Dear Editors,

Thanks for your mail attached with reports on our manuscript (XW10449B).

We appreciate the time and effort that you dedicated to our manuscript. We have addressed all the points raised by the referees, taken all the comments and suggestions into consideration, and modified the manuscript accordingly. The changes in response to the referee reports are marked in red in the manuscript and a summary for the changes is appended after our point-by-point response to the referees. We sincerely hope that both referees will find satisfactory in this revised version.

Best wishes,

Shi Wang, Zhongyuan Qi, Bin Xi, Wei Wang, Shun-Li Yu, and Jian-Xin Li

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**Response to the Referees**

We thank the referees for appreciating our work, and are grateful to the referees for giving the insightful comments and valuable suggestions based on which we can improve our work. We have addressed all the points raised by the referees, taken all the comments and suggestions into consideration, and modified the manuscript accordingly.

**Response to the referee A**

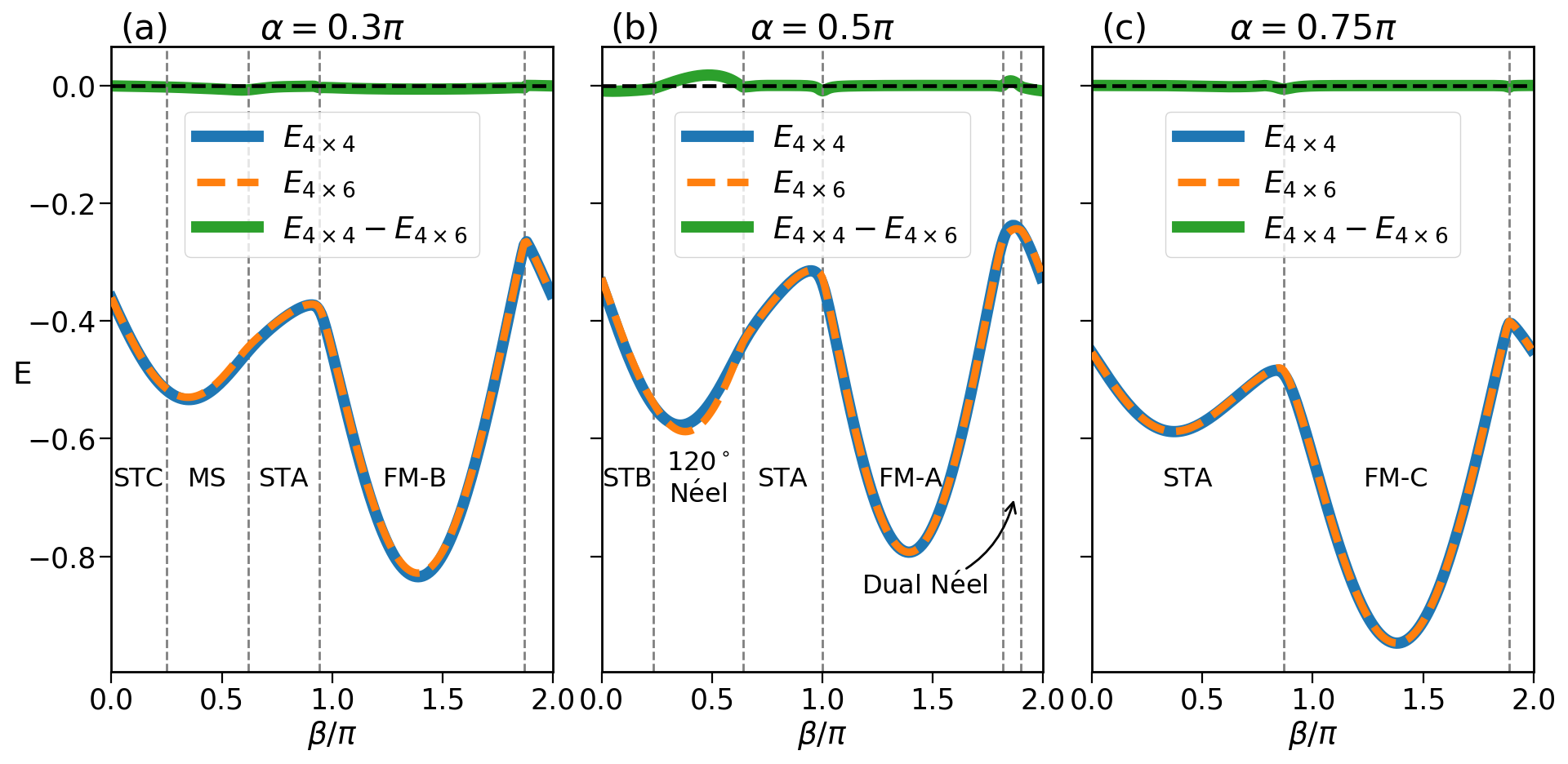
(1) **Comments:** Since the heat-bath method is explained, a few words about the over-relaxation MC method is also needed, just for the sake of self-containment and self-consistency.

**Response:** We thank the referee for suggesting us to include an introduction to the over-relaxation MC method. We have added an explanation of the over-relaxation sampling in the section of the introduction of the classical Monte Carlo method.

(2) **Comments:** What is the effect of an asymmetric system for the ED calculations? Phases which break a real-space symmetry (such as stripes) may have different GS energy in asymmetric cluster when compared to asymmetric one. Does it bias towards a stripe phase? Comments should be given in the main text.

**Response:** To clarify the effect of asymmetry for the ED calculations, we also perform ED calculations on a 4×4 cluster with periodic boundary condition, which is more symmetric than the 4×6 cluster used in the paper. The per-site ground state energy for both 4×4 (blue solid lines) and 4×6 (orange dashed lines) clusters as well as the difference (green solid lines) between them are shown in Fig. 1. When the model parameters are located at the FM or stripe phases, the relative difference is very small which may originate from the finite-size effect, i.e., the 4×4 and 4×6 clusters have different system sizes. Both 4×4 and 4×6 clusters with periodic boundary condition are compatible with FM and stripe magnetic orders, so the asymmetric 4×6 cluster does not bias towards a stripe phase in our ED calculations, although one of the equivalent stripe states will be selected in the calculations due to the asymmetry of the cluster. In addition, the 4×6 cluster is compatible with the 120° Néel and dual Néel order at least in one direction, however, the 4×4 cluster is incompatible with the 120° Néel and dual Néel order in any directions. Thus, the energy differences between the 4×4 and 4×6 clusters in the 120° Néel and dual Néel pahses are relatively larger than those in the FM and stripe phases. In Fig. 2, we show the GS energies and their second derivatives versus β with α fixed to 0.3π, 0.5π and 0.75π. When α = 0.3π and α = 0.75π, the results for both 4×6 and 4×4 clusters are qualitatively consistent, i.e., the singularities corresponding to the quantum phase transitions are in one-to-one correspondence, see Fig. 2(a), 2(d) and Fig. 2(c), 2(f). However, when α = 0.5π, the results for 4×6 and 4×4 clusters are different. For the 4×6 cluster, there are singularities at β = 0.234π and β = 1.9π [indicated by black arrows in Fig. 2(b)] which reveal the phase transitions from Stripe-B to 120° Néel and from dual Néel to Stripe-B. On the other hand, for the 4×4 cluster, we can’t see these singularities. This is owning to the incompatibility between the 4×4 cluster and the magnetic orders. As for the other three peaks in both Fig. 2(b) and 2(e), they correspond one to one. Therefore, under the premise that the current computing power allows, the 4×6 cluster is better than the 4×4 cluster, although the 4×4 cluster is more symmetric.

FIG. 1. Per-site ground state energies for 4×4 and 4×6 clusters. The abbreviations “STA”, “STB”, “STC” and “MS” correspond to stripe-A, stripe-B, stripe-C and modulated stripe, respectively. The vertical gray dashed lines mark the phase transitions. (a) - (c) The model parameter α is fixed to 0.3π, 0.5π and 0.75π, respectively.



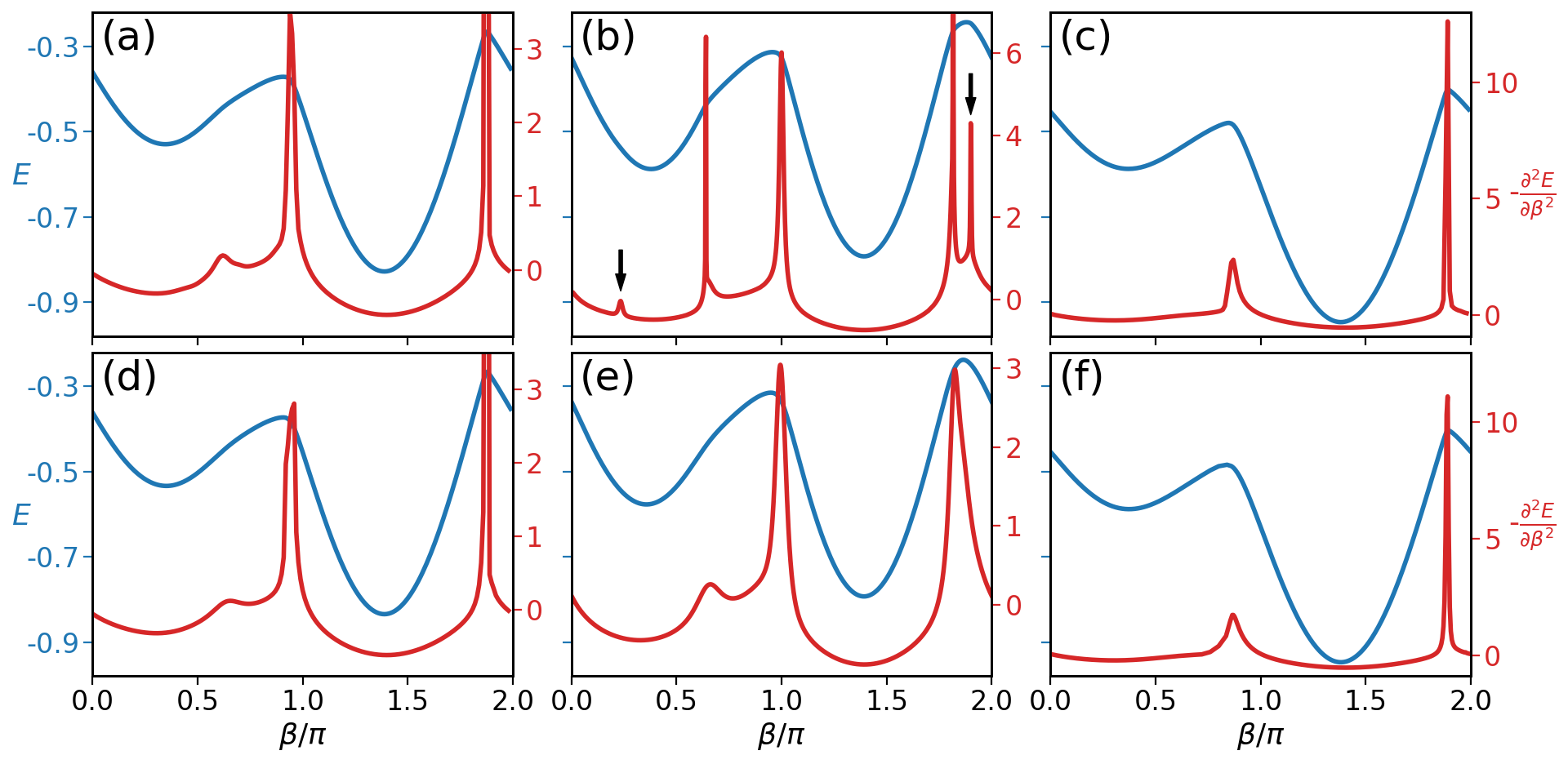


FIG. 2. GS energies and their second derivatives versus β with α fixed to 0.3π, 0.5π and 0.75π. The blue lines are the GS energies and the red lines are the second derivatives. (a) - (c) ED calculations are performed on a 4×6 cluster and α equals to 0.3π, 0.5π and 0.75π, respectively. (d) - (f) ED calculations are performed on a 4×4 cluster and α equals to 0.3π, 0.5π and 0.75π, respectively.

(3) **Comments:** In Sec. 3A and in the caption of Fig. 2, use a more precise description. What is a quantum phase diagram? Is it the zero-T phase diagram or the finite-T phase diagram for the case of quantum spins? What are the triangles in Fig. 4(i)?

**Response:** We are very grateful to the referee for pointing out these imprecise descriptions. The phase diagram we obtained is the ground state phase diagram, i.e., the zero-temperature phase diagram. In the main text, we change the description from “quantum phase diagram” to “zero-temperature quantum phase diagram”.

The red triangles in Fig. 4(i) mark the characteristic wave vectors for the 120° Néel order. In this version, we redraw Fig. 4(i) to explicitly mark the characteristic wave vectors for all of the magnetically ordered phases and add descriptions in the caption. See the main text for details.

(4) **Comments:** In fig. 8, the ideal is that both vertical scales of the energy and of the second derivative are given. In the current version, it seems that the range of the former is small.

**Response:** Following the referee’s nice suggestion, we redraw Fig. 8 with both vertical scales of the energy and of the second derivations are given.

(5) **Comments:** Although the whole analysis is somewhat convincing, there is no consistent study on the second derivatives presented in Fig. 8. Could the authors state their findings of the peaks when compared to smaller systems? Is the finite-size effect consistent with a non-analyticity?

**Response:** Following the referee’s suggestion, we carry out the ED calculations on a 2×6 cluster with periodic boundary condition to study the finite-size effect on the phase diagram. We find that all the peaks of the second derivatives appearing for the 4×6 cluster also exist for the 2×6 cluster. The results for different system sizes are qualitatively consistent, although the phase boundaries slightly shifted depending on the system size. To address this problem more clearly, we have added a new section in the appendix (Appendix B: The effects of system size on the phase boundaries) to present the results for the 2×6 cluster.

(6) **Comments:** The arrows are hard to read in Figs. 10 and 12. Maybe, for clarity, the point sites (which are not necessary) may be dropped out.

**Response:** Following the referee’s nice suggestion, we delete the point sites in Fig. 11, Fig. 12 and Fig. 12 for clarity (In previous version, Fig. 10, Fig. 11 and Fig.12).

(7) **Comments:** I find it important to precisely define the phases found here. What is the order parameter (nematic, stripe, etc)? The ordering vector?

**Response:** The order parameter of FM, stripe, 120° Néel and dual Néel states can be defined as ***m***(***Q***)=∑<Ω|***S***r|Ω>ei***Q***·***r***, where |Ω> is the ground state and ***Q*** is the characteristic wave vector of these magnetically ordered states. Both the ordering vector and the moment direction are the defining characteristic. However, since the cluster ground state in the ED calculations does not spontaneously break the cluster symmetry and corresponds to a superposition of all possible degenerate ground states, the calculated ground-state average cannot give non-zero order parameter. In our paper, we instead calculated the static structure factors to identify the wave vectors of these ordered phases. For these phases with the same type of classical orders, they have the same characteristic wave vectors. The moment directions are extracted by using the spin coherent state in the ED calculation together with the classical analyses. As for the nematic states shown in Fig. 13, their characteristic wave vectors are different from those for the states in the phase diagram in Fig. 2. Thus, we can identify the nematic phases by the characteristic wave vectors, whenever they appear in the calculations. In the revised version, we have added the interpretations about the order parameters in the main text.

(8) **Comments:** I could not see the difference between the SSF of Figs. 4(a), (b), and (d), which are reported to be in different phases. It is only mentioned that these phases are different due to distinct peaks at the M points. I find this vague and confusing. Only much later in the text, different arguments are provided in order to clarify this point. Probably the authors should point out that further arguments will be provided. (Maybe with the clarifications of criticism 7 will help here.)

**Response:** We are very grateful to the referee for pointing out this, and we also realized that the interpretations about the differences between the phases with the same type of order is somewhat too far away from the first discussion of these phases in the text. Following the referee’s suggestion, in order to make our article easier to read, we have added a short explanation of the differences between the phases with the same type of order in the discussion of the phase diagram, but the detailed discussions are still put later in the text for the sake of fluency.

**Response to the referee B**

**Comments:** An optional question is about the QSL phase. The claim that the observed QSL is a Z2 gapped spin liquid is rather weak. DSF is not a critical information for the determination. Can the authors do a parton mean-field theory calculation for this phase?

**Response:** We have to admit that the referee’s criticism is justified, our proposal that the green area in the zero-temperature global phase diagram is a Z2 gapped spin liquid is weak. Parton mean-field theory calculation may provide further evidence for our proposal. However, this is beyond the main focus of this paper. As for the physical properties of that quantum phase, detailed investigations, such parton mean-field theory calculation, DMRG study, etc., may be carried out in a separated paper.

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**Summary for the changes**

**Main text:**

(1) In the “C. Classical Monte Carlo Method” sub-section in the revised manuscript, we include a brief introduction to the over-relaxation Monte Carlo method.

(2) At the beginning of the “A. Global Phase Diagram” sub-section in the revised manuscript, we change the description from “quantum phase diagram” to “zero-temperature quantum phase diagram”.

(3) After the 5th paragraph of sub-section “A. Global Phase Diagram” in the revised manuscript, we add a new paragraph to discuss the order parameter.

**Appendix:**

We add a new appendix section “Appendix B: The effects of system size on the phase boundaries” in the revised manuscript to discuss the effects of system size.

**Figures:**

(1) In the revised manuscript, we redraw Fig. 4(i) to explicitly mark the characteristic wave vectors for the FM, stripe, 120° Néel and dual Néel phases. The caption for Fig. 4(i) is changed from “Characteristic wave vectors for the FM and stripe phases” to “Characteristic wave vectors for the FM (blue circle), stripe (orange square), 120° Néel (red up-triangle) and dual Néel (purple down-triangle) phases”.

(2) Following the referee’s suggestion, we redraw Fig. 8 with both vertical scales of the energy and of the second derivations are given.

(3) Following the referee’s suggestion, we delete the point sites in Fig. 11, Fig. 12 and Fig. 12 for clarity (In previous version, Fig. 10, Fig. 11 and Fig.12).