Topological long-range order for resonating-valence-bond structures

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Resonating-valence-bond structures involving pairings of sites not too distant from one another are considered. It is shown that under fairly general conditions these structures fall into different classes manifesting different "topological" long-range orders. Implications for "winding numbers" resulting from the superposition of pairs of valence-bond structures are noted.

I. INTRODUCTION

Following Anderson's suggestion[1] that resonating-valence-bond (RVB) theory is relevant for high-temperature superconductivity, much work has been published. Of some (possibly crucial) interest in this area has been [2–9] the occurrence of what might be termed a long-range topological order, which actually has also been[10–14] discussed earlier primarily in the rather different context of applications to conjugated hydrocarbons. In all work to date there has always been unnecessary limitations in the generality of results, say, as regards the restriction to special lattices, the limitation to nearest-neighbor RVB structures, the limitation to the discernment only of the parity of a long-range-order quantum number, and limitations in the rigor of proof of various results. Here we wish to remove these various restrictions, insofar as possible, especially as a diversity of applications seems possible.

We emphasize topological graph-theoretic[15] aspects of the results. The first feature of our basic assumptive framework is that of a *graph* $\mathcal{L}$, which is divided into successive *cells* along a long direction. Often $\mathcal{L}$ is a (regular) lattice, but in general it need not be. We assume "cyclic boundary conditions" (in the long direction) and imagine the divisions between cells not to hit any site of $\mathcal{L}$. Edges between sites are only to occur within a cell or between immediately adjacent cells. Sometimes on $\mathcal{L}$ there are quantum states labeled by other graphs in which every site is *paired* exactly to one another. These pairing (or VB) structures may be viewed as having different quantum-mechanical realizations: the well-known covalent singlet structures of the spin-$\frac{1}{2}$ Heisenberg model, extensions[16,17] of this for higher-spin Heisenberg models, mixed covalent and ionic singlet structures[16,18] for Hubbard-type models, the so-called "bond-orbital resonance theory" structures,[19] and yet others. One elegant view[20] is that the nearest-neighbor pairing structures are coherent combinations of a limited number of exact eigenstates to the full Schrödinger Hamiltonian so as to yield as closely as possible the set of classical chemical-bonding patterns. Indeed there is a large body of chemical literature where these RVB structures arise as chemical-bonding patterns in an empirical fashion, for either qualitative[21] or quantitative[22] purposes. The second major feature of our assumptive framework is a restriction on the pairing structures. Each VB structure C is assumed to be of *limited range* $r \geq 1$ in that any two sites paired in C are within a fixed distance, as measured by the minimum number of edges of $\mathcal{L}$ in a path in between the two sites. The range r is assumed to be very much less than the number L of cells of $\mathcal{L}$. As a consequence of our assumptions we may view $\mathcal{L}$ to be embedded in Euclidean space within a torus such that shortest path distances along the long direction roughly correlate to Euclidean distances. Further, a VB structure C is also embedded in the torus in a natural way, with the edges of C of a Euclidean length no greater than $\sim r$. The ideas and results following are framed in terms of these *natural embeddings*.

II. RESONANCE PARITY

Let the parity of the number of sites in the ith cell of the lattice graph $\mathcal{L}$ be denoted by π_i (being $+$ or $-$ as the number is even or odd). In order that $\mathcal{L}$ admit VB structures, the total number of sites must be even and the product over all π_i must be $+$. For a limited-range VB structure C let $\pi_i(C)$ denote the parity of the number of sites to the left of the boundary between cells i and $i+1$ that are paired with sites to the right. Thouless[3] terms $\pi_i(C)$ a *gap parity* (for C a dimer covering), and elsewhere[14] it has been termed a *resonance parity*. Now we have the following.

Theorem A.—For any limited-range VB structure C on $\mathcal{L}$,

$$\pi_{i-1}(C)=\pi_i\pi_i(C) \ .$$

Previously, this has been proved for a few particular cases and asserted several other times. To establish this, for structure C, let π_i(left) and π_i(right), respectively, denote the parities of the numbers of sites of the ith cell paired to sites to the left and right of cell i. Of course the parity of the number of sites internally paired within the cell must be $+$, so that $\pi_i = \pi_i(\text{left})\pi_i(\text{right})$. Now there may be some additional pairings across the cell boundaries of cell i that are not yet counted in π_i(left) and π_i(right). But these additional bonds, which are not paired to a site in i, modify the parities of π_i(left) and π_i(right) by the same factor σ to yield $\pi_i(C)$ and $\pi_{i+1}(C)$, respectively. That is, $\pi_{i-1}(C) = \sigma\pi_i(\text{left})$ and $\pi_i(C) = \sigma\pi_{i+1}(\text{right})$, and one obtains the theorem directly.

As emphasized in earlier work, a result such as that of theorem A implies a type of *long-range* order: If any one $\pi_i(C)$ is given, then all the others are determined. Since there are two conceivable choices ($+$ and $-$) for such a $\pi_i(C)$, there ordinarily are two classes of VB structures. A comparison of two VB structures ϕ and ψ may be made following Pauling[23] via the *superposition* graph, which is that obtained from the union of the edge sets of ϕ and ψ. Such superposition graphs consist of components (i.e., maximal connected pieces) that are either isolated edges or even cycles, here termed *little* and *big islands*, respectively. Inherent "global" characteristics of these islands are[4-7,10,11] relevant, as is now elucidated with a nomenclature introduced by Rokhsar and Kivelson[5] and by Sutherland.[4] For each big island there is a *winding number* giving the number of times it wraps the long way around $\mathcal{L}$. This topological concept (or invariant) is illustrated in Fig. 1. More precisely if one walks along the big island counting $+1$ for each boundary crossing from left to right and -1 for each crossing from right to left, then the island's winding number is the absolute value of the sum of these numbers in proceeding once around the island. The *winding number* for the superposition graph is the sum of those of its (big) islands. Now we can state the following:

Theorem B.—The parity of the winding number for the superposition graph of ϕ and ψ is $\pi_i(\phi)\pi_i(\psi)$, which is independent of i.

To establish this, consider "local" structures which do not contribute to a winding. In particular, consider how two edges of ϕ and/or ψ can cross a given boundary and be connected by a path "entirely to the left" of the boundary. More precisely, consider two boundary-crossing edges in the same island and walk along the edges of the island starting from the right vertex of the other such edge; count $+1$ each time the boundary is crossed right to left and -1 each time it is crossed left to right; then the two edges considered are *left-connected* if and only if the running sum of the crossing numbers is always positive after the first step up until the last step when it is 0. See Fig. 2. A 0-length connecting path to the left can give a little island, clearly giving no winding. In a big island all boundary crossings not left-connected must be of the same sign and thence contribute to the island's winding number. Thus the winding number of the superposition graph is the total number of boundary crossings minus the number that are left-connected. Since the parity of the total number of boundary crossings is $\pi_i(\phi)\pi_i(\psi)$, and since the left-connected edges occur in pairs with net parity $+$, the theorem is established. An independence check may be made via theorem A, which implies $\pi_{i+1}(\phi)\pi_{i+1}(\psi)$ and $\pi_i(\phi)\pi_i(\psi)$ are the same, so that this parity is independent of the boundary i.

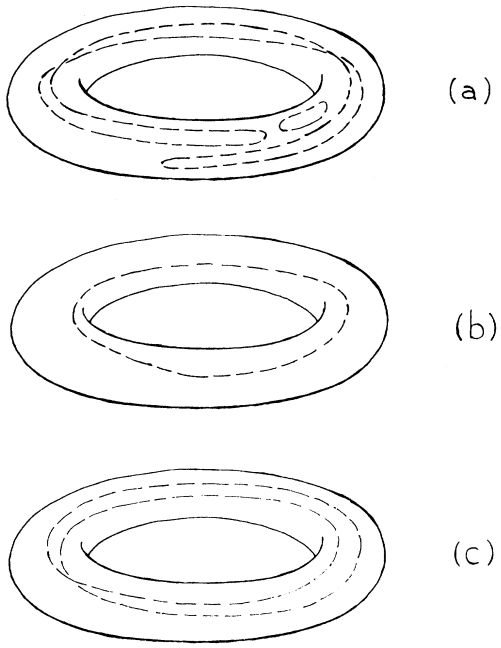

FIG. 1. Illustrations of windings for islands indicated by dashed curves within the toroidal framework of $\mathcal{L}$. Both islands in (a) have winding number 0, that in (b) has winding number 1, and that in (c) is 2.

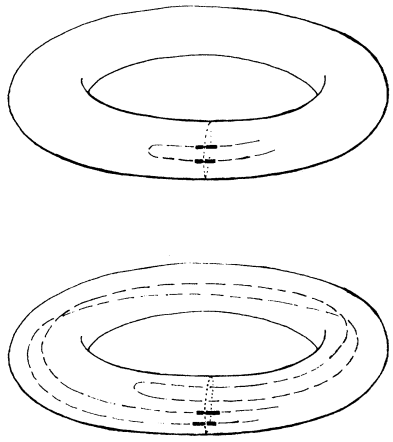

FIG. 2. Two cases where two boldface boundary crossing edges are left-connected. The manner of completion of the islands on the right is irrelevant.

III. TOPOLOGICAL RESONANCE QUANTUM NUMBER

With the imposition of additional conditions, results stronger than those of the preceding section can be obtained. We assume two *bipartitioning* conditions: First the sites of $\mathcal{L}$ are bipartitioned into two equicardinal disjoint sets, A and B, and second the limited-range VB structures are such that each pairing is between sites, one in A and one in B. This second assumption is expected to be more physically reasonable when the dominant pairing-favoring interactions occur between pairs of sites one from each set. That $\mathcal{L}$ admit limited-range VB structures as in the second condition implies that "locally" there are an equal number of A and B sites. If we let Δ_i be the excess of A sites over B sites in cell i, then sums over arbitrarily large sequences of these Δ_i must remain bounded (with the bound tighter for more limited-range VB structures). But even for nearest-neighbor VB structures and translationally symmetric $\mathcal{L}$, these Δ_i need not be 0. In this case they can simply alternate in sign.

We now proceed with some additional useful concepts, and finally some consequences. Let $Q_i^+(C)$ be the number of pairings across the boundary between cells i and $i+1$ such that an A site on the left is paired in C to a B site on the right. Also let $Q_i^-(C)$ be the corresponding number with B sites on the left paired to A sites on the right. Then define a (local, at boundary i) *resonance* quantum number for C as

$$Q_i(C)\equiv Q_i^+(C)-Q_i^-(C)\ . \tag{3.1}$$

Clearly the parity of $Q_i(C)$ is just the pairing parity $\pi_i(C)$ of the preceding section. In analogy to theorem A we have the following.

Theorem A'.—If the bipartitioning assumptions apply, then for any limited-range VB structure C,

$$Q_i(C)=Q_{i-1}(C)+\Delta_i\ .$$

To obtain this, start defining (with regard to C) various numbers $a_i(\cdots)$ and $b_i(\cdots)$ of A and B sites in cell i; the designations "$\cdots$" here may be "left," "right," or "int" as counts refer to sites of cell i paired in C to other sites to the left of cell i, to the right of cell i, or interior to cell i. Then the difference between the number of A and B sites in the ith cell is

$$[a_i(\text{left})+a_i(\text{right})+a_i(\text{int})] -[b_i(\text{left})+b_i(\text{right})+b_i(\text{int})]=\Delta_i\ . \tag{3.2}$$

Since A sites are only paired to B sites, $a_i(\text{int})=b_i(\text{int})$, and these two terms cancel in (3.2). But again, A and B sites pair only to one another, so we identify the remaining terms in brackets in (3.2) as $-Q_{i-1}(C)$ and $+Q_i(C)$, respectively, and the theorem follows.

From this result we see that ordinarily there may now be several long-range order classes, and again comparisons between classes are of interest. In fact, for theorem B, we have an analogous result:

Theorem B'.—The magnitude of the difference between the resonance quantum numbers for two limited-range VB structures satisfying the bipartitioning conditions is a lower bound to the winding number of the associated superposition graph.

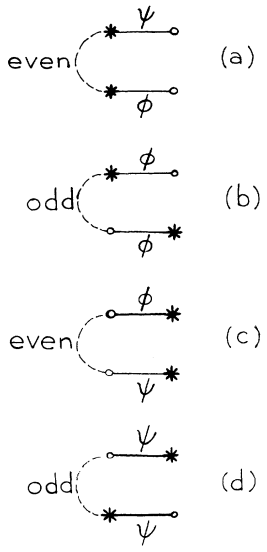

FIG. 3. The four possible manners of left-connecting two boundary-crossing edges from ϕ and/or ψ.

To establish this we use ideas in the proof of theorem B. Given two structures ϕ and ψ we consider the possible manner of left-connecting two edges crossing a fixed boundary between two cells. There are four possible manners indicated in Fig. 3. In this figure A sites are "starred," B sites are "unstarred," and an indication of the parity of the length (i.e., number of edges) of the connecting path off to the left is given. Clearly this parity determines whether the two boundary-crossing edges are in the same or different VB structures (ϕ and ψ). The types of boundary edge-pairs that may be left-connected are indicated in Fig. 4(a), and in Fig. 4(b) are indicated

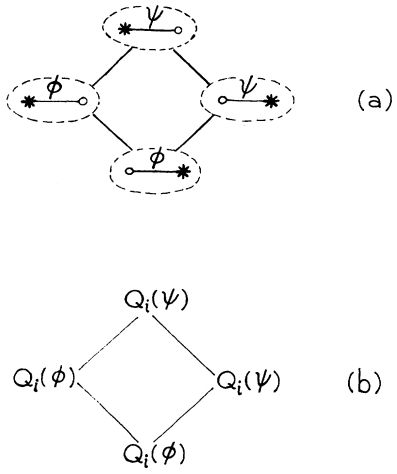

FIG. 4. Types of boundary-crossing edges that may be left-connected. Again, in (a), "starred" and "unstarred" sites indicate those of types A and B, respectively.

the corresponding counts $Q_i^+(\psi), Q_i^+(\phi), Q_i^-(\phi), Q_i^-(\psi)$ to which each type contributes. Thence we see that any boundary pairs of types for $Q_i^+(\psi)$ and $Q_i^+(\phi)$ can be left-connected only to those of types for $Q_i^+(\phi)$ and $Q_i^-(\psi)$. Thus the maximum number of boundary pairs not left-connected is

$$|[Q_i^+(\psi)+Q_i^-(\phi)]-[Q_i^+(\phi)+Q_i^-(\psi)]| = |Q_i(\psi)-Q_i(\phi)| \ . \quad (3.3)$$

As in the proof of theorem B, each of these remaining (not left-connected) boundary pairs evidently must be involved in windings. Thus the present theorem is proved.

IV. FURTHER COMMENTS

Applications envisage the VB structures of the preceding sections as labeling states. Thence the resonance quantum numbers of theorems A and A′ identify long-range-order classes (or subspaces) of states. Two states from two different classes should then differ repeatedly in each of the L cells of the network $\mathcal{L}$, and thence be asymptotically noninteracting, via any interaction mediated by a few-particle operator. That is, we anticipate the following.

Theorem C.—If $|\Phi\rangle$ and $|\Psi\rangle$ are normalized states from two different long-range-ordered subspaces, then $\langle\Phi|\Psi\rangle$ and $\langle\Phi|H|\Psi\rangle$ asymptotically approach zero as $L/r\to\infty$ and where H is a few-particle size-extensive operator (such as the Hamiltonian).

This can often be viewed[4–7,11,12] as a consequence of theorems B or B′ with nonzero winding-number islands "quenching" the matrix elements, as is evident, e.g., from Pauling's[23] formulas, wherein a factor of 2^{2-n} occurs for each big island of $2n$ sites. For H the nearest-neighbor spin-$\frac{1}{2}$ Heisenberg model, Bonesteel[7] has detailed this proof. Essentially the same approach also works for the higher-spin cases of Refs. 16 and 17, as has been indicated[10] for certain $\mathcal{L}$ before. The same approach can be extended to deal with more complicated interactions in H—most easily for local permutation operators, which may be viewed simply to permute any RVB structure in a local fashion and thereby not change its long-range-order class. The result of the theorem "should" hold under much more general circumstances if the state labels are a reasonable reflection of physical reality. The various long-range-order classes identify asymptotically noninteracting subspaces.

In at least one special case there is only one long-range order class. In particular for a "noncyclic" graph our theorems still apply, since $\mathcal{L}$ admits an embedding into a torus. But at the Lth intercell boundary between what are now the left and right sides of $\mathcal{L}$ one has no crossing bonds in a limited-range VB structure C and $Q_L(C)=0$, thereby determining the long-range-order class. Even in this case some applications occur.[12,24]

With the assumption of translational symmetry, so that all cells are equivalent, there are some further consequences. The parity π_i of theorem A is then independent of i, and is abbreviated to π. The excess Δ_i of theorem A′ also has a magnitude Δ independent of i, while its sign alternates, as $(-1)^i$. Then we have the following.

Theorem D.—Granted theorem C and translational symmetry, the eigenenergies of H on the space of limited-range VB structures are asymptotically twofold degenerate if $\pi=-$ or $\Delta\neq 0$.

This follows immediately, since in these indicated cases there are distinct translationally equivalent long-range-order classes. Previously such ideas have been argued to be of relevance for "bond" localization[10–12,14] and "solitonic" excitations.[12–14] Another interesting feature of theorem D as described by Bonesteel[7] is a relation to the theorem of Lieb, Schultz, and Mattis,[25] also indicating either gapless excitations or a twofold asymptotic degeneracy. Further too for a special class of graphs there is a correspondence to another Kekulé-structure parity[26] and interesting consequences are not followed here.

For the case that all $\Delta_i=0$, as often occurs with translationally symmetric lattices, Q is conserved, from one position along the torus to the next. Correspondences of dimer coverings to sets of directed self-avoiding walks often occur and applications[27] to polymer packings may be made. There are mathematical applications[28] also. Though previously this correspondence has only

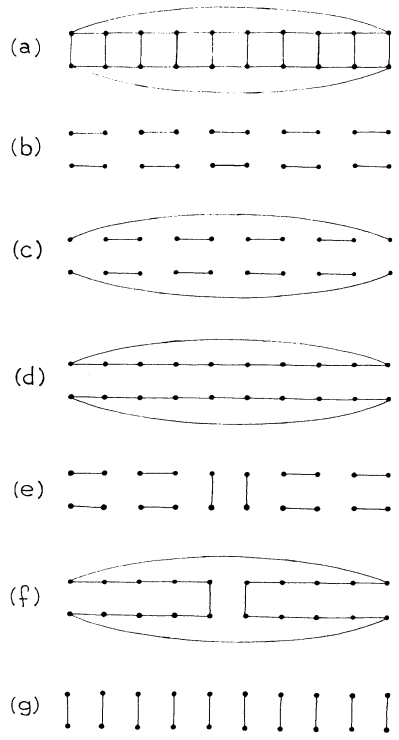

FIG. 5. Illustrations concerning the distinction of long-range-order and winding numbers, for structures on the cyclic ladder graph of (a). The Kekulé structures of (b) and (c) lie in the same long-range-order class, though they have a superposition graph of (d) with winding number 2. The local change from Kekulé structure (b) to (e) gives a superposition graph (f), which is obtained from (c) and (e), and which has winding number 0. Continuing such local changes one reaches (g), from either (b) or (c).

been made for the honeycomb lattice, our considerations allow it for any two-dimensional bipartite lattice as well as for higher-dimensional lattices with vertices of degree 3.

Finally the relation between winding numbers and resonance quantum numbers might be elucidated a little more. Though different resonance quantum numbers imply different winding numbers, the converse is not true. For example, the configurations of Figs. 5(b) and 5(c) give a superposition graph with winding number 2, though both configurations have the same resonance quantum number. This winding number can be modified to 0 by a simple local rearrangement, also as illustrated in Fig. 5. Even more relevant is the fact that a whole sequence of *local* rearrangements will change either of the first two configurations into the other. Hence these two configurations should mix significantly through this "path," with the intermediate configurations also mixing. That is, winding numbers do not break our system state space up into noninteracting classes (or sectors, or subspaces)—rather it is the resonance quantum numbers that do this—generally more finely than the pairing parity of Sec. II.

In conclusion there seems to be a diversity of applications of the basic tools established, clarified, and developed here. Some further discussion of these ideas in the context of the square-planar lattice occurs in the accompanying paper.

ACKNOWLEDGMENTS

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