Letter

Quantum paramagnetism in the decorated square-kagome antiferromagnet $Na_6Cu_7BiO_4(PO_4)_4Cl_3$

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The square-kagome lattice Heisenberg antiferromagnet is a highly frustrated Hamiltonian whose material realizations have been scarce. We theoretically investigate the recently synthesized $Na_6Cu_7BiO_4(PO_4)_4Cl_3$ where a Cu^{2+} spin-1/2 square-kagome lattice (with a six site unit cell) is decorated by a seventh magnetic site alternatingly above and below the layers. The material does not show any sign of long-range magnetic order down to 50 mK despite a Curie-Weiss temperature of −212 K indicating a quantum paramagnetic phase. Our DFT energy mapping elicits a purely antiferromagnetic Hamiltonian that features longer range exchange interactions beyond the pure square-kagome model and, importantly, we find the seventh site to be strongly coupled to the plane. We combine two variational Monte Carlo approaches, pseudofermion/Majorana functional renormalization group and Schwinger-Boson mean field calculations to show that the complex Hamiltonian of $Na_6Cu_7BiO_4(PO_4)_4Cl_3$ still features a nonmagnetic ground state. We explain how the seventh Cu^{2+} site actually aids the stabilization of the disordered state. We predict static and dynamic spin structure factors to guide future neutron scattering experiments.

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Introduction. Magnetic ions forming the kagome lattice, a corner sharing network of triangles, have been the focus of several decades of highly frustrated magnetism research [1]. Kagome lattice antiferromagnets provide some of the most promising examples of highly correlated nonmagnetic ground states [2,3], and are therefore subject of intense experimental efforts while inspiring a wealth of theoretical developments [4,5]. Interestingly, the square-kagome lattice as a differently connected lattice of corner sharing triangles [6] can also support a quantum paramagnetic ground state [7–12]. The precise nature of the ground state is under debate, with proposals spanning a pinwheel valence bond crystal (VBC) [13,14], length six loop VBC [15,16], and (lattice) nematic quantum spin liquid [17]. The field of frustrated quantum magnetism is currently poised with the arrival of new materials based on the square-kagome lattice geometry promising to host exotic nonmagnetic phases at low temperatures [18–21]. In the most prominently studied example $KCu_6AlBiO_4(SO_4)_5Cl$, the Cu^{2+} $S = 1/2$ moments do not show any sign of long-range dipolar magnetic order down to 50 mK despite Curie-Weiss temperatures of −237 K, with indications of gapless quantum spin liquid behavior [22]. Recently, $Na_6Cu_7BiO_4(PO_4)_4[Cl, (OH)]_3$, a novel sodium bismuth oxo-cuprate phosphate chloride containing square-kagome layers of Cu^{2+} ions was synthesized [18]. It contains, besides the six magnetic sites making up the square-kagome lattice, a seventh decorating site which is placed either above or below the square in checkerboard fashion. A study of specific heat indicates that the compound does not order magnetically down to 50 mK [19] despite a large negative Curie-Weiss temperature of −212 K. The scenario in both these compounds is then strikingly similar to the kagome lattice based candidate quantum spin liquid material Herbertsmithite [1].

In this work, we will establish the Hamiltonian of $Na_6Cu_7BiO_4(PO_4)_4Cl_3$ by density functional theory based energy mapping. As it is highly nontrivial to work out the ground state and excited state properties of this complex lattice with three symmetry inequivalent magnetic sites, we apply two types of variational Monte Carlo (VMC), two flavors of functional renormalization group (FRG) calculations

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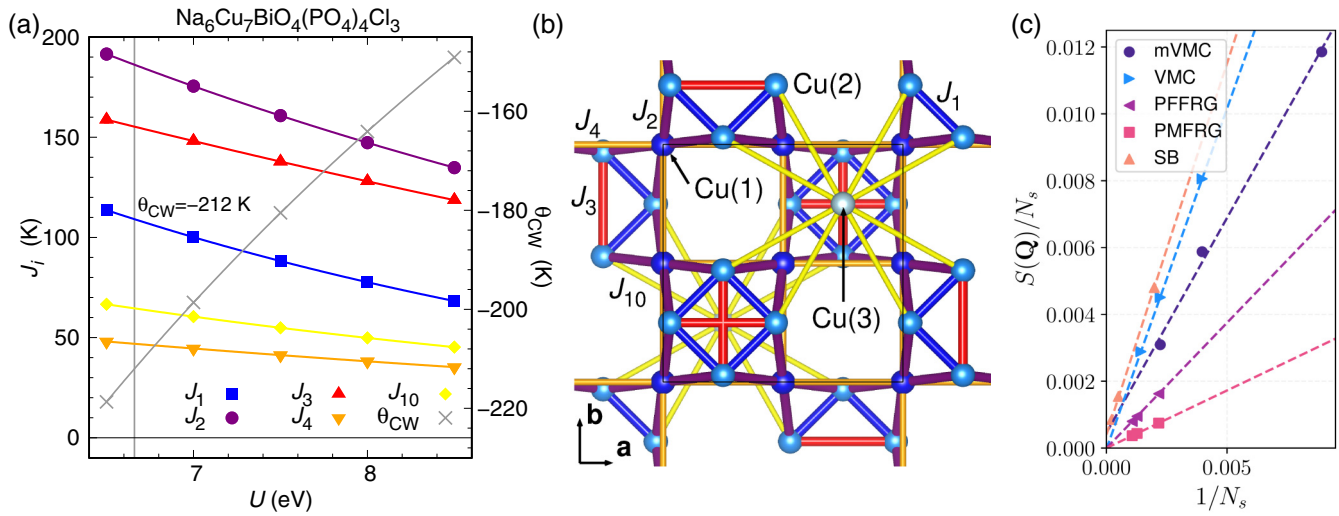

FIG. 1. (a) Heisenberg Hamiltonian parameters of $Na_6Cu_7BiO_4(PO_4)_4Cl_3$ determined by DFT energy mapping as function of onsite interaction strength U (negligible couplings are not shown). The vertical line indicates the U value where the exchange couplings match the experimental [18] Curie-Weiss temperature. The resulting nonnegligible exchange couplings are $J_1 = 109.1(8)\,\mathrm{K}$, $J_2 = 186.2(7)\,\mathrm{K}$, $J_3 = 155.3(1.4)\,\mathrm{K}$, $J_4 = 46.9(4)\,\mathrm{K}$, and $J_{10} = 64.6(2)\,\mathrm{K}$. (b) Relevant exchange paths of $Na_6Cu_7BiO_4(PO_4)_4Cl_3$. (c) Finite-size scaling of the maxima of the equal-time structure factor $S(\mathbf{Q})/N_s$ from many-variable VMC (mVMC), VMC, pseudofermion FRG (PFFRG), pseudo-Majorana FRG (PMFRG) (at $T = 0.2J_2$), and the Schwinger-Boson mean-field theory method. Note that, compared to the other methods, PFFRG and PMFRG use a different definition of the system size N_s, which counts the number of correlated sites around a reference site, likely explaining the quantitative differences of our results. Furthermore, PMFRG results are obtained at a finite temperature $T = 0.2J_2$.

and the Schwinger boson (SB) formalism. We establish that the Hamiltonian of $Na_6Cu_7BiO_4(PO_4)_4Cl_3$ indeed realizes a nonmagnetic ground state, and provide evidence that the seventh magnetic site decorating the square-kagome lattice plays an important role in enhancing the degree of frustration, thus aiding the formation of a magnetically disordered phase in this material. This phase is shown to be a gapped VBC breaking translation symmetry, with a dimer pattern that is periodic in a 2×2 enlarged unit cell. We present its spectroscopic signatures to compare with future neutron scattering experiments.

Heisenberg Hamiltonian. We determined the magnetic interactions of $Na_6Cu_7BiO_4(PO_4)_4Cl_3$ using all electron density functional theory calculations. We use the crystal structure determined in Ref. [18] but simplify it slightly by choosing the majority Na(2) position and by removing O(5) from the Cl(3) position. All 14 Cu^{2+} ions in the primitive unit cell of the tetragonal structure are in square planar coordination with oxygen. The network they form is shown in Fig. 1(b), with the three symmetry inequivalent Cu(1), Cu(2), and Cu(3) shown in different colors. Cu(1) and Cu(2) form a square-kagome lattice, and Cu(3) is decorating this lattice above and below. Spin-polarized calculations show that the Cu^{2+} ions have $S = \frac{1}{2}$ moments, and at $U = 6.5\,\mathrm{eV}$ the system is insulating with a gap of $E_g = 1.7\,\mathrm{eV}$. We use the energy mapping technique that has yielded very good results in other copper based magnets [23,24] to extract the Heisenberg Hamiltonian parameters. Figure 1(a) shows the result of these calculations. Exchange couplings evolve smoothly with the on-site Coulomb repulsion U, and the Hamiltonian reproduces the experimental Curie-Weiss temperature at $U = 6.66\,\mathrm{eV}$ (vertical line). Some couplings that are less than 3% of the largest coupling J_2 are not shown in the plot. Among the couplings we resolve, there is only one, negligibly small, interlayer coupling (see Ref. [25]); as there is a full Na and Cl layer separating the magnetic layers, we expect the deviations from magnetic two-dimensionality to be small and beyond the scope of the present study. The two couplings making up the square-kagome lattice, $J_1 = 0.59J_2$ and J_2, are the third largest and largest coupling, respectively. The diagonal in the squares, $J_3 = 0.83J_2$, is the second largest coupling. One second nearest neighbor of the square-kagome lattice, $J_4 = 0.25J_2$, is also substantial. Furthermore, the $J_{10} = 0.35J_2$ interaction, which couples the square-kagome site Cu(2) to the magnetic decorating sites Cu(3) is found to be important. Strong buckling of the square-kagome lattice means that this coupling is the closest connection between Cu(3) and Cu(2) sites. It has a reasonable superexchange path through a phosphate group. On the other hand, the closer connection of a Cu(3) site to a Cu(1) site turns out to be negligible ($J_5 = 0.03J_2$).

The space group $P4/nmm$ of $Na_6Cu_7BiO_4(PO_4)_4Cl_3$ dictates that the J_1 (light blue) square and the J_2 (purple) triangle couplings are symmetry inequivalent as for the ideal lattice. The isotropic $J_1 = J_2$ Heisenberg antiferromagnet is host to a VBC ground state with a finite spin gap $\Delta \sim 0.04J_1$ [15], whose precise nature is still under debate [13–16]. For $J_2/J_1 \geqslant 2$, classically the system is host to a long-range ferrimagnetically ordered ground state (up-up-down) [10], however, for $S = 1/2$, exact diagonalization studies on $N = 24, 30, 36$ site clusters [10,26,27] find that the system enters the ferrimagnetic ground state for $J_2/J_1 \gtrsim 1.65$. The DFT estimated material couplings with $J_2/J_1 \sim 1.7$ thus precariously places the system in the vicinity of the nonmagnetic-magnetic phase boundary. It is then likely that the significant J_3 diagonal couplings ($J_3/J_1 \sim 1.45$) within the squares generate the necessary frustration to tip the balance in favor of a nonmagnetic

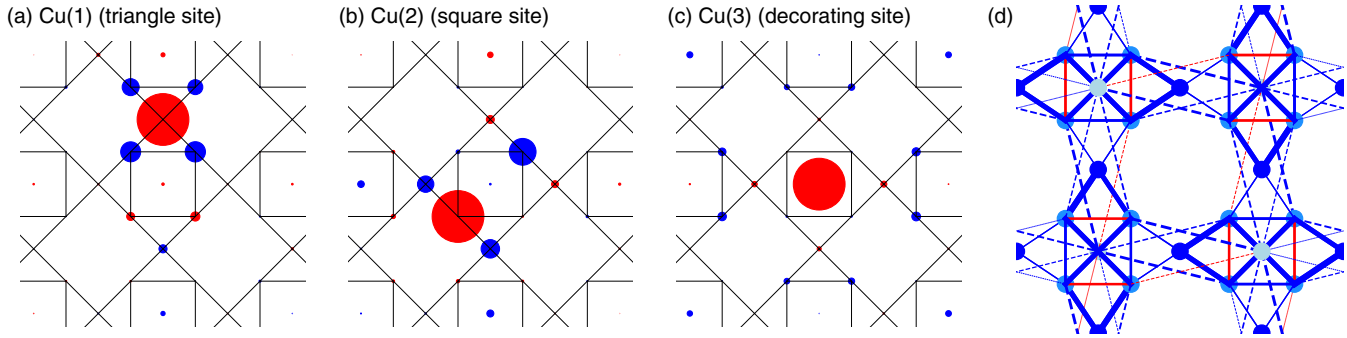

FIG. 2. (a)–(c) The pattern of real space (equal-time) spin-spin correlations $\langle \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \rangle$ from mVMC measured with respect to the three symmetry inequivalent sites. The radius of the circle is proportional to the magnitude of the correlator, and blue (red) denotes antiferromagnetic (ferromagnetic) correlations. The largest red circle corresponds to $i = j$. (d) The pattern of $\langle \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \rangle$ within a 2×2 unit cell showing the pattern of strong/weak bonds in the VBC ground state. The thickness is proportional to $|\langle \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \rangle|$ and blue (red) denotes antiferromagnetic (ferromagnetic) bonds, while the dashed lines denote the J_{10} bonds. Note that the idealized 2D unit cell shown here is rotated by 45° with respect to Fig. 1(b).

ground state. If so, the precise nature of the nonmagnetic state, in the presence of further neighbor coupling J_4 and the coupling J_{10} to the decorating Cu(3) site, needs to be carefully investigated by probing the delicate energetic competition between various quantum spin liquid and VBC ansätze.

Results. We begin our analysis by addressing the issue of the existence of long-range magnetic order in the ground state of the DFT Hamiltonian. Employing state-of-the-art numerical approaches of mVMC [28,29], fermionic VMC [30,31], PFFRG [32–34], PMFRG [35,36], and SB analysis [17,37–40], we compute the static (equal-time) spin structure factor

$$S(\mathbf{q}) = \frac{1}{N_s} \sum_{0 \leqslant i,\, j < N_s} \langle \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \rangle e^{i\mathbf{q}\cdot(\mathbf{r}_i - \mathbf{r}_j)}, \tag{1}$$

where N_s is the number of sites in the lattice, $\mathbf{q}$ is a momentum inside the extended Brillouin zone, and $\mathbf{r}_i$ denotes the site positions (accounting for sublattice displacements), following the convention outlined in the Supplemental Material [25]. Long-range dipolar magnetic order sets in when the maximum of $S(\mathbf{q})$ at $\mathbf{q} = \mathbf{Q}$ scales as $S(\mathbf{Q}) \propto N_s$ for large N_s [41]. The size scaling of $S(\mathbf{Q})/N_s$ [see Fig. 1(c)], yields the magnetization $m^2 \propto \lim_{N_s \to \infty} S(\mathbf{Q})/N_s$, which we consistently find to be zero (within error bars) from different approaches. This provides evidence for a nonmagnetic ground state which is corroborated by the rapid decay of the real space spin-spin correlations seen in Figs. 2(a)–2(c) (see Ref. [25] for results from fermionic VMC and Schwinger boson approaches).

To further elucidate the nature of the nonmagnetic ground state, we perform mVMC simulations with ansätze of different unit cell sizes. Table I shows variational ground state energies for the $Na_6Cu_7BiO_4(PO)_4)_4Cl_3$ Hamiltonian on three different clusters after symmetrization. The energies without the J_{10} coupling to Cu(3) sites are also given. Independent of the system size, the energies for 2×2 enlarged, i.e., 24-site unit cells are slightly (about 2%) lower compared to translation invariant states, i.e., either quantum spin liquids or lattice nematic. Other independent approaches reach similar conclusions, lending support for a translation symmetry broken ground state. These include a fermionic VMC analysis guided by different ansätze, a self-consistent fermionic mean-field analysis of different U(1) and $\mathbb{Z}_2$ spin liquids [42], as well as a Schwinger-Boson mean-field study. The pattern of real space (equal-time) spin-spin correlations is shown in Fig. 2(d), which points to its VBC nature. Here, one observes a checkerboard pattern, whereby the J_1 bonds featuring ferromagnetic (antiferromagnetic) correlations are always complemented by J_2 bonds hosting strong (weak) antiferromagnetic correlations thus forming a staggered horizontal/vertical pattern. The frustrating diagonal bonds inside the squares (J_3) show the strongest (antiferromagnetic) correlations, while the Cu(2) and Cu(3) sites are also found to be strongly correlated via J_{10} bonds, the latter highlighting the decorated nature of the lattice geometry. The VBC pattern possesses only C_2 symmetry.

TABLE I. mVMC energies E/J_2 on the 4×4, 6×6, and 8×8 lattices with a 1×1 and 2×2 unit cell after symmetrization.

Unit cell	4×4	6×6	8×8	
1×1	$-0.4256(1)$	$-0.4205(1)$	$-0.4172(1)$	With Cu(3)
2×2	$-0.4426(1)$	$-0.4304(1)$	$-0.4277(1)$	
1×1	$-0.4729(1)$	$-0.4783(1)$	$-0.4698(2)$	Without Cu(3)
2×2	$-0.4857(1)$	$-0.4816(1)$	$-0.4811(2)$	

Interestingly, we notice that the inclusion of J_{10} interactions in the Hamiltonian increases the frustration thereby enhancing the disordering tendency. This is reflected in an increase of the ground state energy per site [see Table I] and decreasing correlations between Cu(1) and Cu(2) sites in favor of bonds containing a Cu(3) site (see Fig. S7 in Ref. [25]). In particular, the ratio of $J_{10}/J_2 \sim 0.34$ places the material in the vicinity of the high point of frustration (largest ground state energy) (Table S2 in Ref. [25]). In mVMC calculations, the effect of increasing J_{10} is to induce the rotation and reflection symmetry breaking, and broadening of the maxima in S($\mathbf{q}$) leading to a more diffuse signal (see Fig. S4 in Ref. [25]). Thus, an important aspect of this result is that despite an appreciable magnetic coupling of the decorated Cu(3) ions with the square-kagome layers, it does not result in magnetic ordering. Quite the contrary, we show that the presence of these strongly correlated interlayer Cu^{2+} ions aids the

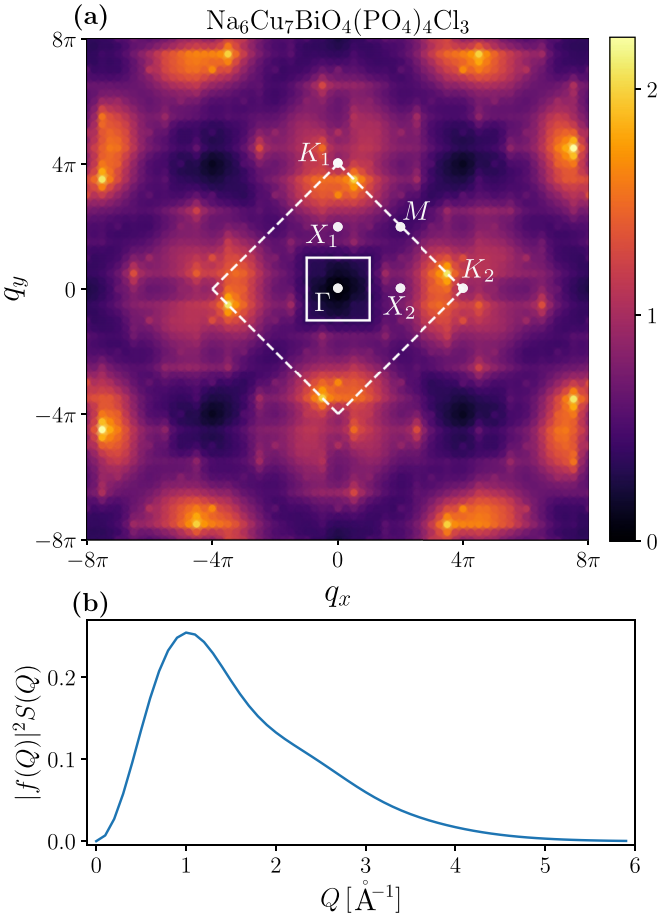

FIG. 3. (a) Static (equal-time) structure factor from mVMC [Eq. (1)] obtained w.r.t. true crystal lattice site positions [25] obtained on a $8 \times 8 \times 7$ site cluster [Note: the $(S\mathbf{q})$ is not periodic, and the Brillouin zones and high-symmetry points of the ideal geometrical lattice are only drawn for illustrative purposes]. (b) The corresponding powder average after accounting for the form factor.

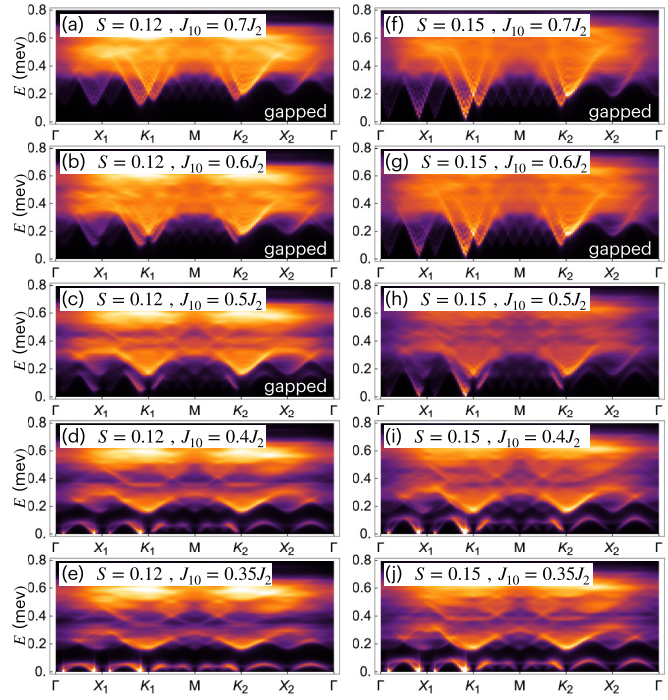

FIG. 4. Dynamical structure factors as function of S and J_{10}. The other parameters are as given in the caption of Fig. 1. As J_{10} increases and/or S decreases, a gap opens and a quantum paramagnet is stabilized. The Bose condensations appear at incommensurate q vectors. In the condensed state, the Cu(3) spins are ordered but the other spins in the square-kagome lattice remain very weakly ordered. This is reflected by the gap between the lower branch excitations and the continuum in the lower panels.

stabilization of a magnetically disordered ground state—thus settling the question raised by specific heat measurements [19]. It is interesting to note that in a related square-kagome material nabokoite $KCu_7TeO_4(SO_4)_5Cl$ which similarly features decorating Cu sites, signatures of long-range ordering have recently been reported [20].

The symmetry breaking manifests itself in the static spin structure factor which is likewise C_2 symmetric [see Fig. 3(a)], as obtained from mVMC. The maxima are located at $\mathbf{q} = \pm 2\pi(3.75, 2.25)$ with the follow-up maxima at $\mathbf{q} = \pm 2\pi(1.75, 0.25)$. The powder average structure factor is presented in Fig. 3(b) in order to facilitate comparison with potential neutron scattering studies.

The flexibility of the SBMFT method allows to efficiently compute the dynamical structure factor,

$$S(\mathbf{q}, \omega) = \frac{1}{N_s} \sum_{i,j} e^{i\mathbf{q}\cdot(\mathbf{r}_i - \mathbf{r}_j)} \int_{-\infty}^{\infty} dt e^{-i\omega t} \langle \hat{\mathbf{S}}_i(t) \hat{\mathbf{S}}_j(0) \rangle, \quad (2)$$

and to extract interesting magnon features with information on the Bose condensations of specific branches. Here, N_s is the total number of sites given by $n_u \times 2 \times l \times l$, where n_u is the number of sites per unit cell [here, 14 in the presence of the Cu(3) atoms], and l is the linear size of the system. This quantity can be compared with with neutron scattering experiments. Within this approach one can artificially tune S to lower values in order to enhance quantum fluctuations [40,43]. Thus, in Fig. 4, we show the dynamical structure factor for two representative spin values $S = 0.12, 0.15$ for which a quantum paramagnetic ground state can be stabilized, and also for various values of Cu(3) coupling, J_{10}, for a system size of $l = 12$ with 4032 spins. This figure displays several features: (i) decreasing J_{10} or increasing S favors Bose condensation of the Cu(3) atom spins, (ii) they appear at incommensurate values of the BZ, (iii) the rotational symmetry breaking is evident from asymmetric excitation spectra around the K_1 and K_2 points, see Fig. 3(a), and (iv) the gap closes at the extracted parameters, but a secondary gap between the lower branch and the continuum appears, reflecting ordered Cu(3) spins, while the others remain weakly ordered. This allows us to reveal the proximity to a phase transition between a quantum paramagnetic state and its Bose condensate counterpart.

As seen in the fermion approaches, the effect of projecting the wave function on exact physical states increases the quantum fluctuations and helps the system to remain disordered even in the presence of the Cu(3) atoms. In the SBMFT, since magnetic orders are more competitive by construction, they are favored at $S = 1/2$. Thus, in order to reach the quantum paramagnet, one has to reduce the spin value.

We can see that the Bose condensation arises on the Cu(3) spins while the others on the square-kagome lattice remain

disordered. This is reflected in Fig. 4 in a clear gap in the lower panels between the condensed branches and the excitations in the continuum (see Fig. S11 of the Supplemental Material [25] for real space spin correlation profiles). As one can see, a clear magnetic order appears on the Cu(3) spins while the spins on the square-kagome lattice are disordered.

Conclusions. We have determined a Heisenberg Hamiltonian for $Na_6Cu_7BiO_4(PO)_4)_4Cl_3$ with five significant antiferromagnetic exchange interactions. While triangle couplings in the square-kagome lattice dominate at about twice the size of the square couplings, the diagonals in the squares are the second largest interaction. The Cu(3) sites decorating the square kagome lattice in $Na_6Cu_7BiO_4(PO)_4)_4Cl_3$ turn out to be substantially coupled to the square sites. Our five numerical techniques all corroborate that the Hamiltonian has a nonmagnetic ground state, in agreement with the fact that experimentally no order was found down to 50 mK. We find the nature of this ground state to be a VBC which breaks translational symmetry. We predict static as well as dynamical structure factors to motivate studies of $Na_6Cu_7BiO_4(PO)_4)_4Cl_3$ with inelastic neutron scattering.

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