Necessity and Experimental Consistency of Antiferromagnetic Ground State without Long-Range Order

G. W. PRATT, JR.
Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts*

(Received December 5, 1960)

A proof is given that long-range order measured as the average value of the z component of sublattice magnetization of an antiferromagnet must be zero in the exact ground state if that state is nondegenerate and the Hamiltonian is invariant under time reversal. The neutron diffraction magnetic cross section is shown not to depend on such a measure of long-range order.

THE purpose of this note is first to point out that the correct quantum mechanical ground state for an antiferromagnet cannot show any long-range order regardless of the spin per atom, lattice geometry, or the nature of the anisotropy as long as the Hamiltonian is invariant under time inversion and the ground state is nondegenerate. All of the Hamiltonians proposed so far in discussing antiferromagnetism in the absence of an external magnetic field fall within this class. The second and essential point here is to show that starting from a ground state showing no long-range order, one can reproduce the elastic magnetic scattering of the completely long-range ordered classical Néel state.

The definition of long-range order used here is the average value of the z component of the total spin of a single sublattice, i.e., $\langle 0|S_A{}^z|0\rangle$, where $|0\rangle$ is the antiferromagnetic ground state. This same definition or one completely equivalent to it has been used by many authors[1] in discussing the subject. An alternate definition has been suggested by Rodriguez,[2] this being essentially $\sum_j\langle 0|\mathbf{S}_j\cdot\mathbf{S}_{j+\lambda}|0\rangle$, where λ is chosen such that the spins $\mathbf{S}_j$ and $\mathbf{S}_{j+\lambda}$ are always on the same sublattice. Long-range order defined in this manner is not necessarily zero as will be evident in the following.

So long as the Hamiltonian[3] is an even function of the angular momentum operators that appear in it, it is invariant under the operation of time reversal, denoted by K, which effectively reverses the direction of all momenta. Therefore, the nondegenerate eigenstates will also be eigenfunctions of K. Consider a portion of the total ground state that exhibits long-range order defined as $\langle 0|S_A{}^z|0\rangle$. Time-reversal symmetry requires that there be another part of the ground state with exactly the opposite long-range order and which appears in the complete eigenstate with the same weight. The net long-range order of these two parts, and hence of the entire state, is zero.[4]

We now show that the Néel state ψ_N corrected for time-inversion symmetry, hence with zero long-range order, leads to exactly the same neutron cross section as the perfectly ordered state ψ_N which we take for simplicity to be

$$\psi_N=\frac{\det}{(2N!)^{\frac{1}{2}}}|u_{1A}(1)\alpha(1)\cdots u_{NA}(N)\alpha(N)u_{1B}(N+1)\times\beta(N+1)\cdots u_{NB}(2N)\beta(2N)|. \quad (1)$$

The time-inversion operator K is

$$K=(i)^{2N}S_{y1A}\cdots S_{yNA}S_{y1B}\cdots S_{yNB}C,$$

C being the operation of complex conjugation. One finds that

$$K\psi_N=(-1)^N\frac{\det}{(2N!)^{\frac{1}{2}}}|u_{1A}(1)\beta(1)\cdots u_{NA}(N)\times\beta(N)u_{1B}(N+1)\alpha(N+1)\cdots u_{NB}(2N)\alpha(2N)|.$$

Eigenstates of K that are even or odd under K are

$$\psi_e=(\psi_N+K\psi_N)/\sqrt{2}, \quad (2)$$

$$\psi_o=(\psi_N-K\psi_N)/\sqrt{2}. \quad (3)$$

ψ_e and ψ_o are, of course, not actual eigenstates but they have the same average energy. The cross section for magnetic scattering of neutrons as used in experiments on antiferromagnets is[5]

$$\frac{d^2\sigma}{d\Omega dE}=\sum_{\epsilon,\eta}\left(\frac{\gamma e^2}{mc^2}\right)^2|f(K)|^2(\delta_{\epsilon\eta}-K_\epsilon K_\eta)\sum_{qq'}P_q\frac{k'}{k}\times\sum_{n,m}\exp[i\mathbf{K}\cdot(\mathbf{n}-\mathbf{m})]\langle q|S_m{}^\epsilon|q'\rangle\times\langle q'|S_n{}^\eta|q\rangle\delta\left\{E_q'-E_q+\frac{\hbar^2}{2m_0}(k'^2-k^2)\right\}. \quad (4)$$

Here the initial state of the scatterer is $|q\rangle$, the probability of this being P_q, and the final state $|q'\rangle$, $S_m{}^\epsilon$ is the $\epsilon(x,y,z)$ component of the spin at the mth lattice site,

* Operated with support from the U. S. Army, Navy, and Air Force.

[1] P. W. Anderson, Phys. Rev. **86**, 694 (1952); P. W. Kasteleijn, Physica **18**, 104 (1952); H. L. Davis, Phys. Rev. **120**, 789 (1960); and T. Nagamiya, K. Yosida, and R. Kubo, Suppl. Phil. Mag. **4**, 1 (1955).

[2] S. Rodriguez, Phys. Rev. **116**, 1474 (1959).

[3] We treat conservative systems here so that the Hamiltonian is not an explicit function of time.

[4] Davis (reference 1) discussed the lack of agreement in the literature as to the existence of long-range order. See also R. Orbach, Phys. Rev. **112**, 309 (1958). L. Walker, Phys. Rev. **116**, 1089 (1959) has stated that it should vanish in the exact ground state, as have T. W. Ruijgrok and S. Rodriguez, Phys. Rev. **119**, 596 (1960).

[5] W. Marshall, unpublished lecture notes.

and $\mathbf{K}$ is the scattering vector. It has apparently been customary to argue that the antiferromagnetic ground state must have long-range order on the basis of the experimental results for elastic scattering. This argument assumes that the system is in the ground state before and after scattering and that since the scattering shows long-range order, so must the ground state. Therefore, the elastic scattering has been treated by setting $|q\rangle = |q'\rangle = \psi_N$ in (4). If we do this and evaluate that part of (4) of interest here, i.e.,

$$\sum_{qq'} \sum_{\epsilon,\eta} (\delta_{\epsilon\eta} - K_\epsilon K_\eta) \langle q | S_m{}^\epsilon | q' \rangle \langle q' | S_n{}^\eta | q \rangle, \tag{5}$$

the result is

$$(1-K_z{}^2)\langle \psi_N | S_m{}^z | \psi_N \rangle \langle \psi_N | S_n{}^z | \psi_N \rangle = (1-K_z{}^2) S_0{}^2 \theta_{mn}, \tag{6}$$

where θ_{mn} is $+1$ if m and n are on the same sublattice and -1 if not and S_0 is the spin per atom. The result of evaluating (5) using ψ_e and ψ_o is

$$\begin{aligned}(1-K_z{}^2)\left\langle \frac{\psi_N + K\psi_N}{\sqrt{2}} \middle| S_m{}^z \middle| \frac{\psi_N - K\psi_N}{\sqrt{2}} \right\rangle \\ \times \left\langle \frac{\psi_N - K\psi_N}{\sqrt{2}} \middle| S_n{}^z \middle| \frac{\psi_N + K\psi_N}{\sqrt{2}} \right\rangle \\ = (1-K_z{}^2)\langle \psi_N | S_m{}^z | \psi_N \rangle \langle \psi_N | S_n{}^z | \psi_N \rangle \\ = (1-K_z{}^2) S_0{}^2 \theta_{mn}.\end{aligned} \tag{7}$$

Therefore, the degenerate pair of states ψ_e and ψ_o, which separately show no long-range order, lead to the same elastic cross section as the ordered state ψ_N. If the actual ground state were degenerate, which is unlikely, these states can always by a unitary transformation be made eigenstates of K, hence with $\langle S_A{}^z \rangle$ zero, without changing the cross section which is invariant under such an operation. Thus it is not at all necessary to require that the ground state be ordered to be consistent with scattering results.

It is of interest to evaluate the magnetic scattering for a more complicated set of states than ψ_e and ψ_o. Thus consider the set of states constructed by taking two sublattices each with maximum spin, i.e., $S_A = NS_0$ and $S_B = NS_0$ and combining $\mathbf{S}_A$ and $\mathbf{S}_B$ to produce a total resultant spin of $\mathbf{S} = 0, 1, 2, \cdots 2NS_0$. Assuming that the system is described by the isotropic exchange Hamiltonian

$$\mathcal{H} = -2J_{AB} \sum_{iA,jB}^{\text{n.n.}} \mathbf{S}_{iA} \cdot \mathbf{S}_{jB}, \tag{8}$$

and for convenience that only the nearest neighbor exchange between spins on different sublattices is nonzero, one finds that the $S=0$ state has the lowest average energy of the set for J_{AB} negative. That this singlet is not an actual eigenstate of (8) is not of paramount concern here. The essential facts are that the singlet has no long-range order and that the $S=1$ member of this set of states has an average energy only $|J_{AB}|/N$ higher than the singlet. This latter result is to be expected since the large sublattice spins can be regarded classically. Because of this infinitisimal energy difference, transitions between the $S=0$ and $S=1$ members of the set must be included in the elastic magnetic scattering.

If (5) is evaluated for this set of states, assuming that initially the scatterer is in the $S=0$ state, then by the Wigner-Eckart theorem only when $|q'\rangle$ of (5) is $\psi(S=1, M_S=1, 0, -1)$ is there a nonzero result. The result is

$$\begin{aligned}\sum_{M_S} \sum_{\epsilon,\eta} (\delta_{\epsilon\eta} - K_\epsilon K_\eta) \langle \psi(s=0) | S_m{}^\epsilon | \psi(s=1, M_S) \rangle \\ \times \langle \psi(s=1, M_S) | S_n{}^\eta | \psi(s=0) \rangle \\ = (3-K^2)(S_0{}^2/3)\theta_{mn}.\end{aligned} \tag{9}$$

This differs from the Néel result (6) in that (9) is isotropic whereas in the Néel state the z axis is implicitly taken as a preferred direction. Thus if we were to make the scattering from ψ_N isotropic by assuming $K_z{}^2 = K^2/3$, (6) and (9) become identical.

The point we wish to make in exhibiting the result of (9) is that starting with states having no long-range order, as it is often defined, and recognizing that there are excited states infinitesimally near the ground state[6] to which transitions can be made (which would only show up experimentally as elastic) one can explain the magnetic scattering. Conversely, it is neither necessary nor correct to infer from the elastic magnetic scattering in antiferromagnets that the ground state must show long-range order. Therefore, the concept of the sublattice orientations interchanging over a period of years, which is equivalent to assuming that the antiferromagnetic ground state is not stationary and that thermal equilibrium is never achieved, is not to be regarded as a physical reality. However, if the degenerate pair $\psi_N \pm K\psi_N$ very nearly represented the actual situation, an ordered linear combination could be used as a matter of convenience to discuss the energy of the ground state and of states nearby. Then this concept becomes a necessary condition that such a discussion be meaningful.

[6] For evidence that this indeed is the case, see L. Hulthén, Proc. Acad. Sci. Amsterdam **39**, 190 (1936); P. W. Anderson, Phys. Rev. **86**, 694 (1952); and R. Orbach, Phys. Rev. **115**, 1181 (1959).