On the possibility of saturated ferromagnetism

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This paper is concerned with the possible degree of spin alignment in the ground state of a system of spin-1/2 fermions whose Hamiltonian (spin independent) consists of the usual kinetic-energy term plus a coordinate-dependent potential energy. It is shown that by suitable choice of the one- and two-body potentials one can rigorously insure that the ground state of a finite system of this sort is one of maximum multiplicity. This result contradicts a contention by Izuyama that when certain very general postulates are fulfilled, a crystal having complete spin alignment in its ground state in the thermodynamic limit will have to have a vanishing or negative spin-wave stiffness. It is shown that there is in fact no conflict with any of Izuyama's several correct expressions for the spin-wave stiffness, provided that in each case the last step in the evaluation is carried out with sufficient care.

I. INTRODUCTION

To what extent can a system of fermions with only coordinate-dependent interactions show a preference for spin alignment in its ground state? Lieb and Mattis[1] have proved a remarkably general theorem that if spin-$\frac{1}{2}$ fermions move in any localized region of a one-dimensional space under an arbitrary nonsingular many-body potential-energy function, the lowest state of spin S always lies lower than the lowest state of spin $S+1$. Although the existence of Hund's-rule atoms shows that there can be no such sweeping theorem in three dimensions, it has occasionally been speculated that even in three dimensions the ground state never has *all* spins parallel. Such a speculation is rendered plausible by the fact that in any number of dimensions the ground state of a system of two electrons must have a nodeless coordinate wave function, hence be a singlet. Through no arguments in support of such a universal speculation have been advanced, Izuyama has, in a recent paper[2] and its sequel, [3-8] undertaken to prove a theorem of similar import for electrons in a metallic or nonmetallic crystal, in the thermodynamic limit, subject to the proviso that the ground state be nondegenerate (except for spin degeneracy).

It is the purpose of the present note to point out (a) that by suitable choice of the one- and two-body potential energy functions one can rigorously insure that certain finite systems of spin-$\frac{1}{2}$ fermions have ground states with maximum multiplicity; (b) that this fact makes overwhelmingly plausible the assertion that there can exist crystalline systems satisfying Izuyama's postulate whose ground states in the thermodynamic limit are saturated ferromagnetic ones—counterexamples to his theorem; (c) that his original proof,[2] though ingenious, contains an inadmissible step; and (d) that the physical plausibility arguments which he has invoked more recently[5,6,8] to circumvent possible objections to this original proof turn out on careful scrutiny to be in no way inconsistent with the existence of saturated ferromagnetic ground states.

II. THREE-FERMION SYSTEMS PREFERRING SPIN ALIGNMENT

Our first point is little more than a buttressing of a fact already well known in the theory of the asymptotically weak exchange coupling of fermions bound to well-separated centers: while pair exchange is, under very general conditions, equivalent to a Heisenberg-type exchange coupling of pairs of spins with antiferromagnetic sign,[9] exchange around an odd-numbered ring favors parallel spin alignment.[10,11] As the rigorous formulation of the theory of direct exchange[9] requires critical attention to a number of approximations, it may be of some value to demonstrate *ab initio* the truth of the statement just made. We shall consider first the problem of three spin-$\frac{1}{2}$ fermions on a ring, i.e., in a one-dimensional space with periodic boundary conditions. The Lieb-Mattis theorem[1] does not apply for such boundary conditions, and in fact we can use essentially their same reasoning to prove the reverse result in certain cases.

Let the Hamiltonian be

$$H=\sum_{i=1}^{3}\frac{p_i^2}{2m}+V(x_1,x_2,x_3)\ , \tag{1}$$

where V is a symmetrical function of its arguments. The eigenfunctions can be classified according to the irreducible representations of the permutation group of three objects; the characters are given in Table I. Here the first column labels representations according to the corresponding Young tableaux (partitions of 3 into a sum of integers); the last column identifies the type of spin functions with which each must be associated to obtain spin-coordinate antisymmetry.[12] Now it is easily verified that the planes in the three-dimensional configura-

TABLE I. Representations of the group of permutations of three particles.

Representation	Identity	Characters of 3 pair interchanges	Characters of 2 cyclic permutations	Associated spin state for spin-$\frac{1}{2}$ fermions
$D(3)$	1	1	1	Nonphysical
$D(2+1)$	2	0	−1	Doublet
$D(1+1+1)$	1	−1	1	Quartet

tion space on which the coordinates of any two of the particles coincide divide this space into two connected regions—I and II—in which the orders of the particles on going around the ring are 123 and 321, respectively. The lowest eigenfunction, which by a well-known theorem[13] must be nodeless, is of the unphysical symmetry type (3). However, in a limiting case where V becomes so rapidly infinite as two particles approach each other that the wave function must vanish on the surface separating regions I and II, the antisymmetric function—symmetry type $(1+1+1)$—defined as equalling the ground-state eigenfunction in region I and its negative in region II, will also be an eigenfunction with the same energy. For this case the lowest eigenfunction of symmetry type $(2+1)$ will necessarily have an energy higher than these two by a finite amount, as it must possess nodal surfaces interior to region I and region II. [To prove this last statement we need only note that by Table I a real eigenfunction of symmetry type $(2+1)$ must be orthogonal to any function which is unchanged under the cyclic permutation of the three particles.] If now we decrease the repulsive potential on the surface dividing regions I and II from infinity to a large finite value, the resulting tunneling between these regions will cause the energies of the completely symmetric and completely antisymmetric states to split slightly apart, with the former of course lying lower; however, if this splitting is small enough, the completely antisymmetric state will continue to lie lower than the one of symmetry type $(2+1)$. In other words, the quartet state will lie lower than either of the doublets.[14]

It is clear that essentially the same reasoning will apply if the three fermions, instead of being constrained to the circumference of a ring, are allowed coordinates in a three-dimensional space, provided the one-body part of the potential-energy function is such as to confine the particles effectively to a close neighborhood of a ring. If in addition the one-body potential energy has pronounced minima at three different places around the ring, the fermions will spend most of their time in configurations with each of them close to a particular one of the minima; i.e., the system can be visualized as one of three one-particle "atoms," weakly exchange coupled to each other. Such an exchange is always describable in terms of an effective spin Hamiltonian,[15] and for three one-particle atoms coupled in this manner the effective spin Hamiltonian must be of the Heisenberg form

$$H_{eff} = -2J_{123}(\vec{s}_1 \cdot \vec{s}_2 + \vec{s}_2 \cdot \vec{s}_3 + \vec{s}_3 \cdot \vec{s}_1) \ , \qquad (2)$$

where $\vec{s}_1$, $\vec{s}_2$, and $\vec{s}_3$ are the spin operators associated with the three "atoms," and where J_{123} is positive.[10,11]

III. MANY-FERMION SYSTEMS PREFERRING COMPLETE SPIN ALIGNMENT

Consider the generalization to many particles of the three-particle system just described: N spin-$\frac{1}{2}$ fermions spend most of their time bound to N discrete centers ("atoms"), between which they occasionally exchange places. One can make very general arguments[15] that the levels of such a system must be the eigenvalues of an effective Hamiltonian operating in the space of N atomic spins, whose terms can be associated with the various types of permutation by which the assignment of fermions to atoms can be altered. If the separations of the atoms are large compared with the radii of their bound wave functions, the terms of the spin Hamiltonian corresponding to permutations of higher and higher order will decrease very rapidly, because they correspond to tunneling to larger and larger distances in configuration space. Under such conditions the spin Hamiltonian tends to be dominated by terms corresponding to pair exchanges or exchanges around the smallest ring paths. (For example, the competition of a dominant antiferromagnetic pair exchange with a subsidiary ferromagnetic exchange via three-atom rings has been much discussed[16] in the theory of solid He^3.)

Since each three-atom ring exchange will contribute a ferromagnetic term of the form (2) to the spin Hamiltonian, it should be possible to construct a completely ferromagnetic crystal by choosing the one-electron part $V^{(1)}$ of the potential energy to be only moderately high along a network of tubes forming triangles connecting all the atoms of the crystal, and very high at other points between the atoms, while suppressing the normally dominant pair exchanges by making the two-electron part $V^{(2)}$ ex-

tremely large when the distance between two particles is comparable with or less than the diameter of a tube or an atom. While there is no rigorous proof that the existence of a very large number of extremely small terms in the spin Hamiltonian, corresponding to high-order permutations, could not alter the ferromagnetic nature of the bound state in the limit of an infinite number of atoms, such a possibility seems extremely unlikely; the extensive literature[17,18] on the "nonorthogonality paradox"[19] in the Heitler-London approximation to direct-exchange theory gives general support to this expectation.

Thus we conclude that by proper choice of $V^{(1)}$ and $V^{(2)}$ one can construct a crystalline array of spin-$\frac{1}{2}$ fermions—e.g., a face-centered-cubic lattice—which will have a non-metallic ground state in which all the spins are aligned parallel.

IV. CRITIQUE OF THE PREDICTION OF VANISHING SPIN-WAVE STIFFNESS

The theorem which Izuyama undertook to prove in his first paper[2] was the following: Let the Hamiltonian of N electrons be of the nonrelativistic (spin-independent) form, with a potential energy $V(x_1, \ldots, x_N)$ having the periodicity of a crystal lattice. Suppose that the ground state of the system is nondegenerate, except for spin degeneracy. Let N and the size of the crystal become infinite together, in such a way that the electron density and the local form of V remain constant. Then the hypothesis that the ground state is, in this thermodynamic limit, one of spin $S = \frac{1}{2}N$ necessarily entails that there exist excited states of spin $S-1$ and wave vector $\vec{q}$ (spin waves) whose energies $\hbar\omega_q$ obey $\hbar\omega_q/q^2 \to 0$ as $q \to 0$. Of Izuyama's subsequent papers, some have discussed the relation of the theorem to the linear response function for a certain fictitious electric field[6,20]; some have treated the analogous theorem for a system whose Hamiltonian is expressed in terms of a hopping matrix between Wannier functions plus site-occupation energies[3-7]; and some have undertaken to buttress the theorem with physical arguments which are presented as compelling even if the mathematical basis of the original proof is defective.[6,8,20]

A Heisenberg-coupled atomic array of the sort discussed in Sec. II—for which the orthodox theory of spin waves should apply—would seem to constitute a counterexample to this theorem. So an examination of the details of Izuyama's arguments is called for. As the starting point for our critique we shall take his expression[21] for the mean energy $\langle\hbar\omega_q\rangle$ of the low-energy components of the wave function obtained by operating on the ground-state wave function Ψ_0 of spin $S^z = \frac{1}{2}N$ with

$$M_-(q) = 2^{-1/2}\sum_i e^{i\vec{q}\cdot\vec{r}_i}(s_i^x - is_i^y) \quad , \tag{3}$$

where $\vec{r}_i$, $\vec{s}_i$ are the position and spin of the ith electron. It is assumed hypothetically that the ground state of the system has the spins of all N electrons aligned parallel. Then, as $q \to 0$, the mean energy of those components of $M_-(\vec{q})\Psi_0$ whose energies are of order q^2 or less is asymptotically

$$\langle\hbar\omega_q\rangle \sim \hbar^2q^2/2m - \langle\overline{\Psi}_0|\mathcal{g}(H-E_0)^{-1}\mathcal{g}|\overline{\Psi}_0\rangle \quad , \tag{4}$$

where H is the Hamiltonian, E_0 is the ground state energy, and

$$\mathcal{g} = \sum_i \frac{\hbar\vec{q}\cdot\vec{p}_i}{m}\left(\frac{1}{2} - s_i^z\right) \quad , \tag{5}$$

$\vec{p}_i$ being the momentum of electron i; $\overline{\Psi}_0$ is the ground-state wave function with $S^z = \frac{1}{2}N - 1$, obtained from Ψ_0 by application of the spin-lowering operator S^- and renormalizing. Though the derivation of this result in a form applicable to metals involves many subtleties and some additional postulates concerning the energy spectrum—which we shall not discuss here—the correctness of the expression (4) is quite obvious for the case of an insulating ferromagnet for which the spectrum of states with $S^z = \frac{1}{2}N - 1$ consists of a single spin-wave branch separated by a finite gap from all other states; it is just the second-order perturbation expression for the ground-state energy of the operator $U^{-1}HU$ in the subspace of states with $S^z = \frac{1}{2}N - 1$ and wave vector 0, where

$$U = \exp\left(i\sum_i \vec{q}\cdot\vec{r}_i(\tfrac{1}{2} - s_i^z)\right)$$

is a unitary operator taking a state with $S^z = \frac{1}{2}N - 1$ and wave vector 0 into one with wave vector $\vec{q}$. The analogy with the $\vec{k}\cdot\vec{p}$ perturbation procedure in the theory of one-electron energy bands[22] is obvious; we shall use this analogy from time to time in the critique to follow. Our remarks on the original proof[2,5] and subsequent plausibility arguments[6,8,20] in support of the theorem that (4) is nonpositive are several:

(i) In his original proof[2] and his Wannier-basis variant of it,[5] Izuyama eliminated the energy denominators from (4) by a procedure reminiscent of the Thomas-Kuhn-Reiche sum rule, writing the $\vec{p}_i/m$ in (5) as

$$\vec{p}_i/m = \dot{\vec{r}}_i = i^{-1}[H, \vec{r}_i] \quad . \tag{6}$$

Averaging the forms obtained by using this first in the left-hand $\mathcal{g}$ and then in the right-hand $\mathcal{g}$ of (4) gives a commutator of $\vec{r}_i$ and $\vec{p}_i$, whose value is independent of the detailed nature of Ψ_0. This procedure yields the universal value $\hbar^2q^2/2m$ for the scalar product in (4); so the right of (4) vanishes.

The fallacy that mars this portion of Izuyama's otherwise sound reasoning is that $\vec{r}_i$ is not a normal operator in crystal wave mechanics. In the commonest formulation of crystal wave mechanics, one uses periodic boundary conditions. Application of $\vec{r}_i$ to a state obeying these boundary conditions gives a state which does not obey them; one gets outside the Hilbert space for which $(H-E_0)^{-1}$ is defined. One can of course define a new operator $\vec{r}_i'$, resembling $\vec{r}_i$ but with the periodicity of the boundary conditions,[23] but if this operator is used instead of $\vec{r}_i$ in (6) an additional term must be added whose consequences in (4) are no longer independent of the nature of Ψ_0.

The nature of the fallacy becomes clearly evident if one tries to apply the same reasoning to the standard $\vec{k}\cdot\vec{p}$ perturbation expression for the effective mass of a single Bloch electron

$$\frac{\hbar^2k^2}{2m^*}=\frac{\hbar^2k^2}{2m}-\sum_{n\neq 0}\frac{|\hbar\vec{k}\cdot\vec{p}_{0n}/m|^2}{\epsilon_n-\epsilon_0}+O(k^4), \tag{7}$$

where ϵ_n is the energy of the state of wave vector 0 in the nth band, ϵ_0 being the ground-state energy, and $\vec{p}_{0n}$ is the matrix element of the momentum between states 0 and n. Use of (6) would give the universal value 0 for the right of (7), clearly a wrong result.

In an apparent response to a communication from Thouless[20] regarding the need to modify the Thomas-Kuhn-Reiche sum rule, Izuyama has, in his more recent papers, reformulated his arguments so as to avoid the error just described, although without elucidating its exact nature in as full detail as has been done above. After developing several intriguing and generally correct transformations of the expression (4) for the spin-wave stiffness, he has used these as a basis for several attempts to show from considerations of physical plausibility that the right of (4) must be either negative or zero.

(ii) In the first of these[6,20] the value of the ground-state expectation value in the second term on the right of (4) for a crystal with periodic boundary conditions—the case to which (4) applies—was compared with that for a finite crystal with boundaries. In the latter case the sum rule applies in the form originally used,[2] and the term in question has the value $\hbar^2q^2/2m$. In each case the term can be shown to equal the response $-\langle\mathcal{J}(t=0)\rangle/A_0$ resulting from application of a perturbation $A_0e^{\epsilon t}\mathcal{J}$, calculated in the framework of linear-response theory and then evaluated in the limit of slow turn-on, $\epsilon\to 0^+$. Here $\langle\mathcal{J}(t=0)\rangle$ is q times the momentum term in the down-spin particle current owing to a weak electric field acting only on down-spin electrons, applied for a long time, and in such sense as to accelerate electrons in the negative-$\vec{q}$ direction. This momentum term is, however, only one of the contributions to the total down-spin particle current, the other being the diamagnetic current, here equal, at time 0, to A_0/m. Thus Izuyama's physically plausible assertion that the particle current resulting from the perturbation is smaller for the finite crystal than for the one with periodic boundary conditions implies that the last term of (4) is larger for the former case than for the latter; his conclusion was the reverse of this, apparently because in hastily adding a note in proof he made a sign error and neglected the diamagnetic current.

(iii) The second argument[6,8] utilized an ingenious transformation by which both terms on the right of (4), for $\vec{q}$ in the x direction, were shown to add up to something proportional to

$$-\langle\bar{\Psi}_0|J_d(x)(H-E_0)^{-1}J_d(-x)|\bar{\Psi}_0\rangle q^2\ , \tag{8}$$

where $J_d(x)$ is the down-spin particle current *density* averaged over the plane $x=$ const., and it must be specified that $x\neq -x$ modulo the periodic boundary conditions. Remarkably enough, (8) can be shown to be independent of x (or the origin of x) for $x\neq -x$. However, Izuyama's preliminary conclusion that (8) is obviously negative for $x\neq -x$ was too hasty; the same reasoning can be carried through for the problem of the effective mass of a single Bloch electron, and the analog of (8) must normally come out positive here.

(iv) In a paper[24] that has appeared subsequently to the original submission of the present Comment, Izuyama has argued that the behavior of the quantity (8) in the magnon problem differs in an essential way from its behavior in the problem of a single Bloch electron. His approach, based on an extension of the linear-response theory used earlier,[6,20] was to express the quantity (8) in terms of the down-spin particle current density $\langle J_d(x)\rangle$ produced on a plane at position x, due to long-time action of a weak electric field E_d, supposed to act on down-spin particles only, and applied to the state $\bar{\Psi}_0$ over a region of space excluding this plane. (Though the region in which $E_d\neq 0$ may be arbitrary, it is convenient to think of it as including all the crystal except for a thin slab centered on the plane at x.) Noting that in the one-electron problem the analogous $\langle J(x)\rangle$ is demonstrably in the direction in which the field accelerates the electron, he argued that this only occurs because the electron can move through the crystal—thus changing its x coordinate—for an infinitely long time without losing the velocity imparted to it by the field. In the magnon case the sea of up-spin electrons is available to scatter the down-spin electron, at least if the crystal is metallic, and so it appears that the ratio of $\langle J_d(x)\rangle$ to $\int E_d\,dt$ will go to zero in the limit of very slow turn-on of E_d.

This inference is not valid, for a subtle reason. In relating (8) to the down-spin current density

produced by a slowly applied field, one first evaluates the linear-response expression for a finite rate of turn-on of the field and then lets the turn-on rate go to zero. The use of linear-response theory implies that one evaluates the limit of

$$\langle J_d(x)\rangle_{t=0}\Big/\int_{-\infty}^{0} E_d\,dt \tag{9}$$

as

$$\int_{-\infty}^{0} E_d\,dt \to 0 \ , \tag{10}$$

before passing to the limit of slow turn-on. Now the effect of E_d on the initial state Ψ_0, the state with one magnon of zero wave vector, is to transform it into a time-dependent state consisting principally of a one-magnon state Ψ_q with a wave vector $q(t)$ given by

$$\hbar dq/dt = -eE_d(t) \ . \tag{11}$$

[This statement is precise for an insulator if E_d is uniform and high-frequency transients can be neglected; for a metal, of course, there are no *exact* magnon eigenstates Ψ_q, but it is generally believed[25,26] there exist magnon-like states Ψ_q whose energy width goes to zero faster than q^2 as $q \to 0$. In such case the meaning of the statement preceding (11) is that in the limit as E_d and hence $q(t)$ become very small the state into which Ψ_0 evolves under E_d will approach such a $\Psi_{q(t)}$.] Thus in the limit (10) we need be concerned only with magnon states of infinitesimal q, and even in a metal the lifetime of such states with respect to scattering by quasiparticles will become infinite in this limit.[25] Since the limit (10) is to be taken before the limit of slow turn-on, the ratio (9) will remain finite even in the latter limit.

The impact of points (i)—(iv) is thus that despite the many ingenious forms into which Izuyama has transformed the expression for the spin-wave stiffness of a hypothetical saturated ferromagnet, there seems to be no reason why such ferromagnetic ground states with all spins aligned should not be in principle possible. Although the arguments of the present comment have referred only to the case of a Hamiltonian with the usual p^2 and $V(\vec{r}$'s) terms, several of Izuyama's papers have applied similar types of reasoning to the Hubbard model, and similar criticisms can be made. For the Hubbard model there are two pieces of suggestive evidence, namely, Nagaoka's[27] exact analysis of states with number of electrons one greater or one less than the number of atoms, and the results of various approximation schemes,[28] which encourage the speculation that in the limit of infinite same-site repulsions there is a finite range of values of the electron-to-atom ratio for which the ground state has all spins parallel. Although this inference has not yet been established rigorously, I would argue that considerations of the type discussed above do not constitute a threat to it.

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