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Ferromagnetic Spin Excitations in a Metallic Loop: The Case of Two Degenerate Bands

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We consider a system of $2N$ electrons in a periodic loop of N lattice sites. These electrons are restricted to occupy the subspace of two degenerate bands. The interaction within a band is limited to nearest-neighbor and on-site Wannier integrals, but is otherwise arbitrary. The interaction between bands contains the *complete* set of intra-atomic terms but no nearest-neighbor term. The spectra of one-spin-flip excitations relative to the saturated ferromagnetic eigenstate are calculated exactly. The results show that it is *necessary* to have the intraband nearest-neighbor exchange coupling in order that the lowest spin-wave branch lie above the saturated ferromagnetic eignestate. Conditions for the local stability of the saturated ferromagnetic eigenstate are given. It is shown that the saturated ferromagnetic eigenstate can still be stable as a result of the nearest-neighbor exchange interaction J, even when the individual atoms would be nonmagnetic when J is turned off.

I. INTRODUCTION

In a previous paper,[1] we considered a system of N interacting electrons in a periodic loop potential which has N lattice sites. A one-band Hamiltonian containing only nearest-neighbor interaction terms was considered, and the one-spin-flip spectra relative to a saturated ferromagnetic eigenstate (the SF state) were calculated exactly.

It was found that the characteristic terms contributing to the spin-wave spectrum are the band term, the "correlation" term ($\sim n\uparrow n\downarrow$), and the intersite exchange term. The local stability of the SF state results from a balance between the ferromagnetic tendency due to the direct intersite exchange term, and antiferromagnetic tendency from indirect effects stemming from the band and other interaction terms. In Ref. 1, the effects of the latter terms on the one-spin-flip spectra were for the first time calculated exactly for a one-band model with nearest-neighbor interactions. It would then be natural to ask how would the local stability of the SF state be changed by having a band degeneracy.

In an early paper, Slater, Statz, and Koster[2] considered the case of two electrons in a nondegenerate band and found that the lowest state is always a singlet state; they then considered two electrons in a doubly degenerate band and found it possible to have a triplet state being the lowest state. As a result, it was proposed that the intra-atomic Hund's rule coupling may be the key mechanism that is responsible for band ferromagnetism. Since then this point of view has been advocated by many authors,[3] and magnon spectra were calculated using various approximations.

In the present article we shall consider a system of $2N$ interacting electrons in a periodic loop potential which has N lattice sites. The Hamiltonian to be considered represents the case of two degenerate bands. It includes the band term, the correlation term, the nearest-neighbor exchange interaction term within a band, and the *complete* interband intra-atomic terms.[4] The explicit form of the Hamiltonian is as follows:

$$H=H_1+H_2+H_{12}, \tag{1.1}$$

where

$$\begin{aligned}H_\lambda=V\sum_{i=1}^{N}\sum_{\sigma}&(C_{i,\sigma}{}^{(\lambda)\dagger}C_{i+1,\sigma}{}^{(\lambda)}\\&+C_{i,\sigma}{}^{(\lambda)\dagger}C_{i-1,\sigma}{}^{(\lambda)})+U\sum_{i=1}^{N}n_{i,+}{}^{(\lambda)}n_{i,-}{}^{(\lambda)}\\&-J\sum_{i=1}^{N}(C_{i,+}{}^{(\lambda)\dagger}C_{i,-}{}^{(\lambda)}C_{i+1,-}{}^{(\lambda)\dagger}C_{i+1,+}{}^{(\lambda)}\\&+C_{i,-}{}^{(\lambda)\dagger}C_{i,+}{}^{(\lambda)}C_{i+1,+}{}^{(\lambda)\dagger}C_{i+1,-}{}^{(\lambda)})\\&-\tfrac{1}{2}J\sum_{i=1}^{N}(n_{i,+}{}^{(\lambda)}-n_{i,-}{}^{(\lambda)})(n_{i+1,+}{}^{(\lambda)}-n_{i+1,-}{}^{(\lambda)}),\quad\lambda=1,2\end{aligned} \tag{1.2}$$

$$\begin{aligned}H_{12}=U_{12}\sum_{i=1}^{N}\sum_{\sigma,\sigma'}&n_{i,\sigma}{}^{(1)}n_{i,\sigma}{}^{(2)}\\&-J_{12}\sum_{i=1}^{N}\sum_{\sigma,\sigma'}C_{i,\sigma}{}^{(1)\dagger}C_{i,\sigma'}{}^{(1)}C_{i,\sigma'}{}^{(2)\dagger}C_{i,\sigma}{}^{(2)}\\&+D_{12}\sum_{i=1}^{N}(C_{i,+}{}^{(1)\dagger}C_{i,-}{}^{(1)\dagger}C_{i,-}{}^{(2)}C_{i,+}{}^{(2)}\\&+C_{i,+}{}^{(2)\dagger}C_{i,-}{}^{(2)\dagger}C_{i,-}{}^{(1)}C_{i,+}{}^{(1)})\\&+W\sum_{i=1}^{N}\sum_{\sigma}(n_{i,\sigma}{}^{(1)}+n_{i,\sigma}{}^{(2)})\\&\times(C_{i,-\sigma}{}^{(1)\dagger}C_{i,-\sigma}{}^{(2)}+C_{i,-\sigma}{}^{(2)\dagger}C_{i,-\sigma}{}^{(1)}),\end{aligned} \tag{1.3}$$

where $C_{i,\sigma}{}^{(\lambda)\dagger}$, $C_{i,\sigma}{}^{(\lambda)}$, and $n_{i,\sigma}{}^{(\lambda)}$ are the creation, annihilation, and number operators of the ith Wannier orbital with band index λ and spin index σ, $\sigma=+,-$. The intraband coefficients V, J, U are taken for simplicity to have the same value for each band.[5]

The degeneracy of the band makes it possible to have four types of one-spin-flip states relative to a saturated ferromagnetic eigenstate which has total and z-component spin quantum number $S=M=N$, and is a respectable candidate for the ground state.

The spectra of all four types of one-spin-flip excitations are calculated exactly. We will first treat only the band term, the correlation term, and the "essential" part of the interband intra-atomic terms, i.e., the U_{12}, J_{12}, and D_{12} terms; these will later be referred to as the Hund's rule terms. The complete Hamiltonian in Eq. (1.1) will then be treated.

There are two central results of the calculation. First, without intraband (nearest-neighbor) exchange coupling, the saturated ferromagnetic eigenstate could never be stable. Second, it is possible for the SF state to be, at least, locally stable as a result of the mutual polarization among atoms through the nearest-neighbor exchange interaction J, even when the individual atoms would be nonmagnetic when J is turned off.[6]

If there is no coupling between the bands, there would be two degenerate spin-wave states plus pairs of degenerate exciton states plus four degenerate excitation bands. When the interband interaction is introduced, the following happens: (1) The pair of degenerate spin waves split into two distinct branches separated by order of magnitude $2J_{12}$; (2) two new exciton states appear, separated also by order of $2J_{12}$, which split off from the continuum; (3) the continua of excited states split into two, separated by order of $4W$; (4) the old excitons are also split by order of $4W$. We shall be interested in this paper in the first three of these effects.

The two spin waves and two new excitons states are very similar to the four states found for one case of two electrons occupying two degenerate levels. For this reason, we start the discussion with a brief description of the two-electron case.

II. TWO-ELECTRON CASE

Let us consider the case of two electrons which are to occupy two degenerate single-particle states Φ and Ψ, i.e., consider the Hamiltonian

$$\hat{H}=\sum_{i=1}^{2}\hat{h}(x_i,p_i)+V(|x_1-x_2|), \tag{2.1}$$

where $\hat{h}$ is the single-particle Hamiltonian, and $V(x_1-x_2)$ is the interaction between the electrons, and

$$\hat{h}\Phi=\epsilon_0\Phi, \qquad \hat{h}\Psi=\epsilon_0\Psi. \tag{2.2}$$

Taking spin and Pauli's principle into account, there are six states to be considered. These states are

$$\begin{aligned}
\Phi^{(\mathrm{up})}&=\tfrac{1}{2}\sqrt{2}[\Phi(x_1)\Psi(x_2)-\Psi(x_1)\Phi(x_2)](+,+),\\
\Phi^{(\mathrm{down})}&=\tfrac{1}{2}\sqrt{2}[\Phi(x_1)\Psi(x_2)-\Psi(x_1)\Phi(x_2)](-,-)\\
\Phi^{(1)}&=\tfrac{1}{2}\sqrt{2}[\Phi(x_1)\Psi(x_2)-\Psi(x_1)\Phi(x_2)]\tfrac{1}{2}\sqrt{2}[(+,-)+(-,+)],\\
\Phi^{(2)}&=\tfrac{1}{2}\sqrt{2}[\Phi(x_1)\Psi(x_2)+\Psi(x_1)\Phi(x_2)]\tfrac{1}{2}\sqrt{2}[(+,-)-(-,+)],\\
\Phi^{(3)}&=\Phi(x_1)\Phi(x_2)\tfrac{1}{2}\sqrt{2}[(+,-)-(-,+)],\\
\Phi^{(4)}&=\Psi(x_1)\Psi(x_2)\tfrac{1}{2}\sqrt{2}[(+,-)-(-,+)],
\end{aligned} \tag{2.3}$$

where $(+,-),\cdots$, etc. represent the spin functions.

The parallel spin states, $\Phi^{(\mathrm{up})}$ and $\Phi^{(\mathrm{down})}$, have total spin quantum number $S=1$. The other four states will be called one-spin-flip states.

Let us solve the energy eigenvalue problem in the subspace spanned by these six states. The matrix elements are calculated to be

$$(H_{ij}-E\delta_{ij})=\begin{bmatrix}
E_0-E & 0 & 0 & 0 & 0 & 0\\
0 & E_0-E & 0 & 0 & 0 & 0\\
0 & 0 & E_0-E & 0 & 0 & 0\\
0 & 0 & 0 & 2J_{12}+E_0-E & W & W\\
0 & 0 & 0 & W & (U+J_{12}-U_{12}+E_0-E) & D_{12}\\
0 & 0 & 0 & W & D_{12} & (U-U_{12}+J_{12}+E_0-E)
\end{bmatrix}, \tag{2.4}$$

where E is the energy eigenvalue, and

$$H_{ij}\equiv\langle\Phi^{(i)}\,|\,\hat{H}\,|\,\Phi^{(j)}\rangle, \qquad i,j=\text{up, down, }1,2,3,4 \tag{2.5}$$

$$E_0\equiv U_{12}-J_{12}+2\epsilon_0, \tag{2.6}$$

and

$$\begin{aligned} U_{12} &\equiv \int dxdy\, \Phi^*(x)\Psi^*(y)\Psi(y)\Phi(x)\, V(x-y), \\ J_{12} &\equiv \int dxdy\, \Phi^*(x)\Psi^*(y)\Phi(y)\Psi(x)\, V(x-y), \\ D_{12} &\equiv \int dxdy\, \Phi^*(x)\Phi^*(y)\Psi(y)\Psi(x)\, V(x-y), \\ U &\equiv \int dxdy\, \Phi^*(x)\Phi^*(y)\Phi(y)\Phi(x)\, V(x-y), \\ W &\equiv \int dxdy\, \Phi^*(x)\Psi^*(y)\Psi(y)\Psi(x)\, V(x-y). \end{aligned} \tag{2.7}$$

In Eq. (2.4) we have assumed, for mathematical simplicity, that Φ and Ψ are real functions, and that

$$\int dxdy\, \Phi^2(x)\Phi^2(y)\, V(x-y) = \int dxdy\, \Psi^2(x)\Psi^2(y)\, V(x-y) \tag{2.8}$$

and

$$\int dxdy\, \Phi(x)\Psi(x)\Psi^2(y)\, V(x-y) = \int dxdy\, \Phi(x)\Psi(x)\Phi^2(y)\, V(x-y). \tag{2.9}$$

Also, using the fact that Φ and Ψ are real functions, we have

$$J_{12} = D_{12}. \tag{2.10}$$

It is clear that the parallel spin states, $\Phi^{(\mathrm{up})}$ and $\Phi^{(\mathrm{down})}$, are themselves energy eigenstates with energy $E^{(\mathrm{up})} = E^{(\mathrm{down})} = E_0$. The one-spin-flip state with $S=1$, the $\Phi^{(1)}$ state, is also an energy eigenstate with energy

$$E_S^{(1)} = E_0. \tag{2.11}$$

The problem now is to solve the 3×3 determinantal equation for the energy eigenvalues of the one-spin-flip states with $S=0$,

$$\begin{vmatrix} 2J_{12}+E_0-E & W & W \\ W & (U-U_{12}+J_{12})+E_0-E & D_{12} \\ W & D_{12} & (U-U_{12}+J_{12})+E_0-E \end{vmatrix} = 0. \tag{2.12}$$

The solutions are, using the fact that $J_{12}=D_{12}$,

$$E_S^{(2)} - E_0 = 2J_{12} + \tfrac{1}{2}\{(U-U_{12}) - [(U-U_{12})^2 + 16W^2]^{1/2}\}, \tag{2.13}$$

$$E_i^{(2)} - E_0 = 2J_{12} + \tfrac{1}{2}\{(U-U_{12}) + [(U-U_{12})^2 + 16W^2]^{1/2}\}, \tag{2.14}$$

$$E_i^{(1)} - E_0 = U - U_{12}. \tag{2.15}$$

The superscripts and subscripts are used to make correspondence with calculations of later sections. In general, $J_{12}>0$, $U-U_{12}>0$; in order that the ferromagnetic ($S=1$) states are the lowest state, the following condition must be satisfied:

$$E_S^{(2)} - E_0 \geq 0$$

or

$$J_{12}^2 + \tfrac{1}{2}J_{12}(U-U_{12}) \geq W^2. \tag{2.16}$$

It is clear that the W term is a source of antiferromagnetic effects, and when $W=0$, the ferromagnetic ($S=1$) states are the lowest states. This is a clear manifestation of the Hund's rule mechanism.

Thus we see that in the two-electron problem (with two degenerate states) there are four distinct excitations relative to the parallel spin states. We will later see that in the band problem a similar separation of electron-hole bound states into four branches occurs; the central bound states of each branch closely correspond to the states found here for two electrons.

III. HUND'S RULE COUPLING WITHOUT NEAREST-NEIGHBOR EXCHANGE

In this section we shall study the effect of the Hund's rule terms versus the band term and the correlation term by calculating the spectra of the one-spin-flip excitations (see Fig. 1). The nature of eigenfunctions will also be considered in some detail. The Hamiltonian $\tilde{H}$ to be used contains the V and U terms of Eq. (1.2), and the U_{12}, J_{12}, and D_{12} terms of Eq. (1.3), and nothing else. (The J and W terms are omitted.)

The method used here is identical to those used in Ref. 1, but we shall go through some of the details to show how degeneracy effects the calculation.

Let $|\Phi_0\rangle$ be the state with all Wannier sites of both bands occupied by down-spin electrons and with no up-spin electron. Then $|\Phi_0\rangle$ is an eigenstate of the Hamiltonian $\tilde{H}$ with eigenvalue

$$\tilde{E}_0 \equiv \langle\Phi_0|\tilde{H}|\Phi_0\rangle = N(U_{12}-J_{12}). \tag{3.1}$$

The state $|\Phi_0\rangle$ will later be referred to as the saturated ferromagnetic eigenstate.

Consider the following four types of one-spin-flip states:

$$\begin{aligned}\Psi_{f,g}^{(1)}&=C_{f,+}^{(1)\dagger}C_{g,-}^{(1)}\mid\Phi_0\rangle,\\ \Psi_{f,g}^{(2)}&=C_{f,+}^{(2)\dagger}C_{g,-}^{(2)}\mid\Phi_0\rangle,\\ \Psi_{f,g}^{(3)}&=C_{f,+}^{(1)\dagger}C_{g,-}^{(2)}\mid\Phi_0\rangle,\\ \Psi_{f,g}^{(4)}&=C_{f,+}^{(2)\dagger}C_{g,-}^{(1)}\mid\Phi_0\rangle,\end{aligned}\tag{3.2}$$

where f, $g=1, 2,\cdots, N$. It can be easily shown that the linear combinations of the sets of functions $\{\Psi_{f,g}^{(1)},\Psi_{f,g}^{(2)}\}$ and $\{\Psi_{f,g}^{(3)},\Psi_{f,g}^{(4)}\}$ are by themselves closed under the operation of $\tilde{H}$, so one can solve the energy eigenvalue problem spanned by these two sets of functions separately.

A. Subspace Spanned by $\{\Psi_{f,g}^{(1)},\ \Psi_{f,g}^{(2)}\}$

Let us first solve the eigenvalue problem in the subspace spanned by $\{\Psi_{f,g}^{(1)},\Psi_{f,g}^{(2)}\}$, i.e., consider the following equations:

$$(\tilde{H}-E)\Psi^{(12)}=0,\tag{3.3}$$

where

$$\Psi^{(12)}=\sum_{f,g=1}^{N}\left(A_{f,g}^{(1)}\Psi_{f,g}^{(1)}+A_{f,g}^{(2)}\Psi_{f,g}^{(2)}\right).\tag{3.4}$$

The set of constants $\{A_{f,g}^{(1)},\ A_{f,g}^{(2)}\}$ to be determined satisfy the periodic boundary conditions

$$A_{f+N,g}^{(\lambda)}=A_{f,g}^{(\lambda)},\qquad A_{f,g+N}^{(\lambda)}=A_{f,g}^{(\lambda)},\tag{3.5}$$

where $\lambda=1$, 2, and f, $g=1, 2,\cdots, N$.

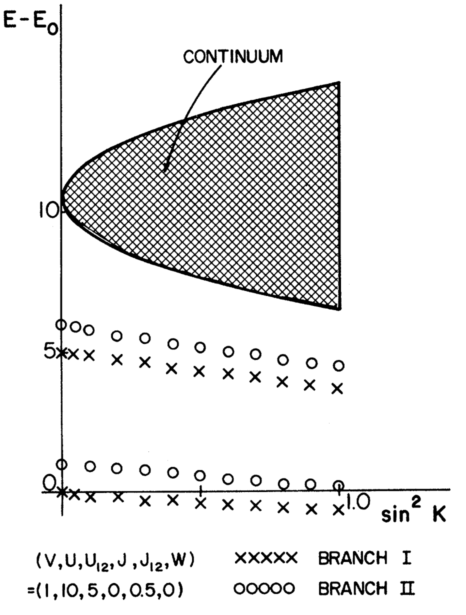

FIG. 1. Bound-state spectra of the system when there is Hund's rule coupling but no nearest-neighbor exchange coupling. There are two spin-wave spectra, separated by order of $2J_{12}$, and two interband exciton spectra, also separated by order of $2J_{12}$. The spin-wave spectra are bending downward, and the ferromagnetic eigenstate is unstable.

To solve the eigenvalue problem, we first calculate $\tilde{H}\Psi_{f,g}^{(1)}$ and $\tilde{H}\Psi_{f,g}^{(2)}$ then substitute the results into Eq. (3.3). Shifting indices under the summation sign and noticing that all the vectors in $\{\Psi_{f,g}^{(1)},\Psi_{f,g}^{(2)}\}$ are linearly independent, we get the following set of equations:

$$V[A_{f+1,g}^{(1)}+A_{f-1,g}^{(1)}-A_{f,g+1}^{(1)}-A_{f,g-1}^{(1)}]\\ -U\delta_{f,g}A_{f,g}^{(1)}-J_{12}\delta_{f,g}A_{f,g}^{(2)}=\tilde{E}'A_{f,g}^{(1)},\tag{3.6}$$

$$V[A_{f+1,g}^{(2)}+A_{f-1,g}^{(2)}-A_{f,g+1}^{(2)}-A_{f,g-1}^{(2)}]\\ -U\delta_{f,g}A_{f,g}^{(2)}-J_{12}\delta_{f,g}A_{f,g}^{(1)}=\tilde{E}'A_{f,g}^{(2)},\tag{3.7}$$

where

$$\tilde{E}'=E-\tilde{E}_0-(U+J_{12}).\tag{3.8}$$

In Eqs. (3.6) and (3.7), both indices f and g run from 1 through N. There are altogether $2N^2$ such equations. To solve these equations, try solutions of the form

$$A_{f,g}^{(\lambda)}=\exp[iK(f+g)]B_{f-g}^{(\lambda)}(K),\qquad\lambda=1,2\tag{3.9}$$

where

$$K=n\pi/N,\qquad n=1,2,\cdots,N.\tag{3.10}$$

Putting Eq. (3.9) into Eqs. (3.6) and (3.7) and simplifying, we get

$$V[e^{iK}B_{r+1}^{(1)}+e^{-iK}B_{r-1}^{(1)}-e^{-iK}B_{r-1}^{(1)}-e^{iK}B_{r+1}^{(1)}]\\ -\eta\delta_{r,0}B_r^{(1)}-J_{12}\delta_{r,0}B_r^{(2)}=\tilde{E}'B_r^{(1)},\tag{3.11}$$

$$V[e^{iK}B_{r+1}^{(2)}+e^{-iK}B_{r-1}^{(2)}-e^{-iK}B_{r-1}^{(2)}-e^{iK}B_{r+1}^{(2)}]\\ -\eta\delta_{r,0}B_r^{(2)}-J_{12}\delta_{r,0}B_r^{(1)}=\tilde{E}'B_r^{(2)},\tag{3.12}$$

where

$$r\equiv f-g,\qquad r=1,2,\cdots,N\tag{3.13}$$

$$\eta\equiv U+2J\cos 2K.\tag{3.14}$$

To solve Eqs. (3.11) and (3.12), let us define the Fourier transform of $B_r^{(1)}$ and $B_r^{(2)}$:

$$b_k^{(\lambda)}=\sum_{r=1}^{N}e^{ikr}B_r^{(\lambda)},\qquad\lambda=1,2\tag{3.15}$$

or

$$B_r^{(\lambda)}=N^{-1}\sum_k e^{-ikr}b_k^{(\lambda)},\qquad\lambda=1,2\tag{3.16}$$

where the set of k values depends on the value of K in such a way that[1]

$$k=\begin{cases}2m\pi/N\\ \\ (2m+1)\pi/N\end{cases}\quad m=1,2,\cdots,N,\\ \text{if}\quad K=\begin{cases}n\pi/N & n\text{ even}\\ \\ n\pi/N & n\text{ odd.}\end{cases}\tag{3.17}$$

Multiply Eqs. (3.11) and (3.12) by $\exp(ik_1r)$ and $\exp(ik_2r)$, respectively, and sum over r; we then have

$$b_{k_1}^{(1)}=-(UB_0^{(1)}+J_{12}B_0^{(2)})/(\tilde{E}'-E_{k_1}),\tag{3.18}$$

$$b_{k_2}^{(2)}=-(UB_0^{(2)}+J_{12}B_0^{(1)})/(\tilde{E}'-E_{k_2}),\tag{3.19}$$

where

$$E_k \equiv 4V \sin K \sin k. \tag{3.20}$$

Substitute Eqs. (3.18) and (3.19) back into Eq. (3.16), and let $r=0$; we have

$$B_0^{(1)} = -(UB_0^{(1)} + J_{12}B_0^{(2)})\tilde{u}, \tag{3.21}$$

$$B_0^{(2)} = -(UB_0^{(2)} + J_{12}B_0^{(1)})\tilde{u}, \tag{3.22}$$

where

$$\tilde{u} \equiv N^{-1} \sum_k [1/(\tilde{E}' - E_k)]. \tag{3.23}$$

Equations (3.21) and (3.22) have nontrivial solutions if and only if

$$\Delta = \begin{vmatrix} 1+U\tilde{u} & J_{12}\tilde{u} \\ J_{12}\tilde{u} & 1+U\tilde{u} \end{vmatrix} = 0. \tag{3.24}$$

The determinantal equation, Eq. (3.24), can be factorized into

$$\Delta = \tilde{F}_1^{(s)} \tilde{F}_2^{(s)} = 0, \tag{3.25}$$

where

$$\tilde{F}_1^{(s)} = 1 + (U + J_{12})\tilde{u} = 0, \tag{3.26}$$

with

$$B_0^{(1)} = B_0^{(2)} \tag{3.27}$$

representing the branch $\mathrm{I}^{(s)}$ solutions, and

$$\tilde{F}_2^{(s)} = 1 + (U - J_{12})\tilde{u} = 0, \tag{3.28}$$

with

$$B_0^{(1)} = -B_0^{(2)} \tag{3.29}$$

representing the branch $\mathrm{II}^{(s)}$ solutions. The superscript s stands for spin wave; it will later be shown that the bound states of these two branches can be identified as spin waves.

In each branch, for a given K, there is a set of continuum states, as indicated by the energy denominator, and a bound state lying below the continuum.[1] Here we are only interested in the energy spectra and total spin of these bound states.

To find the energy spectra of the bound states, we can replace the sum over k by an integral using the correspondence

$$N^{-1} \sum_k \rightarrow \frac{1}{2\pi} \int_{-\pi}^{\pi} dk.$$

For branch $\mathrm{I}^{(s)}$, the energy spectrum and total spin of the bound states are found to be

$$\tilde{E}_s^{(1)} - \tilde{E}_0 = U + J_{12} - [(U + J_{12})^2 + 16V^2 \sin^2 K]^{1/2}, \tag{3.30}$$

$$\begin{aligned} \hat{S}^2 \Psi_s^{(1)}(K) &= N(N-1)\Psi_s^{(1)}(K) \quad \text{if} \quad K \neq 0 \\ &= N(N+1)\Psi_s^{(1)}(K) \quad \text{if} \quad K = 0, \end{aligned} \tag{3.31}$$

where $\Psi_s^{(1)}(K)$ is the eigenfunction corresponding to the branch $\mathrm{I}^{(s)}$ bound state with wave vector K. A similar notation will be used for other branches. It should be noticed that for this branch the bound-state spectrum always lies below $\tilde{E}_0$ except for the $K=0$ state which has total spin N instead of $(N-1)$.

For branch $\mathrm{II}^{(s)}$, the energy spectrum and total spin of the bound states are found to be

$$\tilde{E}_s^{(2)} - \tilde{E}_0 = U + J_{12} - [(U - J_{12})^2 + 16V^2 \sin^2 K]^{1/2}, \tag{3.32}$$

$$\hat{S}^2 \Psi_s^{(2)}(K) = N(N-1)\Psi_s^{(2)}(K). \tag{3.33}$$

B. Subspace Spanned by $\{\Psi_{f,g}^{(3)}, \Psi_{f,g}^{(4)}\}$

The eigenvalue problem in the subspace spanned by $\{\Psi_{f,g}^{(3)}, \Psi_{f,g}^{(4)}\}$ can be solved using exactly the same procedure as that in Sec. III A. The solutions can again be divided into two branches, branch $\mathrm{I}^{(i)}$ and branch $\mathrm{II}^{(i)}$. The superscript i stands for interband, and the bound states are called interband excitons. The reason for this nomenclature is that these bound states occur only when the interband interaction is put on.

In the following we will just write down the energy spectra and the total spin of these excitons:

For branch $\mathrm{I}^{(i)}$,

$$\tilde{E}_i^{(1)} - \tilde{E}_0 = U + J_{12} - [(U_{12} + D_{12})^2 + 16V^2 \sin^2 K]^{1/2}, \tag{3.34}$$

$$\hat{S}^2 \Psi_i^{(1)}(K) = N(N-1)\Psi_i^{(1)}(K). \tag{3.35}$$

For branch $\mathrm{II}^{(i)}$,

$$\tilde{E}_i^{(2)} - \tilde{E}_0 = U + J_{12} - [(U_{12} - D_{12})^2 + 16V^2 \sin^2 K]^{1/2}, \tag{3.36}$$

$$\hat{S}^2 \Psi_i^{(2)} = N(N-1)\Psi_i^{(2)}(K). \tag{3.37}$$

C. Summary

We have thus far calculated the energy spectra of all four types of bound states using the Hamiltonian $\tilde{H}$. We can make the following observations:

(1) The lowest bound-state spectrum, that of branch $\mathrm{I}^{(s)}$, always lies below $\tilde{E}_0$, except for the $K=0$ state which has $E=\tilde{E}_0$, and can be shown to have total spin N instead of $(N-1)$. This shows that the system cannot be ferromagnetic (see Fig. 1).

(2) All four spectra are bending downward as $\sin^2 K$ increases; moreover, the curvatures increase as V increases, indicating that the effect of the kinetic-energy part is against ferromagnetism.

(3) If we identify the coefficients U, U_{12}, J_{12}, D_{12} with those defined in Sec. II, setting $W=0$, then there is a correspondence between the energies of the central bound states ($K=0$ states) and the energy levels of the one-spin-flip states in Eq. (2.11), and Eqs. (2.13)–(2.15); the corresponding members carry the same indices. Furthermore, the total spin of the ferromagnetic eigenstate $|\Phi_0\rangle$ is the same as that of the $\Psi_s^{(1)}(0)$ state, and is one less than that of the other central bound states. This makes the correspondence more vivid.

IV. COMPLETE INTERBAND INTRA-ATOMIC INTERACTION WITH NEAREST-NEIGHBOR EXCHANGE COUPLING

In this section we shall consider the complete Hamiltonian given in Eq. (1.1). The saturated ferromagnetic eigenstate $|\,\Phi_0\rangle$ is again an energy eigenstate with energy

$$E_0=\langle\Phi_0\,|\,H\,|\,\Phi_0\rangle=N(U_{12}-J_{12}-J). \tag{4.1}$$

With the addition of the W term, all four types of one-spin-flip states $\{\Psi_{f,g}{}^{(1)},\Psi_{f,g}{}^{(2)},\Psi_{f,g}{}^{(3)},\Psi_{f,g}{}^{(4)}\}$ are mixed. Following a procedure similar to those used in Sec. III, for a given K, one will arrive at an 8×8 determinantal equation as follows:

$$G=\begin{vmatrix}
1+\frac{1}{2}Jf^{(+)} & \frac{1}{2}Jh^{(+)} & \eta g^{(+)}+Wg^{(-)} & Wg^{(+)}+U_{12}g^{(-)} & Wg^{(+)}+D_{12}g^{(-)} & J_{12}g^{(+)}+Wg^{(-)} & 0 & 0\\
\frac{1}{2}Jh^{(+)} & 1+\frac{1}{2}Jf^{(+)} & -\eta g^{(+)}-Wg^{(-)} & -Wg^{(+)}-U_{12}g^{(-)} & -Wg^{(+)}-D_{12}g^{(-)} & -J_{12}g^{(+)}-Wg^{(-)} & 0 & 0\\
-\frac{1}{2}Jg^{(+)} & \frac{1}{2}Jg^{(+)} & 1+\eta f^{(+)}+Wf^{(-)} & Wf^{(+)}+U_{12}f^{(-)} & Wf^{(+)}+D_{12}f^{(-)} & J_{12}f^{(+)}+Wf^{(-)} & 0 & 0\\
-\frac{1}{2}Jg^{(-)} & \frac{1}{2}Jg^{(-)} & Wf^{(+)}+\eta f^{(-)} & 1+U_{12}f^{(+)}+Wf^{(-)} & D_{12}f^{(+)}+Wf^{(-)} & Wf^{(+)}+J_{12}f^{(-)} & 0 & 0\\
0 & 0 & Wf^{(+)}+J_{12}f^{(-)} & D_{12}f^{(+)}+Wf^{(-)} & 1+U_{12}f^{(+)}+Wf^{(-)} & Wf^{(+)}+\eta f^{(-)} & \frac{1}{2}Jg^{(-)} & -\frac{1}{2}Jg^{(-)}\\
0 & 0 & J_{12}f^{(+)}+Wf^{(-)} & Wf^{(+)}+D_{12}f^{(-)} & Wf^{(+)}+U_{12}f^{(-)} & 1+\eta f^{(+)}+Wf^{(-)} & \frac{1}{2}Jg^{(+)} & -\frac{1}{2}Jg^{(+)}\\
0 & 0 & -J_{12}g^{(+)}-Wg^{(-)} & -Wg^{(+)}-D_{12}g^{(-)} & -Wg^{(+)}-U_{12}g^{(-)} & -\eta g^{(+)}-Wg^{(-)} & 1+\frac{1}{2}Jf^{(+)} & \frac{1}{2}Jh^{(+)}\\
0 & 0 & J_{12}g^{(+)}+Wg^{(-)} & Wg^{(+)}+D_{12}g^{(-)} & Wg^{(+)}+U_{12}g^{(-)} & \eta g^{(+)}+Wg^{(-)} & \frac{1}{2}Jh^{(+)} & 1+\frac{1}{2}Jf^{(+)}
\end{vmatrix}=0, \tag{4.2}$$

where

$$\begin{aligned}
f^{(\pm)}&\equiv(1/2N)\sum_k\left[1/(E'-E_k-2W)\pm1/(E'-E_k+2W)\right],\\
g^{(\pm)}&\equiv(1/2N)\sum_k\left[e^{-ik}/(E'-E_k-2W)\pm k^{-ik}/(E'-E_k+2W)\right],\\
h^{(\pm)}&\equiv(1/2N)\sum_k\left[e^{-2ik}/(E'-E_k-2W)\pm e^{-2ik}/(E'-E_k+2W)\right],
\end{aligned} \tag{4.3}$$

and where

$$E'\equiv E-E_0-(U+2J+J_{12}). \tag{4.4}$$

The determinantal function G in Eq. (4.2) can be factorized into

$$G=F_AF_1F_2=0, \tag{4.5}$$

where

$$F_A=[1+\tfrac{1}{2}J(f^{(+)}+h^{(+)})]^2=0, \tag{4.6}$$

with

$$\begin{gathered}
B_1{}^{(1)}=B_{-1}{}^{(1)},\qquad B_1{}^{(2)}=B_{-1}{}^{(2)},\\
B_0{}^{(1)}=B_0{}^{(2)}=B_0{}^{(3)}=B_0{}^{(4)}=0
\end{gathered} \tag{4.7}$$

representing the branch A solution, and

$$F_1=\begin{vmatrix}
1+\frac{1}{2}J(f^{(+)}-h^{(+)}) & (\eta+J_{12})g^{(+)}+2Wg^{(-)} & 2Wg^{(+)}+(U_{12}+D_{12})g^{(-)}\\
-Jg^{(+)} & 1+(\eta+J_{12})f^{(+)}+2Wf^{(-)} & 2Wf^{(+)}+(U_{12}+D_{12})f^{(-)}\\
-Jg^{(-)} & 2Wf^{(+)}+(\eta+J_{12})f^{(-)} & 1+(U_{12}+D_{12})f^{(+)}+2Wf^{(-)}
\end{vmatrix}=0, \tag{4.8}$$

with

$$\begin{gathered}
B_1{}^{(1)}=-B_{-1}{}^{(1)},\qquad B_1{}^{(2)}=-B_{-1}{}^{(2)},\\
B_0{}^{(3)}=B_0{}^{(4)},\qquad B_i{}^{(1)}=B_i{}^{(2)},\qquad i=0,\pm1
\end{gathered} \tag{4.9}$$

representing the branch I solutions, and

$$F_2=\begin{vmatrix}
1+\frac{1}{2}J(f^{(+)}-h^{(+)}) & (\eta-J_{12})g^{(+)} & (U_{12}-D_{12})g^{(-)}\\
-Jg^{(+)} & 1+(\eta-J_{12})f^{(+)} & (U_{12}-D_{12})f^{(-)}\\
-Jg^{(-)} & (\eta-J_{12})f^{(-)} & 1+(U_{12}-D_{12})f^{(+)}
\end{vmatrix}=0, \tag{4.10}$$

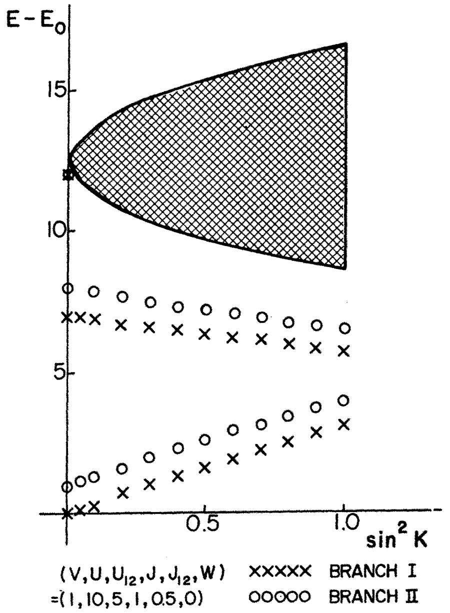

FIG. 2. Demonstration that when the nearest-neighbor exchange coupling is introduced, with the other terms kept the same as in the case of Fig. 1, the spin-wave spectra are turned upward. In particular, since $W=0$, Eq. (2.16) is automatically satisfied, which means that the individual atoms would be magnetic by themselves. The "old" (one-band) excitons appear just below the band at small K, and quickly merge into the band.

with

$$B_1^{(1)}=-B_{-1}^{(1)}, \qquad B_1^{(2)}=-B_{-1}^{(2)},$$

$$B_0^{(3)}=-B_0^{(4)}, \qquad B_i^{(1)}=-B_i^{(2)}, \qquad i=0, \pm 1 \quad (4.11)$$

representing the branch II solutions.

A. The Case $W=0$

It can be easily seen that when $W=0$,

$$f^{(+)}(W=0)=N^{-1}\sum_k (E'-E_k)^{-1}\equiv u,$$

$$g^{(+)}(W=0)=N^{-1}\sum_k (E'-E_k)^{-1}e^{-ik}\equiv v, \qquad (4.12)$$

$$h^{(+)}(W=0)=N^{-1}\sum_k (E'-E_k)^{-1}e^{-2ik}\equiv w,$$

while $f^{(-)}$, $g^{(-)}$, $h^{(-)}$ all vanish. Equation (4.6) then becomes

$$F_A=[1+\tfrac{1}{2}J(u+w)]^2=0, \qquad (4.13)$$

and Eqs. (4.8) and (4.10) can be further factorized into

$$F_1=F_1^{(s)}F_1^{(i)}=0, \qquad (4.14)$$

where

$$F_1^{(s)}=1+(\eta+J_{12})u+\tfrac{1}{2}J(u-w)$$
$$+\tfrac{1}{2}J(\eta+J_{12})(u^2-uw+2v^2)=0, \qquad (4.15)$$

$$F_1^{(i)}=1+(U_{12}+D_{12})u=0, \qquad (4.16)$$

and

$$F_2=F_2^{(s)}F_2^{(i)}, \qquad (4.17)$$

where

$$F_2^{(s)}=1+(\eta-J_{12})u+\tfrac{1}{2}J(u-w)$$
$$+\tfrac{1}{2}J(\eta-J_{12})(u^2-uw+2v^2)=0, \qquad (4.18)$$

$$F_2^{(i)}=1+(U_{12}-D_{12})u=0. \qquad (4.19)$$

These equations are, with the addition of the nearest-neighbor exchange terms ($\sim J$), the modified version of solutions of the branches I$^{(s)}$, I$^{(i)}$, II$^{(s)}$, II$^{(i)}$ in Sec. III. The corresponding members carry the same indices.

The branch A solutions here correspond to the branch A solutions in Ref. 1, and Eqs. (4.15) and (4.18) correspond to the branch B solutions in Ref. 1; there the solutions of these equations have been investigated in detail. We will just list the spectra of the spin-wave states in which we are mainly interested. Under the conditions $U\gg V$, J, J_{12}, the spin-wave spectra may be approximated by

$$E_s^{(1)}-E_0\simeq 4J\sin^2K$$
$$-[8V^2\sin^2K/(U+J_{12}+2J\cos 2K-\tfrac{1}{2}J)], \quad \text{branch I} \qquad (4.20)$$

$$E_s^{(2)}-E_0\simeq 2J_{12}+4J\sin^2K$$
$$-[8V^2\sin^2K/(U-J_{12}+2J\cos 2K-\tfrac{1}{2}J)], \quad \text{branch II.} \qquad (4.21)$$

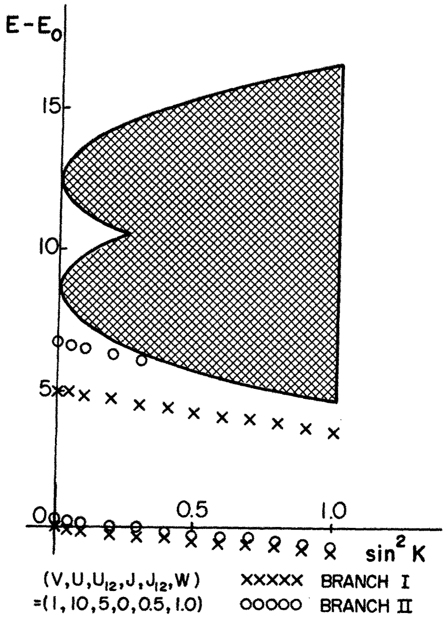

FIG. 3. Case where there is no nearest-neighbor exchange coupling, but there is complete interband intra-atomic coupling. The continuum splits into two parts, and part of the interband exciton spectrum merges into the band. In this case, $W\neq 0$, but Eq. (2.16) is still satisfied. Notice that as $K\rightarrow 0$, the spin wave from branch II still lies above E_0.

Notice the "gap" ($\sim 2J_{12}$) between the two spin-wave spectra. The spectra of bound-state solutions of Eqs. (4.16) and (4.19) are

$$E_i^{(1)}-E_0=U+2J+2J_{12}$$

$$-[(U_{12}+D_{12})^2+16V^2\sin^2K]^{1/2},\quad \text{branch I} \quad (4.22)$$

$$E_i^{(2)}-E_0=U+2J+2J_{12}$$

$$-[(U_{12}-D_{12})^2+16V^2\sin^2K]^{1/2}\quad \text{branch II}. \quad (4.23)$$

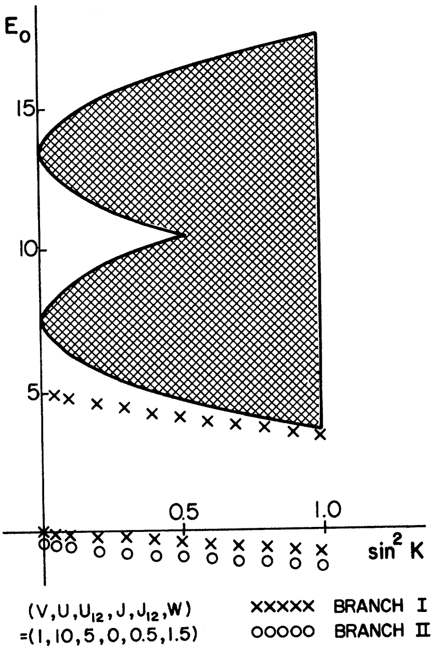

FIG. 4. Similar to Fig. 3, except that in this case Eq. (2.16) is *not* satisfied, which means the atoms would be nonmagnetic by themselves. Here the spin-wave spectrum from branch II lies below E_0 as $K\to 0$. Also, when W becomes larger, the interband excitons from branch II merge into the band.

We can see from Eqs. (4.20) and (4.21) that with the addition of nearest-neighbor exchange coupling, the spin-wave spectra could be concave upward, and hence the saturated ferromagnetic eigenstate could be stable. The conditions for the stability of the saturated ferromagnetic eigenstate are, from Eqs. (4.20) and (4.22), using $J_{12}=D_{12}$,

$$J(U+J_{12}+\tfrac{3}{2}J)\gtrsim 2V^2 \quad (4.24)$$

and

$$(U+2J-U_{12})(U+2J+U_{12}+2J_{12})\geq 16V^2. \quad (4.25)$$

Equation (4.24) is the one that is more likely to be violated.

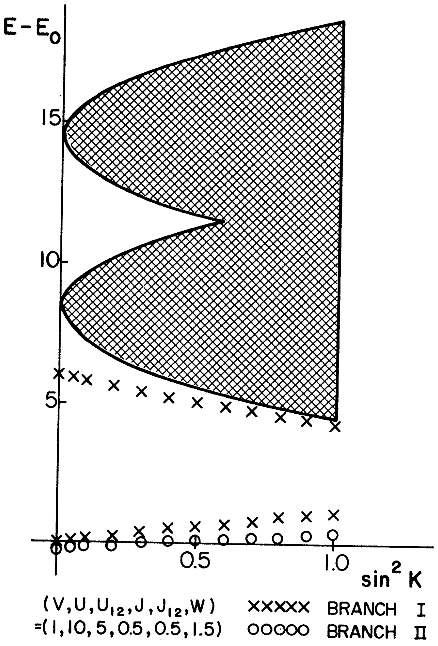

FIG. 5. Case where there is a weak nearest-neighbor exchange coupling, and the individual atoms are nonmagnetic, i.e., Eq. (2.16) is not satisfied. Here the spin-wave spectra are turned upward, but the central part of the spin wave of branch II still lies below E_0. This means that the nearest-neighbor exchange coupling is not strong enough to magnetize the system through mutual polarization. [Equation (4.29) is not satisfied.] The old excitons appear only for extremely small value of $\sin^2K$; they are neglected on this figure.

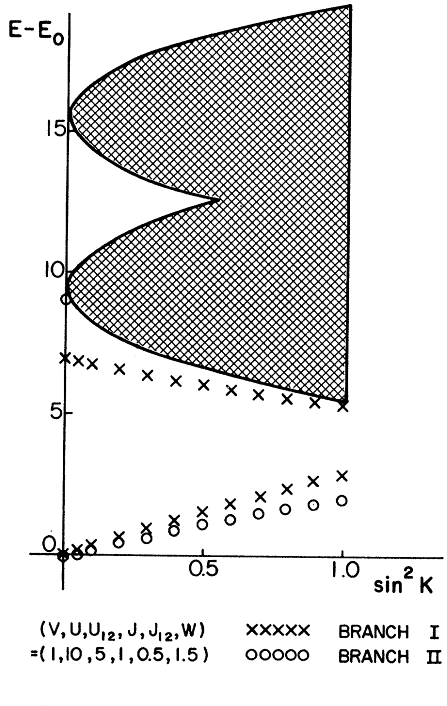

FIG. 6. Similar to Fig. 5 except that the strength of nearest-neighbor coupling (J) is increased a little. The ferromagnetic eigenstate is still not stable.

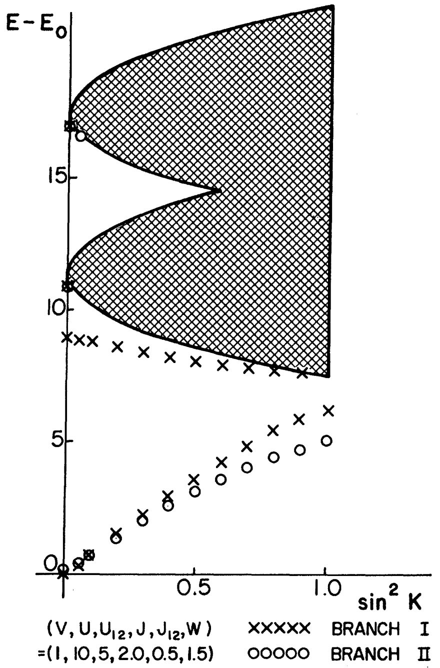

FIG. 7. Case where nonmagnetic atoms are magnetized through mutual polarization. [Equation (2.16) is not satisfied, but Eq. (4.29) is satisfied.] The spin-wave spectrum of branch II again lies completely above the E_0 level.

B. The Case $W \neq 0$

For nonvanishing W, the determinantal functions F_1 and F_2 can be expanded and rewritten in the form

$$F_m = 1 + \sum_k [\rho_m^{(+)}(k)/(E' - E_k - 2W)] + \sum_k [\rho_m^{(-)}(k)/(E' - E_k + 2W)], \qquad m = 1, 2 \qquad (4.26)$$

where $\rho_m^{(+)}(k)$, $\rho_m^{(-)}(k)$ are functions of K and the set of parameters $\{V, U, U_{12}, J, J_{12}\}$. In this form one can see that the continuum states, as indicated by the energy denominators, are subdivided into two subbands. The centers of these two subbands are separated by $4W$. Notice that it is the W term and not U or U_{12} term that splits the continuum. The structure of the continuum is illustrated in Figs. 3–7.

To find the energy spectra of the bound states in which our main interest lies, we again replace $N^{-1}\sum_k$ by $(2\pi)^{-1}\int_{-\pi}^{\pi} dk$, and solve Eqs. (4.8) and (4.10). This is done with the aid of a computer, and the results are presented in Figs. 2–7. Here we have made use of the fact that $J_{12} = D_{12}$. It is seen that the slopes and curvatures of the spin-wave spectra as functions of $\sin^2 K$ are only slightly modified by changing W (comparing Fig. 2 and Fig. 6). At $K = 0$, the energy eigenvalues of the spin-wave states are analytically found to be

$$E_s^{(1)} - E_0 = 0, \quad \text{branch I} \qquad (4.27)$$

$$E_s^{(2)} - E_0 = 2J_{12} + \tfrac{1}{2}\{(U + 2J - U_{12}) - [(U + 2J - U_{12})^2 + 16W^2]^{1/2}\}, \quad \text{branch II.} \qquad (4.28)$$

In order that the central point of the spin-wave spectrum of branch II could lie above the $E = E_0$ state, the following condition has to be satisfied:

$$J_{12}^2 + \tfrac{1}{2} J_{12}(U + 2J - U_{12}) \geq W^2. \qquad (4.29)$$

Equation (4.29) looks very similar to Eq. (2.16), the criterion for the triplet states being the lowest states in the two-electron case discussed in Sec. II; the two equations are in fact the same when $J = 0$, if we identify the corresponding parameters, U_{12}, J_{12}, etc. Yet we see that when the nearest-neighbor exchange coupling (J) is strong enough, Eq. (4.29) can be satisfied even though in the isolated-atom case ($J = 0$) it would not be satisfied (compare Figs. 5 and 6 with Fig. 7). This can be interpreted as follows: Atoms, in the sense of Wannier sites, can be magnetized through mutual polarization, even though individually they would be nonmagnetic.[6]

As mentioned earlier, under the condition $U \gg V$, W, etc., there is only a slight dependence of the slopes and curvatures of the spin-wave spectra on W, hence Eq. (4.24) would be an approximate criterion for the spin-wave spectrum to be concave upward. We propose Eqs. (4.24) and (4.29) as criteria for the ferromagnetic eigenstate to be locally stable.

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[1] J. Chen and M. Bailyn, Phys. Rev. B **1**, 3030 (1970).

[2] J. C. Slater, G. Koster, and H. Statz, Phys. Rev. **91**, 1323 (1953).

[3] See, for example, D. Mattis, *The Theory of Magnetism* (Harper and Row, New York, 1965).

[4] The words "intra-atomic terms" refer to the interaction terms carrying the same Wannier indices.

[5] The parameters characterizing the interband terms are actually defined in terms of Wannier integrals. Numerically, $J_{12} = D_{12}$. There are actually two types of terms formed by the product of three Wannier operators in one band. We have assumed, for mathematical simplicity, that the Wannier integrals associated with these two types of terms are the same, and hence lump the two together to form the W terms. For a classification of interaction terms, see M. Bailyn, Advan Phys. **15**, 179 (1966), Sec. 1.3.3.

[6] A similar "polarization magnetization" was discussed some years ago in another context by one of us: M. Bailyn, Phys. Rev. **139**, A1905 (1965). It was suggested that this is the essence of band magnetization.