Observation of Dynamical Quantum Phase Transition with Correspondence in Excited State Phase Diagram

T. Tian, H.-X. Yang, L.-Y. Qiu, H.-Y. Liang, Y.-B. Yang, Y. Xu, and L.-M. Duan

Center for Quantum Information, IIGS, Tsinghua University, Beijing 100084, PR China

Dynamical quantum phase transitions are closely related to equilibrium quantum phase transitions for ground states. Here, we report an experimental observation of a dynamical quantum phase transition in a spinor condensate with correspondence in an excited state phase diagram, instead of the ground state one. We observe that the quench dynamics exhibits a non-analytical change with respect to a parameter in the final Hamiltonian in the absence of a corresponding phase transition for the ground state there. We make a connection between this singular point and a phase transition point for the highest energy level in a subspace with zero spin magnetization of a Hamiltonian. We further show the existence of dynamical phase transitions for finite magnetization corresponding to the phase transition of the highest energy level in the subspace with the same magnetization. Our results open a door for using dynamical phase transitions as a tool to probe physics at higher energy eigenlevels of many-body Hamiltonians.

Non-equilibrium quantum many-body dynamics have seen a rapid progress in recent years due to deepened theoretical understanding [1–4] and experimental technology advances in systems, such as trapped ions [5, 6], Rydberg atoms [7], ultracold atoms [2, 8, 9, 11], nitrogen-vacancy centers [12], and others [13]. One central question in the field concerns the existence of phase transitions as a system parameter is suddenly varied (referred to as dynamical quantum phase transitions [2–4]). Based on different identification features, such a phase transition can generally be divided into two types. One type refers to the existence of a non-analytical behavior in a long time steady state of a local order parameter with respect to a final Hamiltonian parameter [14, 15]. The other type corresponds to the emergence of a singularity in a global order parameter such as Loschmidt echoes with respect to time after a quench [16, 17]. Both of these two types of dynamical phase transitions are closely related to the ground state quantum phase transition. However, exceptions exist and the Loschmidt echo is allowed to show non-analytical behavior even though a system parameter is quenched within an identical ground state phase [8, 18–21]. Moreover, whether the dynamical phase transition with no correspondence in ground state phase diagram is related to an excited state quantum phase transition is still an open question [2, 22–26].

Similar to the ground state quantum phase transitions, excited state quantum phase transitions refer to the existence of singularities in the energy or an order parameter of an excited energy level [22, 23]. While such a phase transition has been proposed for more than a decade, it has not been experimentally observed in a many-body quantum system. Recently, Ref. [27] has theoretically proposed a dynamical phase transition that is closely related to the quantum phase transition for the highest energy level in a subspace with zero spin magnetization in a spinor condensate. From this perspective, the spinor condensate provides an ideal experimental many-body quantum platform for probing the excited state quantum phase transitions by quench dynamics. In fact, many non-equilibrium phenomena, such as spin domains, topological defects and Kibble-Zurek mechanism, have been experimentally observed in a spinor condensate [28–38]. In addition, the highest energy level in the subspace has an upper bound in energy in a finite system, reminiscent of a state with a negative absolute zero temperature, which has been experimentally realized [39–43].

In this paper, we report the experimental observation of a dynamical quantum phase transition with correspondence in the highest energy level phase diagram in a subspace with fixed spin magnetization in a spinor condensate. Instead of measuring a long time steady value of an order parameter such as the number of atoms with zero spin, we probe the value of the first peak of the time evolution of the atom number appearing in a short time. By preparing a condensate in an antiferromagnetic (AFM) state, we find that the quench dynamics show a non-analytical change as a function of the quadratic Zeeman energy of a final Hamiltonian at \( q_f = 2c_2 \) (\( c_2 \) describes an interaction strength) as \( q \) is suddenly varied from a large negative value to \( q_f \). Our results are beyond the ground state phase transition given the absence of a phase transition at \( q = 2c_2 \). However, our finding is highly related to the phase transition between an AFM and a broken-axisymmetry (BA) phase for the highest energy level in the subspace with zero spin magnetization. We further measure the quench dynamics for finite magnetization and find singular behaviors determined by the phase transition on the upper energy level in the subspace with fixed spin magnetization.

We start by considering a spin-1 BEC described by the following Hamiltonian [44, 45]

\[
\hat{H} = c_2 \frac{\hat{L}_z^2}{2N} + \sum_{m_F=-1}^1 (qm_F^2 - pm_F) \hat{a}_{m_F}^\dagger \hat{a}_{m_F},
\]

(1)

under a widely used single spatial mode approximation, where a spatial wave function \( \Psi(r) \) is approximated to be spin independent so that the atomic field operator can be decomposed as \( \Psi_{m_F}(r) \approx \Phi(r) \hat{a}_{m_F} \) with \( m_F = -1, 0, 1 \) being the magnetic spin quantum number. Here, \( N \) is the total atom number, \( c_2 \) is the spin-dependent interaction energy, \( p(q) \) is linear (quadratic) Zeeman energy, and \( \hat{L}_\mu = \sum_{i,j} \hat{a}_i^\dagger (F_{ij})_{\mu \nu} \hat{a}_j \) is a total spin operator with \( F_{ij} \) being the spin-1 angular momentum matrix along the \( \mu \) direction and \( \hat{a}_j \) (\( \hat{a}_j^\dagger \))
being an annihilation (creation) operator.

To explore dynamical quantum phase transitions, we prepare a condensate of sodium atoms in an AFM state with zero magnetization [equivalent to zero linear Zeeman energy ($p = 0$)] and then suddenly change the quadratic Zeeman energy $q$ to a final value $q_f$ at $t = 0$. As the system evolves under the final Hamiltonian, the quench dynamics can be measured. A non-analytic change in the measured quantity as a function of the final Hamiltonian parameter $q_f$ can be regarded as a signature of dynamical quantum phase transitions. Since the total magnetization is conserved during the time evolution, i.e., $[\hat{H}, \hat{L}_z] = 0$, the quench dynamics is restricted in the subspace with fixed eigenvalue of $\hat{L}_z$. For sodium atoms, which have positive $c_2$, without any linear Zeeman energy, the ground state has a phase transition at $q = 0$ from an AFM phase with equally populated atoms on the $m_F = \pm 1$ levels to a polar phase with all atoms occupying the $m_F = 0$ level [see Fig. 1(a)]. After a quench, the dynamics is restricted in the subspace with zero magnetization. In this subspace, the highest energy level exhibits a phase transition at $q = 2c_2$ between a phase with nonzero population on the $m_F = 0$ level corresponding to the BA phase in the mean-field approximation and an AFM phase and at $q = -2c_2$ between a BA phase and a polar phase [46], similar to rubidium atoms with negative $c_2$, as shown in Fig. 1(a2).

In experiments, directly detectable physical quantities are the number of atoms with spin-$m_F$ divided by the total atom number, i.e., $\rho_{m_F} = \hat{a}_{m_F}^\dagger \hat{a}_{m_F} / N$, and their average $\langle \rho_{m_F} \rangle$ over many experimental ensembles. A dynamical phase transition is usually characterized by an asymptotic long-time steady value of a local order parameter, which in our case can be chosen as $\langle \rho_0 \rangle_\infty = \lim_{T \to \infty} \frac{1}{T} \int_0^T \langle \rho_0 \rangle dt$. Fig. 1(b) shows its increase from zero as $q_f$ is decreased from $2c_2$ (see also Ref. [2]), in stark contrast to the ground state phase diagram without any phase transition at this point. In fact, the dynamical phase transition at $q_f = 2c_2$ corresponds to the quantum phase transition of the highest energy level in the subspace with zero magnetization. This connection can be easily explained in the mean-field approximation. In this approximation, the ground state for $q_1 = -\infty$ and the highest energy state for $q_1 > 2c_2$ share the same wave function since they are both in the AFM phase with zero $\langle \rho_0 \rangle$. It follows that $\langle \rho_0 \rangle$ remains zero when we suddenly vary $q_1$ from $-\infty$ to $q_f$ with $q_f > 2c_2$. Yet, when $q_f < 2c_2$, the time evolved state is no longer an eigenstate of $\rho_0$, leading to the appearance of nonzero values for $\langle \rho_0 \rangle$ as shown in Fig. 2(a). This picture is also valid in the many-body level given that the initial state has a significant probability to overlap with the highest energy state of the final Hamiltonian in the subspace when $q_f > 2c_2$.

In real experiments, it is a significant challenge to observe the long-time average of $\langle \rho_{m_F} \rangle$ as the long-time relaxation dynamics is unavoidable. Fortunately, the model Hamiltonian Eq. (1) actually describes a system of N spin-1 particles with effectively infinite-range interactions [2]; this enables us to characterize the dynamical phase transition by alternative finite-time observables: $\rho_{0,\text{peak}} \equiv \langle \rho_0 \rangle(t = \tau_{\text{peak}})$ and $\delta \rho_{0,\text{peak}} = \delta \rho_0(t = \tau_{\text{peak}})$, the value of $\langle \rho_0 \rangle$ and the standard deviation of $\rho_0$ at the first peak of the spin oscillations, respectively [see Fig. 2] [2]. The occurrence time $\tau_{\text{peak}}$ of the first peak is around several tens of milliseconds, making the experimental observation feasible. Indeed, the dynamical phase transition at $q_f = 0$ reflecting the ground phase transition has been experimentally demonstrated [2]. However, to observe the dynamical phase transition at $q_f = 2c_2$, one needs to reduce the rapid relaxation toward the ground states for large $q_f$. We here solve this challenging problem by significantly reducing the atom number to around $5.8 \times 10^3$ [46].

In experiments, a spin-1 BEC is produced via an all-optical procedure as detailed in Ref. [48]. We then apply a magnetic field gradient to remove the atoms on $|m_F = \pm 1\rangle$ out of the BEC cloud [49], followed by equilibrating the system by holding for 1s. After that, we shine a $\pi/2$-pulse radio frequency radiation to create a nearly AFM state, which has zero magnetization and zero component on the $m_F = 0$ level. Since the experiment is very sensitive to the initial value of $\langle \rho_0 \rangle$ [50], we then immediately apply a microwave pulse for 300 ms with a frequency of 1.7716264 GHz, whose detuning is zero for the clock transition from $|F = 1, m_F = 0\rangle$ to $|F = 2, m_F = 0\rangle$ [the Rabi rate is about 1.9kHz [51] and the applied magnetic field ranges from 0.2 G to 0.373 G for the experiments in Fig. 1(c)]. This pulse allows us to excite the atoms on the hyperfine level $|F = 1, m_F = 0\rangle$.

![FIG. 1.](image-url) (Color online) $\langle \rho_0 \rangle$ as a function of the quadratic Zeeman energy $q$ for (a1) the ground state and (a2) the highest energy state with zero magnetization. The quench dynamics is achieved by suddenly varying $q$ from a large negative value to $q_f$, as schematically shown by the red arrow. Experimentally observed (b) $\rho_{0,\text{peak}}$ and (c) $\delta \rho_{0,\text{peak}}$ with respect to $q_f$, in comparison with the theoretical results (solid lines). $\langle \rho_0 \rangle_\infty$ is plotted as a purple line. (d) Experimentally observed occurrence time $\tau_{\text{peak}}$ of the first peak of $\langle \rho_0 \rangle$ (solid circles), compared with the theoretical results (black line). Here, $c_2/h = 15.2 \pm 0.2$ Hz.
to another level $|F = 2, m_F = 0\rangle$; these atoms then escape from the trap quickly since the latter energy level is quite unstable and the atoms on this state suffer a significant loss. We therefore prepare the initial state with $\rho_0 = 0$ and $m_z = \rho_1 - \rho_{-1} \approx 0 \pm 0.015$. Note that we use a relatively weak microwave field to avoid apparent atom loss.

To study the spin dynamics, the quadratic Zeeman energy $q$ should be suddenly tuned. This can be experimentally achieved by controlling a magnetic field or a microwave pulse, since $q = q_M + q_S$, where $q_M$ and $q_S$ are the quadratic Zeeman energy induced by the microwave pulse and magnetic field, respectively [52–54]. During the preparation of the initial state, we fix the magnetic field so that its contribution to the quadratic Zeeman energy is equal to our final quadratic Zeeman energy $q_f$, i.e., $q_f = q_M \propto B^2$, which can be easily identified by measuring the Zeeman splitting induced by the magnetic field $B$. Simultaneously, we apply a resonant microwave pulse (the same pulse is also used to remove the remaining atoms on the $m_F = 0$ level), generating a large negative quadratic Zeeman energy [52]. To achieve the sudden quench, we quickly switch off the microwave pulse, leading to the final $q_f$.

After that, we perform the measurement of the fractional population $\rho_0$ via the standard Stern-Gerlach fluorescence imaging technique with respect to time. The experiments are repeated for 40 times at each time for each $q_f$, and the average value $\langle \rho_0 \rangle(t)$ and the standard deviation $\delta \rho_0(t)$ are then determined.

In Fig. 1(b) and (c), we show our experimental results of $\rho_0$ and $\delta \rho_0$ as a function of $q_f$, respectively. Both quantities are zero when $q_f > 2c_2$ and then exhibit a linear increase as $q_f$ decreases when $q_f < 2c_2$, which agrees well with our theoretical simulation, predicting the existence of a second-order dynamical phase transition at $q_f = 2c_2$.

In Fig. 2(a) and (b) display the experimentally observed $\rho_0(q_f)$ and $\delta \rho_0$ as a function of $q_f$, respectively. The purple line depicts the theoretically calculated $\rho_0$ versus $q_f$ for the highest energy level with $m_z = 0$, while the purple line denotes this quantity for a time evolved state. Here, $c_2/h = 13 \pm 0.7$ Hz.

FIG. 2. (Color online) Time evolution of $\rho_0$ (blue squares) and $\delta \rho_0$ (red circles) for (a) $q_f = 0.3c_2$ and (b) $q_f = 2.2c_2$. (c) and (d) plot the theoretical predicted results under the same parameters as (a) and (b), respectively. For each $q_f$ and time, we perform 40 times measurements. The background squares show the probability that a measurement outcome occurs. In (c) and (d), the probability is obtained by sampling 40 samples using the Monte Carlo method. Here $c_2/h = 15.2 \pm 0.2$ Hz measured by spin oscillations.

FIG. 3. (Color online) Quasi static measurement of the quantum phase transition in the excited state with $m_z = 0$ achieved by slowly decreasing $q$ across $2c_2$ after $q$ is suddenly changed to $2.3c_2$. The green circles and red squares denote the experimentally observed $\rho_0$ and $\delta \rho_0$, respectively, while the purple and black lines denote the numerical results of the corresponding quantities, respectively. The blue line depicts the theoretically calculated $\rho_0$ versus $q_f$ for the highest energy level with $m_z = 0$, while the purple line denotes this quantity for a time evolved state. Here, $c_2/h = 13 \pm 0.7$ Hz.
tation so that atoms do not share the same spatial wave function, breaking down the single mode approximation. In fact, such a relaxation process is strongly enhanced for larger $m_z$, inevitably involving the energy transfer into the spatial modes. In experiments, we prepare the BEC in an AFM state as previously described. We then apply a microwave pulse for $10 \text{ ms}$ to excite atoms from the hyperfine level $|F = 1, m_F = +1 \rangle$ to $|F = 2, m_F = 0 \rangle$. Since the lifetime of the atoms on the level $|F = 2, m_F = 0 \rangle$ is very short, this operation decreases the number of atoms on $|F = 1, m_F = +1 \rangle$. Using this procedure, we are able to prepare a state with different $m_z$ by tuning the microwave frequency. After that, we immediately apply a microwave pulse for $290 \text{ ms}$ to pump atoms on $|F = 1, m_F = 0 \rangle$ to $|F = 2, m_F = 0 \rangle$; this process removes all atoms on $|F = 1, m_F = 0 \rangle$ for a fixed $m_z$ while keeping the quadratic Zeeman energy a large negative value. Finally, we suddenly switch off the microwave radiation, leading to a sudden change of the quadratic Zeeman energy, and then perform a measurement for $\rho_0$ as time evolves. Our experimental results for three distinct $m_z$ are shown in Fig. 4(e). We see clearly the decrease of the critical phase transition points as $|m_z|$ increases, which agrees well with theoretical prediction.

In summary, we have experimentally studied the dynamical phase transition in a spinor condensate by suddenly tuning the quadratic Zeeman energy. The dynamical phase transition is demonstrated by the appearance of a non-analytical change in the spinor atom number as a function of a final Hamiltonian parameter. We find that the dynamical phase transition has a correspondence with the highest energy level phase transition for both cases of zero and finite magnetization.

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* These authors contributed equally to this work.
FIG. S1. (Color online) Theoretically calculated (a) energy per particle of the highest excited state in a subspace with zero magnetization, (b) its first and (c) second derivative with respect to \( q \). The units of \( E \) and \( q \) are \( c_2 \).

FIG. S2. (Color online) Experimentally measured \( \langle \rho_0 \rangle \) as a function of time for distinct \( c_2 \). As the interaction strength \( c_2 \) is increased by raising the atom number, \( \langle \rho_0 \rangle \) develops nonzero values instead of remaining zero as time progresses, reflecting that the atoms tend to decay into the ground state with \( \rho_0 = 1 \) of the final Hamiltonian. Here, \( q_f \approx 2.1 c_2 \).

SUPPLEMENTAL MATERIAL

In the supplementary material, we will show the presence of singularities in the energy of the highest excited state in a subspace with zero magnetization and show the effects of \( c_2 \) on the relaxation process.

To illustrate the existence of a singularity in the energy of the highest excited state, we plot the level’s energy in Fig. S1. Clearly, the second derivative of the energy with respect to \( q \) exhibits a discontinuous jump at \( q = \pm 2 c_2 \), implying the existence of a second-order excited state quantum phase transition there. This is consistent with the existence of a discontinuous jump for the first derivative of the order parameter \( \langle \rho_0 \rangle \) with respect to \( q \) [see Fig. 1(a2)].

To show the effects of \( c_2 \) on the relaxation process, in Fig. S2, we plot the measured \( \langle \rho_0 \rangle \) as a function of time after \( q \) is suddenly quenched to \( q_f = 2.1 c_2 \) for different \( c_2 \), which is controlled by tuning the atom number \( N \), given \( c_2 \propto N^{2/5} \) under Thomas-Fermi approximation. The figure demonstrates that while \( \langle \rho_0 \rangle \) remains smaller than 0.4% for small \( c_2 \) (there are no observable atoms for \( \rho_0 \) except for the noise of a camera), it increases from zero for sufficiently large values of \( c_2 \), implying that the system decays toward the ground state of the final Hamiltonian with \( \langle \rho_0 \rangle = 1 \). Our results are consistent with previous observation that the relaxation is stronger for larger \( q_f \) and \( c_2 \) [S1, S2].

* These authors contributed equally to this work.
† yongxuphy@tsinghua.edu.cn
‡ lmduan@tsinghua.edu.cn

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