q-1)^2 + \Gamma]$. The model reduces in this limit to the theory of the small polaron [34]. The effective mass indeed undergoes a discontinuous jump as a function of increasing coupling constant at a critical value $\alpha_c \simeq \sqrt{\chi\Gamma}$. From Fig.1(a), as the damping coefficient is reduced, the effective mass discontinuity is reduced and goes to zero at a critical point [35]. According to Fig.1(c), the value of the critical coupling constant for the transition between the large and small polarons is strongly reduced as one approaches the ordering transition of the BEC. This is an important result: the transition from large to small polaron can be induced much easier in a BEC near the ordering transition [36].

$\Delta \lesssim 0$. For negative Δ , the non-linear terms of Eq.2 must be taken into account. It can be shown that only even terms need to be included [5]: $L_{NL} = -u \int d^3 r |\rho(\mathbf{r})|^4 - w \int d^3 r |\rho(\mathbf{r})|^6$. In order to perform the functional integrals for this non-linear case, first expand the free energy $F[\mathbf{R}(s)]$ in a Taylor expansion in powers of the impurity pseudopotential g_{IB} . Then perform, term by term, the functional integrals using the non-linear action. The zero-order term in the expansion is the partition function of the condensate in the absence of the particle. The first order term is

$$F^{(2)}[\mathbf{R}(s)] = \frac{-g_{IB}^2}{2!} \sum_{\mathbf{q}, n} \mathcal{G}_{\mathbf{q}, \omega_n}^{(2)} \int \int_0^{\beta} ds ds' e^{i\mathbf{q} \cdot [\mathbf{R}(s) - \mathbf{R}(s')]} \quad (7)$$

Here, $\mathcal{G}_{\mathbf{q}, \omega_n}^{(2)}$ is the *full* two-point Green’s function of the system without the impurity. The second order term contains the full four-point vertex function. These full correlation functions are obtained by a second expansion, now in powers of

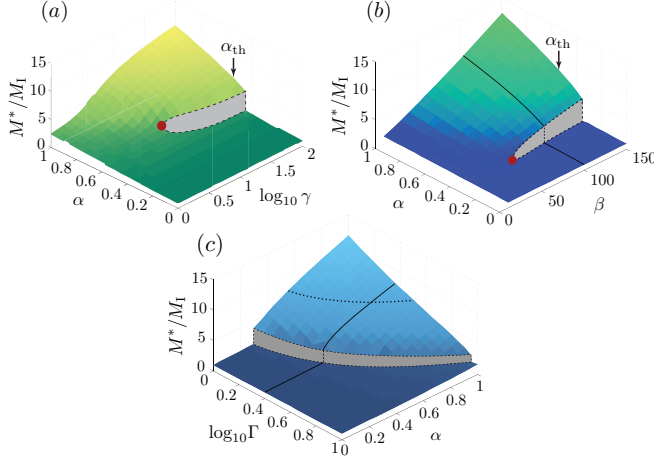


FIG. 1. Effective mass versus dimensionless coupling constant and (a) dissipation, (b) temperature, and (c) distance from the ordering transition. Other parameters are set to: (a) $\beta = \chi = 100$ and $\Gamma = 1$, (b) $\gamma = \chi = 100$ and $\Gamma = 1$, and (c) $\beta = \chi = \gamma = 100$. In (a) and (b) the red dots indicate the critical points where the discontinuity of the effective mass closes. The dark arrows marked by α_{th} correspond to the dotted line in (c), i.e. $\gamma, \beta \rightarrow \infty$, at $\Gamma = 1$.

L_{NL} . The non-linear terms can be included by the renormalization group method [5]. To one-loop order, this leads to a renormalization of Δ to $\bar{\Delta}$ with $\bar{\Delta} \simeq \Delta + \mathcal{P}u \ln(\Delta_c/\bar{\Delta})$, where $\mathcal{P} \propto q_0^2$ and Δ_c a high-energy cutoff. The effective gap $\bar{\Delta}$ of the spectrum remains positive for negative values of Δ . The renormalized quartic coefficient \bar{u} is given by $\bar{u} \simeq u \frac{1-u\Pi}{1+u\Pi}$ with $\Pi = \mathcal{P}/\bar{\Delta}$; so \bar{u} becomes negative if the effective gap $\bar{\Delta}$ drops below $\mathcal{P}u$ (which happens for slightly negative Δ). The functional integral over the renormalized quadratic Lagrangian no longer suffers from strong fluctuations, even for negative \bar{u} . For negative \bar{u} and decreasing $\bar{\Delta}$, a first-order phase transition takes place at $\bar{u}^2 = 4\hbar\bar{\Delta}w/\zeta$ where the modulation amplitude changes discontinuously from zero to $\sqrt{2(\hbar\bar{\Delta}/\zeta|\bar{u}|)^{1/2}}$. The correlation length at the ordering transition is $\xi = \mathcal{R}\sqrt{\lambda/\bar{\Delta}}$. This renormalized action \mathcal{S}_T^R has the same form as \mathcal{S}_T but with u and Δ replaced by the renormalized ones: it can be analyzed by MF theory plus fluctuation corrections.

The simplest case is the strong damping limit, when the condensate density modulation can be treated as quasi-static with respect to the dynamics of the impurity [37]. The MF minimization of \mathcal{S}_T^R leads to a radial density modulation around a static impurity at the origin of the form

$$\rho(r) = \rho_0 \frac{\sin(q_0 r)}{q_0 r} \exp(-r/\xi). \quad (8)$$

Generally $\rho(r)$ retains this form if $q_0 r \gtrsim 1$. Using Eq. 8 as a trial function (with ρ_0 the free parameter), leads to non-linear

MF free energy cost $\mathcal{F}_B(\rho_0)$ for radial density modulations:

$$\mathcal{F}_B(\rho_0) \simeq \frac{1}{\pi^2 q_0^3} \left(\frac{2\hbar\bar{\Delta}(q_0\xi)}{\pi\zeta} |\rho_0|^2 + \bar{u}|\rho_0|^4 + \frac{w}{2} |\rho_0|^6 \right) \quad (9)$$

where $\mathcal{F}_B(\rho_0)$ has in general the form of a double-well potential with one minimum at $\rho_0 = 0$ and a second near the modulation amplitude $\rho^* = \sqrt{2|\bar{u}|/3w}$ of the ordered phase (see Fig.2). The second minimum represents a spherically symmetric deformation of the condensate that interpolates between the ordered state near the origin and the uniform state far from the origin. We will refer to this structure as the *impurity soliton* [38]. At the transition point, the energy cost of the impurity soliton is $\mathcal{F}^* \simeq \frac{8\hbar}{\pi^3 q_0^3 \zeta} (\bar{\Delta}_c |\bar{u}|/w)(q_0\xi)$. If the mass of the impurity is so large ($M_I \gg \hbar^2 q_0^2 / \rho_0 g_{IB}$) that quantum fluctuations of the impurity can be disregarded, the free energy is $\mathcal{F}_I \simeq -|g_{IB}\rho_0|$ as shown in Fig.2(a):

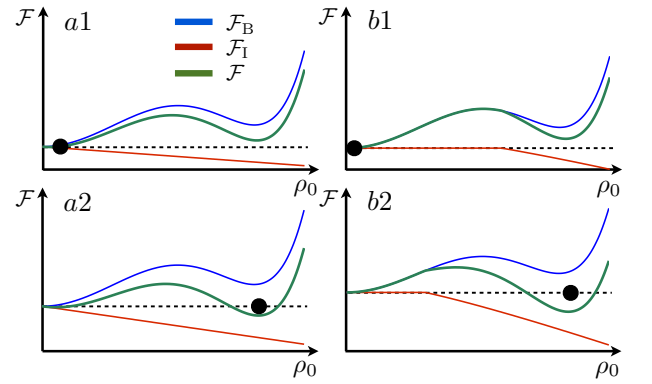


FIG. 2. Variational free energies as a function of the modulation amplitude ρ_0 . The blue, red, and green curves show the condensate free energy $\mathcal{F}_B(\rho_0)$, impurity free energy $\mathcal{F}_I(\rho_0)$, and the total free energy $\mathcal{F}(\rho_0) = \mathcal{F}_B(\rho_0) + \mathcal{F}_I(\rho_0)$, respectively. The absolute minima of total free energies are indicated by black dots in each panel. (a1,a2) Massive impurities: the free energy of the impurity decreases linearly with ρ_0 . For increasing pseudopotential, the absolute minimum shifts from the neighborhood of the Gaussian minimum to that of the vacuum of the ordered phase—corresponding to a transition from the small polaron to the impurity soliton. (b1,b2) include the zero point energy of a bound-state particle, which follows the red curves. The minimum at $\rho_0 = 0$ corresponds to the large polaron (b1). For increasing pseudopotentials (b2), there may be transitions from the large polaron to the small polaron and then to the soliton, or a single transition directly from the large polaron to the impurity-soliton.

For small g_{IB} the absolute minimum of $\mathcal{F}(\rho_0) = \mathcal{F}_B + \mathcal{F}_I$ as a function of ρ_0 is proportional to g_{IB} (Fig. 2(a1)), which corresponds to the small polaron, while there is a metastable minimum near ρ^* that corresponds to the impurity soliton. For larger values of g_{IB} , the absolute minimum jumps near ρ^* (see Fig. 2(a2)). The soliton state has lower energy if

$$\frac{8\hbar}{\pi^3 q_0^3 \zeta} (\bar{\Delta}_c |\bar{u}|/w)(q_0\xi) \lesssim |g_{IB}\rho^*|. \quad (10)$$

Since $\xi \propto \bar{\Delta}^{-1/2}$, the impurity soliton state necessarily has a lower energy than the polaron state for sufficiently large cor-

relation lengths and hence linear-response theory must break down for sufficiently large correlation lengths. When typical values for a pumped BEC are inserted in this inequality, one finds that the polaron-soliton transition should be experimentally accessible [36].

Next, we include quantum fluctuations of the impurity particle. The effective Lagrangian is

$$L_I \simeq \frac{1}{2} M_I |\dot{\mathbf{R}}|^2 + |g_{IB} \rho_0| \frac{\sin(q_0 R)}{q_0 R} \exp(-R/\xi). \quad (11)$$

The impurity free energy \mathcal{F}_I is obtained from L_I by integrating over particle trajectories. This leads to

$$\mathcal{F}_I(\rho_0) \simeq -|g_{IB} \rho_0| + \frac{3}{2} \hbar \Omega(\rho_0) + \dots \quad (12)$$

The first term is the earlier classical limit while the second term is the lowest-order correction due to quantum fluctuations. $\Omega(\rho_0) = \left(\frac{|g_{IB} \rho_0| q_0^2}{3 M_I} \right)^{1/2}$ is the natural frequency of the bound state impurity particle in the effective potential. The bound state disappears if the right-hand side of Eq. 11 is positive, in which case \mathcal{F}_I should be set to zero (the flat part of the red curve in Fig. 2(b1), 2(b2)). The small polaron minimum is replaced by a minimum at $\rho_0 = 0$ that corresponds to the *large* polaron. The condition for the small polaron to survive in the presence of the zero-point fluctuations of the impurity is $g_{IB} > g_{c1} = \sqrt{\frac{\hbar^3 \Delta \xi}{M_I \zeta}}$ or $\alpha_c \simeq \sqrt{\chi \Gamma}$, which is just the earlier criterium for self-trapping of a small polaron. The soliton state is significantly more stable against zero point fluctuations of the impurity, than the small polaron state.

Quantum fluctuations of the condensate are included by treating $\rho_0(t)$ as a time-dependent coordinate with a kinetic energy $K = (2\pi\xi/q_0^2) (d\rho_0(t)/dt)^2$. In the double-well potential, the condensate coordinate now can tunnel between the two minima, allowing for a linear superposition of the small polaron and soliton states. As the strength of the quantum fluctuations of the condensate increases, the impurity soliton state disappears, roughly when the zero point energy of the condensate coordinate exceeds the depth of the well [39].

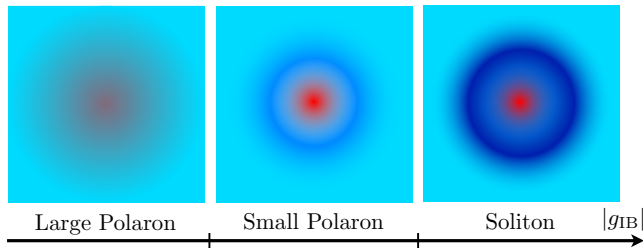


FIG. 3. The different states of the BEC-impurity system are shown schematically. The blue color indicates the condensate modulation amplitude while the red cloud indicates the particle. From left to right, large polaron, small polaron, and the soliton.

An important concern regarding the soliton solutions is their instabilities in spatial dimensions $D > 1$, due to Der-

rick's theorem [40]. We show in Supplementary Materials (3) that the coupling with the particle's degree of freedom stabilizes the soliton. Furthermore, Gaussian fluctuations are shown to be irrelevant in $D \geq 2$.

In summary, the QLB theory predicts that impurity particles generate a variety of structures as the excitation spectrum is progressively depressed. Above the MF transition point, the impurity generates either a large or a small polaron state, depending on the coupling and damping constant. Below the MF transition—but above the actual ordering transition—a new state appears: the soliton with a self-trapped impurity particle (see Fig. 3).

Discussion. In the introduction we posed the question whether impurities in a BEC can serve as a model system for the study of the role of quantum fluctuations and the breakdown of linear response theory in solvation theory. We have found that linear response theory breaks down when a pumped BEC system becomes increasingly correlated on approach of a spontaneous symmetry breaking transition, signaled by the formation of the impurity soliton state. Does this agree, at least qualitatively, with solvation phenomena in conventional fluids? It is believed that the breakdown of linear response theory for small ions in water is related to the formation of partially ordered shells of water molecules (“solvation shells”) around the ion [6]. Water is a highly correlated fluid and solvation shells indeed could be—crudely—viewed as local realizations of the low-temperature ordered phase (i.e., ice). On the other hand, the water molecules surrounding solvated electrons, with much stronger zero-point fluctuations, remain disordered. A recent mixed quantum-classical simulation of electrons solvated in water have a wavefunction that is relatively delocalized (“wet electron”) [7]. According to the theory, an impurity soliton state destroyed by the zero-point motion of the particle would have to be a large polaron. Quantitative experimental studies of impurities in BECs in pumped optical cavities that would verify the association of the breakdown of linear response theory with solitons, should, according to our estimates [36], be possible and would be of great value as a model system to study quantum effects on solvation in a system described by a general theory that does not require detailed assumptions about molecular interactions.

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- [36] See Supplementary Materials (4).
- [37] We take $\gamma \rightarrow \infty$ to approach the classical field limit.
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