Bose-Einstein Condensation of Stationary-Light Polaritons

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We propose and analyze a mechanism for Bose-Einstein condensation of stationary dark-state polaritons. Dark-state polaritons (DSPs) are formed in the interaction of light with laser-driven 3-level Λ-type atoms and are the basis of phenomena such as electromagnetically induced transparency, ultraslow, and stored light. They have long intrinsic lifetimes and in a stationary setup, a 3D quadratic dispersion profile with variable effective mass. Since DSPs are bosons, they can undergo a Bose-Einstein condensation at a critical temperature which can be many orders of magnitude larger than that of atoms. We show that thermalization of polaritons can occur via elastic collisions mediated by a resonantly enhanced optical Kerr nonlinearity on a time scale short compared to the decay time. Finally, condensation can be observed by turning stationary into propagating polaritons and monitoring the emitted light.

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Since the critical temperature of Bose-Einstein condensation (BEC) [1,2] is inversely proportional to the mass of the particles, already early on in the history of the field, quasiparticles were considered as candidates for condensation at high temperatures [3]. Very recently, this subject regained a lot of attention triggered by experimental breakthroughs in microcavity exciton-polaritons [4–6] and thin-film magnons [7]. Yet a major drawback of these systems is the rather short lifetime of the quasiparticles. In addition, the exciton-polariton and magnon systems are two dimensional. In 2D, there is no true long-range off-diagonal order [8], and only quasicondensation can be achieved in a finite system. We here propose a mechanism for true condensation in three spatial dimensions employing a different kind of polaritonic quasiparticles, called dark-state polaritons (DSP) [9]. DSPs [10] emerge in the Raman interaction of lasers with 3-level quantum systems and are the basis of ultraslow [11], stopped [12,13], and stationary light [14]. They are bosons [10] and have considerably longer decoherence times than excitons, exciton polaritons, or magnons, ranging from tens of microseconds in alkali vapors in magneto-optical traps [15], and several milliseconds in condensed atomic gases [13] to seconds in doped glasses [16,17]. Furthermore, they provide a 3D quadratic dispersion with variable mass and have very high condensation temperatures. DSPs can easily be created and thermalization can be achieved on a time scale much shorter than their lifetime. Finally, condensation can easily be observed by transforming stationary DSPs into light pulses. Because of their properties, such as variable mass and long lifetime, DSPs may provide an interesting new approach to many-particle phenomena with quasiparticles [18], BEC being one of the simplest.

If an optically thick ensemble of 3-level quantum systems is irradiated by a coherent coupling laser, it can become transparent for a probe field within a certain frequency range close to the two-photon Raman resonance. Associated with this electromagnetically induced transparency (EIT) [19,20] is the formation of polaritonic eigensolutions, called dark-state polaritons [10], which are superpositions of the probe field and a collective Raman excitation. The mixing angle can be changed by varying the strength of the control laser. At the transparency frequency, the dispersion of DSPs is linear with a slope determined by the mixing angle. This situation changes if two counter-propagating control lasers with comparable intensities are used. The two control lasers lead to a quasi-stationary probe-field pattern, known as stationary light [14,21]. We show that the dark polaritons of stationary light [9] behave like massive particles in 3D and can undergo Bose-condensation at high temperatures.

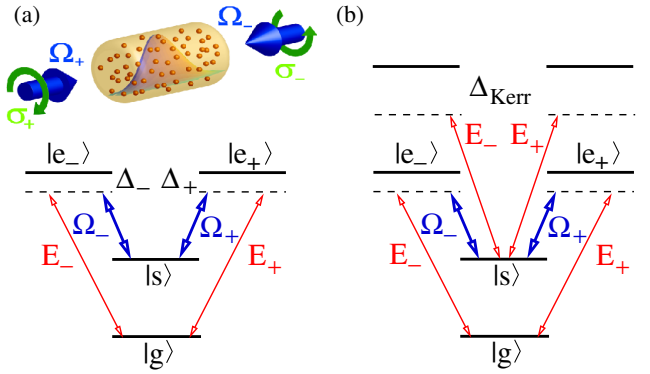

FIG. 1 (color online). (a) Raman interaction of two counter-propagating control lasers $\Omega_\pm$ with opposite circular polarization coupling $|s\rangle - |e_\pm\rangle$ transitions of a double Λ system generates stationary light of Stokes fields $E_\pm$. The stationary DSPs formed by this do not decay radiatively and have a 3D quadratic dispersion profile. (b) Off-resonant coupling of Stokes fields $E_\pm$ with other excited states leads to ac-Stark shift of $|s\rangle$ resulting in a resonantly enhanced Kerr-type self-interaction of $E_\pm$.

We here consider a generalization of the original stationary-light scheme [14], which is shown in Fig. 1(a). It involves two parallel Λ transitions consisting of the common ground levels $|g\rangle$ and $|s\rangle$ and the excited states $|e_\pm\rangle$, coupled to opposite circular polarizations of quantized probe fields $\hat{E}_\pm$ and control lasers described by Rabi-frequencies $\Omega_\pm$ [9]. Both Λ schemes are in two-photon resonance which guarantees EIT. For the present discussion, we will assume that the control fields are homogeneous and constant in time and thus set $\Omega_\pm = \Omega_\pm^*$. The single-photon detunings of the upper states are denoted as Δ_+ and Δ_-, respectively. The fields $\hat{E}_+$ ($\hat{E}_-$) and Ω_+ (Ω_-) propagate in the $+z$ ($-z$) direction with wave numbers k_p ($-k_p$), and k_c ($-k_c$).

We introduce normalized field amplitudes $\hat{\mathcal{E}}_\pm$ that vary slowly in space and time via $\hat{E}_\pm(\mathbf{r}, t) = \sqrt{\frac{\hbar\omega}{2\varepsilon_0}}\{\hat{\mathcal{E}}_\pm(\mathbf{r}, t) \times \exp[-i(\omega_p t \mp k_p z)] + \text{H.a.}\}$, and continuous atomic-flip operators $\hat{\sigma}_{\mu\nu}(\mathbf{r}, t) = \frac{1}{\Delta N}\sum_{j\in\Delta V(\mathbf{r})}\hat{\sigma}^j_{\mu\nu}$, with $\hat{\sigma}^j_{\mu\nu} \equiv |\mu\rangle_{jj}\langle\nu|$ being the flip operator of the jth atom. The sum is taken over a small volume ΔV around $\mathbf{r}$ containing ΔN atoms.

Assuming that in the absence of the probe fields all population is in the ground state $|g\rangle$, the dynamical equations read in the linear response limit, i.e., for a small probe intensity

$$\frac{\partial}{\partial t}\hat{\sigma}_{gs} = i\Omega_+\hat{\sigma}_{ge_+} + i\Omega_-\hat{\sigma}_{ge_-}, \tag{1}$$

$$\frac{\partial}{\partial t}\hat{\sigma}_{ge_\pm} = -\Gamma_{ge_\pm}\hat{\sigma}_{ge_\pm} + i\Omega_\pm\hat{\sigma}_{gs} + ig\sqrt{n}\hat{\mathcal{E}}_\pm, \tag{2}$$

$$\left[\frac{\partial}{\partial t} \pm c\frac{\partial}{\partial z} - i\frac{c}{2k_p}\Delta\right]\hat{\mathcal{E}}_\pm = ig\sqrt{n}\hat{\sigma}_{ge_\pm}. \tag{3}$$

Here, n is the atom number density, and $g = \frac{\wp}{\hbar}\sqrt{\frac{\hbar\omega}{2\varepsilon_0}}$ is the common coupling constant of both probe fields with $\wp$ denoting the dipole matrix element. $\Gamma_\pm = \gamma + i\Delta_\pm$, where γ is the transverse decay rate of the transitions $|e_\pm\rangle - |g\rangle$ and $k_p = \omega_p/c$, is the carrier wave number of the probe field. If the characteristic time- and length-scales of the probe fields T and L are sufficiently large, such that the adiabaticity conditions $T \ll \{L_{\text{abs}}/c, \gamma^{-1}\}$ and $L \gg L_{\text{abs}}$ hold, where $L_{\text{abs}} \equiv \gamma c/(g^2 n)$ defines the resonant absorption length, the system can be described by polariton-like quasiparticles [9]. One of these solutions, called DSP, does not involve excited states and is thus immune to spontaneous emission.

$$\hat{\Psi}(\mathbf{r}, t) = (\cos\theta)[(\cos\phi)\hat{\mathcal{E}}_+(\mathbf{r}, t) + (\sin\phi)\hat{\mathcal{E}}_-(\mathbf{r}, t)] - (\sin\theta)\hat{S}(\mathbf{r}, t) \tag{4}$$

where $\hat{S} \equiv \sqrt{n}\hat{\sigma}_{gs}(\mathbf{r}, t)$ and the mixing angles are defined by $\tan^2\theta = g^2 n/\Omega^2$ with $\Omega^2 = \Omega_+^2 + \Omega_-^2$, and $\tan^2\phi = \Omega_-^2/\Omega_+^2$. The longitudinal dispersion relation of the stationary DSPs is plotted in Fig. 2 for $\tan\phi = 1$ (centermost line). Also shown are the energies of the other eigensolutions. It proves useful to introduce superpositions of these eigensolution termed bright-state polaritons, $\hat{\Phi}_1(\mathbf{r}, t) = -(\sin\phi)\hat{E}_+(\mathbf{r}, t) + (\cos\phi)\hat{E}_-(\mathbf{r}, t)$, and $\hat{\Phi}_2(\mathbf{r}, t) = -(\sin\theta)[(\cos\phi)\hat{E}_+(\mathbf{r}, t) + (\sin\phi)\hat{E}_-(\mathbf{r}, t)] - (\cos\theta)\hat{S}(\mathbf{r}, t)$.

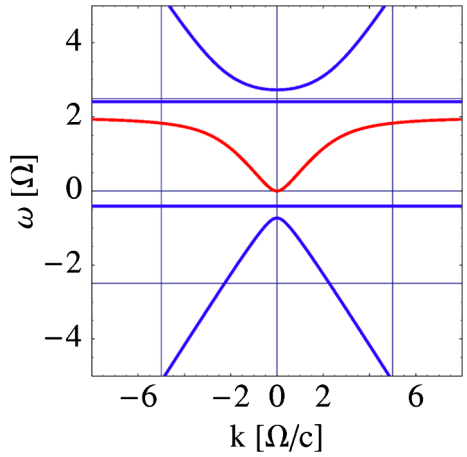

FIG. 2 (color online). Dispersion of stationary DSP (centermost line) and other quasiparticle solutions in propagation direction for $\Delta_+ = \Delta_- = \Delta$, and $\Omega_+ = \Omega_- = \Omega$, $\tan\theta = \tan\phi = 1$, $\Delta/\Omega = 2$. Since only the real parts of the eigenvalues are of interest here, we set $\gamma = 0$. One clearly recognizes a quadratic profile for the dark polariton around $k = 0$. It should be noted that the polariton is defined by the slowly varying envelopes of field and matter variables and k refers to spatial modulations of the slowly varying amplitudes.

It is easy to derive the equation of motion of the DSP up to second order including the transverse directions. One finds for $\Delta_\pm = \Delta$ and $\phi = \pi/4$

$$\left[\frac{\partial}{\partial t} - iv_{\text{gr}}L_{\text{abs}}\left(i + \frac{\Delta}{\gamma}\right)\frac{\partial^2}{\partial z^2} - i\frac{v_{\text{gr}}}{2k_p}\Delta_\perp\right]\hat{\Psi}(\mathbf{r}, t) = 0, \tag{5}$$

where $v_{\text{gr}} = c\cos^2\theta$ is the group velocity of the polariton in the slow-light case, i.e., if only one of the two coupling fields is present. As opposed to microcavity exciton-polaritons, stationary DSPs behave as free, massive Schrödinger particles in all three spatial directions with a tensorial mass, $m_\perp = mv_{\text{rec}}/v_{\text{gr}}$, $m_\parallel^{-1} = m_\perp^{-1}2k_pL_{\text{abs}}(\Delta/\gamma + i)$. Here, we have introduced the recoil velocity of the atom at the probe frequency $v_{\text{rec}} = \hbar k_p/m$ and the mass of the atoms m as comparative scales. The effective mass can be varied via the strength of the control laser, i.e., via the mixing angle θ, and is typically several orders of magnitude smaller than the mass of the atoms. The longitudinal mass $m_\parallel$ is again smaller than the transverse mass by the ratio of resonant wavelength to absorption length and the inverse normalized detuning γ/Δ. It also has a small imaginary component, which describes the absorption of high-k components well known from EIT [20]. As can be seen from Fig. 2, the quadratic dispersion of the dark-state polariton saturates at higher

frequencies giving rise to a finite band whose width is of the order of the single-photon detuning Δ for $|\Delta| \gg \Omega, \gamma$.

Using the well known expression for the critical temperature of condensation for a homogeneous, ideal gas and expressing the polariton mass in terms of the mass of the atoms, one finds

$$T_c = T_c^{\mathrm{atom}} \left(\frac{\rho_{\mathrm{DSP}}}{n}\right)^{2/3} \frac{v_{\mathrm{gr}}}{v_{\mathrm{rec}}} \left(2k_p L_{\mathrm{abs}} \frac{\Delta}{\gamma}\right)^{1/3}. \quad (6)$$

Here, we have introduced the critical temperature of the atoms $k_B T_c^{\mathrm{atom}} = 2\pi n^{2/3}/[\zeta(3/2)^{2/3} m]$ as a comparative scale. ρ_{DSP} is the density of dark-state polaritons, which has to be smaller than the atomic density n in order to stay within the linear response limit. Since the group velocity v_{gr} can be orders of magnitude larger than the recoil velocity of atoms v_{rec} and since $k_p L_{\mathrm{abs}} \gg 1$, the critical temperature can be much larger than that of the atoms. E.g., for $\rho_{\mathrm{DSP}}/n = 10^{-1}$, $v_{\mathrm{gr}} = 1$ km/s, $v_{\mathrm{rec}} = 5$ cm/s, $L_{\mathrm{abs}} = 1$ cm, $\Delta/\gamma = 10$, and $k_p = 2\pi/500$ nm yields: $T_c/T_c^{\mathrm{atom}} \approx 6 \times 10^5$, i.e., a value in the mK regime. On the other hand, the finite frequency window of EIT at large detuning limits the maximum allowed temperature where losses are small to $k_B T_{\mathrm{EIT}} \approx \hbar\Delta$, i.e., to the regime of 0.1–1 mK. The losses outside the EIT transparency window reflected in the imaginary part of the polariton mass may be used for evaporative cooling of polaritons, which is however outside the scope of this Letter [17].

Condensation is achieved differently from atoms or exciton polaritons. Instead of changing the temperature or the density, one can dynamically change the critical temperature of condensation by varying the effective mass. Initial preparation of low-energy DSPs can be realized in various ways [17]. Spontaneous Raman scattering cannot be used since it results in too high polariton momenta on the order of the recoil momentum corresponding to polariton temperatures on the order of $T_{\mathrm{rec}} m/(m_\perp^{2/3} m_\parallel^{1/3})$. Direct RF coupling or storage of a coherent light pulse yield a low-energy coherent polariton distribution. The coherence is then subsequently destroyed by thermalizing collisions. Alternatively, storage of an incoherent light pulse results in the preparation of a nonequilibrium mixed state.

Since the maximum temperature allowed by the finite transparency window is below the temperature range where collisions in a gas cell could provide sufficiently fast thermalization, the latter should occur by elastic collisions between DSPs. These can be induced by resonantly enhanced optical nonlinearities provided by the stationary-light coupling scheme itself. If, e.g., the excited states in Fig. 1 are hyperfine states, then other off-resonant couplings exist, such as those shown in Fig. 1(b). This coupling induces ac-Stark shifts which give rise to a resonantly enhanced optical Kerr nonlinearity [18,22]. If we ignore off-resonant couplings of the control laser fields, which is justified if the energy splitting between the lower states $|g\rangle$ and $|s\rangle$ is larger than $|\Omega_\pm|$, the resulting effective Kerr-interaction reads

$$\hat{H}_{\mathrm{Kerr}} \approx -\hbar \frac{g^2}{\Delta_{\mathrm{Kerr}}} \tan^2\theta \int d^3\mathbf{r} :(\hat{\mathcal{E}}_+^\dagger \hat{\mathcal{E}}_+ + \hat{\mathcal{E}}_-^\dagger \hat{\mathcal{E}}_-)^2: . \quad (7)$$

Δ_{Kerr} is the one-photon detuning from the additional excited states, and ":" denotes normal ordering. Since (7) only effects the electromagnetic component of the polaritons, the optical Kerr interaction involves also nonlinear scattering of dark- into bright-state polaritons. Assuming $\sin\theta \approx 1$, i.e., $v_{\mathrm{gr}} \ll c$, one finds

$$\hat{H}_{\mathrm{Kerr}} = -\frac{\hbar g^2 \cos^2\theta}{\Delta_{\mathrm{Kerr}}} \int d^3\mathbf{r} \hat{\Psi}^\dagger \hat{\Psi}^\dagger \hat{\Psi} \hat{\Psi} + \frac{\hbar g^2 \cos\theta}{\Delta_{\mathrm{Kerr}}} \times \int d^3\mathbf{r} \hat{\Psi}^\dagger (\hat{\Phi}_2^\dagger \hat{\Psi} + \hat{\Psi}^\dagger \hat{\Phi}_2) \hat{\Psi}. \quad (8)$$

The bright polaritons decay rapidly, and thus the nonlinear coupling of polariton modes results in an effective loss. Adiabatic elimination of the fast decaying bright polaritons yields in Born-Markov approximation a Liouville equation for the density matrix of the DSPs with a Hamiltonian part describing elastic two-body collisions and a nonlinear loss part [17].

$$\begin{aligned} \dot{\rho}_\Psi = & -i \frac{g^2 \cos^2\theta}{\Delta_{\mathrm{Kerr}}} \int d^3\mathbf{r} [\hat{\Psi}^\dagger \hat{\Psi}^\dagger \hat{\Psi} \hat{\Psi}, \rho_\Psi] \\ & + 4i \frac{g^2 \Delta \cos^2\theta}{n \Delta_{\mathrm{Kerr}}^2} \int d^3\mathbf{r} [\hat{\Psi}^\dagger \hat{\Psi}^\dagger \hat{\Psi} \hat{\Psi}^\dagger \hat{\Psi} \hat{\Psi}, \rho_\Psi] \\ & + 4 \frac{g^2 \gamma \cos^2\theta}{n \Delta_{\mathrm{Kerr}}^2} \int d^3\mathbf{r} \{\hat{\Psi}^\dagger \hat{\Psi}^\dagger \hat{\Psi} \hat{\Psi}^\dagger \hat{\Psi} \hat{\Psi}, \rho_\Psi\}_+ \\ & - 8 \frac{g^2 \gamma \cos^2\theta}{n \Delta_{\mathrm{Kerr}}^2} \int d^3\mathbf{r} \hat{\Psi}^\dagger \hat{\Psi} \hat{\Psi} \rho_\Psi \hat{\Psi}^\dagger \hat{\Psi}^\dagger \hat{\Psi}. \end{aligned} \quad (9)$$

From these, one can extract the rate of elastic collisions

$$\Gamma_{\mathrm{coll}} = \frac{g^2}{\Delta_{\mathrm{Kerr}}} \cos^2\theta \rho_{\mathrm{DSP}} = \frac{v_{\mathrm{gr}}}{L_{\mathrm{abs}}} \left(\frac{\gamma}{\Delta_{\mathrm{Kerr}}} \frac{\rho_{\mathrm{DSP}}}{n}\right) \quad (10)$$

as well as the rate of collision induced losses

$$\Gamma_{\mathrm{loss}}^{\mathrm{nl}} = \frac{v_{\mathrm{gr}}}{L_{\mathrm{abs}}} \left(\frac{\gamma}{\Delta_{\mathrm{Kerr}}} \frac{\rho_{\mathrm{DSP}}}{n}\right)^2. \quad (11)$$

One notices that the characteristic time scale τ of both processes is determined by the ratio of absorption length to group velocity. For typical vapor values, such as $v_{\mathrm{gr}} = 1$ km/s and $L_{\mathrm{abs}} = 1$ cm, one finds $\tau = 10^{-5}$ s, while for a solid or a condensate of atoms where L_{abs} can be as small as 10 μm, one finds $\tau = 10^{-8}$ s. Since $\gamma/\Delta_{\mathrm{Kerr}}$ as well as ρ_{DSP}/n are small compared to unity, elastic collisions happen always on a much shorter time scale than the nonlinear losses.

In addition to the nonlinear loss, there is also a linear absorption resulting from the imaginary part of the polariton mass. It corresponds to the absorption of high spatial-frequency components in EIT. If L denotes the characteristic longitudinal length scale of the polariton, the linear loss rate can be estimated from Eq. (5):

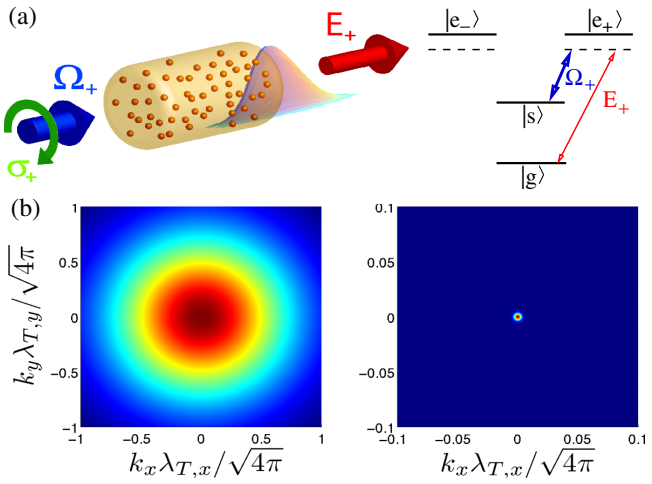

FIG. 3 (color online). (a) Detection of condensation: switching off one of the two control beams transforms the stationary into a propagating polariton which can be observed as emitted light pulse. (b) Transverse emission profile above (left) and much below (right) condensation. An ideal quasihomogeneous Bose-gas was assumed with transversal Gaussian distribution of width 178 $\lambda_{T,x}$ with $\lambda_{T,x}$ being the transversal de Broglie wavelength at temperature T.

$$\Gamma_{\text{loss}}^{\text{lin}} = \frac{v_{\text{gr}}}{L_{\text{abs}}}\left(\frac{L_{\text{abs}}}{L}\right)^2. \tag{12}$$

In order for the elastic collisions to be fast compared to the linear losses, it is necessary that the optical depth $\text{OD} \equiv L/L_{\text{abs}}$ of the medium over the length L of the polariton wave packet fulfills

$$\text{OD} = \frac{L}{L_{\text{abs}}} > \sqrt{\frac{\Delta_{\text{Kerr}}}{\gamma}\frac{n}{\rho_{\text{DSP}}}}. \tag{13}$$

Taking the above example of $v_{\text{gr}} = 1$ km/s and $L_{\text{abs}} = 1$ cm or 10 μm, respectively, and assuming $\Delta_{\text{Kerr}} = 100\gamma$ and $\rho_{\text{DSP}}/n = 0.1$ finally yields an elastic collision rate of $\Gamma_{\text{el}} \approx 10^2$ s^{-1}, or 10^5 s^{-1}. Thus, for sufficiently large elastic collision rates, one needs a large optical depth $\text{OD} > 30$, which is feasible however in solid-state systems or magneto-optical traps.

The signature of condensation is here the macroscopic occupation of the $\mathbf{k} = \mathbf{0}$ momentum state or, respectively, the buildup of spatial coherence. The transition into modes with vanishing transverse momentum $\mathbf{k}_\perp = 0$ can easily be observed by switching off one of the two control laser. In this case, the stationary polariton will be transformed into a moving one and will leave the sample as a light pulse. From the transverse profile of the emitted pulse, one can deduce the transverse coherence length as indicated in Fig. 3.

In summary, we have shown that stationary-light polaritons behave as massive Schrödinger-like particles with small and variable mass. In the presence of a resonantly enhanced optical Kerr nonlinearity, the stationary DSPs undergo elastic collisions at a rate determined by the group velocity divided by the absorption length multiplied by the inverse normalized Kerr detuning. For a sufficiently high optical density of the sample, the elastic collisions are fast enough to mediate a stimulated transition into the $\mathbf{k} = \mathbf{0}$ polariton mode. The critical temperature for this can be orders of magnitude larger than that of atoms. Condensation can be observed by releasing the stationary field into a propagating field and observing the transverse mode profile as well as the temporal coherence. In contrast to exciton-polaritons, no cavity is needed, and the polariton gas is three dimensional.

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