

Two-magnon bound states in itinerant electron ferromagnets*

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A model of a single-narrow-band itinerant-electron ferromagnet is considered. An effective magnon Hamiltonian is introduced to describe magnons and magnon interactions in the itinerant ferromagnet. By the use of the effective magnon Hamiltonian, the problem of two-magnon bound states reduces to solving an integral equation. The equation is solved for a simple cubic lattice using the tight-binding electron energy. The two-magnon bound states are found to exist for values of the total momentum of the interacting magnons close to the Brillouin-zone boundary.

I. INTRODUCTION

In recent years, there has been growing interest in studying the problem of magnon bound states. The importance of the problem is due to the fact that the existence of magnon bound states imposes limits on the range of applicability of the linear or noninteracting spin-wave theory, and is also due to the experimental possibilities of studying the effects of magnon interactions.

Magnon bound states were extensively studied theoretically for the Heisenberg model of ferromagnets and antiferromagnets. The problem was first considered by Bethe¹ in 1931, who showed that in the one-dimensional Heisenberg system of spin $S = \frac{1}{2}$, with positive exchange integral between nearest neighbors, there exist bound states of two, or more, spin waves, their energies being always lower than the sum of energies of free spin waves having the same total momentum \vec{K} . The two-magnon states were then considered by Dyson² in 1956, who proved that in two- and three-dimensional Heisenberg ferromagnets, the two-magnon bound states do not exist for the total momentum $\vec{K} = 0$. In 1963 Hanus,³ for spin $S = \frac{1}{2}$, and Wortis,⁴ for arbitrary S , found that in the simple cubic nearest-neighbor interaction Heisenberg model in the two-dimensional case, two-magnon bound states exist for any $\vec{K} \neq 0$, even for \vec{K} arbitrarily small, whereas in three dimensions there is a region of small \vec{K} for which bound states do not exist; however, they appear near the Brillouin-zone boundary for \vec{K} exceeding a threshold value. The problem of magnon bound states in the Heisenberg spin systems was then discussed in numerous papers.⁵⁻¹¹ In particular, bound states in antiferromagnets were predicted.⁹ Magnon bound states were discovered experimentally in a simple spin system.¹² Various possibilities of experimental investigations of magnon pairing effects were reviewed.¹³

The problem of magnon bound states has an important implication for determining the range of

validity of the linear or noninteracting spin-wave theory. Practical applications of the spin-wave theory are based on the assumption that the superposition of single magnon states holds, approximately. For the regions of values of the total momentum of spin waves for which bound states exist and have energy lower than the sum of energies of free spin waves, the superposition principle breaks down.

Because of the relevance of the magnon bound-state problem to the fundamentals of the spin-wave theory, it is interesting to study the same problem in itinerant-electron ferromagnets. In the itinerant-electron theory of ferromagnetism, a magnon is a bound state of an electron and a hole of opposite spins.^{14,15} The problem of a two-magnon bound state in itinerant-electron ferromagnets is essentially a four-body problem, two electrons and two holes, and is extremely difficult to solve by direct methods. In the present paper, we describe a solution of the problem based on the concept of an effective magnon Hamiltonian,¹⁶ which appeared to be very useful for treating interactions of magnons.^{17,18} The effective magnon Hamiltonian is constructed from combinations of products of electron operators, which in the random-phase approximation (RPA) have the properties of magnon creation and annihilation operators. In the method of the effective Hamiltonian, the task of finding the bound state reduces to solution of a two-body problem, although with a rather complicated kernel in the ensuing integral equation. The equation can be solved only numerically. We consider here the single-narrow-band model of itinerant electrons, use the tight-binding approximation, and consider the simple cubic three-dimensional lattice.

The solution of a similar problem for a one-dimensional case was recently reported.¹⁸ It was shown that for the one-dimensional model, two-magnon bound states exist for all values of the total momentum \vec{K} , so the qualitative behavior is the same as for the Heisenberg one-dimensional sys-

tems.

The format of the paper is as follows. In Sec. II, we outline the method of the effective magnon Hamiltonian; in Sec. III, the integral equation for the two-magnon bound state is derived. The algorithm used for solving this equation for a three-dimensional lattice is described, and results of computations are presented in Sec. IV. Some general remarks on the results are presented in Sec. V.

II. EFFECTIVE MAGNON HAMILTONIAN

Our discussion of magnon bound states in itinerant-electron ferromagnets will be based on the Hubbard¹⁹ model. It is generally believed (e.g., see Ref. 15) that a realistic description of ferromagnetism in itinerant electron $3d$ transition metals like Ni or Co should take into account multiple bands and also include other interactions, not only the leading intra-atomic Coulomb one. For a problem of great mathematical complexity, as the magnon bound-state problem, it is reasonable to start with simplest possible model qualitatively simulating the behavior of itinerant electron ferromagnets.

We consider a system of itinerant electrons in a single narrow band, described by the Hubbard Hamiltonian

$$\mathcal{H} = \sum_{k\sigma} \epsilon_k a_{k\sigma}^\dagger a_{k\sigma} + \frac{I}{N} \sum_{kk'q} a_{k+q}^\dagger + a_{k'-q}^\dagger - a_{k'} - a_{k,+} \quad (1)$$

The standard notation is used: $a_{k\sigma}^\dagger$ ($a_{k\sigma}$) denotes the creation (annihilation) operators for electrons of spin $\hat{\sigma}$ in the Bloch state specified by the wave vector \vec{k} . The Bloch energy is denoted by ϵ_k (if the magnetic field H is present, the Bloch energy has to be replaced by $\epsilon_k \pm \mu_B H$), I is the intra-atomic Coulomb integral, and N is the number of atoms in the crystal.

We assume that the ground state $|\phi_0\rangle$ of the Hubbard Hamiltonian (1) is strongly ferromagnetic with all electrons having spins down. It is well known^{14,20} that in the (RPA), the operator

$$\beta_q^\dagger = \sum_k b_{k+q,k} a_{k+q,+}^\dagger + a_{k,-} \quad (2)$$

generates a one-magnon state in the sense that $\beta_q^\dagger |\phi_0\rangle$ is, within RPA, the eigenstate of \mathcal{H} , and corresponds to a bound state of an electron of reversed spin, and a hole in the Fermi sea. The energy E_q of that bound state or magnon is determined from the equation

$$\frac{I}{N} \sum_k n_k (\epsilon_{k+q} - \epsilon_k + \Delta - E_q)^{-1} = 1, \quad (3)$$

where $\Delta = In$ (or $\Delta = In + 2\mu_B H$) is the exchange (or exchange and Zeeman) splitting, n is the number of itinerant electrons per atom, and $n_k = \langle \phi_0 | a_{k-}^\dagger a_{k-} | \phi_0 \rangle$; obviously, $n_k = 1$ for ϵ_k below the Fermi energy E_F and $n_k = 0$, otherwise.

The coefficients in (2) are

$$b_{k+q,k} = d_q / (\epsilon_{k+q} - \epsilon_k + \Delta - E_q), \quad (4)$$

as can be easily found solving in RPA equation of motion for β_q^\dagger . If the normalization constant d_q is determined from the condition

$$\sum_k |b_{k+q,k}|^2 n_k = 1, \quad (5)$$

the operators β_q^\dagger and their Hermitian adjoint β_q satisfy in RPA the commutation rules $[\beta_q, \beta_{q'}^\dagger]_{\text{RPA}} = \delta_{qq'}$. We also have $[\beta_q^\dagger, \beta_{q'}^\dagger] = 0$.

The set of operators β_q^\dagger and β_q , satisfying (in RPA) boson commutation relations can be interpreted as magnon creation and annihilation operators. For problems of magnon interactions, it is convenient to extract from the general Hamiltonian \mathcal{H} [Eq. (1)] that part which corresponds to the energy of the magnons, and the energy of their interaction. Obviously, such a program cannot be implemented exactly, because magnons are not exact normal modes of the system, even apart from their mutual interaction, since actually a one magnon state $\beta_q^\dagger |\phi_0\rangle$ is not an exact eigenstate of \mathcal{H} as a result of residual interactions of magnons with electrons. However, a consistent approximate procedure working within RPA can be used to derive from \mathcal{H} an effective Hamiltonian which is the energy of the magnons and their interaction. Such procedures have been successfully used in studying magnon relaxation and other problems.¹⁶⁻¹⁸ Similar methods of extracting the energy of boson-type excitations in systems of interacting fermions were used in the theory of nuclear matter.²¹

The effective magnon Hamiltonian is defined as an expansion in powers of the magnon operators β_q^\dagger and β_q . The expansion can be simplified from the outset by taking into account general properties of the primary Hamiltonian of the system of itinerant electrons. The Hubbard Hamiltonian \mathcal{H} [Eq. (1)] conserves the total magnetic moment of the system $\sum_k (a_{k-}^\dagger a_{k-} - a_{k+}^\dagger a_{k+})$. Therefore, the effective magnon Hamiltonian which has to be equivalent to \mathcal{H} in the subspace of magnon states should conserve the total number of magnons, which means that it can contain only products of equal numbers of operators β^\dagger and β . Further restrictions come from the translational symmetry requiring conservation of the crystal momentum: in every term of the effective Hamiltonian the sum of wave vectors of created magnons must be equal to the sum for annihilated magnons. The general expression for the

effective magnon Hamiltonian \mathcal{H}_{eff} equivalent to (1) in the subspace of magnon states compatible with the above mentioned restrictions and with neglect of higher order terms is given by

$$\mathcal{H}_{\text{eff}} = \sum_q K_q \beta_q^\dagger \beta_q + \sum_{kk'q} \Gamma_{kk'}^q \beta_{k+q}^\dagger \beta_{k'-q}^\dagger \beta_k \beta_{k'} + \dots \quad (6)$$

The coefficients are given by the following ground-state averages of multiple commutators¹⁶:

$$K_q = \langle \phi_0 | [\beta_q, [\mathcal{H}, \beta_q^\dagger]] | \phi_0 \rangle, \quad (7)$$

$$\Gamma_{kk'}^q = \frac{1}{4} \langle \phi_0 | [\beta_{k+q}, [\beta_{k'-q}, [[\mathcal{H}, \beta_k^\dagger], \beta_{k'}^\dagger]]] | \phi_0 \rangle. \quad (8)$$

The derivation¹⁶ of the relations (7), (8) is based on equivalence of the ground-state averages of commutators like

$$[\beta_q, [\mathcal{H}_{\text{eff}}, \beta_q^\dagger]] \quad \text{and} \quad [\beta_q, [\mathcal{H}, \beta_q^\dagger]].$$

From (7) follows the expression

$$K_q = \sum_k (\epsilon_{k+q} - \epsilon_k + \Delta) |b_{k+q,k}|^2 n_k - \left(\frac{N}{I}\right) |d_q|^2$$

which, on using (4) and (3), reduces to $K_q = E_q$, where E_q is the magnon energy determined by Eq. (3), as it should be for consistency. The coefficients of the interaction terms in the effective magnon Hamiltonian (6) are given by¹⁶

$$\Gamma_{kk'}^q = \frac{1}{4} (B_{kk'}^q + B_{kk'}^{-q} + E_{kk'}^{k'-k-q} + B_{kk'}^{-k'+k+q}), \quad (9)$$

$$B_{kk'}^q = \frac{I}{N} \sum_{pp'} (b_{p+k',p} - b_{p'+k',p'}) \times b_{p+k+q,p+q} b_{p+k'-q,p} b_{p+k+q,p} n_p n_{p'}. \quad (10)$$

Higher-order terms in the effective Hamiltonian, if necessary for a particular problem, can be calculated by the same procedure, although with much

more labor.

From now on we are working in the space of magnon states, the effective Hamiltonian (6) is of the same form as the magnon Hamiltonian for the Heisenberg ferromagnets. This formal analogy will be used now to study the problem of two-magnon bound state by methods developed for the Heisenberg ferromagnets.^{9,4}

III. TWO-MAGNON BOUND STATE

Let us consider two magnons of wave vectors $\frac{1}{2}\vec{K} + \vec{k}$ and $\frac{1}{2}\vec{K} - \vec{k}$. The total wave vector \vec{K} is the constant of motion and is used as the quantum number for a general two-magnon state defined by

$$|K\rangle = \sum_k g_k \beta_{K/2+k}^\dagger \beta_{K/2-k}^\dagger | \phi_0 \rangle, \quad (11)$$

where the summation on the relative momentum of the pair extends over the first Brillouin zone. The coefficients g_k of the superposition (11) and the energy ω of the two-magnon bound state are calculated from the Schrödinger equation $\mathcal{H}_{\text{eff}} |K\rangle = \omega |K\rangle$ which leads to the following integral equation:

$$[\omega - \Omega(K, k)] g_k = \sum_q V(K, k, q) g_q. \quad (12)$$

The solution of Eq. (12) for a one-dimensional itinerant-electron system was reported recently.¹⁸ Now we solve (12) for a three-dimensional, simple cubic lattice, using as previously the tight-binding approximation (with one parameter) for the band energy of electrons. Since the coefficients $g_k = g(k_x, k_y, k_z)$ have the following symmetry properties:

$$g(k_x, k_y, k_z) = g(-k_x, k_y, k_z) \\ = g(k_x, -k_y, k_z) = g(k_x, k_y, -k_z),$$

we have

$$\Omega(K, k) = \frac{1}{4} \sum_{\alpha_x, \alpha_y, \alpha_z = \pm 1} E_{\vec{K}/2 + (\alpha_x k_x, \alpha_y k_y, \alpha_z k_z)}, \quad (13)$$

and

$$V(K, k, q) = \frac{1}{8} \sum_{\substack{\alpha_x, \alpha_y, \alpha_z \\ \beta_x, \beta_y, \beta_z = \pm 1}} \Gamma_{\vec{K}/2 + (\beta_x q_x, \beta_y q_y, \beta_z q_z), \vec{K}/2 - (\alpha_x q_x, \alpha_y q_y, \alpha_z q_z)}, \quad (14)$$

where the expressions in parentheses denote vector components, and each of the summation indices runs independently over a set of only two elements (+1, -1).

IV. DETAILS OF COMPUTATIONS

In order to solve (12) we have to first calculate the energies of free magnons E_q from (3), and then

the interaction matrix elements $\Gamma_{\mathbf{k},\mathbf{k}}$, from (9). All these calculations have to be done numerically. Calculations for a realistic band structure would require very large amounts of computer time. Therefore, the computations were performed for a model of an itinerant-electron ferromagnet, namely, for a band structure given by the simplest tight-binding formula, and for the simple cubic lattice. We assume

$$\epsilon_{\mathbf{k}} = \frac{1}{6} W(3 - \cos ak_x - \cos ak_y - \cos ak_z), \quad (15)$$

where a denotes the lattice constant. Values of the material constants were taken as appropriate for ferromagnetic nickel. The bandwidth W or rather its ratio to the Fermi energy, W/E_F is determined by the number of itinerant electrons per atom $n = N^{-1} \sum_{\mathbf{k}} n_{\mathbf{k}}$. Taking $n = 0.6$, as for Ni, we obtain $E_F/W = 0.559$. For $E_F = 0.3$ eV, as appropriate for Ni,^{15,22} we have the value $W = 0.536$ eV for the bandwidth. For the exchange splitting $\Delta = nI$, the value $\Delta = 0.57$ eV was taken according to recent estimates^{23,24} for Ni.

The free magnon energies E_q calculated for the directions [100], [110], and [111] are plotted in Fig. 1. For the values of material parameters adopted in our calculations, the magnons exist for all wave vectors \vec{q} in the first Brillouin zone, i.e., the magnon energy E_q always lies below the continuum of the Stoner excitations $E_q < \min_{\mathbf{k}}(\epsilon_{\mathbf{k}+\mathbf{q}} - \epsilon_{\mathbf{k}} + \Delta)$. High anisotropy of the magnon energy is a feature of our simple model and makes our final conclusions only qualitatively applicable to nickel.

In order to solve the integral equation (12), it is necessary first to compute the kernel $V(K, \mathbf{k}, q)$ for a given \vec{K} and for all \vec{k} and \vec{q} in the first Brillouin

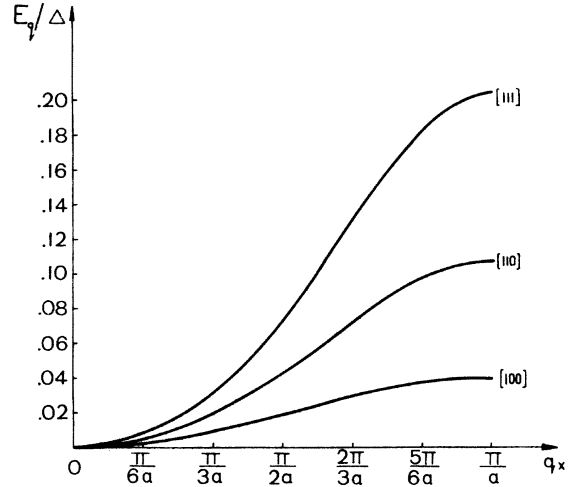


FIG. 1. Magnon energies for the high-symmetry directions [100], [110], [111].

zone. We restrict the calculations only to the case of \vec{K} parallel to the direction [111]. Computations of $V(K, \mathbf{k}, q)$ for all points \vec{k} and \vec{q} for a reasonably fine mesh which spanned the first Brillouin zone would need enormous computer capacity to give sufficiently accurate results. Therefore, the following interpolation procedure was used: for a fixed value of \vec{K} , values of the function $V(K, \mathbf{k}, q)$ were calculated for several values of \vec{k} and \vec{q} , for \vec{k} and \vec{q} parallel to the high symmetry directions [100], [110], and [111]. Then an interpolation analytic formula for $V(K, \mathbf{k}, q)$ was proposed in a form of an expansion into symmetry invariants

$$\begin{aligned} V(K, \mathbf{k}, q) = & A_0 + A_1(x+y+z) + A_2(X+Y+Z) + A_3(xX+yY+zZ) + A_4[x(Y+Z)+y(Z+X)+z(X+Y)] + A_5(xy+xz+yz) \\ & + A_6(XY+XZ+YZ) + A_7[xX(Y+Z)+yY(X+Z)+zZ(X+Y)] + A_8(xYZ+yXZ+zXY) \\ & + A_9[xy(X+Y)+xz(X+Z)+yz(Y+Z)] + A_{10}(xyZ+xzY+yzX) + A_{11}xyz + A_{12}XYZ + A_{13}(x+y+z)XYZ \\ & + A_{14}(xyXY+xzXZ+yzYZ) + A_{15}[xyZ(X+Y)+xzY(X+Z)+yzX(Y+Z)] + A_{16}xyz(X+Y+Z) \\ & + A_{17}(xy+xz+yz)XYZ + A_{18}xyz(XY+XZ+YZ) + A_{19}xyzXYZ, \end{aligned} \quad (16)$$

where the following abbreviations are introduced:

$$\begin{aligned} x &= \cos ak_x, & y &= \cos ak_y, & z &= \cos ak_z, \\ X &= \cos aq_x, & Y &= \cos aq_y, & Z &= \cos aq_z. \end{aligned}$$

The values of the coefficients A_i for different $\vec{K} = K_x(1, 1, 1)$, were determined by the least-squares fit to calculated directly values of $V(K, \mathbf{k}, q)$. The interpolation formula reproduced

$V(K, \mathbf{k}, q)$ within an accuracy better than 5%.

The kernel $V(K, \mathbf{k}, q)$ given by the interpolation formula (16) is degenerate, it can be written in the form

$$V(K, \mathbf{k}, q) = \sum_i a_i(K, \mathbf{k}) b_i(K, q), \quad (17)$$

with $1, x, y, z, xy, yz, zx, xyz$ as a_1, \dots, a_8 and b_i easy to find from (16), and the integral equation

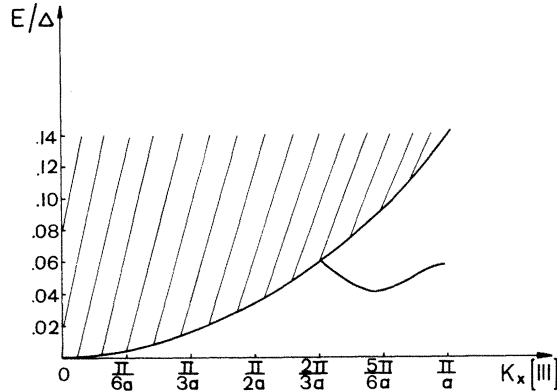


FIG. 2. Energy spectrum of the two-magnon bound states for $\vec{K} = K_x(1, 1, 1)$. The shaded area represents part of the quasicontinuum of free two-magnon states, its lower boundary is given by $\min_k (E_{K/2+k} + E_{K/2-k})$.

(12) reduces to solving the set of linear algebraic equations

$$\sum_j [\delta_{ij} - G_{ij}(\omega)] \sum_q b_j(K, q) g_q = 0, \quad (18)$$

where

$$G_{ij}(\omega) = \sum_q \frac{a_i(K, q) b_j(K, q)}{\omega - \Omega(K, q)}. \quad (19)$$

The energy eigenvalue ω , which is the energy of the two-magnon bound state, is calculated from the usual condition of the vanishing of the determinant $D(\omega) = \det[\delta_{ij} - G_{ij}(\omega)]$ of the system (18). For a fixed value of \vec{K} we compute $G_{ij}(\omega)$ and then $D(\omega)$ for consecutive values of ω , locating finally that value of ω for which $D(\omega) = 0$.

The computations showed that in the interval of K_x ranging from 0 to about $2\pi/3a$ there are no so-

lutions of (18), whereas for each K_x from $2\pi/3a < K_x \leq \pi/a$ there is one solution for energy $\omega = \omega(\vec{K})$ lying below the quasicontinuum of energies of free magnons. The calculated energy spectrum $\omega(\vec{K})$ of the two-magnon states is exhibited in Fig. 2.

V. CONCLUSIONS

The energy spectrum of the two-magnon bound states in our model of an itinerant-electron ferromagnet has some resemblance to the bound-states spectrum in the Heisenberg ferromagnets.⁴ In both models, the two-magnon bound states exist for the total momentum K greater than a critical value K_c , typically, K_c is near the Brillouin zone boundary (in our case, for \vec{K} parallel to the [111] direction, $K_c = 2\pi/a\sqrt{3}$). There are also differences. We have found only one branch of the two-magnon energy spectrum, whereas in three-dimensional Heisenberg ferromagnets there are two branches,⁴ one of them being doubly degenerate. By our method of computations we are unable to assess any eventual degeneracy of the two-magnon spectrum. Besides, we have found a shallow minimum in the dispersion curve for the two-magnon states, whereas the corresponding curves for the Heisenberg ferromagnets are monotonic. This result might well be only a feature of our simplified model.

Our simplified model of the band structure exhibits some unrealistic features as applied to nickel. In nickel, the experimentally determined magnon energies enter into the continuum of the Stoner excitations at a critical value q_s of the magnon wave vector.²⁵ Therefore, in nickel, the additional conditions $|\frac{1}{2}\vec{K} \pm \vec{k}| < q_s$ should be imposed, and the two-magnon bound states may only exist for K from the interval $K_c < K \lesssim 2q_s$. Unfortunately, the experimental value²⁵ of $2q_s$ is quite close to K_c .

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