We report on the experimental observation of the quantum phase transition from a para- to a ferromagnetic state in a spatially extended atomic condensate in the presence of a coherent coupling between two internal spin states. The nature of the transition is experimentally assessed by looking at the phase diagram as a function of the control parameters, at hysteresis phenomena, and at the magnetic susceptibility and the magnetization fluctuations around the critical point. Agreement with mean-field calculations is shown. We develop experimental protocols to deterministically generate domain walls that separate spatial regions of opposite magnetization in the ferromagnetic state. Thanks to the enhanced coherence properties of our atomic superfluid system compared to standard condensed matter systems, our results open the way towards the study of different aspects of the relaxation dynamics in isolated coherent quantum systems.

I. INTRODUCTION

Superfluidity in many-body quantum systems leads to interesting and notable transport and coherence properties [1–3]. Such properties are due to a thermal second-order transition from a normal to a superfluid state, a transition which is formally characterised by the spontaneous breaking of the $U(1)$ symmetry related to particle number conservation. Since such a transition is driven by the Bose statistics, atom-atom interactions are not needed to observe it. Still, they play an important role in stabilizing the superfluid phase against disturbances, e.g. guaranteeing a finite compressibility and a finite critical Landau velocity for superfluidity.

On top of this, superfluids can also have internal degrees of freedom, leading to order parameters with a non-trivial spinor or vector structure [4]. In this case, non-spin-symmetric interactions may lead to ground states with a very different spinor structure of the order parameter depending on the interactions. A natural question is therefore whether the transition between different states can be described as a quantum phase transition (QPT), and, if so, which universality class such a QPT belongs to, what is the interplay between the superfluid nature of the system and the QPT, and whether the QPT in these system survives at low yet finite temperature [5]. The paradigmatic model for (continuous) QPT is the so-called quantum Ising model [6, 7], where the ferro- (or antiferro-) magnetic interactions along one spin direction of the standard Ising model compete with a transverse magnetic field. The field theory describing the order parameter dynamics near the critical point is the so-called $\phi^4$ theory, which represents the prototypical Ginzburg-Landau functional for a continuous phase transition, with its iconic single- to double-well energy landscape transition upon the change of an external parameter [8].

Combining these ideas, recent theoretical work (see review in [9]) has anticipated a phase transition in the quantum Ising universality class in a two-component atomic Bose-Einstein condensate (BEC) subject to an external field that coherently couples the two components [10]: at zero temperature, mean-field theory predicts an interaction-driven transition from a paramagnetic (PM) to a $\mathbb{Z}_2$-symmetry breaking ferromagnetic (FM) state and the low-energy magnetic fluctuations are described by a $\phi^4$ theory.

In this work, we investigate this magnetic QPT in a superfluid cloud of sodium atoms. We experimentally demonstrate the ferromagnetic nature of our two-component superfluid by providing a detailed characterization of the phase diagram and of the associated hysteresis phenomena. Moreover, we show the possibility of deterministically generating domain walls between different magnetic states.

Besides providing a clear illustration of an exciting fundamental physics, our results highlight the potential of two-component BECs as a new platform where to explore QPTs. As compared to previous works on spinor quantum gases, our elongated geometry allows us to depart from the 0-dimensional magnetic models often realized in this context [11] and to take advantage of the rich manipulation and detection toolset available for cold atoms to explore the critical physics of the phase transition in spatially extended systems. As compared to usual solid-state systems, our platform features important advantages: on the one hand, the cold atom platform allows for a microscopic description of all the interaction processes taking place in the system and, therefore, is amenable to a quantitative comparison with theory. On the other hand, the superfluid nature of the cloud and the smoothness of the trapping potential remove all those complications that normally stem from the unavoidable disorder of solid-state systems and their fast incoherent relaxation process, hence allowing to focus on the intrinsic many-body
properties of the stationary states. On a longer run, we expect that this feature will be of extreme importance in view of using our platform to experimentally study coherent relaxation phenomena in isolated quantum systems, such as the time-dependent motion of domain walls or the quantum-induced bubble-mediated decay of metastable states.

The structure of the article is the following: In Section II, we illustrate the properties of the PM-FM phase transition and define the relevant quantities in our atomic system. We describe our novel atomic platform and the experimental protocol in Section III. Section IV shows the experimental results, as well as a comparison with mean field theory, and Section V is devoted to the controlled generation and observation of magnetic domain walls. Conclusions and future perspectives are reported in Section VI.

II. PARA- TO FERROMAGNETIC PHASE TRANSITION

As discussed in the Introduction, our goal is the realization of the PM to FM phase transition in a coherently-coupled two-component superfluid. With the aim of clarifying the analogy between our platform and a magnetic system, in this Section we briefly recall the relevant properties of such a transition in a spin chain and we then map the magnetic model onto our atomic system.

A. The magnetic model

A textbook model of a FM to PM transition at zero temperature is based on a spin chain, subject to an external magnetic field and to internal spin-spin interactions. Within a mean-field approach, the energy of a ferromagnetic material can be written [12] in terms of the local
The magnetic system

$E(S) \propto - \int \left( B \cdot S - \frac{1}{2} \nabla \cdot (S \times \vec{K} S) - \frac{1}{2} \nabla S^2 \right) dV. \quad (1)$

In the previous expression, $B$ is the external field, $\vec{K}$ is a diagonal matrix describing the anisotropic magnetic interactions in the material due to, e.g., the sample crystalline structure, and the last term is the exchange energy, which accounts for the tendency of having a spatially uniform magnetization. In the absence of any damping, the dynamics of the local spin is given by the (dissipationless) Landau-Lifshitz equation \[ \partial_t S = -\mathbf{H}_{\text{eff}} \times S, \] i.e., a non-linear precession around the effective field $\mathbf{H}_{\text{eff}} = -\delta E/\delta S$.

For the later analogy with our two-component superfluid platform, we consider a translationally invariant ferromagnet of spin density $n = |S|$ with uniaxial magnetic anisotropy such that the only non-zero element of the magnetic interactions is $K_{33} = \alpha < 0$, which sets the easy axis along the axial direction 3. The magnetic field is uniform and has components along the axial direction ($B_3$) and in the transverse plane ($B_1$). The ground state solutions are characterized by homogeneous profiles, and a uniform effective magnetic field $\mathbf{H}_{\text{eff}} = (B_1, 0, B_3 - \alpha S_3)$. In this case, the energy of the system can be written as

$E(Z, \phi) \propto -B_3 Z - \frac{|\alpha|n}{2} Z^2 - B_1 \sqrt{1 - Z^2} \cos \phi, \quad (2)$

where $Z = S_3/n$ is the relative magnetization and $\phi = \arctan(S_2/S_1)$ is the angle of the spin in the plane. The ground state is obtained by minimizing the energy $E(Z, \phi)$ with respect to $Z$ and $\phi$. Assuming $B_1 > 0$, all ground states have $\phi = 0$. The relative magnetization $Z$ is, instead, a function of $B_3/B_1$ and $|\alpha|/B_1$, as shown in Fig. 1. The energy profiles computed using Eq. (2) are shown for eight different points in the phase diagram (see panels A-H). If $B_3 = 0$, the energy landscape shows a transition from a single minimum (paramagnet) with $Z = 0$, when $|\alpha|/B_1 < 1$, to a symmetric double minimum (ferromagnet) with $Z \neq 0$, when $|\alpha|/B_1 > 1$, corresponding to the $Z_2$ symmetry breaking, $Z \leftrightarrow -Z$ (see bottom grey panel in Fig. 1).

In the presence of a finite $B_3$, the energy minimum is shifted to a finite magnetization in the PM phase, while in the FM region an energy splitting is observed between the two minima, corresponding to the absolute ground state and to a metastable state, whose lifetime is expected to depend on the height of the barrier between the two minima [14]. For very strong $B_3$ beyond some critical value (panels D,H), one of the two minima disappears leading to a saturated ferromagnet (S-FM).

B. The atomic system

The magnetic model discussed above can be used to describe the spin sector of an atomic superfluid mixture of two spin states $|\uparrow\rangle$ and $|\downarrow\rangle$. The correspondence is based on identifying the spin vector components with the population difference $S_3 = n_\uparrow - n_\downarrow$ and the intercomponent coherences with $S_1$ and $S_2$. Given the positive intra- and intercomponent scattering lengths $a_{\uparrow\uparrow}$, $a_{\uparrow\downarrow}$ and $a_{\downarrow\downarrow}$, we focus on a configuration with $a_{\uparrow\uparrow}^2 > a_{\downarrow\downarrow} a_{\uparrow\downarrow}$, which, in the absence of a coupling between the two states, is unstable against spatial phase separation of the two components. The mixture is stabilized by the presence of a coherent radiation with amplitude $\Omega_R$ and detuning $\delta_B$, which allows for state inter-conversion. The detuning $\delta_B$ corresponds to the frequency difference between the hyperfine splitting of the two internal levels including the linear Zeeman energy shift, and the frequency of the driving microwave.

Table I illustrates how the Rabi coupling and interaction unbalances map into the components of an effective field in the magnet model (more details can be found in the Appendix A). The role of the transverse field, $B_1$, is played by $\Omega_R$. The axial component of the external field, $B_3$, has two contributions: the detuning $\delta_B$ and the imbalance of the intra-component atomic interaction energy in the two states $n\Delta \propto (a_{\downarrow\downarrow} - a_{\uparrow\uparrow})n$. The difference between intra- and intercomponent scattering lengths $\kappa \propto [(a_{\downarrow\downarrow} + a_{\uparrow\uparrow})/2 - a_{\downarrow\downarrow}]$ represents the anisotropic magnetic interactions in the material, uniaxial along direction 3. Therefore, the resulting effective magnetic field is

$\mathbf{H}_{\text{eff}} = (\Omega_R, 0, \delta_B - \kappa n Z), \quad (3)$

where $\delta_B = \delta_B + n\Delta$. In the following, we will use this atomic parameter notation to describe the phase diagram. The precise definition of the parameters $\Delta$ and $\kappa$, which takes into account the geometry of our sample, as well as their experimental estimation, can be found in the Appendices A and D.

| Physical Quantity | Magnetic System | Atomic System |
|-------------------|----------------|--------------|
| Anisotropy Interactions | $\alpha n$ | $\kappa n$ |
| Axial field | $B_3$ | $\delta_B + n\Delta$ |
| Transverse field | $B_1$ | $\Omega_R$ |
| Spin States | $|\uparrow\rangle$ | $|2, -2\rangle$ |
| | $|\downarrow\rangle$ | $|1, -1\rangle$ |
| Magnetization | $S(|S| = n)$ | $Z = S_3/n$ |

TABLE I. Mapping between magnetic and atomic system.

III. THE EXPERIMENT

A. Atomic sample

In contrast to recent works that investigate dynamical properties across a QPT [15, 16] using a rubidium two-component spin mixtures, we realize our two-level
system, choosing sodium atoms and selecting the hyperfine spin states \( |F, m_F⟩ = |2, -2⟩ \equiv |↑⟩ \) and \( |1, -1⟩ \equiv |↓⟩ \), where \( F \) is the total angular momentum and \( m_F \) its projection. This yet unexplored spin combination has interesting features for our purposes.

First of all, it is stable against spin-changing collisions and possesses intra- and intercomponent scattering lengths \( a_{11} = 54.5 \text{ a}_0 \), \( a_{1\uparrow} = 64.3 \text{ a}_0 \), \( a_{1\downarrow} = 64.3 \text{ a}_0 \), being \( \text{ a}_0 \) the Bohr radius \([17]\), that make it an immiscible mixture in the absence of coherent coupling \( \Omega_R \) and, for generic \( \Omega_R \), allows to describe in terms of the magnetic model. By combining the chosen spin mixture, sufficiently large peak density \( n \), and high magnetic field stability guaranteed by a dedicated magnetic shield \([18]\), we are able to investigate the static properties of the system across the QPT. In fact, if the typical Zeeman shift associated to the residual magnetic field fluctuations is \( \Delta E/\hbar \ll |\kappa| n \), then the ratio between spin interaction energy and the coupling energy \( |\kappa| n/\Omega_R \) can be finely tuned above and below unity, while keeping the mixture coherent during the whole duration of the measurement.

**B. Sample preparation**

We prepare condensates with typical total atom numbers \( N = 10^6 \) and peak densities of \( n = 10^{14} \text{ atoms/cm}^3 \) in a hybrid trap \([19]\) inside a magnetic shield that allows for a field stability at the few \( \mu \text{G} \) level \([18]\). We set an external magnetic field bias of 1.3 G, necessary to split the magnetic sublevels. A microwave radiation around 1.769 GHz is used to coherently couple the two states \( |↑⟩ \) and \( |↓⟩ \) with tunable intensity \( \Omega_R \). The detuning \( \delta_B \) is controlled by finely tuning the external field.

The degenerate sample is trapped in an elongated optical harmonic trap, where the trapping frequencies \( \omega_x/2\pi = 2 \text{ kHz} \) and \( \omega_y/2\pi = 20 \text{ Hz} \) are chosen to ensure an effective one-dimensional spin dynamics \([20]\). In this configuration, the BEC is cigar-shaped and presents an inhomogeneous axial density profile with a characteristic parabolic shape (see Fig. 2), typical of a harmonically-trapped system in the Thomas-Fermi (TF) regime, with a TF radius \( R_s \approx 200 \text{ \mu m} \).

Given the external trapping, our system is tightly confined along the transverse directions \( y, z \) and the use of an effective one-dimensional density \( n(x) \) is justified. Thanks to the smooth density variation, we can make use of a local density approximation (LDA) for the effective magnetic field \( H_{\text{eff}} \) through the replacement \( n \rightarrow n(x) \).

Since the parameter characterizing the phases of the equivalent magnetic system is \( |\kappa| n(x)/\Omega_R \), the spatial dependence of \( n(x) \) allows us to observe a spatially resolved phase diagram with different magnetic phases coexisting in the same sample. The tilted yellow lines in Fig. 1 represent the regions of the phase diagram which can be experimentally accessed in a single-shot experiment for different choices of \( \delta_B \).

**C. Experimental protocol**

In order to experimentally characterize the phase diagram presented in Fig. 1, it is important to make sure that the system is always in its local energy minimum. In all our experiments, we initially prepare the system in a fully polarized state with a large detuning \( \delta_B \), and then slowly ramp \( \delta_B \) to adiabatically rotate the state to the desired final configuration.

In a first set of experiments, we initialize the system in the \( |↓⟩ \) state and linearly ramp \( \delta_B \) towards positive values with a constant speed of about 100 Hz/ms (forward ramp). In a second set of experiments, a reversed procedure is performed starting from a fully polarized state.
\[|\uparrow\rangle \] \textit{(backward ramp)} and lowering the value of \(\delta_B\).

We expect the local magnetization in the low-density tails of the cloud, for which \(|\kappa|n(x) < \Omega_R\), to smoothly change sign as a function of \(\delta_B\) (left grey panel of Fig. 1), behaving as a PM. On the contrary, the high-density central part of the cloud, for which \(|\kappa|n > \Omega_R\), should remain longer in the initial state during the \(\delta_B\) ramp, starting from an initially S-FM configuration, then entering the proper FM phase, and eventually rotating the spin to the other S-FM state once the FM region is over (right grey panel of Fig. 1). In the FM region, the presence of a double well allows for the magnetization to have opposite signs depending on the preparation protocols. The bifurcation shown in the lower grey panel of Fig. 1 cannot be observed in a single realization with given \(\Omega_R\) and \(\delta_B\), because the condition \(\delta_{\text{eff}} = 0\) is fulfilled only locally.

For each experimental run, information about the spatial spin state is gathered from absorption images of the \(|\uparrow\rangle\) and \(|\downarrow\rangle\) population, with protocols similar to those reported in Ref. [20]. Since the tight radial confinement suppresses any transverse spin excitation, as it is clear from absorption images, we focus on the spatial dependence of the relative magnetization \(Z\) along the \(x\) direction, which is obtained by integrating the magnetization of the two-dimensional raw pictures along the \(y\) direction.

Examples are shown in Fig. 2(a-b), where we plot only the left part of the cloud to show how \(Z\) changes in space for increasing \(n\). Labels 1, 2 and 3 correspond to three different configurations of the system for increasing \(\delta_B\), starting from the system in the ground state \(|\downarrow\rangle\) with large negative \(\delta_B\). The experiment is repeated also starting from the \(|\uparrow\rangle\) state with large positive \(\delta_B\) that is decreased towards negative values (backward 4, 3 and 2).

\section*{IV. EXPERIMENTAL OBSERVATION OF THE PHASE TRANSITION}

As a first quantitative measurement, we employ the experimental set-up presented in the previous Section to observe and characterize the quantum phase transition from the PM to the FM state as theoretically presented in Section II. Our study here will address the typical properties of the system stationary state, such as the phase diagram in Section IV A and the magnetic response and fluctuation properties in Section IV B.

\subsection*{A. Phase diagram and hysteresis phenomena}

In order to determine the phase diagram, panels (c) and (d) in Fig. 2 show the experimental 1D magnetization as a function of the final applied detuning \(\delta_B\) for forward and backward ramps keeping a constant Rabi frequency \(\Omega_R = 2\pi \times 400 \text{ Hz}\).

The location of the \(Z = 0\) line along a parabolic-like curve in the \((x, \delta_B)\) plane is easily explained: in a fully paramagnetic sample the zero magnetization line would coincide with the locus of points satisfying \(\delta_{\text{eff}}(x) = 0\), hereafter referred to as local resonance (indicated by the dash-dotted line in Fig. 2); with a harmonic trap in the TF regime, this curve corresponds to a parabola, due to the density-dependent detuning. In addition to this, the ferromagnetic nature of the cloud shifts the \(Z = 0\) line from the local resonance and pushes it towards the edges of the hysteresis region, the direction of the shift depending on the sign of the slope of the \(\delta_B\) ramp.

\begin{figure}[h]
\centering
\includegraphics{figure3.pdf}
\caption{Magnetic hysteresis. (a)-(b) Experimental magnetization data from Fig. 2(c-d), rescaled according to the \(|\uparrow\rangle\)-\(|\downarrow\rangle\) asymmetry and to the density profile, see main text. White regions in the bottom-left corner are due to a lack of data that manifests when applying the vertical-axis rescaling. (c)-(d) One dimensional mean-field numerical simulations for the experimental parameters of Fig. 2(c-d). The dotted black and white lines in panels (c-d) mark the border of the hysteresis region calculated from theory. Yellow dashed lines mark experimental shots shown in panel (a), corresponding to number 1-4, as in Fig. 1. (e) The width of the hysteresis \(\delta_{\text{hys}}\) is calculated as explained in the Appendix. Green points are experimental data with their uncertainties resulting from the binning procedure and systematic errors. The dotted line stands for theory, while the purple points are results from numerical simulations.}
\end{figure}
At fixed $\delta_B$, the interface spatially lags behind the local resonance: towards the tail of the cloud (see (3)) for a forward ramp, and towards the center for a backward one (see (2)). Thus, along the $x$ direction, the relative magnetization $Z$ zeros between the S-FM region (exterior) and the FM internal region. On the other hand, at fixed $x$ [see $x \sim 0$ in Fig. 2(c)-(d)], the interface is pushed towards higher values of $\delta_B$, with respect to the local resonance, in the case of a forward ramp and towards lower values for a backward one, as it was pictorially represented in the right panel of Fig. 1. This behaviour marks the evidence of a hysteresis cycle, observable both as a function of $x$ and $\delta_B$.

The raw data in Fig. 2(c-d) qualitatively agree with the expected behaviour. For a more quantitative comparison with the phase diagram in Fig. 1, we need to rescale the vertical axis and plot the observed magnetization in terms of $\delta_{eff}/\Omega_R$, to properly take into account the difference between the scattering lengths. The results of the rescaling for a set of measurements acquired at constant $\Omega_R$ are shown in Fig. 3(a)-(b). This resembles the experimental data one would gather in a system with $a_{1\downarrow} = a_{1\uparrow}$ and spatial dependent density.

We compare our measurements to a mean-field calculation [panels (c) and (d) of Fig. 3] based on one-dimensional two coupled Gross-Pitaevskii equations for the spinor superfluid order parameter $Ψ = (ψ_↑, ψ_↓)$ (see Appendix A for more details). Within this formalism, we can properly take into account both the trapping potentials and time sequence used in the experimental protocols. The local spin is given by $S = Tr(\hat{σ}Ψ^\dagger Ψ^\dagger)$, with $\hat{σ}$, the Pauli matrices. The numerical simulations confirm the observation of a hysteretic region and show a good agreement with the experimental data also for what concerns small structures resulting from the experimental protocol.

Fig. 3(e) shows the hysteresis width $\delta_{hys}$ (see Appendix for definition and calculation) as a function of $|κ|n/\Omega_R$, that has been computed analytically [see Eq. (A12)], numerically [from the simulations in panels (c-d)] and experimentally (by averaging over more than a thousand shots obtained for different $\Omega_R$). Remarkably, the ultra-stable magnetic environment ensures that uncertainty in the value of $\delta_B$ is negligible as compared to the relevant parameters of the system, leading to small experimental error on the $\delta_{hys}/\Omega_R$ axis.

The discrepancy between the numerical points and the theoretical expectation, attributed to beyond-LDA effects, is reduced by considering slower detuning ramps: if the evolution is not truly adiabatic, spin currents, which are included in the simulations, play a small, although observable, role. For what concerns the experimental data, discrepancies can be ascribed to the departure from adiabaticity in the preparation, finite temperature effects that could shift the critical point to lower values of $|κ|n/\Omega_R$, and loss of coherence. Additionally, systematic errors can be introduced while subtracting the detuning $n(x)Δ$, especially in the regions at low values of $|κ|n/\Omega_R$ that are located near the tails of the sample. Nevertheless, both simulation and experiment capture the growth of the hysteresis region predicted by the theory above the critical point.

Finally, it is worth pointing out that the hysteresis phenomena observed in [21] referred to the completely different case of a zero-dimensional single component condensate with attractive interactions in a tunable double-well potential. The crucial novelty introduced by our setup resides on the spontaneous emergence of hysteresis due to strong atom-atom interactions in a spatially extended system, which opens the way to study the interplay of hysteresis with the spatial dynamics.

### B. Magnetic susceptibility and magnetic fluctuations

In the vicinity of the phase transition many quantities characterizing the system’s response to external parameters diverge. One of these is the magnetic susceptibility $χ$, which we can extract as variation of magnetization against variation of $δ_{eff}$ as

$$χ = \frac{\partial Z}{\partial δ_{eff}}|_{δ_{eff}=0}. \tag{4}$$

Within the universality class of Landau theory, the susceptibility has a finite value at large transverse field where the magnetization follows the applied field, it goes to zero when strong interactions fix the magnetization to $|↑⟩$ or $|↓⟩$ state, and it diverges at the critical point $|κ|n(x)/\Omega_R = 1$, where small variations of the effective field lead to strong changes in $Z$.

Within the homogeneous mean-field approximation (see Eq. (2)), the susceptibility can be written as

$$\frac{1}{χ} = \left| \frac{\partial δ_{eff}}{\partial Z} \right|_{δ_{eff}=0} = |κ|n \begin{cases} \frac{Ω_R}{|κ|n} - 1 & |κ|n < Ω_R, \\ \frac{1}{|κ|n} Ω_R^2 - 1 & |κ|n > Ω_R, \end{cases} \tag{5}$$

with the typical asymmetric behaviour of a $Z_2$ phase transition in the PM and in the FM region [7]. This behaviour is well captured by the experimentally measured $χ$, shown in Fig. 4, where it is compared with the Eq. (5) (red lines) and the numerical solution of Gross-Pitaevskii equations. To suppress spurious effects arising from inhomogeneity, we restrict the analysis to regions of the sample where the density is nearly constant. Again, we attribute the mismatch between theory and experimental finite temperature effects [22] and non perfect adiabaticity in the preparation of the cloud. In particular, at low $|κ|n/Ω_R$ in the PM region, large values of the coupling $Ω_R$ lead to fast spin decoherence in the vicinity of $Z = 0$, reducing the overall contrast of the magnetization and, consequently, the value of $χ$. In the FM region, where the system is more robust against decoherence as
fixing the detuning $\delta_{\text{eff}}$ to the local resonance in the central part of the cloud, we measure the variance $\sigma^2$ of $Z$ for different values of $\Omega_R$. We select the central part of our condensate, where the density profile is almost flat, to minimize density-related effects on $\sigma^2$.

In detail, the protocol for the fluctuation measurements consists in acquiring up to 100 realizations for about 20 different values of $\Omega_R$. As a first step, we calculate $|\langle n \rangle| / \Omega_R$ for each realization by taking into account the measured atom number in the shot. The calculation of magnetic fluctuations is performed by computing the standard deviation of the 1D magnetization in a region $w_x = 120 \text{ pixel} \approx 123 \mu \text{m}$ wide. To suppress spurious effects due to the limited resolution of the imaging, we perform the $\sigma^2$ analysis by grouping $N_p$ pixels. The variance $\sigma^2$ so obtained corresponds to:

$$\sigma^2 = \left\langle \frac{1}{w_x / N_p} \frac{w_x / N_p}{|\langle n \rangle| / \Omega_R} \sum_i \left( Z_i - \frac{\sum_j w_x / N_p Z_j}{w_x / N_p} \right)^2 \right\rangle_{N_p} ,$$

where $Z_i$ is the relative magnetization of the $i$-th grouping element and $\langle \ldots \rangle_{N_p}$ is the average over different grouping sizes. The final results plotted in Fig. 4(b) are obtained by binning the fluctuation data by means of a fixed $|\langle n \rangle| / \Omega_R$ interval, where the uncertainties is taken as a combination of the standard deviation of the fluctuation between different shots and different binning. They clearly show how the measured variance is maximal at the critical point and reflects the behavior observed for the susceptibility.

It is worth noticing that, in general, for a large homogeneous system, due to the fluctuation-dissipation theorem, the fluctuations of the magnetization of the system and its susceptibility are strictly related. In our system, however, the variation of the number of atoms from shot-to-shot, the finiteness of the system and its inhomogeneity prevent us from a proper quantitative analysis of their relation.

### V. Deterministic Creation of Ferromagnetic Domain Walls

Another fundamental feature characterizing ferromagnetism is the possibility of forming spatial domains with opposite magnetization. This can take place in a stochastic way via the Kibble-Zurek mechanism during a sudden quench across the PM to FM phase transition [25–28], or by directly engineering the domains with suitable protocols. Different FM domains are separated by domain walls (DW), which constitute low-energy and long-lifetime excitations of the ferromagnet. A review of such investigations in the field of solid-state magnetism can be found in [29]. Recently, the spontaneous and deterministic creation of DWs in an effective ferromagnetic BEC under a periodic driving was shown in [30, 31]. In these works, however, the ferromagnetic DW was not...
FIG. 5. Deterministic creation of FM domain walls. (a) Experimental protocol used to create DW through a ramp on $\delta_B$. 
(b) Absorption images of the two components (left half of the system only) at the initial point (I) and after a wait time of 25 ms (IV), when $\delta_B = \delta_{ref}$ [dashed line in panel (a)]. (c) Absorption images of the two components corresponding to the solid line ramp in panel (a), where $\delta_B$ reaches $\delta_{DW}$ (II) and is then ramped back down to $\delta_{ref}$ (III). PM, S-FM and FM regions are illustrated in the line between the absorption images. The third (III) image in panel (c) shows the presence of a DW between two FM domains with opposite magnetization. (d) Continuous dependence of the position $x_{DW}$ of the DW with respect to the initial interface position $x_{ref}$ (in units of $R_x$) as a function of $(\delta_{DW} - \delta_{ref})/\Omega_R$. The red line is the theory prediction, whereas the grey area represents the position of the interface extracted from Fig. 2. Error bars show the experimental uncertainties (horizontal axes) and the shot-to-shot standard deviation (vertical axes).

In our system, we are able to control the size of the FM region of the cloud and, inside it, to deterministically create in a precise yet flexible way DWs where the magnetization $Z$ changes sign, and then control their position at will. To this purpose, we exploit the dependence of the spatial boundaries of the FM region on the applied detuning $\delta_B$ to control both the position of the DW and the extension of the FM area. Our protocol [Fig. 5(a)] consists in the following steps: 1) ramping $\delta_B$, as for the data in Fig. 2, to values $\delta_{DW}$ for which part of the system is in the FM regime; 2) waiting for 25 ms to let the system relax; 3) ramping back to a fixed detuning $\delta_{ref} = 2.5\Omega_R$ with $\Omega_R = 400 \times 2\pi$ Hz.

Figure 5(b) and Fig. 5(c) show the absorption images of the two states for the left half of the sample in case of the two different ramps illustrated in Fig. 5(a). As illustrated by the grey-black-light grey bar between the absorption images, the location where the PM region ends remains fixed at the position in the BEC where $|\kappa| n/\Omega_R = 1$. During the ramp of $\delta_B$, the interface between S-FM and FM moves accordingly, i.e., the FM region initially shrinks (step 1) and then re-enlarges going back to the initial size (step 3). However, during the ramp in step 3, the size of the FM domain in the $|\downarrow\rangle$ state remains unchanged, and a FM domain in the $|\uparrow\rangle$ state forms in the remaining FM region. In this manner, the $|\uparrow\rangle$-$|\downarrow\rangle$ interface, that previously separated S-FM from the FM, becomes a ferromagnetic DW within the FM region, at a position determined by the final detuning $\delta_{DW}$.

In Fig. 5(d), we show the experimentally measured position of the created DW as a function of $\delta_{DW}$. The theory prediction (red line) and the expected position at the beginning of the 25 ms, as extracted from Fig. 2 data (grey area), are also shown. The smooth and linear dependence observed in this figure demonstrates how the smoothness of the confinement potential allows for the continuous and deterministic control of the DW position via $\delta_{DW}$, without it being pinned by external disorder as it often happens in solids. This key result showcases the promise of our set-up in view of future studies of the quantum relaxation dynamics of the domain wall.

The slight position shift of the DW (green dots) from the magnetic interface at the end of the first ramp (gray shaded area) can be attributed to some spurious nonadiabatic spin dynamics. In fact, at fixed spatial position the disappearance of the FM metastable state, previously occupied due to the presence of hysteresis, into the S-FM one, occurs via a non adiabatic spin rotation. Thus, spin excitations are generated no matter how slow the system is manipulated [see for instance the ripples in Fig. 2(c-d) and in Fig. 3(a-d)]. Their interaction with the DW is responsible for the observed shift in its position. A similar spatial drift of the magnetization jump towards the bulk of the FM region is also visible by comparing the top and bottom panels of Fig. 5(b).
VI. CONCLUSIONS AND OUTLOOK

In this work, we explored the zero-temperature magnetic phase diagram of a two-component superfluid gas subject to an external coherent Rabi coupling. In addition to the critical region where enhancement of both magnetic susceptibility and fluctuations was detected, a special attention was paid to the ferromagnetic state where metastability and hysteresis features are observed, and domain walls separating different magnetic states are deterministically generated.

The comparison of our results (density profiles, phase diagram, susceptibility) with a zero-temperature mean-field theory seems to indicate that the finite temperature of the superfluid system does not quantitatively affect the behaviour of the QPT, and that the transition is mean-field like. In this respect, it would be interesting to study the same system in true one and two dimensions to observe the expected breaking of the mean-field character of the transition.

Our studies highlight the power of the specific two-component atomic superfluid platform employed here, for a number of key open problems. As a natural first step, one can take advantage of the non-conservation of magnetization in the system and the subsequent reinforced quantum fluctuations to analyse their scaling as a function of the subsystem size in the critical region.

Besides the investigation of the static properties of the system in its ground or metastable state immediately after the preparation, the challenge is now to extend the study to the quantum many-body dynamics. The superfluid nature of the atomic gas suggests the possibility to investigate magnetism in a novel dissipationless and collisionless regime where the coherence of the two-component superfluid is not affected on the timescale of the experiment by thermal collisions nor by the trap imperfections [32, 33]. The combination of robust isolation from the environment, and long-lasting quantum coherence in the system will open to explorations of the quantum relaxation dynamics in metastable spinor superfluid.

For instance, in the initial presence of domain walls separating ferromagnetic domains in different states, spin current may develop through the domain wall, so to push the metastable state towards its ground state. The underlying microscopic process may include dissipating the extra energy into the collective excitations of the superfluid, such as spin- or density-phonons [9, 34]. In the absence of initial ferromagnetic domains, on the other hand, relaxation of the metastable spin superfluid involves, as a preliminary step, a stochastic local spin rotation under the effect of quantum fluctuations and the subsequent spontaneous formation of ground state bubbles. The latter should grow, then, according to the previous mechanism, eventually bringing the whole system to its ground state. Beyond its intrinsic interest for quantum statistical mechanics, observing this mechanism will pave the way to the experimental study of false vacuum decay phenomena [14, 35] and will shine light on processes of crucial cosmological interest [36–38].

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Appendix A: Theoretical framework

At the mean field level, a one-dimensional superfluid spin mixture is described by two coupled GPEs for the two order parameters $\psi_\uparrow, \psi_\downarrow$:

$$i\hbar \partial_t \psi_\uparrow = \left( -\frac{\hbar^2 \nabla^2}{2m} + V + g_{\uparrow\downarrow} n_\downarrow + g_{\uparrow\uparrow} n_\uparrow \right) \psi_\uparrow - \frac{\hbar \Omega_R}{2} \psi_\uparrow$$

$$i\hbar \partial_t \psi_\downarrow = \left( -\frac{\hbar^2 \nabla^2}{2m} + V - \hbar \delta_B (t) + g_{\downarrow\uparrow} n_\uparrow + g_{\downarrow\downarrow} n_\downarrow \right) \psi_\downarrow + \frac{\hbar \Omega_R}{2} \psi_\downarrow$$

where $m$ is the sodium mass, $g_{\uparrow\uparrow}, g_{\downarrow\downarrow}$ and $g_{\uparrow\downarrow}$ are intra- and inter-component interactions, linked to the $s$-wave scattering lengths by:

$$g_{ij} = \frac{4\pi \hbar^2}{m} a_{ij}$$

Strength and detuning of the coherent couplings are indicated with $\Omega_R$ and $\delta_B$, while $V$ is the external harmonic potential trapping the atoms.

In view of analysing the magnetic properties of the mixture, it is convenient to define the spinor $\Psi = (\psi_\uparrow, \psi_\downarrow)^T$ and the density matrix $\rho = \Psi \otimes \Psi^\dagger$. The state of the mixture can be then represented on a Bloch sphere of radius $n = Tr(\rho)$ and encoded in a spin vector

$$\mathbf{S} = Tr(\sigma \rho) = n \left( \sqrt{1 - Z^2} \cos \phi, \sqrt{1 - Z^2} \sin \phi, Z \right),$$

where $\sigma$ is the Pauli matrices vector, $\phi$ the relative phase of the two components and the relative magnetization $Z$ is defined as $nZ = n_\uparrow - n_\downarrow$. The equation governing
the dynamics of the spin vector can be derived directly from Eq. (A4), Eq. (A1), Eq. (A2). Imposing that the density is uniform and the total (density) current is zero one obtains [9, 39]:

\[
\partial_t \mathbf{S} = - \mathbf{H}_{\text{eff}}(\mathbf{S}) \times \mathbf{S}. \tag{A5}
\]

The state-dependent nonlinear external field

\[
\mathbf{H}_{\text{eff}}(\mathbf{S}) = (\Omega_R, 0, \delta_B + \Delta n - \kappa n Z) + \frac{\hbar}{2 m n} \nabla^2 \mathbf{S} \tag{A6}
\]
depends on the interaction constants as:

\[
\Delta \equiv \frac{g_{\downarrow\downarrow} - g_{\uparrow\uparrow}}{2\hbar} < 0 \tag{A7}
\]
\[
\kappa \equiv \frac{g_{\downarrow\uparrow} + g_{\uparrow\downarrow}}{2\hbar} - \frac{g_{\uparrow\uparrow}}{\hbar} < 0 \tag{A8}
\]

and from a density-dependent effective detuning:

\[
\delta_{\text{eff}} \equiv \delta_B + n\Delta. \tag{A9}
\]

In the special case of uniform systems, kinetic contributions, which can be ascribed to quantum mechanical currents, can be neglected: hence the stationary condition \( \mathbf{H}_{\text{eff}}(\mathbf{S}) \times \mathbf{S} = 0 \) translates to

\[
\begin{cases}
(\delta_{\text{eff}} - \kappa n Z)\sqrt{1 - Z^2} - \Omega_R Z = 0 \\
\sin \phi = 0
\end{cases} \tag{A10}
\]

and coincides with the minimization of the energy of the system with respect to both the relative phase and the polarization:

\[
E(Z, \phi) \propto -\delta_{\text{eff}} Z + \frac{\kappa n}{2} Z^2 - \Omega_R \sqrt{1 - Z^2} \cos \phi \tag{A11}
\]

This formula, with the constraint \( \phi = 0 \), is used to calculate the energy profiles shown in the different insets in Fig. 1, and the corresponding minimizing polarization in the main graph of Fig. 1. The function \( E(Z, \phi = 0) \) is symmetric with respect to polarization only at resonance \( \delta_{\text{eff}} = 0 \). Moreover, it shows a single minimum at \( Z = 0 \) if \( |\kappa| n < \Omega_R \) whereas two degenerate minima at \( Z = \pm \sqrt{1 - (\Omega_R/|\kappa| n)^2} \) in the opposite regime \( |\kappa| n > \Omega_R \).

At the critical point \( |\kappa| n = \Omega_R \), a ferromagnetic QPT takes place, as witnessed by the non-zero value of the polarization, which plays the role of the order parameter for such a transition.

The criticality of the point \( (\delta_{\text{eff}}, |\kappa| n) = (0, \Omega_R) \) is also confirmed by the divergence of measurable physical quantities, such as the magnetic susceptibility \( \chi \), given by Eq. (5). At finite detuning \( \delta_{\text{eff}} \), the energy profile Eq. (A11) may show two non-degenerate minima: the absolute one describes the ground state of the system, while the local one is associated to a metastable excited state. A hysteresis cycle can therefore be observed by slowly varying the effective detuning from positive to negative values and vice versa. The width of the hysteresis region is:

\[
\delta_{\text{hys}} = 2\Omega_R \left[ \left( \frac{|\kappa| n}{\Omega_R} \right)^{2/3} - 1 \right]^{3/2}. \tag{A12}
\]

As already mentioned in the main text, the experimental system is confined in a harmonic trap, and consequently has a non-uniform density profile. Since the gas is tightly confined along two out of three spatial directions, it is legitimate to integrate out these two degrees of freedom in order to focus on the effective one-dimensional dynamics along the longitudinal axis, hereafter indicated as \( x \)-axis. Assuming \( Z \) is only function of \( x \) and integrating Eq. (A5) in the \( yz \) plane, one obtains a one-dimensional equation formally identical to Eq. (A5) with an effective density profile (see, for instance, Ref. [40]):

\[
n(x) = \frac{2}{3} n_0^{3D} \left( 1 - \frac{x^2}{R_x^2} \right)^2 \tag{A13}
\]

\( n_0^{3D} \) and \( R_x \) being the 3D density in the center of the trap and the longitudinal Thomas-Fermi radius, respectively.

Due to the non-uniformity of the system, the ferromagnetic condition \( |\kappa| n(x) > \Omega_R \) is only verified at specific real-space positions: there is always a PM region close to the edges of the trap, while the density is smaller. The same holds for the resonant condition \( \delta_{\text{eff}}(x) = \delta_B + n(x)\Delta = 0 \), due to the 2D symmetry breaking term \( \Delta \neq 0 \). More specifically, the locus of resonant points is a parabola in the plane \( (\delta_B, x) \), shown as a black dashed line in Fig. 2.

When a detuning ramp is applied to the superfluid mixture, the system does not adiabatically follow the global ground state in the ferromagnetic region, but rather stays in the local metastable minimum until the edge of the hysteresis cycle. In other words, the jump in polarization takes place when \( \delta_{\text{eff}}(x) = \pm \delta_{\text{hys}}(x)/2 \), the sign depending on the direction of the ramp, as shown in Fig. 2.

**Appendix B: Numerical simulations**

Numerical simulations are performed by exactly solving the one-dimensional GPEs Eq. (A1) and Eq. (A2) in the external harmonic trapping potential. The ground state is found through imaginary-time evolution via the Euler algorithm, while the following real time dynamics is obtained via a split-step algorithm. The parameters are chosen to reproduce those of the experiment, taking into account the geometrical renormalization; in particular, \( (2/3)|\kappa| n_0^{3D}/\hbar \sim (2/3)|\Delta| n_0^{3D}/\hbar \sim 1.2 \text{ kHz} \) and \( L \sim 200 \mu\text{m} \). A small energy-loss term is necessary to stabilize the system against turbulence at long times: this is realized by adding a small imaginary part to the time-step, \( \text{d}t \rightarrow (1 + i\gamma)\text{d}t \). We verified that the value of the loss rate \( \gamma \) does not affect the relevant physical
properties of the system; for the simulations shown in the main text, $\gamma = 0.1$.

For any given space position $x$, the polarization $Z(\delta_{\text{eff}}(x)) = Z(\delta_B + n(x)\Delta)$, obtained from the real-time dynamics calculation, is fitted with an arctan function. The hysteresis width is given by the shift of the sigmoid derivative at $\delta_{\text{eff}} = 0$. See the grey panels of Fig. 1 at $|n/B_1| = 0, 3$ for illustrative examples of sigmoid functions.

**Appendix C: Image analysis**

The separated image of the two states are taken following similar protocol to the one explained in Ref. [20]. After a time of flight of 1 ms, state $|\uparrow\rangle$ is imaged by standard resonant absorption imaging. Residual atoms in $|\uparrow\rangle$ are blown away by a short push laser pulse. After an additional 1ms, $|\downarrow\rangle$ atoms are transferred in $|\uparrow\rangle$ by using repumping light and then imaged as before. To calibrate the spin-selective imaging, we assure that the total atom number remain constant during an adiabatic transfer between the states. We also radially rescale the image of state $|\uparrow\rangle$ to match the extension of the image of state $|\downarrow\rangle$, by taking into account the extra expansion time. Differential axial expansion can be neglected thanks to the large trap frequency difference ($\omega_{\perp} \ll \omega_{\parallel}$) and the small TOF.

The imaging process includes a first spatial integration of the density in each of the two spin states along the transverse $z$-axis. From a first two-dimensional bimodal fit, we determine the size of the condensate. Then the contribution from thermal atoms is removed from the total population profile and the condensate fraction is integrated along $y$. The two population $n_\uparrow$ and $n_\downarrow$ are used to calculate the relative magnetization

$$Z(x) = \frac{n_\uparrow(x) - n_\downarrow(x)}{n_\uparrow(x) + n_\downarrow(x)} \quad (C1)$$

**Appendix D: Experimental calibration of $n$**

The determination of $|\kappa|n$ and $n\Delta$ is critical to determine the parameter $|\kappa|n/\Omega_R$ and, more important, to locate the resonance $\delta_{\text{eff}} = 0$. Due to a fortunate coincidence in collisional parameters of our mixture, $\Delta$ and $\kappa$ differ only at the $10^{-3}$ level. For the two involved hyperfine states, coupled channel calculations provide $a_{11} = 54.5a_0$, $a_{22} = 64.3a_0$ and $a_{12} = 64.3a_0$ [17]. $n\Delta$ can be experimentally determined either through spectroscopic protocols [20] or by locating the resonance position $\delta_{\text{eff}} = 0$ at the center of the cloud in the PM regime, with large $\Omega_R$, so that hysteresis is absent. We verify the consistency between the two methods and the direct determination of $|\kappa|n$ from the experimentally measured atom number and trap frequencies together with geometrical consideration [40].

**Appendix E: Experimental susceptibility**

In our measurement of $\chi$, we used thousands of experimental scans performed for different values of $\Omega_R$ with either forward or backward ramps, as the ones presented in 2(c)-(d). To evaluate $\chi$, we make use of the fact that the derivative of the magnetization with respect to $\delta_{\text{eff}}$ is equivalent to the derivative with respect to $\delta_B$.

$$\chi = \frac{1}{n} \left. \frac{\partial s_z}{\partial \delta_{\text{eff}}} \right|_{\delta_{\text{eff}}=0} = \frac{1}{n} \left. \frac{\partial s_z}{\partial \delta_B} \right|_{\delta_{\text{eff}}=0} = \left. \frac{\partial \delta_B}{\partial \delta_{\text{eff}}} \right|_{\delta_{\text{eff}}=0} \quad (E1)$$

As a first step, for noise reduction, we spatially average the magnetization $Z$ as well as the total density within a series of 10-pixel-wide windows. For each window, we obtain the value of the magnetization as a function of $\delta_B$ and we perform an arctan plus linear model fit. The estimate for $\chi$ is then extracted as the value of the derivative of the arctan-fit at $\delta_{\text{eff}} = 0$. Associated $|\kappa|n$ is obtained from an averaged density profile of the experimental shot with $\delta_{\text{eff}}$ closest to zero. This procedure results in significant uncertainties for points in the tails of the cloud where the density gradient is large. For this reason we chose to exclude the outer points from the final binning.

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