

the angular dependence of the resonance fields and the static torques with this value of  $\Delta_{\text{ex}}$ . For such a small value of  $\Delta_{\text{ex}}$  one cannot neglect the coupling of states differing in  $S_{\theta} = m$  by the effective Hamiltonian given by Eq. (6). Apart from a less important splitting of the doublets for  $\vec{H}_{\text{ex}}$  close to the [111] direction this coupling gives an angular-dependent second-order energy, which is equal for the states  $|E_{\epsilon}, 2\rangle$  and  $|E_{\theta}, 2\rangle$ :

$$\delta E = -(\delta^2/4\Delta_{\text{ex}})[1 + 11(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2)]. \quad (20)$$

With  $\Delta_{\text{ex}} = 62 \text{ cm}^{-1}$  this gives at low temperatures, where  $kT \ll \Delta_{\text{ex}}$ , a negative  $K_1$  of  $7.5 \text{ cm}^{-1}$  per  $\text{Fe}^{2+}$  in addition to the positive anisotropy as given by Eq. (10). However with such a large negative  $K_1$  the angular dependence of the torques and the resonance fields cannot be fitted. A correct fit at 4.2 K can only be obtained with  $\Delta_{\text{ex}} \geq 200 \text{ cm}^{-1}$ , which confirms the estimate from the Curie temperature of  $\text{FeCr}_2\text{S}_4$  (Sec. II). An exchange splitting of that magnitude gives a negative  $K_1$  com-

parable to the positive  $K_1$  expected from the  $\text{Cr}^{3+}$  ions, which anisotropy could be neglected (Sec. IVA).

The value of  $13 \text{ cm}^{-1}$  of the parameter  $\delta = 6(\lambda^2/\Delta_c + \rho)$  compares nicely with the value of  $12 \text{ cm}^{-1}$  from the Mössbauer spectrum of  $\text{Cd}_{0.98}\text{Fe}_{0.02}\text{Cr}_2\text{S}_4$ .<sup>13</sup>

In conclusion, the anisotropy of  $\text{Cd}_{1-x}\text{Fe}_x\text{Cr}_2\text{S}_4$  at helium temperatures can be described very well by the single-ion anisotropy of the ferrous ion on the tetrahedral site. Along the [111] direction, where the doublet levels of the ferrous ion are expected to come to a near crossover, the simple model fails below 4 K. This may be due to a small splitting of the lowest doublet and to relaxation between the doublet levels.

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## Anomalous Damping of Spin Waves in Magnetic Metals\*

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An analysis of long-wavelength magnon damping in magnetic metals is presented in the context of an itinerant-electron picture of ferromagnetism. The decay is into single-particle excitations, and is a result of spin-orbit forces. The predicted dependence of the damping constant on the electron mean free path is in accord with recent experiments on ferromagnetic resonance. In particular, for pure metals it increases as the temperature is lowered, and then saturates. This anomalous rise is closely related to the anomalous electrical conductivity.

### I. INTRODUCTION

Ferromagnetism in the  $d$ -band metals is a phenomenon which has resisted complete understanding for many years, despite the technological importance of these materials and the fundamental chal-

lenge to theorists which has long been recognized.<sup>1</sup>

A major problem for the theory is to provide a model which accounts for the main features of the ground-state energetics. In any many-body theory, however, the discussion of the elementary excitations also plays a central role. Fortunately, it is

not always necessary to have a completely adequate theory of the ground state before developing theories of the elementary excitations. We shall rely on this circumstance in the present work, which does not bear on the problem of the ground-state properties.

The elementary excitation most characteristic of a magnet is, of course, the spin wave or magnon. As compared with the insulating magnets, the main complication encountered in the case of metals is that there are present over the same energy range the single-particle excitations associated with the conduction electrons. This circumstance is ultimately responsible for the relatively short lifetime of magnons in metals. Models in which localized magnetic electrons interact with nearly free itinerant electrons are quite common, and indeed offer a good account of the rare-earth ferromagnets. Such models do not literally apply to the  $d$ -band ferromagnetic metals, since there is strong evidence that the magnetic electrons are themselves itinerant, although they retain some properties of localized spins. It is this localized-vs-itinerant dichotomy of the magnetic electrons which has so far resisted theoretical understanding.<sup>1,2</sup>

We shall in this work adopt the band model of ferromagnetism, even though it fails to account correctly for the localized aspect of itinerant electrons. A localized-magnetic-electron-itinerant-conduction-electron model also gives qualitatively similar results, and we believe that the Landau-Fermi liquid model<sup>3</sup> should do the same. In the problem to which we address ourselves the localization of the electrons does not play a conspicuous role.

What we wish to study is the interaction of the spin wave with the single-particle excitations. Whatever the spin wave is, it certainly has a strong interaction via exchange with the conduction electrons. Then, at the least, the spin wave is strong-

ly renormalized, most simply through the process in which the magnon becomes a virtual spin-up particle spin-down hole pair.

In fact, we shall regard the spin wave as being nothing but a sequence of virtual spin-up particle spin-down hole pairs, successively scattered by means of the exchange potential. The Feynman-graph description of this is given in Fig. 1.

This treatment of the exchange interaction (the random-phase approximation) gives the dominant features of the spin-wave dispersion and has already been widely studied.<sup>1,4</sup> It cannot account, however, for the main features of the real decay of the spin wave into other excitations, particularly into the single-particle excitations.

The fact is that the exchange interaction conserves spin, and therefore the infinite-wavelength spin wave cannot decay at all through this mechanism. At finite wave number, diffusionlike processes can occur which give damping rates proportional to  $q^4$ . Much effort has been expended on the evaluation of such terms but they are generally known to be negligible under the experimental conditions prevailing in the study of ferromagnetic metals.<sup>5,6</sup>

Thus it is necessary to invoke the spin-orbit interaction if one wishes to study the possibility of decay of spin waves into other elementary excitations in the long-wavelength case.

This program has been worked on for some time, but no mechanism in agreement with experiment has been put forward until the present work.<sup>7-9</sup> The main problem is that the spin-orbit coupling is quite difficult to handle, being off-diagonal in both spin and space variables. In order to get large enough spatial matrix elements of the spin-orbit interaction, one has to deal with a nontrivial band structure. What is more, one has to take into account real imperfection or phonon scattering in order to dissipate the momentum carried by the

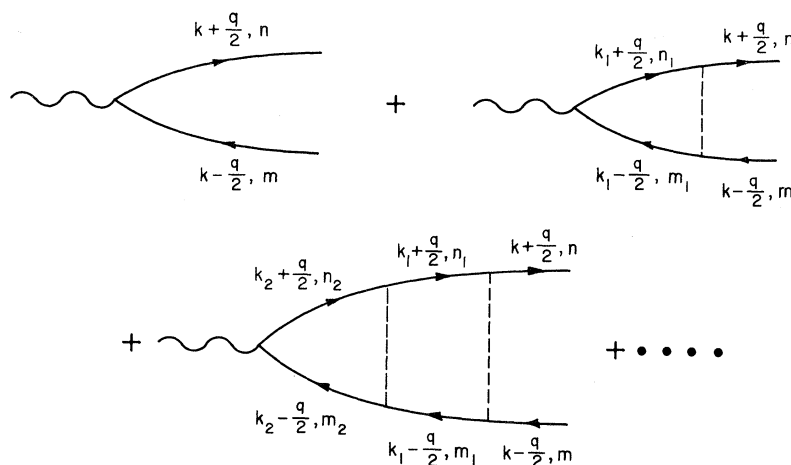


FIG. 1. Diagrammatic representation of the random-phase approximation for the magnon propagator. Solid lines represent electrons, dashed lines the screened Coulomb interaction  $V_C$ .

spin wave.

The only mechanisms which seem to have a chance of giving acceptably large decay rates are based on the process of the spin wave decaying into an electron-hole pair, but without the conservation of spin. The spin nonconservation can come about either through a real scattering in which spin-orbit effects are involved, or through a virtual modification of the exchange interaction, introducing an effective exchange interaction which no longer conserves spin. The former case does not seem to be in agreement with experiment, as the real scattering is bound to have an important temperature dependence of a type which is not observed.

The latter possibility is the subject of the present paper, and it gives rise to a decay whose temperature (i. e., mean-free-path) dependence is in qualitative agreement with experiment.

The propagation of a magnon and its damping are studied experimentally by observing the response of a magnetic system to an imposed (microwave) magnetic field (ferromagnetic resonance).<sup>10</sup> The experiments are analyzed in terms of the phenomenological Landau-Lifshitz-Gilbert<sup>11</sup> equation, which describes the evolution of long-wavelength slowly varying spin deviations. This equation is written

$$\frac{\partial \vec{M}}{\partial t} = \gamma \vec{M} \times \left( \vec{H} + \frac{D}{\gamma M_s} \nabla^2 \vec{M} \right) + \frac{\lambda}{\gamma M_s^2} \left( \vec{M} \times \frac{\partial \vec{M}}{\partial t} \right), \quad (1)$$

where  $\gamma$  is the gyromagnetic ratio,  $M_s$  is the saturation magnetization, and  $D$  is the exchange stiffness. The damping is given in terms of the parameter  $\lambda$ , which is the main object of our attention. Alternative phenomenological equations are in much worse accord with experiment,<sup>10</sup> and our theory, in fact, predicts that Eq. (1) is correct. Equation (1) gives the linear response of the magnetization to the local magnetic field. This response is a dynamic wave number and frequency-dependent susceptibility, which is then used to connect  $\vec{B}$  and  $\vec{H}$  in Maxwell's equations. Maxwell's equations are subsequently solved with appropriate boundary conditions to yield the required system response.

Although the actual magnons excited are more complicated, it is sufficient to study the response to a right circularly polarized local microwave field, which drives a pure right circularly polarized spin wave. According to Eq. (1) the magnetic response to such a field  $h^*(\vec{q}, \Omega)$  is

$$M^*(\vec{q}, \Omega) = \chi^*(\vec{q}, \Omega) h^*(\vec{q}, \Omega), \quad (2)$$

where

$$\chi^*(\vec{q}, \Omega) = \frac{-\gamma M_s}{\Omega - \gamma H - Dq^2 - i\Omega\lambda/\gamma M_s}. \quad (3)$$

Clearly, Eq. (3) describes the propagation of a

spin wave, with a damping term characterized by the parameter  $\lambda$ . Generally, in a given metal,  $\lambda$  is a constant, independent of frequency, magnetic field, and temperature over a wide range.<sup>10</sup> Our theory is concerned with an anomalous contribution to the damping,<sup>12</sup> which can still be written as a contribution to  $\lambda$ . This damping is anomalous in the sense that it consists of a plateau in  $\lambda$ , regarded as a function of temperature, which adds to the usual constant value only in pure nickel and cobalt at low temperatures, and which vanishes at higher temperatures (Fig. 2). It is also, as we shall see, anomalous in precisely the same sense that the "anomalous skin effect" is an anomaly, namely, the wavelengths of the excited spin waves are shorter than the mean free path, which means there are wave-number-dependent effects which are independent of mean free path.

This effect is definitively established in the case of nickel,<sup>12</sup> where it seems to have been confirmed by independent measurement.<sup>13</sup> Preliminary but quite convincing data indicate that it is also present in cobalt.<sup>14</sup> No such effects are observed in iron<sup>15</sup> or in alloys.<sup>16</sup>

The remainder of the paper is organized as follows. In Sec. II we develop the formalism required and consider the situation in the absence of spin-orbit effects. In Sec. III we include the spin-orbit terms and obtain an expression for the damping parameter  $\lambda$ . In Sec. IV we discuss the results and how they compare, at this stage, with experiment. A preliminary report of this work has already appeared.<sup>17</sup>

## II. FORMAL THEORY

We consider the band theory as a model of ferromagnetism. It is imagined that the band theory is carried out in the full Hartree-Fock approximation, or in various other approximations. In any case, the results of such a theory are the single-particle wave functions, together with the corresponding single-particle energies. In all such band models the states are labeled by the crystal momentum  $\vec{k}$  and a band index  $n$ .

As we are primarily interested in the decay of the spin wave, it is essential to keep the spin-orbit-interaction terms. This interaction is off-diagonal in both spin and space variables. Naturally, the screened Coulomb interaction, which is responsible for the exchange mechanism, is also essential.

As we shall soon see, in order to arrive at reasonable results it is necessary to treat the periodic lattice potential in a nontrivial approximation. This is not only because the existence of the ferromagnetism itself depends, for example, on having a density of states quite different from that of free electrons,<sup>1</sup> but more directly because the effect of the spin-orbit interaction is much reduced when

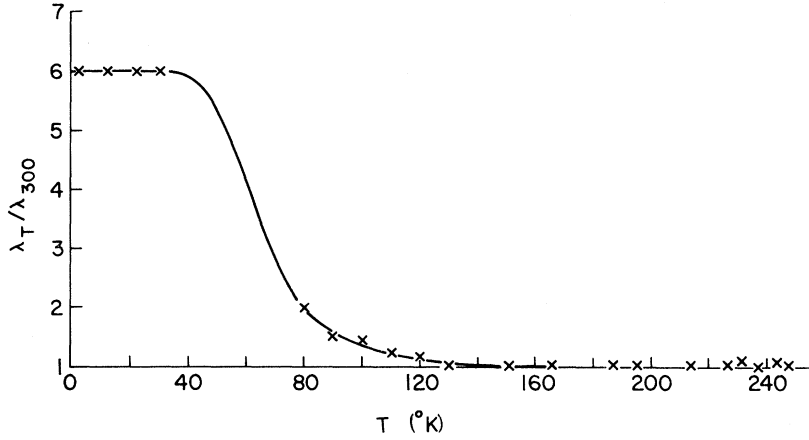


FIG. 2. Experimental values of the damping constant  $\lambda$ , in pure nickel, normalized to its value at 300 °K. From Ref. 12.

the single-particle wave functions have too great spatial symmetry. Therefore, the interactions of interest would be lost if we were to oversimplify the band aspect of the problem.

The effect of the spin-orbit interaction is twofold. On the one hand, in its absence the spin is conserved, and so it is directly responsible for the decay of the spin waves. On the other hand, it gives rise to magnetic anisotropy, which affects the real spin-wave dispersion relation as well as the  $g$  values, etc. We shall not be overly concerned with these latter effects in the present paper.

Since the spin-orbit interaction is relatively weak, it is natural to think of treating it in perturbation theory. However, straightforward perturbation theory is nontrivial, and we wish to search for a formulation which will isolate the influence of the spin-orbit interaction on the spin-wave decay. We may then be entitled to treat the other spin-orbit effects, which are formally included exactly, in such approximations as can be conveniently handled. As will be seen, the spin-wave lifetime can, in this fashion, be expressed as the square of an expression which can be interpreted as a real magnetic anisotropy effect and, in fact, one which has already been discussed in the literature.<sup>18</sup>

The Hamiltonian for the electrons is thus chosen to be

$$H = H^0 + V_{so} + \frac{1}{2} \sum_{\alpha, \beta} \int d\vec{r} d\vec{r}' V_C(\vec{r} - \vec{r}') \times [\psi_\alpha^\dagger(\vec{r}) \psi_\beta^\dagger(\vec{r}') \psi_\beta(\vec{r}') \psi_\alpha(\vec{r}) - 2\psi_\alpha^\dagger(\vec{r}) \psi_\alpha(\vec{r}) \langle \psi_\beta^\dagger(\vec{r}') \psi_\beta(\vec{r}') \rangle] - \frac{1}{2} \gamma \sum_{\alpha\beta} \int d\vec{r} \psi_\alpha^\dagger(\vec{r}) \vec{\sigma}_{\alpha\beta} \cdot [\vec{H} + \vec{h}(\vec{r}, t)] \psi_\beta(\vec{r}), \quad (4)$$

where  $H^0$  includes the kinetic energy, potential energy of the lattice, and Hartree energy;  $V_{so}$  is the spin-orbit interaction; and  $V_C$  is the screened Coulomb interaction. From the latter term we have subtracted the Hartree energy, as it is already included in  $H^0$ . The remaining term represents

the interaction of the electronic spins with the external magnetic field, which we have broken up into a static and dynamic component, the latter to be treated in lowest order. The orbital interaction of the electrons with the magnetic field is neglected. The annihilation operator  $\psi_\alpha(\vec{r}, t)$  is in the Heisenberg representation with  $\alpha$  the spin index.

We now assume the Hartree-Fock problem to be solved in the static case with  $\vec{h} = 0$ . We shall, however, write out the equations in an arbitrary band representation since we wish to consider bands in which the spin-orbit force may or may not be included. In this case the Hartree-Fock energies can be labeled by  $\vec{k}$ , but are not generally diagonal in the band index. They may thus be labeled by two band indices or, alternatively, be regarded as a matrix in the band labels. We shall denote such matrices with a caret. Thus, the Hartree-Fock energy is

$$[\hat{E}(\vec{k})]_{mn} = E_{mn}(\vec{k}). \quad (5)$$

We introduce as well the spin matrix  $\hat{S}$  defined by

$$\hat{S}_{mn}(\vec{k} | \vec{q}) = \sum_{\alpha, \beta} \int d\vec{r} u_{\vec{k} + \vec{q}/2, m}^* \alpha(\vec{r}) \vec{\sigma}_{\alpha\beta} u_{\vec{k} - \vec{q}/2, n} \beta(\vec{r}) \quad (6)$$

and the overlap function  $\hat{\Lambda}$ , where

$$\Lambda_{mn}(\vec{k}, \vec{k}' | \vec{G}) = \sum_{\alpha} \int d\vec{r} u_{\vec{k}, m}^* \alpha(\vec{r}) e^{i\vec{G} \cdot \vec{r}} u_{\vec{k}', n} \alpha(\vec{r}) \quad (7)$$

and the integrals are over the unit cell of the lattice. In these definitions,  $u_{\vec{k}, n}^\alpha(\vec{r})$  is the Bloch function, the label  $n$  gives the band index (which cannot in general be separated into space and spin parts), and  $\alpha$  labels the spin component.

It is convenient to introduce the abbreviation

$$\hat{S}_0(\vec{k}) \equiv \hat{S}(\vec{k} | 0). \quad (8)$$

It may easily be shown that  $\hat{S}_0$  satisfies the relation

$$\hat{S}_0(\vec{k}) \hat{\Lambda}(\vec{k}, \vec{k}') = \hat{\Lambda}(\vec{k}, \vec{k}') \hat{S}_0(\vec{k}'). \quad (9)$$

Next we introduce a shorthand for the operation

of multiplication by the Fourier transform of the screened Coulomb potential, integration over the first Brillouin zone, and summation over the reciprocal lattice. This operation will be denoted  $\mathfrak{S}$ , where

$$\mathfrak{S} \equiv \sum_{\mathbf{G}} \int \frac{d\mathbf{k}'}{(2\pi)^3} V_C(\mathbf{k} - \mathbf{k}' + \mathbf{G}). \quad (10)$$

Using this notation, the Hartree-Fock equation is

$$\hat{E}(\mathbf{k}) = E^0(\mathbf{k}) - \frac{1}{2} \gamma \vec{H} \cdot \vec{S}_0(\mathbf{k}) - \mathfrak{S} \hat{\Lambda} \hat{n}(\mathbf{k}') \hat{\Lambda}^\dagger, \quad (11)$$

where the electron density  $\hat{n}$  is given in terms of the single-particle Green's function at equal times by

$$\hat{n}(\mathbf{k}) = -i \hat{G}(\mathbf{k} | t, t^+). \quad (12)$$

The problem of the band theory is stated in the present nomenclature as the problem of the self-consistent diagonalization of Eq. (11).

The response function to a right circularly polarized magnetic field is of direct interest. This function is defined by

$$\chi^*(\vec{\mathbf{r}}t, \vec{\mathbf{r}}'t') = \left. \frac{\delta \langle m^*(\vec{\mathbf{r}}t) \rangle}{\delta h^*(\vec{\mathbf{r}}'t')} \right|_{h^*=0}, \quad (13)$$

where we have used coordinate-space nomenclature. The magnetization is given by the Green's function according to

$$\langle m^*(\vec{\mathbf{r}}t) \rangle = -\frac{1}{2} i \gamma \sum_{\alpha\beta} G_{\beta\alpha}(\vec{\mathbf{r}}t, \vec{\mathbf{r}}t) \sigma_{\alpha\beta}^+. \quad (14)$$

This response function is essentially the magnon propagator. It also contains a transient response, consisting of high-energy single-particle excitations, and is of course renormalized; but in the long-wavelength low-frequency case of interest here, the magnon pole dominates, and we shall ignore the transient terms.

As we have noted, in the absence of the spin-orbit coupling there is no decay of the magnon in the long-wavelength limit. This fact is manifested by a pole on the real frequency axis in  $\chi^*$ , and is a consequence of the conservation laws. Therefore, in order to describe correctly the effects of the breakdown of the spin-conservation law due to the spin-orbit interaction, we must use an approximation in which the conservation laws are maintained. The appropriate approximation consistent with the use of the Hartree-Fock approximation for the single-particle energies may be represented by the "ladder" sum of Feynman graphs shown in Fig. 1. An integral equation for this sum is as follows: Define

$$M_{\beta\alpha}(\vec{\mathbf{r}}_1 \vec{\mathbf{r}}_2 t, \vec{\mathbf{r}}' t') = \left. \frac{-i \gamma \delta G_{\beta\alpha}(\vec{\mathbf{r}}_1 t; \vec{\mathbf{r}}_2 t^+)}{\delta h^*(\vec{\mathbf{r}}' t')} \right|_{h=0}. \quad (15)$$

Applying the usual finite-temperature formalism

at temperature  $T$ , taking matrix elements with respect to the band wave functions, and Fourier transforming with respect to  $\vec{\mathbf{r}}'$ ,  $t'$ , we may obtain an integral equation for  $\hat{M}(\mathbf{k} | \vec{\mathbf{q}} \Omega)$  in the band representation, where  $\Omega$  is the frequency and  $\vec{\mathbf{q}}$  is the wave number of the magnon. Since  $\vec{\mathbf{q}}$ ,  $\Omega$  are fixed, we generally suppress the dependence on them. We also introduce the notation  $\hat{n}'(\mathbf{k}) = \hat{n}(\mathbf{k} + \frac{1}{2}\vec{\mathbf{q}})$  and  $\hat{n}''(\mathbf{k}) = \hat{n}(\mathbf{k} - \frac{1}{2}\vec{\mathbf{q}})$  in which the dependence on the momentum variable ( $s$ ) is shifted by  $\pm \frac{1}{2}\vec{\mathbf{q}}$ . A similar notation is used for other functions.

The integral equation is, then, in this notation,

$$\hat{M}(\mathbf{k} | \vec{\mathbf{q}} \Omega) = T \sum_{\omega_s} \hat{G}'(\omega_s + \Omega) \left[ \frac{1}{2} i \gamma^2 \hat{S}^- + \mathfrak{S} \hat{\Lambda}^\dagger \hat{M}(\mathbf{k}') \hat{\Lambda}^\dagger \right] \hat{G}^r(\omega_s) \quad (16)$$

and the magnon propagator is obtained in turn as

$$\chi^*(\vec{\mathbf{q}} \Omega) = \frac{1}{2} \int \frac{d\mathbf{k}}{(2\pi)^3} \text{Tr} \hat{S}^+(\mathbf{k} | -\vec{\mathbf{q}}) \hat{M}(\mathbf{k} | \vec{\mathbf{q}} \Omega) \quad (17)$$

and the trace is over the band indices.

Let us now imagine for the moment that we have solved the static Hartree-Fock problem and obtained the eigenvalues. In the band representation defined by this solution  $\hat{E}$ ,  $\hat{G}$ , and  $\hat{n}$  are diagonal and we may perform the "temperature" sum over  $\omega_s$ . This may be followed by the analytic continuation of  $\Omega$  to real frequency. Denoting the diagonal elements with a single index, the result is

$$+ iT \sum_{\omega_s} G_m^i(\omega_s + \Omega) G_n^r(\omega_s) = \frac{n_m^i - n_n^r}{\Omega - E_m^i + E_n^r + i/\tau}, \quad (18)$$

where, in fact, the density is explicitly given by

$$n_m(\mathbf{k}) = (e^{[E_m(\mathbf{k}) - \mu]/T} + 1)^{-1}, \quad (19)$$

and  $\tau$  is the ordinary scattering lifetime.

We insert this result in Eq. (16) after which we multiply through by the denominator of Eq. (18). It is easy to see that the result may be written in a form independent of band representation. In fact, we may write

$$\Omega \hat{M} - [\hat{E}, \hat{M}]_q = \frac{1}{4} \gamma^2 [\hat{n}, \hat{S}^-]_q + \mathfrak{S} [\hat{n}(\mathbf{k}), \hat{\Lambda}^\dagger \hat{M}(\mathbf{k}') \hat{\Lambda}^\dagger]_q, \quad (20)$$

where we have introduced the generalized commutator

$$[\hat{C}(\mathbf{k}), \hat{D}]_q = \hat{C}^i \hat{D} - \hat{D} \hat{C}^r. \quad (21)$$

Note that  $[\hat{C}, \hat{D}]_q$  becomes the ordinary commutator at  $q=0$ .

It is now easy to verify that these equations in the absence of spin-orbit interaction indeed predict that the infinite-wavelength spin wave does not decay. Let us make the ansatz

$$\hat{M}(\mathbf{k} | \vec{\mathbf{q}} \Omega) = [\hat{n}(\mathbf{k}), \hat{A}(\mathbf{k} | \vec{\mathbf{q}} \Omega)]_q. \quad (22)$$

Using the fact that  $\hat{n}$  and  $\hat{E}$  commute, we may rewrite Eq. (20) as

$$[\hat{n}, (\Omega\hat{A} - [\hat{E}, \hat{A}]_q - \mathcal{G}\hat{\Lambda}^\dagger[\hat{n}, \hat{A}]_q\hat{\Lambda}^{\dagger\dagger} - \frac{1}{4}\gamma^2\hat{S}^-)]_q = 0. \quad (23)$$

Now, in the absence of spin-orbit interaction, the single-particle Hamiltonian conserves spin just as does the interaction Hamiltonian. Thus, in this case, the commutation relation

$$[\hat{E}_0(\vec{k}), \hat{S}_0(\vec{k})] = 0 \quad (24)$$

holds. Therefore, at  $q=0$  it is natural to look for a solution in the form

$$\hat{A}(\vec{k}|0, \Omega) = g(\Omega)\hat{S}_0^-(\vec{k}). \quad (25)$$

It is easy to see that, by virtue of relation (9), the Fock terms in the expression for  $\hat{E}$  cancel against the  $\mathcal{G}$  term, in Eq. (23). Thus we readily find that

$$g(\Omega) = \frac{1}{4}\gamma^2/(\Omega - \gamma H). \quad (26)$$

This leads at once to the usual expression for the magnon propagator,

$$\chi^*(0, \Omega) = -\gamma M_s/(\Omega - \gamma H), \quad (27)$$

where  $M_s$  is the magnetization. Furthermore, the exchange-stiffness contribution to the magnon energy can be found by keeping the  $q$  dependence to order  $q^2$ .

### III. SPIN-ORBIT INTERACTION

In the presence of spin-orbit forces, the solution Eq. (25) can no longer be correct, since the relation (24) does not hold. In the previous simple case, the solution for  $\hat{A}$  had the property that it was off-diagonal in the Hartree-Fock representation, since  $\hat{S}^-$  is off-diagonal in spin, and there is a trivial connection between spin and band labels when the spin is conserved. Once spin-orbit forces are taken into account this will no longer be true, and  $\hat{A}$  and  $\hat{M}$  will have diagonal elements in the Hartree-Fock representation. These diagonal elements are the essence of the decay process which we envisage. In fact, the off-diagonal nature of  $\hat{A}$  in the absence of spin-orbit forces corresponds to the fact that the intermediate states in the Feynman-graph ladder sum of Fig. 1 consist of electron-hole pairs which are split by the exchange interaction; i. e., they are in different bands. Thus energy cannot be conserved in the decay of the spin wave directly into these pairs.

On the other hand, diagonal terms in  $\hat{M}$  would mean that electron-hole pairs in the same band could exist as intermediate states so that energy- and momentum-conserving final quasiparticle states could exist for the decay process.

Therefore, we make the more general ansatz

$$\hat{M} = \hat{M}^d + [\hat{n}, \hat{A}]_q, \quad (28)$$

where  $\hat{M}^d$  is diagonal in the Hartree-Fock representation and  $\hat{A}$  is off-diagonal. Because  $\hat{M}^d$  can

exist only by virtue of the weak spin-orbit force, we are justified in treating it as small.

Inserting this ansatz into Eq. (20) we obtain two equations for the parts which are off- and on-diagonal in the Hartree-Fock representation, respectively. Writing out the equations in this representation we find

$$[\hat{n}, \Omega\hat{A} - [\hat{E}, \hat{A}] - \mathcal{G}\hat{\Lambda}[\hat{n}, \hat{A}]\hat{\Lambda}^\dagger - \mathcal{G}\hat{\Lambda}\hat{M}^d\hat{\Lambda}^\dagger] = [\hat{n}, \frac{1}{4}\gamma^2\hat{S}^-] \quad (29)$$

and

$$[\Omega - \vec{q} \cdot \vec{V}_m(\vec{k}) + i/\tau]M_m^d = \vec{q} \cdot \frac{dn_m}{d\vec{k}} [\frac{1}{4}\gamma^2\hat{S}^- + \mathcal{G}\hat{\Lambda}(\hat{M}^d + [n, A])\hat{\Lambda}^\dagger]_{mm}, \quad (30)$$

where we can put  $q=0$  in the off-diagonal equation (except for the dependence of  $\hat{M}^d$  on it). Clearly, we must keep the wave-number dependence in the equation for the diagonal elements, but we have kept it only to lowest order. The velocity  $dE_n(\vec{k})/d\vec{k}$  is written  $\vec{V}_n(\vec{k})$ .

In order to solve Eqs. (29) and (30), we note that it is safe to neglect the  $\hat{M}^d$  which appears under the  $\mathcal{G}$  sign in Eq. (30). We may then solve for  $\hat{M}^d$  to obtain

$$M_m^d = \vec{q} \cdot \frac{dn_m}{d\vec{k}} \frac{\frac{1}{4}\gamma^2 S_{mm}^- + (\mathcal{G}\hat{\Lambda}[\hat{n}, \hat{A}]\hat{\Lambda}^\dagger)_{mm}}{\Omega - \vec{q} \cdot \vec{V}_m(\vec{k}) + i/\tau}. \quad (31)$$

If the expression just found for  $M^d$  is substituted into Eq. (29), an inhomogeneous linear integral equation for  $\hat{A}$  is obtained. Clearly, we may expect the solution of this equation to have a pole near  $\Omega = \gamma H$  which will be shifted by a small complex value. The real part of the shift is the anisotropy contribution to the spin-wave frequency, whereas we are primarily interested in the imaginary part of the shift. There will also be a change in the residue of the pole, but this is of secondary importance.

Since the inhomogeneous equation has an isolated pole, its solution in the neighborhood of the pole will be proportional to the solution of the corresponding homogeneous equation, and the position of the pole will be determined by the eigenvalue of the homogeneous equation.

In order to find an expression for the imaginary shift, we may employ perturbation theory as follows. Consider first the homogeneous equation for  $\hat{A}$ , which we denote by Eq. (29'). This equation is obtained from Eq. (29) by replacing the right-hand side,  $\frac{1}{4}\gamma^2[\hat{n}, \hat{S}^-]$ , by zero, and regarding  $\hat{M}^d$  as given by Eq. (31) but with the term  $\frac{1}{4}\gamma^2 S_{mm}^-$  replaced by zero. We then do perturbation theory in  $\hat{M}^d$ .

The zeroth-order solution is denoted  $\hat{\Psi}_0$  and satisfies

$$[\hat{n}, \Omega_0\hat{\Psi}_0 - [\hat{E}, \hat{\Psi}_0] - \mathcal{G}\hat{\Lambda}[\hat{n}, \hat{\Psi}_0]\hat{\Lambda}^\dagger] = 0. \quad (32)$$

The zeroth-order eigenvalue  $\Omega_0$  is not just  $\gamma H$  since

$\hat{E}$  contains spin-orbit effects, but it is nevertheless real. This may be seen by defining an inner product. Multiplying Eq. (32) by  $\hat{\Psi}_0^\dagger$ , taking the trace over the band indices, and integrating over momenta in the first Brillouin zone gives

$$\Omega_0 = \text{Tr} \int d\vec{k} \{ \hat{\Psi}_0^\dagger [\hat{n}, [\hat{E}, \hat{\Psi}_0]] - \hat{\Psi}_0^\dagger \mathcal{G} [\hat{n}, \hat{\Lambda} [\hat{n}_0 \hat{\Psi}_0] \hat{\Lambda}^\dagger] \} \\ \times [ \text{Tr} \int d\vec{k} \hat{\Psi}_0^\dagger [\hat{n}, \hat{\Psi}_0] ]^{-1}. \quad (33)$$

It is straightforward to verify, using the properties of the trace, the hermiticity of  $\hat{n}$ ,  $\hat{E}$ , and the fact that they commute, the definition of the operator  $\mathcal{G}$  and the relation  $\hat{\Lambda}^\dagger(\vec{k}' | \vec{G}) = \hat{\Lambda}(\vec{k}' | -\vec{G})$ , that  $\Omega_0$  is real. Further, with this definition of inner product, the linear operator of Eq. (29) is Hermitian. In fact, our motivation for obtaining an equation for the operator  $\hat{A}$  rather than for  $\hat{M}$  directly is just this possibility of forming an appropriate inner product.

We now are in a position to write down the expression for the shift in eigenfrequency  $\Delta\Omega$  using ordinary first-order perturbation theory in  $\hat{M}^d$ . The result is

$$\Delta\Omega = \frac{\text{Tr} \int d\vec{k} \hat{\Psi}_0^\dagger(\vec{k}) [\hat{n}(\vec{k}) \mathcal{G} \hat{\Lambda} \hat{M}^d(\vec{k}') \hat{\Lambda}^\dagger]}{\text{Tr} \int d\vec{k} \hat{\Psi}_0^\dagger [\hat{n}, \hat{\Psi}_0]} \quad (34)$$

Again using the properties mentioned in the preceding paragraph, this may be rewritten

$$\Delta\Omega = \sum_m \int d\vec{k} \frac{\gamma^2 |\epsilon_m(\vec{k})|^2 \vec{q} \cdot \vec{V}_m(\vec{k}) (dn_m/dE_m)}{\Omega - \vec{q} \cdot \vec{V}_m + i/\tau} \\ \times \left[ \text{Tr} \int d\vec{k} [\hat{\Psi}_0, \hat{\Psi}_0^\dagger \hat{n}] \right]^{-1}, \quad (35)$$

where we have introduced

$$\gamma \epsilon_m(\vec{k}) \equiv [\mathcal{G} \hat{\Lambda} [\hat{n}, \hat{\Psi}_0] \hat{\Lambda}^\dagger]_{m,m}. \quad (36)$$

Clearly,  $\Delta\Omega$  vanishes in the absence of spin-orbit coupling since  $\hat{M}^d$  does. [In order to verify this directly from Eq. (36) one must invoke the properties of  $\hat{\Lambda}$ ,  $\hat{n}$ ,  $\hat{\Psi}_0$  in the absence of spin-orbit coupling.] We will return to the interpretation of  $\epsilon_m(\vec{k})$  in the following.

The integration in Eq. (35) is a Fermi-surface average of a type often encountered in calculations of the conductivity. In view of the fact that the magnitude of  $\vec{q}$  is determined by the skin depth (i. e.,  $q \sim 10^5 \text{ cm}^{-1}$ ), we may conclude that  $\Omega \ll q V_m(\vec{k})$ . If we also assume inversion symmetry, so that  $\vec{V}_m(\vec{k}) = -\vec{V}_m(-\vec{k})$ , the imaginary part of the expression for  $\Delta\Omega$  is

$$\Delta\Omega = -\Omega \gamma^2 \sum_m \int d\vec{k} \frac{|\epsilon_m(\vec{k})|^2 (dn_m/dE_m)}{\vec{q} \cdot \vec{V}_m(\vec{k}) - i/\tau} \\ \times \left[ \text{Tr} \int d\vec{k} [\hat{\Psi}_0, \hat{\Psi}_0^\dagger \hat{n}] \right]^{-1}. \quad (37)$$

This would be precisely the conductivity, up to an over-all factor, if  $|\epsilon_m(\vec{k})|$  were replaced by the transverse velocity. These "velocity" factors usually are not considered to have a dominating influence on the behavior of the integral, and they can be treated in some average sense. It is the denominator which is of importance. Thus, as in the case of the conductivity, we have two extreme cases, corresponding to the "normal" conductivity and to the "anomalous" case. These cases are defined by  $q V_F \ll 1/\tau$  and  $q V_F \gg 1/\tau$ , respectively. An interpolation formula, based on a spherical Fermi surface, gives the following expression:

$$\Delta\Omega = -i\Omega \langle N(E_F) | \epsilon_m(E_F) |^2 \rangle \frac{\tan^{-1} q V_F \tau}{q V_F} \\ \times \frac{\gamma^2}{\text{Tr} \int d\vec{k} [\hat{\Psi}_0, \hat{\Psi}_0^\dagger \hat{n}]} \quad (38)$$

The angular brackets are meant to indicate an appropriate average over the Fermi surface and  $N(E_F)$  is the density of states. This expression is definitely oversimplified in that, as we shall see,  $\epsilon(\vec{k})$  is probably a quite strongly varying function of position on the Fermi surface, and, of course, the Fermi surfaces are far from spherical. Nevertheless, the main dependences of the damping on the experimental variables and on the strength of the spin-orbit coupling can be made manifest by means of Eq. (38).

The next question to be resolved is how to evaluate  $\epsilon_m(\vec{k})$  and the denominator. To study this, we again note that the solution to the inhomogeneous equation (29) for a frequency in the neighborhood of the eigenfrequency is proportional to the solution of the homogeneous equation (29'); that is, at such frequencies the pole term in the solution to the inhomogeneous equation dominates. Since the normalization of the solution drops out of expression (37) we conclude that a correct expression for  $\hat{\Psi}_0$  is  $\hat{A}(\vec{k} | 0, \Omega)$  for  $\Omega$  very near  $\Omega_0$ .

Now, from Eqs. (12) and (15) we see that

$$\hat{M}(\vec{k} | \vec{q}, \Omega) = \frac{\gamma \delta \hat{n}(\vec{k})}{\delta h^+ (\vec{q}, \Omega)}; \quad (39)$$

i. e.,  $\hat{M}$  is the operator change of  $\hat{n}$  in response to an applied transverse field. Furthermore, neglecting  $\hat{M}^d$ , we have from Eq. (28)

$$\hat{M}(\vec{k} | 0, \Omega) = [\hat{n}, \hat{A}(\vec{k} | 0, \Omega)]. \quad (40)$$

Then, from Eqs. (11) and (36) we find that  $\epsilon_m(\vec{k})$  is the shift in the exchange energy with transverse field, at frequency  $\Omega_0$ . As a practical matter, the pole at  $\Omega_0$  is so isolated from the other singularities of  $\hat{A}$ , which occur at frequencies greater than the exchange splitting frequency, that it is sufficient to find the response to a static transverse

field. If we make the further simplifying assumption that the exchange energy depends on the direction of magnetic field only through the magnetization, we find

$$\epsilon_m(\vec{k}) = \frac{\delta E_m(\vec{k})}{\delta h^*} = \frac{dE_m(\vec{k})}{dm^*} \chi^*(\Omega) . \quad (41)$$

The denominator may be evaluated easily because small spin-orbit effects are unimportant there. We may use the solution of Eq. (25) therefore to find

$$\text{Tr} \int \frac{d\vec{k}}{(2\pi)^3} [\hat{A}^\dagger, \hat{A}] \hat{n} = \frac{\gamma [\chi^*(\Omega)]^2}{M_s} . \quad (42)$$

Putting these results together, and comparing with the Gilbert equation, we finally have

$$\lambda = \gamma^2 \left\langle N(E_F) \left| M_s \frac{dE_m(k)}{dm^*} \right|^2 \right\rangle \tan^{-1} \frac{q V_F \tau}{q V_F} . \quad (43)$$

#### IV. DISCUSSION

Equation (43), although somewhat oversimplified as compared with Eq. (37), will be used as a basis for discussion. In fact, using a crude model, Kambersky<sup>9</sup> obtained expression (43) with the important difference that he replaced  $\tan^{-1} q V_F \tau / q V_F$  by  $\tau$ ; i. e., Eq. (43) in the normal-skin-effect limit.

Kambersky's model, whose physical basis is similar to ours, is based on the observation that since the exchange energy is a function of the direction of the magnetization, so will be the Fermi surface. Thus, as the spin wave propagates, the shape of the Fermi surface is distorted periodically in space and time. The efforts of the electrons to repopulate the changing Fermi sea account for the damping. We have simply found a formal procedure to justify this intuitive approach and, in addition, have generalized it to the anomalous case.

Kambersky has already discussed the over-all magnitude of the effect. This is not easy to estimate reliably. In general, one can expect two kinds of contributions to  $dE_m/dm^*$ . One is a sum of small contributions coming from the bulk of the Fermi surface, and the other is a sum of larger contributions coming from special regions of the Fermi surface. Which is the more important is a question to be resolved by detailed band-structure calculations.

One may note however, that if plane waves are used as an approximation to the band states, the effect is expected to be small. Because of the high symmetry of these wave functions, there are strong selection rules on the matrix elements of the exchange energy which prohibit a dependence of the exchange energy on the spin-orbit potential in lowest order.

The possibility that special regions of the Fermi surface play an important role comes from the fact that there may exist band-crossing degeneracies near the Fermi energy which are lifted by the spin-orbit interaction.<sup>18</sup> In such a case, the exact position of the Fermi surface depends sensitively on the spin-orbit force, since degenerate perturbation theory must be used and, as a consequence, the exchange energy in this region will depend strongly on the direction of the magnetization. There is strong evidence that such is the case in nickel<sup>18</sup> (near the point *X* in the Brillouin zone), some preliminary evidence that it might be the case in cobalt,<sup>19</sup> and little evidence for its possibility in iron.<sup>20</sup> This would immediately explain the fact that anomalies have been observed in Ni and Co, but not in Fe. {An additional consideration is that the exchange stiffness [*D* in Eq. (1)] is much larger in Fe than in Ni, though not in Co, so that contributions to the observed linewidth of ferromagnetic resonance for Fe are less easily extracted from experiment. Thus an existing small anomaly of  $\lambda$  in Fe might have gone undetected.}

If a special region of the Fermi surface is involved, then obviously the density of states, velocities, and free times in Eq. (43) should refer to this region. Thus, the velocity may be quite small and the free time not characteristic of the conductivity as a whole. Indeed, considerable variations in free time, not to speak of Fermi velocities, have been found in Fermi surfaces of neighboring nonmagnetic materials, such as Cu.<sup>21</sup>

A detailed comparison with experiment is also complicated by the necessity to recompute the solutions to Maxwell's equations,<sup>22</sup> using a wave-number-dependent  $\lambda$ . This is being studied at present.

Nevertheless, we may agree with Kambersky that the theoretical magnitude of  $\lambda$  could possibly be in agreement with experiment. One may determine an average value of  $q$  to be used in the evaluation of  $\lambda$  by considering it to be given by a non-self-consistent calculation of the skin depth. Then  $q$  is roughly constant below 150 °K for Ni. As the temperature is lowered,  $\tau$  becomes longer so the factor  $\tan^{-1} q V_F \tau / q V_F$  becomes larger and saturates, as observed. The temperature region above which this saturation occurs indeed corresponds to the onset of the anomalous skin effect in the ordinary conductivity. The level of saturation is independent of residual resistivity, as observed, provided the resistance ratio is large enough.<sup>12,16</sup>

For small enough resistance ratio, no anomaly should be observed, which agrees with the absence of an anomaly in alloys. Single crystals with a variety of resistance ratios, studied over the difficult temperature range 20–100 °K, would allow a detailed study of the correctness of the formula



(43). Such experiments are tedious and difficult, however, and have not so far been carried out.

There still remains the unexplained constant contribution to  $\lambda$ , independent of temperature and mean free path. It is difficult to conceive of this as arising from the decay of the spin wave into other elementary excitations, since such a process would surely show a temperature dependence<sup>8,9</sup> over the temperature range 100–600 °K. Therefore, we feel it likely that this source of damping

is some sort of inhomogeneous line broadening, perhaps arising from a surface phenomenon. No detailed mechanism for this constant contribution has been found to date, however.

In summary, we have concluded that, by virtue of the spin-orbit interaction, a spin wave can decay directly into a particle-hole pair, the decay not conserving spin. This particular source of damping has an anomalous character and depends on detailed properties of the Fermi surface.

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## Evidence for Fisher's Droplet Model in Simulated Two-Dimensional Cluster Distributions

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The distribution function  $n_l$  of clusters with  $l$  reversed spins has been calculated for two square  $N \times N$  Ising lattices ( $N=55$  and  $110$ ) at various temperatures  $T < T_c$  using periodic boundary conditions. These calculations strongly support the cluster model proposed by Fisher and yield the exponent  $\tau=2.1 \pm 0.1$ . However, for small  $l$ ,  $T$  not close to  $T_c$ , and  $H \neq 0$ , we find considerable deviations.

Fisher's cluster model<sup>1-3</sup> is a semiphenomenological description of critical phenomena in Ising spin systems, leading to a physical interpretation of the static scaling relations.<sup>3,4</sup> Within the framework of the lattice-gas terminology it can also be applied to the gas-liquid phase transition. These results agree surprisingly well with ex-

periment for gases<sup>5-7</sup> and have also been used in the nucleation problem<sup>8-10</sup> to calculate the formation rate of large droplets in a supersaturated vapor. Perhaps this model can also contribute to a better understanding of the phenomena near tricritical points.<sup>11-13</sup> It should be noted, however, that it fails above the critical temperature  $T_c$ .<sup>6</sup>