Quasispin model of itinerant magnetism*

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(Received 5 January 1976)

The problem of spin waves in itinerant ferromagnets and antiferromagnets is discussed from the band-theory point of view. In the ground state, the energy bands are spin split and there is a spin-density cloud around each ion site. When a spin wave is excited, we postulate that the spin cloud of each ion precesses as a rigid unit, i.e., a quasispin. This gives rise to a modulation of the effective exchange potential, and the difference between the modulated potential and the ground-state exchange potential has the form of the s-d interaction. Here the s part includes all the conduction electrons and the d part arises from the precession of the quasispins. The magnon energy, electron band splitting, and the magnon-magnon interaction are discussed. In every case the agreement between the result and the experiment is as good as or better than the existing theories.

I. INTRODUCTION

A difficult problem in the itinerant theory of ferromagnetism in real transition metals is how to deal with the Hund's-rule correlation of the electrons in the many d orbitals.[1] Using iron as an example, we can find direct experimental evidence that the band description of the Bloch electron states is valid at low temperatures.[2] On the other hand, the iron atoms retain their local moments in the paramagnetic phase. This means that the electronic structure of paramagnetic iron is not described by spin degenerate d bands as the mean-field theory suggests. Instead, a correct theory should preserve the strong Hund's-rule correlation within each ion. The same problem is present in the spin-wave theory.[1,3] In the current theory of spin waves, the one-magnon state is constructed as a linear combination of Slater determinants of Bloch states. Each Slater determinant has one spin deviation from the ground state. This type of wave function is very convenient for handling correlations in the momentum space, but not for the Hund's-rule problem because this involves real space correlations. In this paper we develop a different description of the spin waves in an itinerant system. In the ground state there is a spin cloud around each atomic site. When excited, the spin cloud around each site is assumed to precess rigidly as a unit, a quasispin. A spin wave is then a coherent precession of quasispins. Thus, the on-site Hund's-rule correlation is strictly retained. The two seemingly divergent points of view represent two extreme limits of the actual situation, the current theory is the weak-coupling limit and the quasispin theory is the strong-coupling limit. We will show that the conclusions of the strong-coupling theory agree with the data on nickel and chromium better than the weak-coupling theory, although these two metals are largely regarded as weak-coupling systems.

The starting point of any theory of itinerant magnetism is the theory of the ground state. There have been important advances in the determination of energy bands, Fermi surface, and wave functions for ferromagnetic and antiferromagnetic ground states.[4-14] Some of these calculations are first-principles calculations because they attempt to construct the crystal potential from atomic or crystal charge densities by using the Hartree-Slater approximation.[6-14] It was generally found that when the calculation is carried out self-consistently, the band splitting, the Fermi-surface geometry, and the magnetization come out to agree well with the experiments. The spin distribution obtained from the crystal wave functions also agrees with that inferred from the neutron form factor.[5,9] We take the band-calculation method as the starting point because it offers the following advantages: (i) the full set of s, p, and d bands come out naturally from the calculation, and (ii) the exchange screening and the Hund's-rule correlation are accounted for in the same approximation as in an atomic Hartree-Slater calculation.

We then treat the spin waves as perturbations on the ground state. The presence of a spin-wave modifies the exchange potential in the one-electron problem, and the modification is readily determined in the rigid spin approximation. Since one magnon produces only a small perturbation on the ground state, the modified one-electron problem can be solved by the standard method. The added energy to the entire electron system due to one magnon is identified as the magnon energy. The electron-magnon and magnon-magnon interactions can also be discussed in this manner. Thus, the properties of the low-lying excited states are cal-

culated from ground-state electron levels, wave functions, and potentials without introducing additional parameters.

II. FERROMAGNETIC SPIN WAVES

The energy-band problem for the ferromagnetic ground state is defined by

$$H_\sigma \phi_{n\vec{k}\sigma}(\vec{r}) = E_{n\vec{k}\sigma}\phi_{n\vec{k}\sigma}(\vec{r}), \tag{2.1}$$

where $E_{n\vec{k}\sigma}$ is the energy eigenvalue of band n, wave vector $\vec{k}$, and spin σ, $\phi_{n\vec{k}\sigma}(\vec{r})$ is the spatial part of the band wave function which is normalized within the Wigner-Seitz cell. The one-electron Hamiltonian is given by

$$H_\sigma = \frac{p^2}{2m} + V_\sigma(\vec{r}), \tag{2.2}$$

where $V_\sigma(\vec{r})$ is the periodic potential for an electron with spin σ. The potential consists of the following terms:

$$V_\sigma(\vec{r}) = V_n(\vec{r}) + V_H(\vec{r}) + V_{x\sigma}(\vec{r}), \tag{2.3}$$

where the nuclear potential is

$$V_n(\vec{r}) = -\sum_i \frac{Ze^2}{|\vec{r}-\vec{R}_i|},$$

the Hartree potential is

$$V_H(\vec{r}) = e^2 \int \frac{\rho(\vec{r}')}{|\vec{r}-\vec{r}'|} d^3r',$$

and the exchange potential is

$$V_{x\sigma}(\vec{r}) = -\alpha(3e^2)[(3/4\pi)\rho_\sigma(\vec{r})]^{1/3}. \tag{2.4}$$

In the above equations, Z is the atomic number of the metal, $\vec{R}_i$ are the positions of the atoms, the electron density

$$\rho_\sigma(\vec{r}) = \frac{1}{N}\sum_{n\vec{k}} |\phi_{n\vec{k}\sigma}(\vec{r})|^2 f_{n\vec{k}\sigma}, \tag{2.5}$$

where $f_{n\vec{k}\sigma}$ is the Fermi distribution function, N is the number of atoms in the specimen, and

$$\rho(\vec{r}) = \rho_\uparrow(\vec{r}) + \rho_\downarrow(\vec{r}). \tag{2.6}$$

The factor α takes care of corrections from the Slater exchange.[15-17]

In the end of the calculation, one finds the spin density

$$\vec{s}(\vec{r}) = \frac{1}{2N}\sum_{n\vec{k}} [|\phi_{n\vec{k}\uparrow}(\vec{r})|^2 f_{n\vec{k}\uparrow} - |\phi_{n\vec{k}\downarrow}(\vec{r})|^2 f_{n\vec{k}\downarrow}], \tag{2.7}$$

and the moment per atom

$$S = \int_c \vec{s}(\vec{r})\, d^3r = \frac{1}{2N}\sum_{n\vec{k}} (f_{n\vec{k}\uparrow} - f_{n\vec{k}\downarrow}), \tag{2.8}$$

where the integration is carried out in the unit cell, assuming one atom per cell. The moment per atom is in general nonintegral. The exchange splitting for the band n at the wave vector $\vec{k}$ is

$$2\Delta_{n\vec{k}} = E_{n\vec{k}\downarrow} - E_{n\vec{k}\uparrow}. \tag{2.9}$$

This quantity is in general dependent on n and $\vec{k}$.

Now we construct a theory of ferromagnetic spin waves based on the band-theory formalism. We write

$$V_\sigma(\vec{r}) = V(\vec{r}) - \sigma^z W(\vec{r}),$$

where

$$\begin{aligned} V(\vec{r}) &= \tfrac{1}{2}[V_\uparrow(\vec{r}) + V_\downarrow(\vec{r})], \\ W(\vec{r}) &= \tfrac{1}{2}[V_\downarrow(\vec{r}) - V_\uparrow(\vec{r})], \end{aligned} \tag{2.10}$$

and σ^z is 1 or -1 for spin-up and spin-down electrons. The spin-dependent part of the exchange potential is $W(\vec{r})$. In the ground state it has the same periodicity as the lattice. When a spin wave is excited, the spin-dependent part $W(\vec{r})$ undergoes a periodic variation. In a rigid spin model we postulate that the spin direction of the atom at $\vec{R}_i$ is shifted from the z direction so that it is given by the unit vector $\hat{\eta}_i$. Then the perturbing potential in the one electron Hamiltonian is

$$H_1 = -\sum_i W_0(\vec{r}-\vec{R}_i)(\hat{\eta}_i\cdot\vec{\sigma} - \sigma^z), \tag{2.11}$$

where $W_0(\vec{r}-\vec{R}_i)$ is the exchange potential in the Wigner-Seitz cell around $\vec{R}_i$, and is zero outside that cell. The Pauli matrices for the band electron is denoted by $\vec{\sigma}$.

The quantization of the quasispins is a little more subtle because the size of the spins is in general not an integer or a half odd integer. The ground state of the spin on each site may be viewed, according to the interconfiguration fluctuation model,[18] as a mixture of a number of spin configurations, all in the state of maximum z spin component. Thus,

$$\psi = \sum_n b_n |S_n, S_n\rangle,$$

where $\sum_n |b_n|^2 = 1$. The size of the quasispin is $S = \sum_n |b_n|^2 S_n$. The ground state of the entire spin system is then

$$|0\rangle = \prod_i \psi(i),$$

where i is the site index. The one-magnon state with wave vector $\vec{q}$ is defined as

$$|\vec{q}\rangle = C_{\vec{q}} S^-_{\vec{q}} |0\rangle,$$

where $C_{\vec{q}}$ is the normalization constant and $S^\pm_{\vec{q}} = S^x_{\vec{q}} \pm iS^y_{\vec{q}}$

$$\vec{S}_{\vec{q}} = \frac{1}{\sqrt{N}} \sum_i \vec{S}_i e^{i \vec{q} \cdot \vec{R}_i} .$$

The normalization constant can be readily found to be

$$|C_{\vec{q}}|^{-2} = \langle 0 | [S^+_{-\vec{q}}, S^-_{\vec{q}}] | 0 \rangle = 2S.$$

Then the creation and annihilation operators of the magnons are found to be

$$a^\dagger_{\vec{q}} = (2S)^{-1/2} S^-_{\vec{q}}, \quad a_{\vec{q}} = (2S)^{-1/2} S^+_{-\vec{q}} .$$

The spin deviation operators are related to the magnon operators by

$$a_i = \frac{1}{\sqrt{N}} \sum_{\vec{q}} a_{\vec{q}} e^{-i \vec{q} \cdot \vec{R}_i} .$$

Therefore,

$$S^x_i = (\tfrac{1}{2}S)^{1/2} (a^\dagger_i + a_i), \quad S^y_i = i (\tfrac{1}{2}S)^{1/2} (a^\dagger_i - a_i).$$

At this point, we approximate the spin deviation operators as boson operators. Then to satisfy the spin commutation relations in the lowest order, we must have

$$S^z_i = S - a^\dagger_i a_i .$$

This form of S^z_i satisfies the additional condition that

$$\sum_i S^z_i = NS - \sum_{\vec{q}} a^\dagger_{\vec{q}} a_{\vec{q}} ,$$

which means that each magnon reduces the z component of the total spin of the crystal by one unit.

Now the unit vectors $\hat{\eta}_i$ in the perturbing potential are readily related to the spin deviation operators

$$\eta^z_i = 1 - a^\dagger_i a_i / S , \quad \eta^x_i = (2S)^{-1/2} (a^\dagger_i + a_i) , \quad \eta^y_i = i (2S)^{-1/2} (a^\dagger_i - a_i) . \tag{2.12}$$

In terms of the electron operators $C^\dagger_{n\vec{k}\sigma}$, $C_{n\vec{k}\sigma}$ and spin-wave operators, we can write

$$\begin{aligned} H_1 = \frac{1}{NS} \sum_{nn'} \sum_{\vec{k}} \sum_{\vec{q}\,\vec{q}'} a^\dagger_{\vec{q}} a_{\vec{q}'} \big(& \langle n', \vec{k} + \vec{q}' \uparrow | W_0 | n, \vec{k} + \vec{q} \uparrow \rangle C^\dagger_{n', \vec{k}+\vec{q}', \uparrow} C_{n, \vec{k}+\vec{q}, \uparrow} \\ & - \langle n', \vec{k} + \vec{q}' \downarrow | W_0 | n, \vec{k} + \vec{q} \downarrow \rangle C^\dagger_{n', \vec{k}+\vec{q}', \downarrow} C_{n, \vec{k}+\vec{q}, \downarrow} \big) \\ - \left(\frac{2}{NS}\right)^{1/2} \sum_{nn'} \sum_{\vec{k}} \sum_{\vec{q}} \big(& \langle n', \vec{k} + \vec{q} \downarrow | W_0 | n \vec{k} \uparrow \rangle a_{\vec{q}} C^\dagger_{n', \vec{k}+\vec{q}, \downarrow} C_{n \vec{k}, \uparrow} + \langle n', \vec{k} + \vec{q} \uparrow | W_0 | n \vec{k} \downarrow \rangle a^\dagger_{-\vec{q}} C^\dagger_{n', \vec{k}+\vec{q}, \uparrow} C_{n \vec{k}, \downarrow} \big) . \end{aligned} \tag{2.13}$$

The matrix elements are evaluated by integrating over one unit cell.

One may be tempted to work out a deformable spin model by postulating a continuous spin distortion $\hat{\eta}(\vec{r})$. It is questionable whether the spin quantization procedure can be carried through for a deformable spin. Besides, recent neutron scattering measurement of the spin-wave form factor of chromium seems to favor the rigid spin model.[19]

The perturbing Hamiltonian in Eq. (2.13) is identical to the s-d Hamiltonian that was extensively studied a decade ago. The only difference is in the physical meaning. Here the band electrons include not only the s electrons but also the d electrons. The spin waves are generated by a periodic precession of the spin clouds in the Wigner-Seitz cells.

The consequences of the perturbing Hamiltonian may be immediately calculated by using the diagrammatic rules of Izuyama and Kubo.[20] For example, the magnon energy is given by the implicit equation[20,21]

$$\omega = \Pi(\vec{q}, \omega) , \tag{2.14}$$

where $\Pi(\vec{q}, \omega)$ is the magnon self-energy which, in the lowest order, is given by the Feynman diagrams in Fig. 1. Straightforward evaluation of these diagrams yields

$$\begin{aligned} \omega = \frac{1}{NS} \sum_{n\vec{k}} \big(& \langle n \vec{k} \uparrow | W_0 | n \vec{k} \uparrow \rangle f_{n\vec{k}\uparrow} \\ & - \langle n \vec{k} \downarrow | W_0 | n \vec{k} \downarrow \rangle f_{n\vec{k}\downarrow} \big) \\ + \frac{2}{NS} \sum_{nn'\vec{k}} & \frac{f_{n\vec{k}\downarrow} - f_{n', \vec{k}+\vec{q}\uparrow}}{E_{n\vec{k}\downarrow} - E_{n', \vec{k}+\vec{q}\uparrow} - \omega} \\ & \times |\langle n', \vec{k} + \vec{q} \uparrow | W_0 | n \vec{k} \downarrow \rangle|^2 . \end{aligned} \tag{2.15}$$

This result differs from that of Nagaoka[21] in that a number of interband terms do not appear here. They arise from higher-order diagrams not included in Fig. 1. In Appendix A, we show in de-

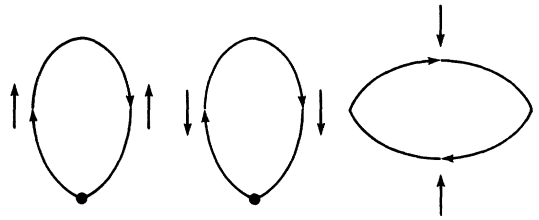

FIG. 1. Self-energy diagrams for the ferromagnetic spin waves.

tail that the spin-wave energy at zero wave vector vanishes. For small wave vectors the magnon energy must depend quadratically on the wave vector $\vec{q}$.

We now compare our theory with some of the known results of other theories. If we ignore the magnon energy in the energy denominator in Eq. (2.15), which is justified for large values of $\vec{q}$, the equation

$$\omega = \Pi(\vec{q}, 0)$$

is precisely what one would obtain by first calculating the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the spins in each cell and then obtaining the magnon spectrum from the spin pair interaction Hamiltonian.[22-24] This strongly suggests that the large-q part of the magnon spectrum, which reflects the near-neighbor coupling, is given by the RKKY theory. This point of view has been very convincingly advocated by Stearns.[25]

If we ignore ω on the left-hand side and at the same time assume only one band and approximate all matrix elements by a constant Δ, we find the following equation for the magnon spectrum:

$$1 + \frac{\Delta}{NS} \sum_{\vec{k}} \frac{f_{\vec{k}\downarrow} - f_{\vec{k}+\vec{q},\uparrow}}{E_{\vec{k}\downarrow} - E_{\vec{k}+\vec{q}\uparrow} - \omega} = 0 ,$$

which is the same equation as found from treating the magnon problem by the random-phase approximation.[1] This suggests that in the weak-coupling limit where the random-phase approximation (RPA) applies, our theory must break down. This is understandable because when the coupling is weak and the magnetization itself is very small, even one magnon produces too large a perturbation. On the other hand, when the exchange interaction is strong and there are many bands, we propose that our method of treating the moments on the atoms as quasispins takes better account of the exchange screening and Hund's-rule correlation within the spins.

For small $\vec{q}$, the spectrum may be solved in the constant matrix element model to give

$$\omega_{\vec{q}} = \frac{\Delta}{2} \left(1 + \frac{\Delta}{NS} \sum_{nn'} \sum_{\vec{k}} \frac{f_{n\vec{k}\downarrow} - f_{n',\vec{k}+\vec{q}\uparrow}}{E_{n\vec{k}\downarrow} - E_{n',\vec{k}+\vec{q}\uparrow}} \right) , \tag{2.16}$$

which is smaller than the RKKY and the RPA theory results by a factor of 2.[26]

In addition, in the constant matrix element approximation, the equation for the magnon spectrum may be written

$$\omega = [\chi_{+-}(\vec{q}, \omega)]^{-1} ,$$

where $\chi_{+-}(\vec{q}, \omega)$ is the dynamic transverse susceptibility of the electron gas. For large $\vec{q}$, the magnon energy may be neglected in the right-hand side, and we obtain the Landau-Lifshitz[27] and Herring-Kittel[28] theory for the magnon energy of itinerant ferromagnets. For small $\vec{q}$, our treatment puts a dynamic correction into the theory.

The magnon damping due to electron-magnon interaction follows directly from Eq. (2.15). If we write the complex magnon energy as $\omega_{\vec{q}} - i\Gamma_{\vec{q}}$, then

$$\Gamma_{\vec{q}} = \frac{2\pi}{NS} \sum_{nn'} \sum_{\vec{k}} |\langle n', \vec{k}+\vec{q}\uparrow | W_0 | n\vec{k}\downarrow \rangle|^2 \times (f_{n',\vec{k}+\vec{q}\uparrow} - f_{n\vec{k}\downarrow}) \times \delta(E_{n\vec{k}\downarrow} - E_{n',\vec{k}+\vec{q}\uparrow} - \omega_{\vec{q}}) . \tag{2.17}$$

At low temperatures, the damping becomes nonvanishing after the magnon dispersion curve enters the Stoner threshold.

One may obtain more qualitative feeling for these results by studying a particularly simple model of one parabolic band with effective mass m and constant matrix elements Δ. Figure 2 compares the spin-wave spectrum calculated from Eq. (2.15) with those calculated from RKKY theory and RPA.

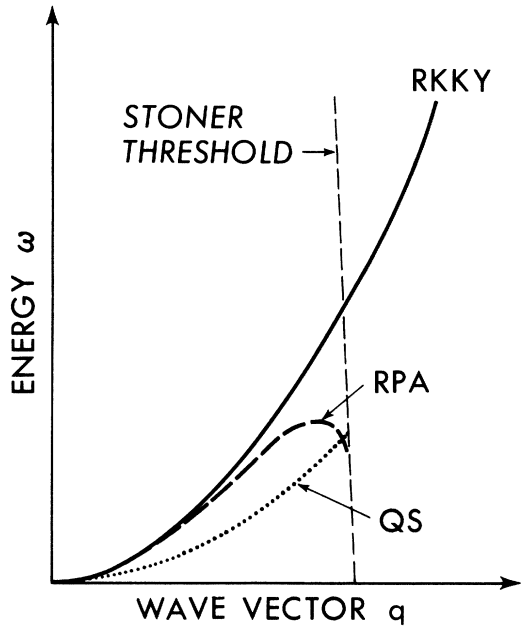

FIG. 2. Qualitative sketches of the dispersion relation of ferromagnetic spin waves according to various theories. The RKKY theory gives a dispersion curve which extends into the Stoner mode region. The random-phase approximation (RPA) predicts that the dispersion curve bends over prior to reaching the Stoner threshold. The quasispin model (QS) predicts that the dispersion curve approaches the Stoner threshold without bending, but becomes overdamped after crossing the threshold.

In the RPA result, the dispersion curve dips down upon reaching the Stoner threshold.[1,29] The RKKY theory and our result do not show this dip. The condition for the existence of a dip near the Stoner threshold was discussed by Nagaoka.[21] He showed that it has to do with the ratio of the exchange splitting and the Fermi energy. When this ratio is small, no dip is expected. So, unlike the RPA, our theory does not predict that a dip always exists. Recent neutron scattering experiments on Ni do not seem to indicate such a dip.[30]

After passing the Stoner threshold, the magnon damping is found to be

$$\Gamma_q \cong (m^2 V_0 \Delta^2/\pi S q)\,\omega_q ,$$

where V_0 is the volume of the unit cell. The factor in front of the magnon energy is approximately 1.5 near the Stoner threshold. So the magnon mode is so overdamped that it becomes unobservable. Of course different band models may give more or less damping, but the ratio of $\Gamma_q/\omega_q \cong 1$ should hold.

Similar to the work of Izuyama and Kubo[20] we can calculate the electron energy, and in particular, the band splitting as a function of temperature. We will present the results here for the one band, constant matrix element model with the following interaction Hamiltonian:

$$H_1 = \frac{\Delta}{NS}\sum_{\vec{q}\,\vec{q}'}\sum_{\vec{k}} a^{\dagger}_{\vec{q}}\, a_{\vec{q}}\,(C^{\dagger}_{\vec{k}+\vec{q}\uparrow} C_{\vec{k}+\vec{q}'\uparrow} - C^{\dagger}_{\vec{k}+\vec{q}\downarrow} C_{\vec{k}+\vec{q}'\downarrow}) - \frac{2\Delta}{NS}\sum_{\vec{k}}\sum_{\vec{q}} (a_{\vec{q}} C^{\dagger}_{\vec{k}+\vec{q}\downarrow} C_{\vec{k}\uparrow} + a^{\dagger}_{-\vec{q}} C^{\dagger}_{\vec{k}+\vec{q}\uparrow} C_{\vec{k}\downarrow}) . \tag{2.18}$$

The electron self-energy diagrams are given in Fig. 3. By taking the difference of the energies of opposite spin states near the Fermi level, we obtain the temperature dependence of the band splitting

$$\Delta(T) = \Delta + BT^{5/2} , \tag{2.19}$$

where the constant B is expressed in terms of the band parameters as

$$BT^{5/2} = \frac{4}{NS}\sum_{\vec{q}} n_{\vec{q}}[\,\omega_{\vec{q}} + (1/\Delta)(\vec{q}\cdot\nabla_{\vec{k}} E_{\vec{k}})^2|_{E_F}] , \tag{2.20}$$

where $n_{\vec{q}}$ is the Bose distribution function for the magnons. Furthermore, the self-consistent relation between $\Delta(T)$ and the electron distribution gives the well-known T^2 term to the band splitting. Thus,

$$\Delta(T) = \Delta - AT^2 + BT^{5/2} . \tag{2.21}$$

The surprising result is that the $T^{5/2}$ term is positive. So, not only is the band splitting dependent on a higher power law than $T^{3/2}$, the temperature-dependent terms are opposite in sign and tend to cancel. The net result is that the T dependence of the band splitting is extremely weak, in agreement with the experimental result of Lonzarich and Gold.[31] The theory of Edwards predicts a negative sign for the $T^{5/2}$ term.[32] The origin of the negative sign can be traced to his assumption on the form of the electron-magnon interaction. Lonzarich refined Edward's argument according to the itinerant theory and obtained the same sign as ours.[33]

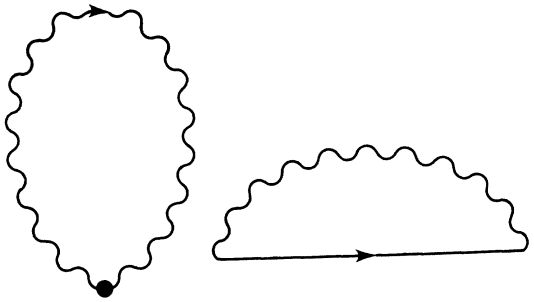

FIG. 3. Diagrams of electron self-energies.

Finally we discuss the magnon energy renormalization effects. At elevated temperatures there is the well-known T^2 term in the magnon energy due to the excitation of electrons near the Fermi level. The energy renormalization due to magnon-magnon interactions is calculated from the sum of the group of diagrams in Fig. 4. We keep only those

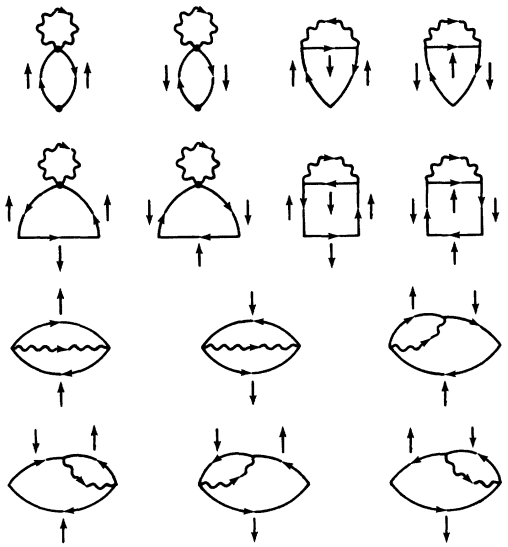

FIG. 4. Diagrams of magnon self-energy due to magnon-magnon interaction.

terms which are proportional to $n_{\vec{q}}$. It is not difficult but rather tedious to show that the temperature dependence of this term is indeed $T^{5/2}$ as shown by Izuyama and Kubo from very general physical grounds.[20,34] The expression for the energy shift came out slightly different from that given in Ref. 20, conceivably because we have cut off the interaction at the two-magnon term whereas Izuyama and Kubo included some third-order terms. We find, for the one-band model

$$\omega_{\vec{q}}(T) = \omega_{\vec{q}}(0) + \delta\omega_{\vec{q}} ,$$

where

$$\delta\omega_{\vec{q}} = -\frac{1}{8S}\frac{1}{N}\sum_{\vec{q}_1} n_{\vec{q}_1}\frac{1}{N}\sum_{\vec{k}}\{\delta(E_{\vec{k}\uparrow}-E_F)[(\vec{q}\cdot\nabla_{\vec{k}})^2E_{\vec{k}}-(1/\Delta)(\vec{q}\cdot\nabla_{\vec{k}}E_{\vec{k}})^2][(\vec{q}_1\cdot\nabla_{\vec{k}})^2E_{\vec{k}}-(1/\Delta)(\vec{q}_1\cdot\nabla_{\vec{k}}E_{\vec{k}})^2]$$
$$+\delta(E_{\vec{k}\downarrow}-E_F)[(\vec{q}\cdot\nabla_{\vec{k}})^2E_{\vec{k}}+(1/\Delta)(\vec{q}\cdot\nabla_{\vec{k}}E_{\vec{k}})^2][(\vec{q}_1\cdot\nabla_{\vec{k}})^2E_{\vec{k}}+(1/\Delta)(\vec{q}_1\cdot\nabla_{\vec{k}}E_{\vec{k}})^2]\} . \tag{2.22}$$

The conclusion of these rather lengthly calculations is that our model predicts the expected temperature power laws for all the energy shifts. Furthermore, the energy shifts are accessible to numerical calculation without the need of introducing new parameters.

III. ANTIFERROMAGNETIC SPIN WAVES

The itinerant theory of ferromagnetism predicts a spin-wave energy which is a complicated function of the band parameters. In contrast, the theory of itinerant antiferromagnetism gives a very simple prediction, namely that the magnon dispersion relation is linear and the magnon velocity is $(\frac{1}{3}\langle v_F^2\rangle)^{1/2}$, $(\langle v_F^2\rangle)^{1/2}$ being the average Fermi velocity over the nested part of the Fermi surface.[35] The magnon velocity has been estimated for chromium based on band-calculation results and compared with experiments.[36,37] It was found that the experimental result is lower than the theoretical value by about a factor of 2. The mystery of this discrepancy was compounded when the calculated velocity was found to explain the spin-diffusion rate above the Néel temperature.[36,38] We will show in this section that a calculation analogous to the ferromagnetic spin waves in Sec. II gives a correction to the previous result for the magnon velocity. This correction factor depends on the interaction strength and it brings the theory and experiment into good agreement.

In pure Cr, the spin-density-wave (SDW) state has a periodicity which is incommensurate with the lattice. However, in all the inelastic neutron scattering experiments, a few percent of Mn was added to Cr so that the spin-density wave is locked on to the lattice.[39] In this case the magnetic moment is the same on every atom in the solid except that half the moments point up and the other half point down. The periodicity may be characterized by a wave vector $\vec{Q}$ which is equal to one-half of a primitive reciprocal-lattice vector and is perpendicular to the spin direction. Once locked on to the lattice the SDW shows remarkable stability in structure when the Mn concentration is varied over a range of a few percent. The moment per atom is also larger than that of pure Cr. We will extend the spin-wave theory to this transverse, commensurate spin structure.

In the SDW state, the spin density has a spatial dependence

$$\rho_\sigma(\vec{r}) = \rho_\sigma(\vec{r}) + \sigma^z\sum_i \delta\rho(\vec{r}-\vec{R}_i)e^{i\vec{Q}\cdot\vec{R}_i}, \tag{3.1}$$

where $\delta\rho(\vec{r}-\vec{R}_i)$ is defined in the Wigner-Seitz cell around $\vec{R}_i$. Then the exchange potential is also spatially dependent,

$$V_{x\sigma}(\vec{r}) = V_x(\vec{r}) + V_x(\vec{r})\sigma^z\sum_i \frac{\delta\rho(\vec{r}-\vec{R}_i)}{3\rho_0(\vec{r})}e^{i\vec{Q}\cdot\vec{R}_i} \tag{3.2}$$

and

$$V_x(\vec{r}) = -\alpha(3e^2)[(3/4\pi)\rho_0(\vec{r})]^{1/3}. \tag{3.3}$$

We treat the last term in Eq. (3.2) as a perturbation on the band Hamiltonian

$$H_0 = p^2/2m + V(\vec{r}), \tag{3.4}$$

where $V(\vec{r})$ is the sum of $V_n(\vec{r})$, $V_H(\vec{r})$ defined in Eq. (2.4) and $V_x(\vec{r})$ in Eq. (3.3). The eigenstates and eigenvalues of H_0 are defined by

$$H_0\psi_{n\vec{k}\sigma} = E_{n\vec{k}}\psi_{n\vec{k}\sigma}, \tag{3.5}$$

where $\psi_{n\vec{k}} = \phi_{n\vec{k}}(\vec{r})\xi_\sigma$, ξ_σ is the Pauli spinor. The perturbing Hamiltonian is

$$W(\vec{r}) = -\sigma^z\sum_i W_0(\vec{r}-\vec{R}_i)e^{i\vec{Q}\cdot\vec{R}_i}, \tag{3.6}$$

where

$$W_0(\vec{r}) = -V_x(\vec{r})[\delta\rho(\vec{r})/3\rho_0(\vec{r})], \tag{3.7}$$

which is defined within the Wigner-Seitz cell. Since the periodicity of $W_0(\vec{r})$ differs from that of $V(\vec{r})$, the perturbing potential will link together

states $|n\vec{k}\sigma\rangle$ and $|n,\vec{k}+\vec{Q},\sigma\rangle$. If a large number of states near the Fermi level can be linked by the same wave vector $\vec{Q}$, the reduction in exchange energy due to this pairwise correlation stabilizes the SDW state. In pure Cr the wave vector Q is rather close to one-half of a reciprocal-lattice vector. The addition of a few percent of Mn shifts the Fermi level slightly to enable the SDW to lock on to the lattice.[40]

In the following discussion, we will use the Fedders-Martin model for the bands.[35] The band 1 is an electron band of mass m,

$$E_{1\vec{k}} = k^2/2m. \tag{3.8}$$

The Fermi wave vector is k_F and the Fermi energy is $E_F = k_F^2/2m$. The band 2 is a hole band of equal mass,

$$E_{2\vec{k}} = 2E_F - (1/2m)[(k_x - Q)^2 + k_y + k_z^2]. \tag{3.9}$$

The Fermi surfaces of the bands are two equal spheres separated by $\vec{Q}$, assumed to be in the x direction. Then the most important part of the total Hamiltonian is

$$\begin{aligned} H = &\sum_{\vec{k}\sigma}(E_{1\vec{k}}C^{\dagger}_{1\vec{k}\sigma}C_{1\vec{k}\sigma} + E_{2\vec{k}}C^{\dagger}_{2\vec{k}\sigma}C_{2\vec{k}\sigma}) \\ &- \sum_{\vec{k}}[\langle 1\vec{k}|W_0|2,\vec{k}+\vec{Q}\rangle \\ &\times(C^{\dagger}_{1\vec{k}\uparrow}C_{2,\vec{k}+\vec{Q}\uparrow} - C^{\dagger}_{1\vec{k}\downarrow}C_{2,\vec{k}+\vec{Q}\downarrow}) + \text{H.c.}], \end{aligned} \tag{3.10}$$

where

$$\langle 1\vec{k}|W_0|2,\vec{k}+\vec{Q}\rangle = \int \phi^*_{1\vec{k}}(\vec{r})W_0(\vec{r})\phi_{2,\vec{k}+\vec{Q}}(\vec{r})\,d^3r, \tag{3.11}$$

and the integral is over the Wigner-Seitz cell. The wave functions are also normalized in the unit cell. We will also replace the matrix elements of $W_0(\vec{r})$ by their average over the Fermi level

$$\Delta = \langle 1\vec{k}|W_0|2,\vec{k}+\vec{Q}\rangle_{\text{av}}. \tag{3.12}$$

Then the quasiparticle states are easy to find. The energies of these states are

$$E^{(\pm)}_{\vec{k}} = E_F \pm (\epsilon_k^2 + \Delta^2)^{1/2}, \tag{3.13}$$

where $\epsilon_k = E_{1\vec{k}} - E_F = E_F - E_{2,\vec{k}+\vec{Q}}$. In particular the off-diagonal correlation function may be calculated

$$\begin{aligned} &\langle C^{\dagger}_{1\vec{k}\sigma}C_{2,\vec{k}+\vec{Q},\sigma}\rangle \\ &\quad = \pm[\Delta/2(\epsilon_k^2+\Delta^2)^{1/2}]\tanh[\tfrac{1}{2}\beta(\epsilon_k^2+\Delta^2)^{1/2}]. \end{aligned} \tag{3.14}$$

The sinusoidally modulated part of the charge density is found to be

$$\delta\rho_\sigma(\vec{r}) = \frac{1}{N}\sum_{\vec{k}}[\phi^*_{1\vec{k}}(\vec{r})\phi_{2,\vec{k}+\vec{Q}}(\vec{r})\langle C_{1\vec{k}\sigma}C_{2,\vec{k}+\vec{Q},\sigma}\rangle + \text{c.c.}]. \tag{3.15}$$

Then Eqs. (3.7) and (3.12) give

$$\Delta \cong \frac{U}{N}\sum_{\vec{k}}\frac{\Delta}{2(\epsilon_k^2+\Delta^2)^{1/2}}\tanh[\tfrac{1}{2}\beta(\epsilon^2+\Delta^2)^{1/2}], \tag{3.16}$$

where

$$U = -\langle 1\vec{k}|\frac{V_x(\vec{r})}{3\rho_0(\vec{r})}\frac{1}{N}\sum_{\vec{k}}[\phi^*_{1\vec{k}}(\vec{r})\phi_{2,\vec{k}'+\vec{Q}}(\vec{r}) + \text{c.c.}] \times|2,\vec{k}+\vec{Q}\rangle_{\text{av}}, \tag{3.17}$$

with $\vec{k}$ averaged over the Fermi surface. The equation for Δ, Eq. (3.16), may be recognized as the gap equation, with 2Δ as the energy gap.[35] For the band model assumed here, the gap at low temperatures is given by

$$\Delta = 2E_F\exp[-1/N(E_F)U], \tag{3.18}$$

where $N(E_F)$ is the density of states per band per spin per atom. In the ground state the total spin on the site i is given by $Se^{i\vec{Q}\cdot\vec{R}_i}$, where

$$\begin{aligned} S &= \frac{1}{2}\int_c(\delta\rho_\uparrow - \delta\rho_\downarrow)\,d^3r \\ &= \frac{1}{N}\sum_{\vec{k}}\frac{\Delta}{2(\epsilon^2+\Delta^2)^{1/2}} \\ &\quad\times\left(\int_c \phi_{1\vec{k}}(\vec{r})\phi_{2,\vec{k}+\vec{Q}}(\vec{r}) + \text{c.c.}\right)d^3r. \end{aligned} \tag{3.19}$$

When spin waves are excited the added perturbing Hamiltonian is

$$H_1 = -\sum_i W_0(\vec{r}-\vec{R}_i)(\hat{\eta}_i\cdot\vec{\sigma} - \sigma^z)e^{i\vec{Q}\cdot\vec{R}_i}. \tag{3.20}$$

The quantization of H_1 in terms of spin-wave operators is facilitated by the transformation to the local spin coordinates. We define

$$\begin{aligned} &\eta_i^x = \eta_i^\alpha, \quad \eta_i^y = \eta_i^\beta e^{i\vec{Q}\cdot\vec{R}_i}, \\ &\eta_i^z = \eta_i^\gamma e^{i\vec{Q}\cdot\vec{R}_i}. \end{aligned} \tag{3.21}$$

In the ground state the local spin always points in the γ direction. Then

$$\begin{aligned} \eta_i^\gamma &= 1 - \frac{1}{NS}\sum_{\vec{q}\vec{q}'}a^{\dagger}_{\vec{q}}\,a_{\vec{q}'}\,e^{i(\vec{q}'-\vec{q})\cdot\vec{R}_i}, \\ \eta_i^\alpha &= \frac{1}{(2NS)^{1/2}}\sum_{\vec{q}}(a^{\dagger}_{-\vec{q}} + a_{\vec{q}})e^{i\vec{q}\cdot\vec{R}_i}, \\ \eta_i^\beta &= -\frac{i}{(2NS)^{1/2}}\sum_{\vec{q}}(a^{\dagger}_{-\vec{q}} - a_{\vec{q}})e^{i\vec{q}\cdot\vec{R}_i}. \end{aligned} \tag{3.22}$$

Then in the average matrix element approximation

$$H_1 = \frac{\Delta}{NS}\sum_{\vec{q}\vec{q}'} a^\dagger_{\vec{q}}\, a_{\vec{q}'}\sum_{\vec{k}} (C^\dagger_{1,\vec{k}-\vec{q}\uparrow}C_{2,\vec{k}-\vec{q}'+\vec{Q}\uparrow} - C^\dagger_{1,\vec{k}-\vec{q}\downarrow}C_{2,\vec{k}-\vec{q}+\vec{Q}\downarrow} + \text{H.c.})$$
$$- \Delta\frac{1}{2NS}\sum_{\vec{k}\vec{q}}[(a^\dagger_{-\vec{q}} + a_{\vec{q}})(C^\dagger_{1,\vec{k}+\vec{q}\uparrow}C_{1\vec{k}\downarrow} + C^\dagger_{2,\vec{k}+\vec{q}\uparrow}C_{2\vec{k}\downarrow} + \text{H.c.})$$
$$- i(a^\dagger_{-\vec{q}} - a_{\vec{q}})(C^\dagger_{1,\vec{k}+\vec{q}\uparrow}C_{2,\vec{k}+\vec{Q}\downarrow} - C^\dagger_{1,\vec{k}+\vec{q}\downarrow}C_{2,\vec{k}+\vec{Q}\downarrow} + \text{H.c.})]\,. \tag{3.23}$$

For an antiferromagnet the low-energy magnons have wave vectors near $\vec{Q}$. Consequently, we define a two-component spin-wave operator

$$A_{\vec{q}} = \begin{pmatrix} a_{\vec{Q}+\vec{q}} \\ a^\dagger_{-\vec{Q}-\vec{q}} \end{pmatrix}, \tag{3.24}$$

and a matrix Green's function

$$\tilde{D}(\vec{q}, t) = -i\langle TA_{\vec{q}}(t)A^\dagger_{\vec{q}}\ (0)\rangle\rangle\,, \tag{3.25}$$

where $q \ll Q$. The Fourier transform of $\tilde{D}$ satisfies the Dyson equation

$$[\tilde{D}(\vec{q}, \omega)]^{-1} = [\tilde{D}^{(0)}(\vec{q}, \omega)]^{-1} - \tilde{\Pi}(\vec{q}, \omega)\,, \tag{3.26}$$

where the zeroth-order Green's function is

$$[\tilde{G}^{(0)}(\vec{q}, \omega)]^{-1} = \begin{pmatrix} \omega & 0 \\ 0 & -\omega \end{pmatrix}. \tag{3.27}$$

The diagrams for the self-energy matrix $\tilde{\Pi}$ are shown in Fig. 5. The two-magnon terms in H_1 give the graphs in Fig. 5(a). They contribute to the diagonal part of the self-energy matrix

$$\Pi^{(2)}_{11}(\vec{q}, \omega) = \Pi^{(2)}_{22}(\vec{q}, \omega)$$
$$= \frac{\Delta}{NS}\sum_{\vec{k}} (\langle C^\dagger_{1\vec{k}\uparrow}\, C_{2,\vec{k}+\vec{Q}\uparrow}\rangle$$
$$- \langle C^\dagger_{1\vec{k}\downarrow}\, C_{2,\vec{k}+\vec{Q}\downarrow}\rangle + \text{c.c.})$$
$$= 4\Delta^2/US\,. \tag{3.28}$$

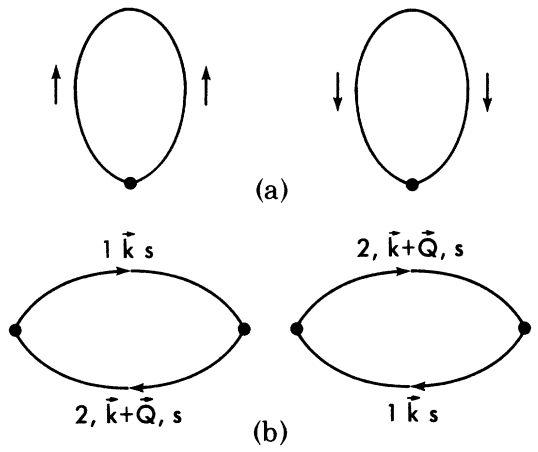

FIG. 5. Self-energy diagrams for the antiferromagnetic spin waves.

The one-magnon terms in H_1 give the diagrams in Figs. 5(b) and 5(c). The rules for computing these diagrams are given by Fedders and Martin.[35] We find the following diagonal elements:

$$\Pi^{(1)}_{11}(\vec{q}, \omega) = \Pi^{(1)}_{22}(\vec{q}, \omega) = I(\vec{q}, \omega)\,,$$

where

$$I(\vec{q}, \omega) = -\frac{2\Delta^2}{NS}\sum_{\vec{k}} \frac{1}{4}\left(1 + \frac{\epsilon_{\vec{k}}\epsilon_{\vec{k}+\vec{q}} + \Delta^2}{E_{\vec{k}}E_{\vec{k}+\vec{q}}}\right)$$
$$\times \frac{2}{E_{\vec{k}} + E_{\vec{k}+\vec{q}} - \omega}\,; \tag{3.29}$$

and the following off-diagonal elements:

$$\Pi^{(1)}_{12}(\vec{q}, \omega) = \Pi^{(1)}_{21}(\vec{q}, \omega) = -I(\vec{q}, \omega)\,. \tag{3.30}$$

Putting these results into Eq. (3.26) we find that the poles of $\tilde{D}(\vec{q}, \omega)$ are solved from

$$\begin{vmatrix} \omega - 4\Delta^2/US - I(\vec{q}, \omega) & I(\vec{q}, \omega) \\ I(\vec{q}, \omega) & -\omega - 4\Delta^2/US - I(q, \omega) \end{vmatrix} = 0 \tag{3.31}$$

or

$$\omega^2 = 2(4\Delta^2/US)[2\Delta^2/US + I(\vec{q}, \omega)]\,.$$

We expand $I(\vec{q}, \omega)$ for small q and ω. The result is

$$I(\vec{q}, \omega) = -(2\Delta^2/US)\{1 + [N(E_F)U/4\Delta^2](\omega^2 - \tfrac{1}{3}v_F^2 q^2)\}\,, \tag{3.32}$$

where $v_F = \hbar k_F/m$ is the Fermi velocity. Then, in the lowest order in q, the magnon dispersion relation is found to be $\omega = cq$, where

$$c = [\xi/(1+\xi)]^{1/2} v_F/\sqrt{3} \tag{3.33}$$

and $\xi = 4\Delta^2 N(E_F)/US^2$. The magnon velocity is lower than the Fedders-Martin result by the factor $[\xi/(1+\xi)]^{1/2}$.

There are a number of uncertainties in estimating the size of ξ. The energy gap and the moment per atom for the 2%-Mn alloy can be taken from the experimental data, $\Delta = 0.2$ eV,[41] and $S = 0.28$.[39] The product $N(E_F)U$ has been estimated as 0.43.[37] It is difficult to pin down the values of $N(E_F)$ and U separately because it is not certain how much of the real Fermi surface can nest. However, for most transition metals U is of the order 2–3 eV. Within this uncertainty, we esti-

mate $\xi = 0.10-0.22$, or the reduction factor $[\xi/(1+\xi)]^{1/2} = 0.30-0.42$. The estimated band velocity is $v_F = 5.1\times10^7$ cm/sec, with the phonon mass enhancement factor included.[36] So the estimated magnon velocity is $c = (0.9-1.2)\times10^7$ cm/sec. This is consistent with the experimental value 1.3×10^7 reported by Sinha *et al.*[36] and 1.55×10^7 reported by Als-Nielsen *et al.*[38] Thus, the quasispin theory removes a long-standing discrepancy between theory and experiment.

ACKNOWLEDGMENTS

The author is indebted to Professor L. M. Falicov, Professor M. L. Cohen, Professor S. K. Sinha, Professor C. Stassis, and Professor B. N. Harmon for discussions and criticisms. He also wishes to thank Professor Falicov for inviting him to spend a year at the University of California, Berkeley.

APPENDIX A

We prove here that the magnon frequency given by Eq. (2.15) is zero at zero wave vector. This is accomplished by showing that $\Pi(0,0)=0$. We define

$$\psi_{n\vec{k}\sigma} = \phi_{n\vec{k}\sigma}(\vec{r})\xi_\sigma ,$$

where ξ_σ is the Pauli spinor. Then $\psi_{n\vec{k}\sigma}$ is an eigenstate of the Hamiltonian

$$H = p^2/2m + V(r) - \sigma^z W(\vec{r}) .$$

The self-energy $\Pi(0,0)$ may be written

$$\Pi(0,0) = \frac{1}{NS}\sum_{n\vec{k}\sigma} f_{n\vec{k}\sigma}\langle\psi_{n\vec{k}\sigma}|W_0\sigma^z|\psi_{n\vec{k}\sigma}\rangle + \frac{1}{2NS}\sum_{n\vec{k}\sigma}\sum_{n'\vec{k}'\sigma'}\frac{f_{n\vec{k}\sigma}-f_{n'\vec{k}'\sigma'}}{E_{n\vec{k}\sigma}-E_{n'\vec{k}'\sigma'}} \times|\langle\psi_{n\vec{k}\sigma}|W_0\sigma^-|\psi_{n'\vec{k}'\sigma'}\rangle|^2 .$$

There is the identity

$$[H,\sigma^-] = 2\sigma^- W(\vec{r}) .$$

We take the matrix element in a unit cell to obtain

$$\langle\psi_{n\vec{k}\sigma}|W_0\sigma^-|\psi_{n'\vec{k}'\sigma'}\rangle = \tfrac{1}{2}(E_{n\vec{k}\sigma}-E_{n'\vec{k}'\sigma'})\langle\psi_{n\vec{k}\sigma}|\sigma^-|\psi_{n'\vec{k}'\sigma'}\rangle .$$

The last term of $\Pi(0,0)$ is equal to

$$\frac{1}{4NS}\sum_{n\vec{k}\sigma}\sum_{n'\vec{k}'\sigma'}(f_{n\vec{k}\sigma}-f_{n'\vec{k}'\sigma'})\langle\psi_{n\vec{k}\sigma}|\sigma^-|\psi_{n'\vec{k}'\sigma'}\rangle \times\langle\psi_{n'\vec{k}'\sigma'}|W_0\sigma^+|\psi_{n\vec{k}\sigma}\rangle = \frac{1}{4NS}\sum_{n\vec{k}\sigma} f_{n\vec{k}\sigma}\langle\psi_{n\vec{k}\sigma}|W_0[\sigma^-,\sigma^+]|\psi_{n\vec{k}\sigma}\rangle = -\frac{1}{NS}\sum_{n\vec{k}\sigma} f_{n\vec{k}\sigma}\langle\psi_{n\vec{k}\sigma}|W_0\sigma^z|\psi_{n\vec{k}\sigma}\rangle ,$$

which exactly cancels the first term. This completes the proof.

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*Part of this work was done at Ames Laboratory—U. S. ERDA under Contract No. W-7405-eng-82.

†On leave of absence from Ames Laboratory—ERDA and Dept. of Physics, Iowa State University, Ames, Ia. 50010.

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