Local-band theory of itinerant ferromagnetism. III. Nonlinear Landau-Lifshitz equations*

V. Korenman, J. L. Murray,[†‡] and R. E. Prange
Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742
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To provide insight into the results of the two preceding papers (I and II), the Landau-Lifshitz equation has been analyzed. It is found, as in II, that the magnon energy, close to and above the Curie temperature T_C, is approximately $DQ^2-2D|\vec{a}|^2_{av}$, for $Q>|\vec{a}|$, where $|\vec{a}|^2_{av} = 1/4(\nabla\hat{M})^2_{av}$. This is predicted to be the dispersion observed by neutron scattering, with an inhomogeneous broadening, also of order $D|\vec{a}|^2_{av}$. At low temperature, the first nonlinear approximation is made to see how this result can be reconciled with the usual form given in the Landau theory.

I. INTRODUCTION

The Landau-Lifshitz equation[1] (LLE) is the basic macroscopic equation of motion of ferromagnetism. Although the mechanism of the possible damping terms in this equation is not completely settled,[2] there is little doubt that the nondissipative part of the equation is correct, and not only in its linearized version. Certainly the developments of the first two papers of this series[3-5] provide support for this view.

In II, one of the main new results was a downward shift in the magnon dispersion, $-2D|\vec{a}|^2_{av}$, which numerically seems to account for the major part of observed softening of the magnons with temperature. This expression is valid in the limit of magnon wave number $Q \gg |\vec{a}|$ where $|\vec{a}|^2_{av} = \frac{1}{4}(\nabla\hat{M})^2_{av}$. The simplicity of this result and lack of dependence on the underlying band structure, as well as its novelty, has led us to investigate whether this term already appears in the LLE. It is the purpose of this note to show that it does, and thereby to clarify the meaning of the term. It is not expected on physical grounds that the damping term derived in II will be a consequence of the LLE. However, the LLE sheds light on the inhomogeneous broadening which also contributes to the observed linewidth. We may also hope to learn something about the transition region between propagation and diffusion from the LLE.

The LLE is (with $\partial/\partial t$ represented by a dot)

$$\dot{\vec{M}} = C\vec{M}\times\nabla^2\vec{M}+\gamma\vec{M}\times\vec{H}. \tag{1}$$

We choose units of length, time, and magnetic field so as to make $C=M=D=\gamma=1$. We rewrite Eq. (1):

$$\dot{\vec{S}} = \vec{S}\times\nabla^2\vec{S}+\vec{S}\times\vec{H}, \tag{2}$$

where $\vec{S}(rt)$ is a unit vector. The LLE makes $|\vec{S}|$ constant in time, so nothing is gained by allowing the magnitude to vary in space.

We treat H as small (it may represent the interaction with a neutron) and seek a solution of the form $\vec{S}=\vec{S}_B+\vec{s}$, where $\vec{S}_B$ is a thermal background magnetization which satisfies

$$\dot{\vec{S}}_B = \vec{S}_B\times\nabla^2\vec{S}_B \tag{3}$$

and $\vec{s}$ is the linear response to $\vec{H}$. It is perpendicular to $\vec{S}_B$. Then

$$\dot{\vec{s}} = \vec{S}_B\times\nabla^2\vec{s}+\vec{s}\times\nabla^2\vec{S}_B+\vec{S}_B\times\vec{H}. \tag{4}$$

The middle term on the right-hand side of Eq. (4) may be suspected of giving the shift $-\frac{1}{2}(\vec{\nabla}\vec{S}_B)^2$. As we shall see it gives twice the final shift as there is a cancellation from the first term.

Because $\vec{S}_B$ itself is space and time dependent, indeed in a way which is more or less random, it will not be possible to solve Eq. (4) exactly. We shall therefore make several approximations, notably that the wave number and frequency of H are large compared with those of $\vec{S}_B$. We shall also consider the case of weak nonlinearity.

II. LOCAL ROTATION

We study Eq. (4) by going to a frame of reference in which $\vec{S}_B$ is constant. We call A the 3×3 rotation matrix carrying the unit vector in the z direction, $\hat{e}_z\equiv\hat{e}_0$, into the direction $\vec{S}_B$, given by polar angles θ,ϕ. That is,

$$\vec{S}_B = A\hat{e}_0. \tag{5}$$

We also introduce unit vectors $\hat{e}_x$, $\hat{e}_y$; and $\hat{e}_\pm=(\hat{e}_x\pm i\hat{e}_y)/\sqrt{2}$. An expression for A is

$$A\equiv\exp(-iL_z\phi)\exp(-iL_y\theta)\exp(-iL_zb), \tag{6}$$

where b is an arbitrary third Euler angle, the L_i are 3×3 matrices satisfying angular momentum commutation rules and such that $L_i\hat{e}_j=i\hat{e}_i\times\hat{e}_j$. We shall also use $L_\pm=L_x\pm iL_y$, where $L_\pm\hat{e}_0=\mp\sqrt{2}\,\hat{e}_\pm$. (Note we do not use the Condon and Shortly phase convention.) Putting

$$\vec{s} = A\vec{\sigma}, \quad (7a)$$

$$\vec{H} = A\vec{h} \quad (7b)$$

and using the pseudovector property of the cross product, $A(\vec{K}\times\vec{L}) = A\vec{K}\times A\vec{L}$ we find

$$\dot{\vec{\sigma}} + A^{-1}\dot{A}\vec{\sigma} = \hat{e}_0 \times A^{-1}\nabla^2(A\vec{\sigma}) + \vec{\sigma}\times A^{-1}\nabla^2 A\hat{e}_0 + \hat{e}_0\times\vec{h}. \quad (8)$$

It is easy to show that $A^{-1}\nabla^2(A\vec{\sigma}) = [\nabla + (A^{-1}\nabla A)]^2\vec{\sigma}$, for any A and σ. A simple calculation gives

$$A^{-1}\dot{A} = -2i\tilde{g}L_z + i\tilde{a}^* L_+ + i\tilde{a}L_-, \quad (9)$$

$$A^{-1}\nabla A = -2i\vec{g}L_z + i\vec{a}^* L_+ + i\vec{a}L_-, \quad (10)$$

where

$$\vec{a} = \tfrac{1}{2}(\nabla\phi\sin\theta - i\nabla\theta)e^{-ib}, \quad (11a)$$

$$\vec{g} = \tfrac{1}{2}(\nabla b + \cos\theta\nabla\phi), \quad (11b)$$

$$\tilde{a} = \tfrac{1}{2}(\dot{\phi}\sin\theta - i\dot{\theta})e^{-ib}, \quad (12a)$$

$$\tilde{g} = \tfrac{1}{2}(\dot{b} + \cos\theta\dot{\phi}). \quad (12b)$$

The notation conforms to I. Equation (3) gives the LLE in terms of θ, ϕ, which is

$$i\tilde{a} = -(\vec{\nabla} + 2i\vec{g})\cdot\vec{a}. \quad (13)$$

Using (9) and (10) in (8), it is found that $\vec{\sigma}$ obeys

$$\dot{\vec{\sigma}} + (-2i\tilde{g}L_z + i\tilde{a}^*L_+ + iaL_-)\vec{\sigma} = \hat{e}_0\times(\nabla - 2i\vec{g}L_z + i\vec{a}^*L_+ + i\vec{a}L_-)^2\vec{\sigma} + \vec{\sigma}\times(\nabla - 2i\vec{g}L_z + i\vec{a}^*L_+ + i\vec{a}L_-)^2\hat{e}_0 + \hat{e}_0\times\vec{h}. \quad (14)$$

Expressing σ as

$$\vec{\sigma} = (\Sigma_-\hat{e}_+ + \Sigma_+\hat{e}_-)/\sqrt{2} \quad (15)$$

with $(\Sigma_+)^* = \Sigma_-$, it is possible to read off the equation for Σ_+ from Eq. (14)

$$\left(\frac{i\partial}{\partial t} - 2\tilde{g}\right)\Sigma_+ - \left[\left(\frac{1}{i}\nabla + 2\vec{g}\right)^2 - 2|\vec{a}|^2\right]\Sigma_+ = -2\vec{a}\cdot\vec{a}\Sigma_- + h_+. \quad (16)$$

The driving term h_+ is defined analogously to Eq. (15). It is given by

$$e^{ib}h_+ = \cos^2(\tfrac{1}{2}\theta)e^{-i\phi}H_+ - \sin^2(\tfrac{1}{2}\theta)e^{i\phi}H_- - \sin\theta\, H_0, \quad (17)$$

where $H_0, H_\pm$ are components of H in the laboratory frame.

III. SUSCEPTIBILITY

Restricting considerations to the case $T \geq T_C$, we wish to calculate the susceptibility, χ, where

$$\langle\vec{s}\rangle_{S_B} = \chi\vec{H} \quad (18)$$

and $\vec{H} = \mathrm{Re}\vec{H}_0\exp(i\vec{Q}\cdot\vec{R} - i\Omega t)$. The brackets indicate a thermal average over $\vec{S}_B$. It is clearly sufficient to take $\vec{H}_0$ to be in the $\hat{e}_0$ direction (in the laboratory frame). Equation (17) is thus simplified by setting $H_+ = H_- = 0$.

To solve Eq. (16) we wish to make the approximation that $|\vec{a}|^2_{av}$ is small compared with Q^2. We note that the term $\vec{a}\cdot\vec{a}\Sigma_-$ may be neglected as it makes corrections of higher order [besides which $\langle\vec{a}\cdot\vec{a}\rangle_{S_B} = 0$]. We may also replace $|\vec{a}|^2$ by $|\vec{a}|^2_{av} + (|\vec{a}|^2 - |\vec{a}|^2_{av})$, and include the bracketed term in $\tilde{g}$. This term vanishes in the thermal averaging and makes corrections in a higher order than we are keeping.

Let $K(\vec{r}t, \vec{r}'t')$ be the Green's function satisfying

$$\left(\frac{i\partial}{\partial t} - 2\tilde{g} + (\nabla + 2i\vec{g})^2 + 2|\vec{a}|^2_{av}\right)K(\vec{r}t, \vec{r}'t') = \delta(\vec{r} - \vec{r}')\delta(t - t'). \quad (19)$$

Then

$$\Sigma_+(\vec{r}t) = -\int K(\vec{r}t, \vec{r}'t')\sin\theta(\vec{r}'t') \times \exp[-ib(\vec{r}'t')]H_0(\vec{r}'t')\,d^3r'\,dt'. \quad (20)$$

To calculate $\vec{s}$, we form $\vec{\sigma}$ according to Eq. (15) and use Eq. (7a). We write down only the component in the $\hat{e}_0$ direction, as the others vanish upon averaging. Then

$$\langle s_0\rangle_{S_B} = \mathrm{Re}\langle\sin\theta(\vec{r}t)\exp[ib(\vec{r}t)]K(\vec{r}t, \vec{r}'t') \times \exp[-ib(\vec{r}'t')]\sin\theta(\vec{r}'t')\rangle_{S_B}H_0(\vec{r}'t'). \quad (21)$$

The kernel K depends on $\vec{g}, \tilde{g}$ in two ways. As pointed out in I and II, $2\tilde{g}$ is a scalar potential and $-2\vec{g}$ a vector potential corresponding to an electric field, $\vec{e} = \nabla\phi\,\partial\cos\theta/\partial t - \nabla\cos\theta\,\partial\phi/\partial t$ and a magnetic field $\vec{h} = -\nabla\cos\theta\times\nabla\phi$.

These fields are weak, being of cubic and quadratic order, respectively, in spatial gradients. Further, they vanish on averaging so come into the final result in higher order. They can thus be neglected.

Paradoxically, the potentials have an effect even in the absence of fields. Normally, of course, there is no physical effect if the electric and magnetic fields vanish, as the existence of potentials merely alters the Green's function by a phase factor. Here the phase factor is important. Because the spin rotation is space and time dependent, the wave number and frequency is not the same in the laboratory and locally rotated frames. The extra phase factors contribute to this Doppler shift, and result in an inhomogeneous broadening of the observed spectrum.

The combination $e^{ib}Ke^{-ib'}$ is gauge invariant. To estimate it one may express K as a sum over paths[6]

$$K=\int d(\text{paths})\exp\left(i\int_{t'}^{t}d\tau\,(\tfrac{1}{4}v^2-2\vec{v}\cdot\vec{g}-2\tilde{g})\right). \quad (22)$$

Neglecting the potentials $\vec{g},\tilde{g}$, the path integral may be performed to obtain

$$K_0(\vec{r}t,r't')=\frac{\Theta(t-t')}{[e^{-\pi i/2}2\pi(t-t')]^{3/2}}\times\exp\left|\frac{i(\vec{r}-\vec{r}')^2}{4(t-t')}+i2|\vec{a}|^2_{av}(t-t')\right|. \quad (23)$$

The minimizing classical path in this case is a straight line, $\vec{r}(\tau)=\vec{r}'+\vec{v}(\tau-t')$ with $\vec{v}=(\vec{r}-\vec{r}')/(t-t')$. Retaining this path alone already gives the dominating singular exponential in Eq. (23).

The fields $\tilde{e},\tilde{h}$ derived from the potentials $\vec{g},\tilde{g}$ are weak and do not appreciably deflect the "particle" traveling in its path between $\vec{r}'t'$ and $\vec{r}t$. This happens because we restrict ourselves to high momentum and frequency components, so that $\vec{r}t$ and $\vec{r}'t'$ are close together. We may thus approximate the potential terms in Eq. (22) by keeping only the most important path, to obtain

$$e^{ib}K(\vec{r}t,\vec{r}'t')e^{-ib'}=K_0(\vec{r}t,\vec{r}'t')\exp\left(-i\int_{t'}^{t}\cos\theta(\vec{r}(\tau),\tau)\frac{d}{d\tau}\phi(\vec{r}(\tau),\tau)\,d\tau\right). \quad (24)$$

We are thus faced with evaluating

$$W=\langle\sin\theta(\vec{r}t)\exp\left(i\int_{t'}^{t}\cos\theta\,d\phi\right)\sin\theta(\vec{r}'t')\rangle_{SB}. \quad (25)$$

This correlation function vanishes for either large space or large time separation. It is needed for relatively small separation, for which the form

$$W=\tfrac{2}{3}\exp|-\gamma_1|\vec{a}|^2_{av}|\vec{r}-\vec{r}'|^2-\gamma_2|\tilde{a}|^2_{av}(t-t')^2| \quad (26)$$

is plausible. We have shown elsewhere[7] that $\gamma_1=\frac{1}{3}$, $\gamma_2=1$.

The Fourier transform of Eq. (23) is

$$K(\vec{Q},\Omega)=1/[\Omega-(Q^2-2|\vec{a}|^2_{av})+i\eta]. \quad (27)$$

The Fourier transform of the magnon creation part of the susceptibility χ will be given by Eq. (27) convoluted with the Fourier transform of W. This convolution gives an approximately Gaussian line shape with a width of order $(Q^2|\vec{a}|^2_{av})^{1/2}$ for $Q^2\gg|\vec{a}|^2_{av}$. This is inhomogeneous broadening, and does not represent decay of the magnon state. In addition, the decay into quasiparticle states found in II, and the neglected higher-order terms of this paper give additional contributions to the observed linewidths.

IV. WEAK NONLINEARITY

In this section we take up the case of weak nonlinearity, which would be applicable at low temperature. In this case, the energy

$$\mathcal{E}=\sum Dq^2N_q+\frac{1}{2}\sum h_{qq'}q^2q'^2N_qN_{q'}+\frac{1}{2}\sum h'_{qq'}\vec{q}\cdot\vec{q}'N_qN_{q'} \quad (28)$$

is usually postulated.

We consider a two-spin density wave problem, with a symmetrical treatment of the waves. We will not find the term in $h_{qq'}$ of Eq. (28) above, as the LLE is quadratic in the gradients. However, the result obtained is instructive.

Writing the LLE as

$$-i\dot{S}_+=S_z\nabla^2S_+-S_+\nabla^2S_z, \quad (29)$$

we assume $S_z=(1-S_+S_-)^{1/2}$ and calculate the corrections which are relatively of second order in S_+.

Putting $S_+=S_1+S_2+S_3$ with $S_1=\alpha(t)e^{i\vec{q}_1\cdot\vec{r}}$, $S_2=\beta e^{i\vec{q}_2\cdot\vec{r}}$, and S_3 the third-order correction, we have

$$i\dot{\alpha}/\alpha=q_1^2(1-\tfrac{1}{2}|\alpha|^2)-|\beta|^2\vec{q}_1\cdot\vec{q}_2, \quad (30a)$$

$$i\dot{\beta}/\beta=q_2^2(1-\tfrac{1}{2}|\beta|^2)-|\alpha|^2\vec{q}_1\cdot\vec{q}_2, \quad (30b)$$

$$i\dot{S}_3+\nabla^2S_3=\tfrac{1}{2}\{(q_2^2-2\vec{q}_1\cdot\vec{q}_2)\alpha^2\beta^*\exp[i(2\vec{q}_1-\vec{q}_2)\cdot\vec{r}]+(q_1^2-2\vec{q}_1\cdot\vec{q}_2)\alpha^*|\beta|^2\exp i(2\vec{q}_2-\vec{q}_1)\cdot\vec{r}\}. \quad (30c)$$

Let $\alpha=\alpha_0e^{-i\omega_1t}$, $\beta=\beta_0e^{-i\omega_2t}$,

with
$$\omega_1=q_1^2(1-\tfrac{1}{2}\alpha_0^2)-\vec{q}_1\cdot\vec{q}_2\,\beta_0^2, \quad (31a)$$

$$\omega_2=q_2^2(1-\tfrac{1}{2}\beta_0^2)-\vec{q}_1\cdot\vec{q}_2\alpha_0^2. \quad (31b)$$

Note that ω_i vanishes with q_i. We find

$$S_+ = S_1 + S_2 - \tfrac{1}{4} S_1^2 S_2^* [1 - q_1^2/(\vec{q}_1 - \vec{q}_2)^2] - \tfrac{1}{4} S_2^2 S_1^* [1 - q_2^2/(\vec{q}_1 - \vec{q}_2)^2]\,. \tag{32}$$

This solution can now be compared with the solution of Eq. (16) (with $h^+ = 0$). We assume $\beta_0 \ll \alpha_0 \ll 1$. We rotate through angles θ, ϕ, where to first approximation $\alpha_0 = \theta$, $\phi = \vec{q}_1 \cdot \vec{r} - q_1^2(1 - \frac{1}{2}\theta^2)t$, then $\vec{S}_B \sim \hat{e}_z + S_1 \hat{e}_- /\sqrt{2}$. The leading term of Σ_+ has the exponential dependence $\exp(i\vec{Q}\cdot\vec{r} - i\Omega t)$ with $\Omega = Q^2 - \frac{1}{2} q_1^2 \theta^2$ $(Q \gg q_1)$. Transforming back to the laboratory frame gives $s \approx S_2 \propto \exp[i(\vec{Q} + \frac{1}{2}\theta^2 \vec{q}_1)\cdot\vec{r} - i(\Omega + \frac{1}{2} q_1^2\theta^2)t]$. With $\vec{q}_2 = \vec{Q} + \frac{1}{2}\theta^2\vec{q}_1$, $\Omega + \frac{1}{2} q_1^2\theta^2 = (\vec{q}_2 - \frac{1}{2}\theta^2\vec{q}_1)^2 = \omega_2$.

The energy Ω, in addition to giving the frequency of the excitation in the rotated frame, is related to the mean energy of the excitation. The total energy per unit volume $\mathcal{E} = \frac{1}{2} V^{-1} \int d^3r [\nabla S_+ \cdot \nabla S_- + (\nabla S_z)^2]$ is

$$\mathcal{E} = \tfrac{1}{2} [q_1^2 \alpha_0^2 + q_2^2 \beta_0^2 + \tfrac{1}{2} (\vec{q}_1 - \vec{q}_2)^2 \alpha_0^2 \beta_0^2]\,. \tag{33}$$

The magnon number $N = N_1 + N_2$ is defined as

$$N = 1 - V^{-1} \int d^3r\, S_z\,. \tag{34}$$

It is assumed that $N \ll 1$. Then N to fourth order is

$$N = \tfrac{1}{2}(\alpha_0^2 + \beta_0^2) + \tfrac{1}{8}(\alpha_0^4 + 4\alpha_0^2\beta_0^2 + \beta_0^4)\,, \tag{35}$$

from which we see that

$$N_1 = \tfrac{1}{2}\alpha_0^2 (1 + \tfrac{1}{4}\alpha_0^2 + \tfrac{1}{2}\beta_0^2)\,. \tag{36}$$

Solving for α_0 we find

$$\tfrac{1}{2}\alpha_0^2 = N_1(1 - \tfrac{1}{2} N_1 - N_2)\,. \tag{37}$$

The free energy is, in terms of N_i,

$$\mathcal{E} = \sum q_i^2 N_i (1 - \tfrac{1}{2} N_i) - \sum_{i \neq j} N_i N_j \vec{q}_i \cdot \vec{q}_j\,. \tag{38}$$

This is of the form Eq. (28) although the diagonal terms of h' play a role not previously anticipated. (Usually the term in h' is dropped on the grounds that the magnon distribution is isotropic.) It is seen that $(\partial\mathcal{E}/\partial N_i)_{N_j} = \omega_i$ so there is internal consistency with the definition of N_i.

If the calculation in the locally rotated frame is carried to the next order, it is found that

$$\sin^2\theta = 2N_1(1 - \tfrac{1}{2}N_1 + N_2)\,. \tag{39}$$

Thus θ is dependent not only on N_1 but on N_2 as well. Keeping θ fixed means that an increase ∂N_2 is accompanied by a decrease $-N_1\partial N_2$ in N_1. Hence

$$\left(\frac{\partial\mathcal{E}}{\partial N_2}\right)_\theta = q_2^2 - N_1 q_1^2 = q_2^2 - \tfrac{1}{2} q_1^2 \sin^2\theta = q_2^2 - 2|\vec{a}|^2\,. \tag{40}$$

Considering excitations in the locally rotated frame is equivalent to fixing θ, so the result of Eq. (40) is gratifying.

V. CONCLUSION

As in I, the same excitation is described by two different energies. Both are correct, as they refer to different frames of reference.

The energy measured by neutron scattering is the pole in the susceptibility. At low temperature this is clearly the laboratory-frame energy of the elementary excitations. At higher temperatures, the nonlinear interaction spoils the concept of elementary excitation in the laboratory frame. In the locally rotated frame it is still valid, however, at least at short enough wave lengths. Because of the nonlinearity, the elementary excitations do not map onto a single wave number in the laboratory. Close to and above T_C the energy observed by neutrons should be that of the local frame, $D(Q^2 - 2|\vec{a}|^2_{av})$ with an inhomogeneous broadening comparable to the shift.

Below T_C, an interesting transition region awaits further study. As $|\vec{a}|$ becomes smaller, and the fluctuations become concentrated toward smaller θ, the form of the observed spectrum should change. In fact, the observed spectrum may depend on the polarization of the exciting neutron. This could happen because the neutron does not excite an elementary excitation in the laboratory frame. Rather, it excites something with a more or less definite wave-number–frequency relation in the rotated frame. Because the thermal fluctuations are nonlinear, different polarization components can have different shifts and broadening in going from local frame to laboratory frame.

In a similar way, as in I, the energies describing thermal occupations will change their character as the fluctuations become nonlinear. This is a more consequental problem in the case of nonlinear magnons than it was for the quasiparticles of I. It deserves fuller study.

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‡Present address: National Bureau of Standards, Washington, D.C. 20234.

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