Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In their manuscript, Yamamoto et al. study frustrated quantum magnetism in a triangular lattice. The first part consists of a Gutzwiller analysis (static and time-dependent) of the Bose Hubbard model on a triangular lattice. This includes a proposed preparation scheme for the Gutzwiller state at negative-zero temperature. The second, significantly shorter, part of the work is an advanced DMRG-based analysis of the chiral-superfluid to Mott transition in 2D, which includes state-of-the-art numerics. Both parts deserve to be published, and in my view the paper mostly fulfills the acceptance criteria detailed at https://www.nature.com/commsphys/referees/guide-to-referees#criteria. Before I can fully recommend publication, the authors should comment on the following point:

The proposed preparation scheme is motivated by the Gutzwiller analysis performed by the authors. They also benchmark their protocol using a time-dependent Gutzwiller approach. I am unsure how reliable this method is, if strong correlations and entanglement are involved. Indeed, the authors propose a protocol that transitions from a weakly to a strongly correlated regime, and the DMRG calculations show some significant deviations from the simpler Gutzwiller predictions. Are there at least some regimes, where we can definitely trust the time-dependent Gutzwiller analysis?

A thorough methods section is provided.

Additional comments to the authors:

- A brief but precise definition of the term "isosceles-type anisotropy" would be useful if the paper is written for a broad physics audience.

- At the end of the introduction, the authors argue that "the effort in realizing frustrated quantum systems with cold atoms has not yet paid off even though many theoretical proposals have been made." Personally, I find this statement too pessimistic. Surely, many obstacles remain that have to be overcome. But the new perspective afforded by quantum gas experiments has already lead to significant insights and sparked new numerical studies. Astonishing experimental achievements have been made in a comparatively short time.

- I was wondering whether a Gutzwiller analysis of this problem, at least in equilibrium, has been done before (it seems like a natural step). Can the authors provide references in this context?

- At the beginning of the discussion of the Gutzwiller analysis, it is somewhat unclear where U = 0 is considered, and where U \neq 0 is studied. Maybe the authors can make this more explicit.

- In Fig. 3 (c), it remains unclear which time t is considered in the plot.

- In the caption of Fig. 3, it would be useful to clarify that all results have been obtained by Gutzwiller analysis.

- Below Fig. 3, the authors write: "the values of U and V have to be changed as … in order to adjust the energy scale in consideration of the reduction of the kinetic energy due to the frustration". More information should be provided to allow readers to easily follow the argument.

- In (the caption of) Fig. 4, it would be useful if the authors could indicated the method used for the calculations.

Reviewer #2 (Remarks to the Author):

(i) Some numerical details of time dependent Gutzwiller approximation calculations are not clear. Determination of large number of on-site superfluid parameters in a self-consisting way is a difficult numerical problem, especially in case of strong frustration, site-dependent trapping potential and non-equilibrium dynamics. In such case the convergence of self-consisting mean-field calculations, at given time step, may be slow or not converge at all, depend on variations of initial conditions or result in multiple solutions. Unfortunately no information about stability and convergence of self-consisting solution is presented by authors.

(ii) Since relevant energy scale for superfluidity in frustrated system is much lower than for non-frustrated initial system, there is a problem of temperature scale which has to be reached in non-frustrated system prior to quench to negative temperature state. For example important question would be impact of such temperature on dynamical stability of evolving non-equilibrium CSF state.

See attached file for additional comments.

Reviewer #3 (Remarks to the Author):

In the manuscript entitled "Frustrated quantum magnetism with Bose gases in triangular optical lattices at negative absolute temperatures" by Dr. D. Yamamoto et al., the authors proposed a new experimental method to realize geometrically frustrated quantum states with ultracold Bose atoms in a triangular optical lattice, and carefully investigated the validity with numerical simulations.

Geometrical frustration is one of the important topics on many-body quantum physics. The ground state is highly entangled and sometimes has large residual entropy even at the zero temperature. As a result, exotic states such as spin liquid are highly theoretically discussed despite difficulties on numerical calculation of geometrically frustrated quantum systems.
Experimental studies are also difficult because geometrical frustration require a highly clean and ideal quantum system. Cold atoms system is one of the promising targets, however, a geometrically frustrated quantum state using cold atoms has not experimentally realized because it requires lower temperature to reach a regime of the geometrically frustrated quantum physics.

The main novelty of this manuscript is the application of idea of "negative temperature" to Bose atoms in a triangular optical lattice. To my best knowledge, the proposed method mentioned in the manuscript is new and important. Previous research by the Prof. Sengstock group reported geometrically frustrated "classical" system because of inevitable heating to realize the geometrically frustrated triangular system. Heating during the proposed method is expected to be smaller than the previous research and experimental study on the geometrically frustrated "quantum" regime is promised. Theoretical and numerical analysis at the zero temperature which support the proposal are also described.

In summary, I believe that the present manuscript reaches the criteria of Communications Physics and recommend for publication at Communications Physics.

Minor questions and comments:
A)
The authors exclude influence of atom interaction during the phase-imprinting. Because of inhomogeneous atom density profile, interaction energy depends on the position. This energy shift might disturb perfect phase-imprinting. The inhomogeneous interaction energy is negligible?

B)
One of the important features of the frustrated system discussed in the manuscript is that it has Z2 symmetry (chirality) and spontaneous symmetry breaking (q=Q or -Q) or the other entangled states are expected, In contrast, symmetry of the created state after the proposed three operations shown in Fig.3(b) is naturally broken because the phase-imprinting break the symmetry. In this meaning I wonder the created state is the “real” ground state. Nevertheless, is it possible to capture the signature of the CSF-MI quantum phase transition?

C)
In Fig.4(a), labels of the tics along the Y axis are missing.

In the paper authors propose a new method of realising quantum simulator of geometrically frustrated strongly-correlated boson system. Quantum simulators made of ultra-cold gases in optical traps are one of the most active frontiers of quantum many-body physics. However simulating some of the most interesting Hamiltonians, including geometrically frustrated models of quantum magnetism, at the temperatures relevant for ground state phases remains a challenge. The authors study Bose-Hubbard model and propose to use sudden change of sign of boson-boson interaction U and trapping potential in order to create quantum state at negative temperature, which is equivalent to a sought of quantum state of inverted sign Hamiltonian at positive temperature. This way one can avoid a problem of direct engineering negative hopping amplitudes needed for geometrical frustration. To my knowledge this is a novel approach which possible realisations are of broad interest.

Authors consider conditions to obtain negative temperature state with maximum energy which is equivalent to simulating low positive temperature state. To this end they propose to use phase imprinting to change BEC momentum, and rescaling of absolute values of U and trapping potential. Validity of the proposed protocol is checked by numerical calculations based on Gutzwiller approximation (mean-field). First, rough ground state diagram is determined by restricted Gutzwiller calculations for translationally invariant case. Authors restrict considered superfluid phases to the analogues of known phases of XY Hamiltonian. This allows to find suitable BEC momentum, which is to be set by phase imprinting . Main part of calculations uses time dependent unrestricted (site dependent) Gutzwiller approach for case in a presence of trapping potential. Although this is uncontrolled approximation , it is state of the art approach for problem considered as more precise numerical methods do not work well in a presence of both strong frustration and time evolution. However some numerical details are not clear. Determination of large number of on-site superfluid parameters in a self-consisting way is a difficult numerical problem, especially in case of strong frustration, site-dependent trapping potential and non-equilibrium dynamics. In such case the convergence of self-consisting mean-field calculations, at given time step, may be slow or not converge at all, depend on variations of initial conditions or result in multiple solutions. Unfortunately no information about stability and convergence of self-consisting solution is presented by authors.

Finally authors use advanced numerical calculations based on cluster mean-filed with DMRG solver to precisely determine superfluid-Mott insulator critical point in equilibrium translationally invariant case. Although this method is far superior to Gutzwiller calculations I find its use and results rather subsidiary in this particular paper. While details of the CSF-MI transition are new and interesting, this transition is not a main problem for experimental realisation of proposed quantum simulation. On the other hand, since relevant energy scale for superfluidity in frustrated system is much lower than for non-frustrated initial system, there is a problem of temperature scale which has to be reached in non-frustrated system prior to quench to negative temperature state. For example important question would be impact of such temperature on dynamical stability of evolving non-equilibrium CSF state.

In conclusion this paper presents novel and broadly interesting approach for simulation of geometrically frustrated quantum magnetic systems. The proposed experimental protocol is substantiated by numerical calculations . However some numerical issues concerning convergence of self-consisting Gutzwiller solutions and impact of finite temperatures could be better addressed.

I. SUMMARY OF CHANGES

1. All changes in the manuscript text file are highlighted in red.
2. In Figs. 3c and 4a, "$0 \leq t \leq 200\hbar U^{-1}$" has been changed to "$t = 200\hbar U^{-1}$."
3. In Fig.4a, the labels of the tics along the Y axis have been properly attached.
4. Figure 7 has been newly added, and Figs. 7 and 8 of the previous manuscript have been renumbered to be Figs. 8 and 9, respectively.
5. Refs. 39, 40, 46-50, 56, 61 have been newly added.

II. RESPONSE TO REFEREE #1

We thank Referee #1 for his/her detailed attention to our new findings and for recognizing that the paper mostly fulfills the acceptance criteria of Communications Physics. Please find our answers to his/her comments below.

Referee: "The proposed preparation scheme is motivated by the Gutzwiller analysis performed by the authors. They also benchmark their protocol using a time-dependent Gutzwiller approach. I am unsure how reliable this method is, if strong correlations and entanglement are involved. Indeed, the authors propose a protocol that transitions from a weakly to a strongly correlated regime, and the DMRG calculations show some significant deviations from the simpler Gutzwiller predictions. Are there at least some regimes, where we can definitely trust the time-dependent Gutzwiller analysis?"

Response: The Gutzwiller approximation gives the exact wave function in the weak and strong coupling limits, and thus is believed that it is reliable in the deep-SF and deep-MI regimes. Therefore, our analysis that showed the dynamical stability of the negative-temperature frustrated CSF state should be trustworthy since no strong correlations exist in the deep-SF regime. The stability of the negative-temperature state is indeed one of the most core issues for our protocol proposed in the paper.

We also gave a demonstration of the CSF-to-MI transition when the ratio $|U/J|$ is increased, which should, on the other hand, be taken as a qualitative prediction because, as the referee pointed out, strong correlations and entanglement are involved in the vicinity of the critical point. (We would like to remind that no quantitative numerical method on classical computers is currently available for studying such a strongly-correlated dynamics in 2D frustrated systems.) Therefore, whether the physics beyond the TDGA takes place in the quantum dynamics is indeed an important subject in future experiments. We believe that our TDGA analysis on the CSF-MI transition provides a qualitative prediction on the transition dynamics, and will drive future quantum simulation studies attacking the fundamentally interesting challenge in frustrated quantum many-body systems. The quantitative estimation of the CSF-MI critical point determined by the CMF+DMRG calculations (in equilibrium) must be also helpful for those future experimental studies.

In the revised manuscript, we added comments on the above points in the last paragraph of "The CSF-to-MI phase transition" and in the Discussion. Please see the parts highlighted in red (lines 321-340, 413-418).

Referee: "A brief but precise definition of the term "isosceles-type anisotropy" would be useful if the paper is written for a broad physics audience. "

Response: We added more precise definition of the hopping anisotropy in the Introduction and in the first paragraph of "The Bose-Hubbard model on triangular lattice" (lines 87-90, 110-118).

Referee: "At the end of the introduction, the authors argue that "the effort in realizing frustrated quantum systems with cold atoms has not yet paid off even though many theoretical proposals have been made." Personally, I find this statement too pessimistic. Surely, many obstacles remain that have to be overcome. But the new perspective afforded by quantum gas experiments has already lead to significant insights and sparked new numerical studies. Astonishing experimental achievements have been made in a comparatively short time. "

Response: We agree that the statement was too pessimistic. We have revised the sentence by removing the pessimistic expression and just writing the facts.

Referee: "I was wondering whether a Gutzwiller analysis of this problem, at least in equilibrium, has been done before (it seems like a natural step). Can the authors provide references in this context? "

Response: At first, we had thought the same as the referee. However, we could not find any similar Gutzwiller analysis on frustrated Bose-Hubbard models. To the best of our knowledge, it has not been published in the literature.

Referee: "At the beginning of the discussion of the Gutzwiller analysis, it is somewhat unclear where $U = 0$ is considered, and where $U \neq 0$ is studied. Maybe the authors can make this more explicit. "

Response: We have made this point clearer in the revised manuscript (lines 124-128).

Referee: "Below Fig. 3, the authors write: the values of U and V have to be changed as ... in order to adjust the energy scale in consideration of the reduction of the kinetic energy due to the frustration. More information should be provided to allow readers to easily follow the argument. "

Response: To address this point, we have revised the texts in the section "Negative absolute temperature." Please see the sentences highlighted in red (lines 234-258). We have also added some comments in the last paragraph of "Gutzwiller analysis" and in the Methods:"The GA analysis for finite-momentum BEC states" about the reduction of the kinetic energy scale due to frustration (lines 177-190, 556-563). We believe that the explanations have become much clearer now, thanks to the referee's comment.

Referee: "In Fig. 3 (c), it remains unclear which time t is considered in the plot. " "In the caption of Fig. 3, it would be useful to clarify that all results have been obtained by Gutzwiller analysis. " " In (the caption of) Fig. 4, it would be useful if the authors could indicated the method used for the calculations. "

Response: We have changed "$0 \leq t \leq 200\hbar U^{-1}$" to "$t = 200\hbar U^{-1}$" in Fig. 3 (c) and added "within the TDGA" in the caption of Figs. 3 and 4.

III. RESPONSE TO REFEREE #2

We thank Referee #2 for his/her work in carefully reading our manuscript and for recognizing that our protocol for realizing quantum simulators of frustrated quantum systems is novel and of broad interest. We hope that the answers to the referee's questions below resolve his/her remaining concerns.

Referee: "(i) Some numerical details of time dependent Gutzwiller approximation calculations are not clear. Determination of large number of on-site superfluid parameters in a self-consisting way is a difficult numerical problem, especially in case of strong frustration, site-dependent trapping potential and non-equilibrium dynamics. In such case the convergence of self-consisting mean-field calculations, at given time step, may be slow or not converge at all, depend on variations of initial conditions or result in multiple solutions. Unfortunately no information about stability and convergence of self-consisting solution is presented by authors. "

Response: We thank the referee for pointing out the lack of the numerical details of time-dependent Gutzwiller approximation (TDGA) calculations. In the Methods section of the revised manuscript, we have presented more detailed procedure of the TDGA.

In the TDGA method, one does not perform self-consistent convergence of mean fields at each time step. Starting from an initial state, the time evolution is carried out step-by-step with short time interval Δt using the values of mean fields $\psi_i(t)$ at the time step before, in a deterministic fashion. Therefore, it does not encounter the problem of multiple solutions. Please see the section "The TDGA simulation in a trap potential" highlighted in red in Methods for the calculation procedure (lines 581-605).

Note that the initial state at $t = 0$ (shown in Fig. 3a of the paper) is calculated by the static GA method, which has to be done in a self-consistent way to determine a large number of on-site superfluid parameters in a trap potential. This is, however, not a very hard task. To achieve the convergence, taking the uniform solution in the absence of the trap potential as the initial condition, we solve the set of self-consistent equations for ψ_i on the entire lattice sites with the standard Newton-Raphson method. In the revised manuscript, we have briefly explained the GA calculations in the section "The GA analysis for finite-momentum BEC states" of Methods (lines 564-579).

Referee: "(ii) Since relevant energy scale for superfluidity in frustrated system is much lower than for non-frustrated initial system, there is a problem of temperature scale which has to be reached in non-frustrated system prior to quench to negative temperature state. For example important question would be impact of such temperature on dynamical stability of evolving non-equilibrium CSF state. "

Response: Since the quench to the negative-temperature state is performed in the deep-SF regime in our protocol, the difference in the temperature scale between unfrustrated and frustrated systems is expected not to be a big problem. In such a regime with rigid order parameters with no strong fluctuations, the energy scale of frustrated systems are actually lower than that of unfrustrated systems but only by the factor of ~ 2 or 3: As a typical, related example, while the transition temperature of the square-lattice XY model is given by $T_c/zJ \approx 0.22$ (scaled by the coordination number z and the exchange interaction J), the frustrated triangular-lattice XY model yields $T_c/zJ \approx 0.09$ [H.-J. Xu and B. W. Southern, J. Phys. A: Math. Gen. **29**, L133 (1996); M. Hasenbusch, J. Phys. A: Math. Gen. **38** 5869 (2005)]. Therefore, we believe that our proposal for creating the dynamically-stable CSF state is realistic given the currently-achievable low temperatures of Bose-gas systems.

IV. RESPONSE TO REFEREE #3

We thank Referee #3 for recommending our paper for publication at Communications Physics. Please find our answer to his/her comments below.

Referee: "Minor questions and comments: A) The authors exclude influence of atom interaction during the phase-imprinting. Because of inhomogeneous atom density profile, interaction energy depends on the position. This energy shift might disturb perfect phase-imprinting. The inhomogeneous interaction energy is negligible? "

Response: In the previous experiments using the phase-imprinting techniques (e.g. [S. Taie *et al.*, Sci. Adv. **1**, e1500854 (2015)]), the imprinting has been carried out in a much shorter time scale than the relaxation time by hoppings and with a much larger potential gradient than the interaction and potential energy scales; namely, $\delta E \gg J_{ij}^{-1}, U, V$. In such a situation, the inhomogeneity in the local interaction energy does not become a problem, because only the potential difference due to the potential gradient is dominant during the phase-imprinting process. Since such experiments have already been succeeded, we just assumed in the simulations of the paper that the perfect phase imprinting was achieved. In the revised manuscript, we have added some remarks on this point in the second paragraph of "Negative absolute temperature" (lines 220-233).

For the convenience of the readers, we also added a new section "The phase imprinting operation" in Methods (lines 608-629). As can be seen in Fig. 7, when δE exceeds $\sim 10U_0$, the imprinting becomes almost perfect with no changes other than the local phase distribution.

Referee: "B) One of the important features of the frustrated system discussed in the manuscript is that it has Z2 symmetry (chirality) and spontaneous symmetry breaking (q=Q or -Q) or the other entangled states are expected, In contrast, symmetry of the created state after the proposed three operations shown in Fig.3(b) is naturally broken because the phase-imprinting break the symmetry. In this meaning I wonder the created state is the real ground state. Nevertheless, is it possible to capture the signature of the CSF-MI quantum phase transition? "

Response: In the protocol we proposed in the paper, the negative-temperature state is created in the deep-SF regime, in which the ground state is known to be the CSF state with chiral ($\mathbb{Z}_2$) symmetry breaking from the GA calculations. Although, strictly speaking, the GA is exact only in the limit of $U \approx 0$, the appearance of the other (more exotic) entangled states is unlikely in the deep-SF regime, because even in the spin-1/2 triangular-lattice Heisenberg model, which subjects to stronger quantum correlation effects due to the higher SU(2) symmetry, the ground state is the 120° order with chiral symmetry breaking. For another example, the quantum simulations of the classical triangular XY model by Sengstock's group [Science **333**, 996 (2011)] indeed observed that the chiral symmetry breaking in the deep-SF regime.

In our protocol, which chirality ($\boldsymbol{Q}$ or $-\boldsymbol{Q}$) is chosen is determined explicitly by the phase imprinting, as the referee points out. However, whether the chirality is determined explicitly or *spontaneously* (by nature) does not affect the properties of the created symmetry-broken order and its phase transition phenomena. When approaching the CSF-MI transition, the collective modes that tend to restore the $\mathbb{Z}_2$ (and $U(1)$) symmetry become developed, and give rise to particular quantum phase transition phenomena associated with the restoration of the $U(1) \times \mathbb{Z}_2$ symmetry. This does not depend on whether the chirality is determined explicitly or *spontaneously*.

Referee: "C) In Fig.4(a), labels of the tics along the Y axis are missing."

Response: We have added the labels.