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Electron-phonon interaction and itinerant-electron ferromagnetism

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Starting from a consideration of how the phonon frequency would change with the spin splitting of the conduction-electron energy bands in an itinerant-electron ferromagnet, we find that in the paramagnetic region the effect of the electron-phonon interaction makes the temperature dependence of the spin susceptibility Curie-Weiss-type, with the Curie temperature T_C much lower than in the Stoner theory. Further, for $T < T_C$ we find that the phonon effect on magnetization can be large enough to account for the anomalous temperature dependence of spontaneous magnetization often observed in itinerant-electron ferromagnets, especially in Invar alloys. To confirm such a conclusion, with the same model and approximation we discuss also the very closely related problem of the magnetic field effect on the sound velocity in an itinerant-electron ferromagnet, and our theory is found to explain satisfactorily the varied observed results for both $T > T_C$ and $T < T_C$.

I. INTRODUCTION

One of the fundamental problems concerning itinerant-electron ferromagnetism[1] is to explain the Curie-Weiss-type behavior of the paramagnetic spin susceptibility widely observed in itinerant-electron ferromagnets. Even within the Stoner theory we can derive a Curie-Weiss-type spin susceptibility but the Curie-Weiss-type behavior thus obtained is restricted to the vicinity of the Curie temperature T_C, quite contrary to actual observations, and furthermore, the Curie temperature the Stoner theory gives is generally too high by more than an order of magnitude. Another fundamental problem which confronts the Stoner theory is concerned with the temperature dependence of the spontaneous magnetization below T_C; in itinerant electron ferromagnets the observed characteristic temperature dependence of magnetization is different for different materials; for example, as we increase temperature toward T_C the magnetization decreases much faster in Fe-Ni Invar alloys[2] than in pure Ni. Such difficulty in understanding temperature dependence of magnetization has been recently emphasized.[3]

As for the first problem of explaining the observed Curie-Weiss-type spin susceptibility, very intensive progress is being made in various directions.[4–17] As for the second problem of understanding the variety in the temperature dependence of spontaneous magnetization, however, there seems to be much less effort.

The purpose of the present paper is to present an entirely new approach and results on both of the above two fundamental problems in itinerant-electron ferromagnetism; we demonstrate that the effect of the electron-phonon interaction can be responsible both for the Curie-Weiss-type paramagnetic spin susceptibility, with T_C much lower than in the Stoner theory, and the anomalous temperature dependence of the magnetization.

As for the possible effect of the electron-phonon interaction on the magnetic properties of a metal, however, it is widely believed that it does not play any significant role. Such general belief seems to have been derived from some earlier trials[18] to show that the spin susceptibility of a metal is not affected by the electron-phonon interaction. Those earlier discussions are by no means complete nor comprehensive enough, however. Thus, even after the very influential critical review by Herring there have been continuing efforts to reexamine the possible effect of the electron-phonon interaction on the spin susceptibility.[19–22] The origin of such drastic difference between those earlier results and ours is that we include the effects of the exchange interaction between electrons fully self-consistently in dealing with the electron-phonon interaction; based on the jellium model with some extension, we treat electrons and ions on the same footing in handling the dynamics of the coupled system of electrons and ions.

Let us briefly summarize what we will show in the present paper. Firstly, as for the spin susceptibility, the following mean-field approximation result is familiar:

$$\chi_0=\frac{2\mu_B^2 N(0)}{1-\tilde{V}N(0)}\ , \tag{1.1}$$

where $N(0)$ is the electronic density of states per spin at the Fermi surface and $\tilde{V}$ is the effective-exchange interaction. As discussed by Hopfield[19] and Enz and Matthias,[21] the effect of the electron-phonon interaction may be considered to modify the exchange interaction from $\tilde{V}$ to $\tilde{V}+J_{\rm ph}$ in Eq. (1.1). According to the prevailing view, however, the size of such a phonon effect is estimated as

$$O(|J_{\rm ph}|N(0))\simeq\hbar\omega_D/\epsilon_F\simeq 10^{-2}\ , \tag{1.2}$$

where ϵ_F and ω_D are, respectively, the Fermi energy and the Debye frequency. Thus, the phonon effect can be important only in the very special situations where either the magnitude of $\tilde{V}N(0)$ is only slightly smaller than unity, for example, by ~ 0.01, or $\hbar\omega_D/\epsilon_F$ happens to be as large as ~ 0.1, as conjectured for $ZrZn_2$.[19–21] Recently, however, we showed[23] that the size of the phonon effect on the spin susceptibility is enhanced from the magnitude of Eq. (1.2) by the Stoner exchange-enhancement factor,

$$O(|J_{\rm ph}|N(0))\simeq 1/[1-\tilde{V}N(0)](\hbar\omega_D/\epsilon_F)\ . \tag{1.3}$$

Furthermore in our theory, the magnitude and sign of $J_{\rm ph}$ are related to the electronic density of states near the Fermi surface of a metal in a very simple way; $J_{\rm ph}$ can be either positive or negative.

What we will show still further in this paper is that $J_{\rm ph}$ becomes proportional to T,

$$J_{\rm ph}(T)N(0)\propto k_B T/\epsilon_F\ . \tag{1.4}$$

The relation holds from high temperatures to quite low temperatures of $\sim 0.2\Theta$, where Θ is the Debye temperature. As can be easily envisaged, the temperature dependence of Eq. (1.4) leads to the Curie-Weiss-law spin susceptibility.

Secondly, let us summarize our result on the phonon effect on the magnetization behavior for $T<T_C$. Naively, corresponding to Eq. (1.2), the possible effect of the electron-phonon interaction on magnetization $M_{\rm ph}$ per atom is expected to be

$$O(|M_{\rm ph}|/N\mu_B)\simeq\hbar\omega_D/\epsilon_F\simeq 10^{-2}\ , \tag{1.5}$$

N being the total number of atoms in the system. Since generally the magnetization per atom is $\sim 1\mu_B$, the phonon effect as estimated in Eq. (1.5) cannot be important. Here again, however, we find that the phonon effect on magnetization is enhanced by the effect of the exchange interaction between electrons by a factor as large as $\sim 10^2$ to make $|M_{\rm ph}|/N\mu_B\sim 1$.[24,25] Furthermore, in our model numerical calculation, the sign of the phonon effect turns out to be negative; the magnetization is reduced by the effect of the electron-phonon interaction as is required to explain the observations such as mentioned above on Fe-Ni alloys.

As summarized in the above, our conclusion in this paper is that the effect of the electron-phonon interaction on the magnetic properties of a metal is far more important than generally thought. How could such a conclusion of ours be confirmed experimentally? In the course of our discussion we find that the present problem of how the magnetic properties of a ferromagnetic metal is influenced by the effect of the electron-phonon interaction is very closely related to the problem of how the sound velocity or the phonon frequency of the ferromagnetic metal is influenced by an external magnetic field, both above and below T_C; we can treat both problems quite in parallel, exactly with the same model and approximations. Success in the latter problem with our approach would be a strong support to our conclusion on the former problem; our theory offers a first quantitative explanation on the observed magnetic-field effect on sound velocity both for $T>T_C$ (Ref. 26) and $T<T_C$ (Ref. 27). Thus, in order to make our new conclusion on the former problem persuasive, and simultaneously show the importance of studying magnetic field dependence of sound velocity, we discuss both problems together in this paper.

In Sec. II we show generally how the phonon contribution to the free energy becomes magnetization dependent and how the spin susceptibility for $T>T_C$ and the magnetization for $T<T_C$ are related to the external field or the magnetization dependence of the sound velocity in each temperature region. After such general preparation, detailed discussions for the temperature regions $T>T_C$ and $T<T_C$ are given, respectively, in Secs. III and IV. Concluding remarks are given in Sec. V. In the Appendix we briefly derive the basic starting point of our whole discussion, how the phonon frequency changes with the spin splitting of the conduction-electron bands.

II. PHONONS AND PHONON FREE ENERGY IN FERROMAGNETIC METALS

Our discussion in this paper proceeds as follows: The free energy of a metal can be divided into the electron part F_e and the phonon part $F_{\rm ph}$,

$$F=F_e+F_{\rm ph} . \tag{2.1}$$

The phonon part is given in terms of the phonon frequency ω_q as

$$F_{\rm ph}=\tfrac{1}{2}\sum_q \hbar\omega_q + k_BT\sum_q \ln[1-\exp(-\hbar\omega_q/k_BT)] , \tag{2.2}$$

where the polarization of phonons is understood to be included in q. The magnetization M below T_C of the metal is determined by the following minimization condition:

$$\frac{\partial F_e(M)}{\partial M}+\frac{\partial F_{\rm ph}(M)}{\partial M}=0 . \tag{2.3}$$

Correspondingly, the spin susceptibility χ for $T>T_C$ is obtained as

$$\frac{1}{\chi}=\left.\frac{\partial^2 F_e(M)}{\partial M^2}\right|_{M=0}+\left.\frac{\partial^2 F_{\rm ph}(M)}{\partial M^2}\right|_{M=0} . \tag{2.4}$$

What, then is the physical origin of the magnetization dependence of phonon free energy $F_{\rm ph}(M)$? As can be seen from Eq. (2.2), it is through the magnetization dependence of the phonon frequency ω_q. The phonon frequency depends on the magnetization and/or magnetic field as follows: The ion-ion interaction in a metal is screened by the conduction electrons, and the screening behavior of the electrons is modified by the magnetization of the screening electrons. Thus our whole discussion starts from studying how the ion-ion interaction is screened and how the screening behavior is affected by the spin splitting of the energy bands of the conduction electrons.

From the nature of our discussion, it is essential to treat the electrons and phonons on the same footing. Under such a requirement, at present it may be the only possible choice to use the jellium model, with some extension, and the mean-field approximation. With such a model and approximation, as discussed in detail elsewhere,[28] we have only the longitudinal-acoustic phonons, and the phonon frequency ω_q is obtained in the form (see Appendix)

$$\omega_q^2=\Omega_q^2-|g(q)|^2\times\frac{\tilde F_+(q)+\tilde F_-(q)}{1+V(q)[\tilde F_+(q)+\tilde F_-(q)]} , \tag{2.5}$$

where Ω_q is the bare-phonon frequency, $g(q)$ is the electron-phonon interaction constant, $V(q)=4\pi e^2/q^2$ is the long-range Coulomb repulsion, and $\tilde F_\pm(q)$ are the exchange-enhanced static Lindhard functions of $\pm$ spin electrons,

$$\tilde F_\pm(q)=\frac{F_\pm(q)}{1-\tilde V(q)F_\pm(q)} , \tag{2.6}$$

$F_\pm(q)$ being given as

$$F_\pm(q)=\sum_k \frac{f(\epsilon_{k,\pm})-f(\epsilon_{k+q,\pm})}{\epsilon_{k+q,\pm}-\epsilon_{k,\pm}} , \tag{2.7}$$

where $\epsilon_{k,\pm}$ is the one-particle energy of an electron with wave number k and $\pm$ spin [see Eq. (3.1)], and $f(\epsilon)$ is the Fermi distribution function. Finally, $\tilde V(q)$ is the effective-exchange interaction (see the Appendix).

The expression for the phonon frequency of Eq. (2.5) can be used for the ferromagnetic state of a metal where $F_+(q)\neq F_-(q)$, as well as for the paramagnetic state. In the paramagnetic state where

$$F_+(q)=F_-(q)=F(q) ,$$

Eq. (2.5) is simplified to

$$\omega_q^2=\Omega_q^2-|g(q)|^2\frac{\chi_0(q)}{1+V(q)\chi_0(q)} \tag{2.8}$$

$$=\Omega_q^2-|g(q)|^2\frac{2F(q)}{1+[2V(q)-\tilde V(q)]F(q)} , \tag{2.8'}$$

where $\chi_0(q)$ is the familiar wave-number-dependent exchange-enhanced spin susceptibility (with $\mu_B^2=1$),

$$\chi_0(q)=\frac{2F(q)}{1-\tilde V(q)F(q)} . \tag{2.9}$$

With $\tilde V(0)=\tilde V$ and $F(0)\cong N(0)$, Eq. (2.9) reduces to Eq. (1.1).

If we neglect the exchange effect by setting $\tilde V(q)=0$ in Eq. (2.8'), it reduces to the familiar result.[29] Even the expression of the form of Eq. (2.8') has long been known. Note, however, that it is not a trivial exercise to write the phonon frequency in the form of Eq. (2.8) in terms of $\chi_0(q)$ rather than in the form of Eq. (2.8'). In the paramagnetic state the entire magnetic properties of a metal are embodied in the spin susceptibility $\chi_0(q)$, and the phonon frequency is determined by this $\chi_0(q)$. We can immediately see that the larger the spin susceptibility is, the softer the phonon be-

comes. Actually such observations have long been made[30] but not adequately understood by including the exchange-interaction effect as in Eq. (2.8).

As for the phonon frequency in the ferromagnetic state of a metal, Eq. (2.5) shows how it depends on the magnetization; as the spin splitting of the bands changes with magnetization, $F_{\pm}(q)$ and, accordingly, $\tilde{F}_{\pm}(q)$ change in Eq. (2.5). Recently,[31] starting from Eq. (2.5) we studied numerically how sensitively the characteristic magnetization dependence of the sound velocity is related to the location of the Fermi energy in the electronic density of states; depending upon the location of the Fermi energy in a given density of states, the magnetization dependence of sound velocity can be either large or small. Our numerical result is included in Fig. 6. Actually, in some ferromagnetic alloys, such as the Fe-Ni and Fe-Pt alloys,[32] the elastic constant as measured through sound velocity is observed to change with magnetization quite drastically, as much as by an order of magnitude. On the other hand, in pure Ni, for instance, the elastic properties are much less related to the magnetization.

It is instructive and convenient to rewrite Eq. (2.5) in the following form:

$$\omega_q^2=[\Omega_q^2-|g(q)|^2/V(q)] + \frac{|g(q)|^2/V(q)}{1+V(q)[\tilde{F}_+(q)+\tilde{F}_-(q)]} . \tag{2.5'}$$

Remember that in the genuine jellium model the bare-phonon frequency Ω_q coincides with the ionic-plasma frequency $\Omega_{\rm pl}$ and that[29]

$$\Omega_{\rm pl}^2=|g(q)|^2/V(q) .$$

We have

$$\Omega_q^2-|g(q)|^2/V(q)=0$$

for the jellium model. Thus the first term on the right-hand side of Eq. (2.5') is understood to represent the non-point-charge part of the ion-ion interaction. Correspondingly, the second term shows how the point-charge part of the ion-ion interaction is screened by the conduction electrons.

In the actual calculation, given below, of the phonon frequency to be inserted in Eq. (2.2), we use the Debye approximation

$$\omega_q=sq , \tag{2.10}$$

where s is the velocity of sound to be obtained from the $q\to 0$ limit of Eq. (2.5). In handling Eq. (2.5) with such a limit let us put deviations from the jellium model in terms of a parameter ξ as

$$\Omega_q^2-|g(q)|^2/V(q)\equiv \xi s_0^2 q^2 , \tag{2.11}$$

where $s_0=\Omega_{\rm pl}/[8\pi e^2 N(0)]^{1/2}$ is the Bohm-Staver sound velocity. ξ can be either positive or negative and for the jellium model $\xi=0$. Then the sound velocity is obtained as

$$\left[\frac{s}{s_0}\right]^2=\xi+\frac{2N(0)}{\tilde{F}_+(0)+\tilde{F}_-(0)} . \tag{2.12}$$

For low temperatures, such as

$$(k_B T/\epsilon_F)^2 \ll 1 ,$$

$$F_{\pm}(0)=\lim_{q\to 0} F_{\pm}(q)=-\sum \frac{\partial f(\epsilon_{k,\pm})}{\partial \epsilon_{k,\pm}} \cong N_{\pm}(0) , \tag{2.13}$$

where $N_{\pm}(0)$ is the density of states of $\pm$ spin electrons at the Fermi surface.

Note that for $T>T_C$ the sound velocity becomes related to the uniform spin susceptibility of Eqs. (1.1) or (2.9) in a very direct way,

$$\left[\frac{s}{s_0}\right]^2=\xi+\frac{2N(0)}{\chi_0(0)} . \tag{2.14}$$

According to Eq. (2.14), as the temperature approaches T_C from above, the sound velocity decreases. The relative size of such temperature dependence, however, depends on the magnitude of ξ quite sensitively. Since $2N(0)/\chi_0(0)$ varies only between unity and zero, if $\xi \gg 1$ the expected relative change of the sound velocity is small. Experimentally,[32] as the temperature approaches T_C from above, a large decrease of sound velocity was observed in Fe-Ni and Fe-Pt Invar alloys, while such a tendency was not observed in pure Ni. Such a difference in the temperature dependence may be attributed to the difference in the magnitude of ξ; $\xi<1$ for Fe-Ni and Fe-Pt alloys, but ξ seems to be sufficiently larger than unity in Ni.

Another important fact to be considered concerning the characteristic difference between the behaviors of Ni and Fe-Ni Invar alloys is the effect of thermal expansion on ξ. Since ξ represents the non-point-charge part of the ion-ion interaction which is generally short-ranged, such as the core-core interaction, the value of ξ is expected to be very sensitive to a volume change; if ξ is to represent the effect of the core-core repulsive interaction, the value of ξ would increase very rapidly with decreasing ion-ion distance. Therefore, al-

though from the second term $2N(0)/\chi_0$ alone of Eq. (2.14) the sound velocity in a ferromagnet is expected to decrease as the temperature is lowered toward T_C, such a tendency is countered by an increase in the magnitude of $\xi(T)$ caused by thermal contraction.

In this respect, note that the ferromagnetic Invar alloys, by definition, thermal expansion is much smaller than in non-Invar ferromagnets; owing to small or even negative thermal expansion coefficients in Invar alloys, the elastic softening caused by the temperature dependence in $2N(0)/\chi_0(T)$ is not overridden by the temperature dependence in $\xi(T)$. In contrast, with Ni, apart from the fact that ξ is larger than unity, the increase in $\xi(T)$ owing to thermal contraction is faster than the decrease in $2N(0)/\chi_0(T)$. We think this is the reason why the elastic softening is distinctly observed in the Invar alloys but not in non-Invar ferromagnets like Ni as we lower the temperature toward T_C.

Here let us note that our later discussions on the effect of the electron-phonon interaction on the magnetic properties of a metal are based on Eqs. (2.12) and (2.14), but it does not matter essentially whether elastic softening is observed or not. As will be shown, what matters is the magnetic field or magnetization dependence of phonon frequency. In the discussion of the phonon mechanism of the Curie-Weiss-law spin susceptibility, for instance, whether phonons soften or not is of importance only through the factor $[s_0/s(0)]^2$ in Eq. (3.20) and clearly our result is not governed by this factor in any essential way.

As described at the beginning of this section what we need to know in our discussion is how the phonon frequency of the sound velocity depends on the magnetization or the external magnetic field. Our simple equation (2.12) gives a basis for such study, as will be shown in Secs. III and IV, respectively, for $T > T_C$ and $T < T_C$.

Once we know the magnetization and/or the external magnetic field dependence of sound velocity, the phonon part of the free energy Eq. (2.2) can be written as

$$F_{\rm ph}(\eta)=F_{\rm ph}(0)+\Delta F_{\rm ph}(\eta) . \tag{2.15}$$

In the thermal equilibrium, half the spin splitting of the band η is related to the magnetization M as

$$\eta=\mu_B H+\tfrac{1}{2}\tilde{V}M , \tag{2.16}$$

$$M=n_+-n_- , \tag{2.17}$$

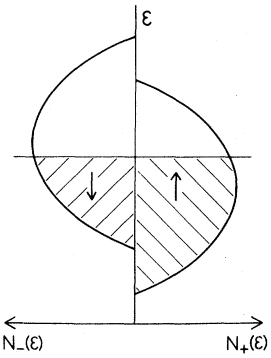

FIG. 1. Density of states of electrons with $\pm$ spins, split by either spontaneous magnetization and/or an external magnetic field. The occupied states are shaded, and throughout this paper we assume the majority electrons are with + spin.

where $n_\pm$ is the total number of electrons with $\pm$ spins, and throughout this paper we assume the majority spin electrons are with + spins as shown in Fig. 1. Note that the η to be used in the variational calculation with Eq. (2.15) is different from Eq. (2.16); the η to be used in Eq. (2.15) is related to M only through the electron-number conservation and in the paramagnetic state it is given as

$$\eta=M/[2F(0)]\simeq M/[2N(0)] . \tag{2.16'}$$

In the Debye approximation the η-dependent part of the phonon free energy is expressed as

$$\Delta F_{\rm ph}(\eta)=Nk_B\Theta P(T/\Theta)\Delta s(\eta)/s(0) , \tag{2.18}$$

where we set

$$s(\eta)=s(0)+\Delta s(\eta) , \tag{2.19}$$

$$P(x)=\frac{3}{8}+xD\left[\frac{1}{x}\right] , \tag{2.20}$$

$D(y)$ being the Debye function

$$D(y)=\frac{3}{y^3}\int_0^y dz\frac{z^3}{e^z-1} .$$

Note that the function $P(T/\Theta)$ does not depend on η; the η dependence comes only through that of the sound velocity $\Delta s(\eta)$. We discuss the temperature dependence of the function $P(T/\Theta)$ in detail in the next section. $\Delta s(\eta)$ is studied in Secs. III and IV, respectively, for $T > T_C$ and $T < T_C$.

III. PHONON EFFECT ON SPIN SUSCEPTIBILITY ABOVE T_C; PHONON MECHANISM OF CURIE-WEISS LAW

A. Magnetic field dependence of sound velocity for $T > T_C$

According to the prescription given in Sec. II, in order to calculate the size of the phonon effect on the paramagnetic spin susceptibility of a metal, we must know how the sound velocity would change with magnetization M and/or external magnetic field H. For that purpose, the only thing that we must do is study how the zero wave-number limit of the Lindhard function $F_{\pm}(0)$ would change with the spin splitting of the energy bands of the metal in Eq. (2.12).

Under magnetization and an external magnetic field H in the direction of the negative z axis (see Fig. 1), the one-particle energy of an electron as appears in Eqs. (2.7) and (2.13) is given as (see the Appendix)

$$\begin{aligned}\epsilon_{k_\pm} &= \epsilon_k - \tilde{V} n_\pm \mp \mu_B H \\ &= \epsilon_k \mp \tfrac{1}{2}\tilde{V}(n_+ - n_-) \mp \mu_B H \\ &\quad - \tfrac{1}{2}\tilde{V}(n_+ + n_-) \\ &= \epsilon_k \mp \eta \ , \end{aligned} \tag{3.1}$$

where η is half the spin splitting of the bands as already defined in Eq. (2.16). In the last equation we neglected the constant quantity

$$-\tfrac{1}{2}\tilde{V}(n_+ + n_-) \ .$$

The Fermi distribution function is affected by the spin splitting in the following fashion:

$$f(\epsilon_{k_\pm}) = \{1 + \exp[\beta(\epsilon_k - \mu \mp \eta - \Delta\mu)]\}^{-1} \ , \tag{3.2}$$

where $\Delta\mu$ is the change in the chemical potential due to the spin splitting. Then from Eq. (2.13) it is straightforward to obtain the $q \to 0$ limit of the static Lindhard function $F(0;\eta)$ under the spin splitting 2η as

$$\begin{aligned}\lim_{q\to 0} F_\pm(q;\eta) &\equiv F_\pm(0;\eta) \\ &= F_\pm(0) + F'_\pm(0)(\mp\eta - \Delta\mu) \\ &\quad + \tfrac{1}{2}F''_\pm(0)(\mp\eta - \Delta\mu)^2 + \cdots \ , \end{aligned} \tag{3.3}$$

where we set $F_\pm(0;0) = F_\pm(0)$ and

$$F_\pm^{(n)}(0) = -\left[\int d\epsilon_{k_\pm} N(\epsilon_{k_\pm}) \frac{\partial^{(n+1)} f(\epsilon_{k_\pm})}{\partial \epsilon_{k_\pm}^{n+1}}\right]_{\eta=0} . \tag{3.4}$$

In the present situation of the paramagnetic state we can ignore the subscripts $\pm$ in the right-hand sides of Eqs. (3.3) and (3.4), but we retain them to use these equations also for the ferromagnetic state to be discussed in the next section.

The shift $\Delta\mu$ in the chemical potential due to the spin splitting of the bands is obtained from the requirement of the electron-number conservation:

$$\sum_{k,\sigma} f(\epsilon_{k\sigma}) = \sum_{k\sigma}[f(\epsilon_k) + f'(\epsilon_k)(-\sigma\eta - \Delta\mu) + \tfrac{1}{2}f''(\epsilon_k)(-\sigma\eta - \Delta\mu)^2 + \cdots] = 2\sum_k f(\epsilon_k) \ , \tag{3.5}$$

where $\sigma = +$ or $-$. Retaining up to the second order in η, we obtain

$$\Delta\mu = \frac{\frac{1}{2}\sum_k f''(\epsilon_k)}{\sum_k f'(\epsilon_k)}\eta^2 \ . \tag{3.6}$$

By putting Eq. (3.6) into Eq. (3.3) and retaining terms up to $O(\eta^2)$ we obtain finally

$$F_\pm(0;\eta) = F(0) \mp F'(0)\eta + \frac{1}{2}\left[F''(0) - \frac{[F'(0)]^2}{F(0)}\right]\eta^2 \ . \tag{3.7}$$

Note that at low temperatures such as

$$\begin{aligned}&(k_B T/\epsilon_F)^2 \ll 1 \ , \\ &F'(0) = \int N'(\epsilon) f'(\epsilon) d\epsilon \cong -N'(0) \ , \\ &F''(0) = -\int N''(\epsilon) f'(\epsilon) d\epsilon \cong N''(0) \ , \end{aligned} \tag{3.8}$$

where $N'(0)$ is the derivative of the density of states with respect to the energy at $\epsilon = \mu$.

The sound velocity under the spin splitting of 2η is obtained by putting Eq. (3.7) into Eq. (2.12),

$$\left[\frac{s(\eta)}{s_0}\right]^2 = \xi + \frac{1}{\Phi(\eta)} \ , \tag{3.9}$$

$$\Phi(\eta) = D_0 \frac{F(0)}{N(0)}\left[1 + D_0 K\left[\frac{\eta}{W}\right]^2\right] , \tag{3.10}$$

where $D_0=1/[1-\tilde{V}F(0)]$ is the Stoner exchange-enhancement factor, W is the width of the electron-energy band, and we set

$$K=\left\{\frac{1}{2}\left[\frac{N''(0)}{N(0)}-\left[\frac{N'(0)}{N(0)}\right]^2\right]+\bar{V}D_0\left[\frac{N'(0)}{N(0)}\right]^2\right\}W^2\,, \tag{3.11}$$

$$\bar{V}\equiv\tilde{V}N(0)\,. \tag{3.12}$$

In Eq. (3.11), except in D_0, $F^{(n)}(0)$ are approximated by Eq. (3.8) and we used the relation $1+\tilde{V}F(0)D_0=D_0$. Finally, noting

$$|\Delta s(\eta)/s(0)|\ll 1,$$

the relative change in the sound velocity as required in Eq. (2.18) is obtained as

$$\frac{\Delta s(\eta)}{s(0)}\cong\frac{1}{2}\left[\left[\frac{s(\eta)}{s_0}\right]^2-\left[\frac{s(0)}{s_0}\right]^2\right]\Big/\left[\frac{s(0)}{s_0}\right]^2$$

$$=-\tfrac{1}{2}[s_0/s(0)]^2K(\eta/W)^2\,, \tag{3.13}$$

where $[s(0)/s_0]^2=(s/s_0)^2$ is given by Eq. (2.14).

The observed magnetic field dependence of sound velocity in some itinerant-electron ferromagnets is quite large. In Fe-Ni and Fe-Pt alloys,[32] for instance, a magnetic field of less than 10 kG was found to change the sound velocity as much as $\sim 1\%$, both above and below T_C. Since the size of the relative change in the sound velocity due to a magnetic field H is expected to be $O(\mu_B H/W)^2$ for $T>T_C$ and $O(\mu_B H/W)$ for $T<T_C$, such an observation is beyond our simple intuition; with $H=10$ kG, $W=1$ eV, and accordingly $\mu_B H/W \simeq 10^{-4}$, we need very large enhancement factors of $\sim 10^6$ for $T>T_C$ and $\sim 10^2$ for $T<T_C$. Furthermore, we are required to explain the fact that the observed change in the sound velocity of those systems is positive for $T<T_C$ and negative for $T>T_C$. The magnetic field effect on sound velocity is not always large, however; in pure Ni the magnetic field effect appears much less than in Fe-Ni and Fe-Pt alloys both for $T>T_C$ and $T<T_C$. In the following let us show how such a variety of observations for $T>T_C$ can be understood from Eq. (3.13). As for the problem for $T<T_C$, we will discuss it in Sec. IV.

The external magnetic field dependence of sound velocity is obtained by setting

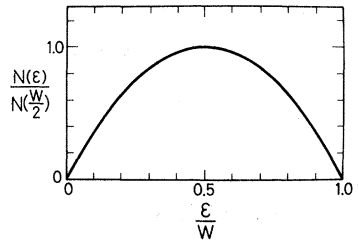

FIG. 2. Electronic density of states defined by Eq. (3.16).

$$\eta=\mu_B H(1+\tfrac{1}{2}\tilde{V}\chi_0)=D_0\mu_B H \tag{3.14}$$

in Eq. (3.13), where we assumed Eq. (2.9) for the paramagnetic spin susceptibility, as

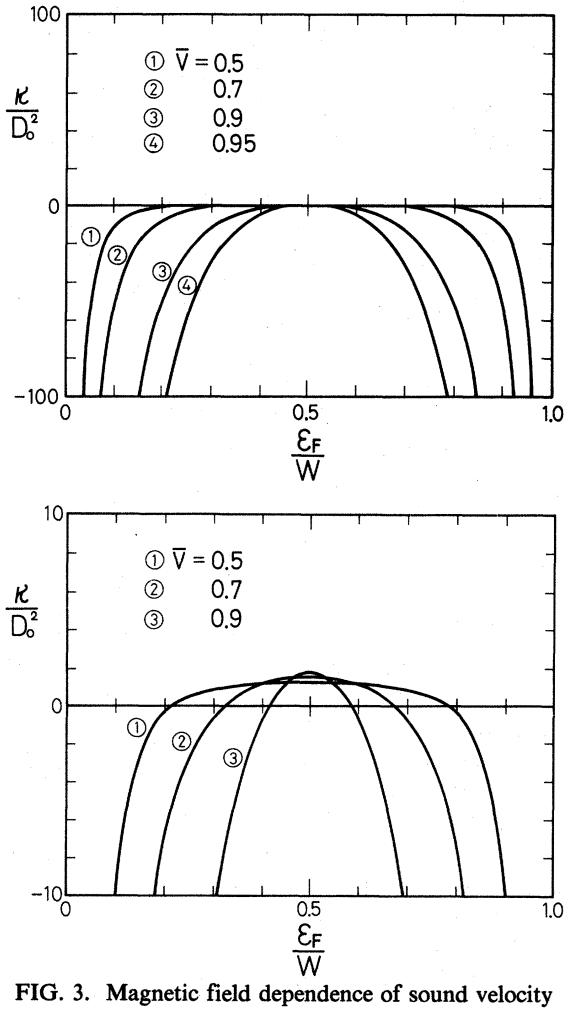

FIG. 3. Magnetic field dependence of sound velocity in the paramagnetic state of metals as defined in Eq. (3.15), for different locations of the Fermi energy in the density of states of Fig. 2, for different values of $\bar{V}$, and with $\xi=1$.

$$\frac{\Delta s(H)}{s(0)} = -\frac{1}{2}\left[\frac{s_0}{s(0)}\right]^2 D_0^2 K \left[\frac{\mu_B H}{W}\right]^2 \equiv \kappa \left[\frac{\mu_B H}{W}\right]^2 . \quad (3.15)$$

Note that the exchange-enhancement factor D_0 appears within K; thus $\Delta s(H)/s(0) \propto D_0^3$.

In order to see how the magnetic field dependence of the sound velocity in a metal is related to the electronic structure and the exchange-enhancement factor of the metal, we carry out a numerical calculation of the quantity κ by assuming the following form of parabolic density of states:

$$N(\epsilon) = (N/6W^3)\epsilon(W-\epsilon) , \quad (3.16)$$

which is shown in Fig. 2.

The result of Fig. 3 shows that the size of the magnetic field dependence of sound velocity is quite sensitive to the values of $\bar{V}$ and ϵ_F/W, ϵ_F being the Fermi energy measured from the bottom of the band in the state without spin splitting. If the exchange interaction is weak, κ is small; in the case of $\bar{V}=0.5$, where $D_0^2=4$, $|\kappa| < 10$ except at the ends of the band. The magnitude of κ is drastically enhanced by the exchange-interaction effect; in the case of $\bar{V}=0.95$, where $D_0^2=400$, $\kappa \cong -10^5$ for $\epsilon_F/W=0.2$ or 0.8. In order to account for the experimental observation on Fe-Ni and Fe-Pt alloys[32] κ is required to be negative and as large as $\sim 10^6$. Such a situation is quite conceivable from our results.

Even if $\bar{V}$ is close to unity, the magnitude of κ becomes small if ϵ_F is located near the peak of the density of states; in the case of $\bar{V}=0.95$, the magnitude of κ for $\epsilon_F/W=0.5$ is smaller than that for $\epsilon_F/W=0.2$ or 0.8 by more than 2 orders of magnitude. The absence of appreciable magnetic field effects on the sound velocity in Ni, in contrast to the case of Fe-Ni and Fe-Pt alloys, strongly suggests that in Ni ϵ_F is located near the peak of the density of states.

The effect of magnetic field on the sound velocity in a paramagnetic metal was earlier discussed by Dieterich and Fulde[33] without including the effect of the exchange interaction between electrons; their result is reproduced by setting $\bar{V}=0$ in our results of Eqs. (3.15) and (3.11). The first difference between the results of Dieterich-Fulde and ours is the factor D_0^2 in Eq. (3.15); this factor represents the fact that the external magnetic field an electron "feels" is exchange enhanced from H to $D_0 H$. The second and the more important difference is the second term on the right-hand side of Eq. (3.11); this term totally disappears for $\bar{V}=0$. Without this term, the magnitude of κ/D_0^2 would have been ~ 1, as suggested by the case of $\bar{V}=0.5$ in Fig. 3. Without our new term the difference between the behaviors of pure Ni and Fe-Ni alloys, for instance, cannot be understood. More importantly, it is this new term that makes the phonon contribution to the paramagnetic spin susceptibility interesting, as will be shown in Sec. III B.

B. Effective exchange interaction due to phonons

Putting Eq. (3.13) into Eq. (2.18), we obtain the η-dependent part of the phonon free energy $F_{\rm ph}(\eta)$. Then from the prescription of Eq. (2.4), the phonon contribution to the spin susceptibility is immediately obtained. Note that here we have to use Eq. (2.16′) for η.

For electrons, the following mean-field approximation result is familiar[4]:

$$\frac{\partial^2}{\partial M^2} F_e(M)\Big|_{M=0} = \frac{1}{2F(0)} - \frac{1}{2}\tilde{V} . \quad (3.17)$$

As for phonons, let us define

$$J_{\rm ph} = -2\frac{\partial^2}{\partial M^2} F_{\rm ph}(M)\Big|_{M=0} , \quad (3.18)$$

which can be understood as the effective exchange interaction due to phonons. Thus, for the total spin susceptibility we obtain

$$\chi = \frac{2F(0)}{1-(\tilde{V}+J_{\rm ph})F(0)} . \quad (3.19)$$

The effective exchange interaction due to phonons is obtained from Eqs. (2.16′), (2.18), (3.13), and (3.18) as

$$J_{\rm ph} = \tfrac{1}{2}(N/W)[1/F(0)]^2[s_0/s(0)]^2 \times KP(T/\Theta)(\hbar\omega_D/W) . \quad (3.20)$$

It would be more convenient to rewrite Eq. (3.20) in the following form,

$$J_{\rm ph}N(0) = [N(0)/F(0)]^2 L(\bar{V},\epsilon_F/W)[s_0/s(0)]^2 \times P(T/\Theta)(\hbar\omega_D/W) , \quad (3.21)$$

where we set

$$L(\bar{V},\epsilon_F/W) = \tfrac{1}{2}[N/N(0)W]K . \quad (3.22)$$

Before entering a detailed study of $J_{\rm ph}$ in Secs.

III C and III D, let us first briefly estimate the possible size of $J_{\rm ph}N(0)$. Earlier discussions, which did not properly include the effect of the exchange interaction between electrons on the screening of the electron-phonon interaction, were unanimous in concluding that

$$O(|J_{\rm ph}|N(0))\simeq\hbar\omega_D/W\simeq 10^{-2}\ .$$

$|J_{\rm ph}|N(0)$ can be much larger than 10^{-2}, however.

In the present approximation of Eq. (3.8), the quantity L is independent of temperature. The two following factors, $[s_0/s(0)]^2$ and $P(T/\Theta)$ in Eq. (3.21), are temperature dependent but always positive and of the order of ~ 1. Thus we can set

$$J_{\rm ph}N(0)\simeq L(\hbar\omega_D/W)\ .$$

As for L, as can be seen from Eqs. (3.22) and (3.11), it can be estimated as

$$|L|\simeq D_0[N'(0)/N(0)]^2W^2\simeq D_0\ . \tag{3.23}$$

Thus we obtain

$$O(|J_{\rm ph}|N(0))\simeq D_0(\hbar\omega_D/W)\ . \tag{3.24}$$

If $D_0\simeq 10$, then $O(|J_{\rm ph}|N(0))\simeq 0.1$; if the phonon effect on the spin susceptibility is of this size it cannot be neglected.

C. Sign and size of the effective exchange interaction due to phonons

There may be situations where the phonon softening (or, hardening) factor $[s_0/s(0)]^2$ plays a very important role in Eq. (3.21), but for simplicity, for the present let us set

$$[s_0/s(0)]^2\simeq 1\ . \tag{3.25}$$

Similarly, the temperature dependence in $F(0)$ can be important when the band is narrow and the temperature is high, but in Eq. (3.19) let us set [see Eq. (2.13)]

$$F(0)\simeq N(0)\ . \tag{3.26}$$

Shortly, in Sec. III D, we will see that the temperature dependence in $P(T/\Theta)$ is much more sensitive than that of $[s_0/s(0)]^2$ and $F(0)$. Under these two approximations, Eqs. (3.19) and (3.21) are simplified to

$$\chi=\frac{2N(0)}{(1-\bar{V})-J_{\rm ph}N(0)}\ , \tag{3.19'}$$

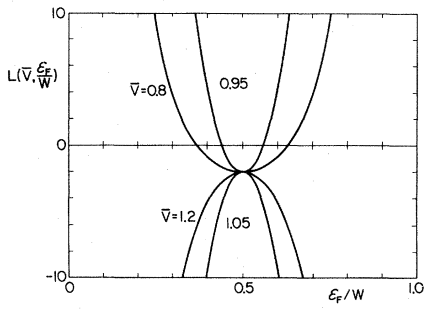

FIG. 4. Calculation of $L(\bar{V},\epsilon_F/W)$ defined by Eq. (3.22) for different values of ϵ_F/W in the electronic density of states of Fig. 2 and for different values of $\bar{V}$.

$$J_{\rm ph}N(0)=L(\bar{V},\epsilon_F/W)P(T/\Theta)\hbar\omega_D/W\ . \tag{3.21'}$$

Thus, in order to fully study the electron-phonon interaction effect on the paramagnetic spin susceptibility we must discuss the quantities L and $P(T/\Theta)$; L determines the size and sign of the effective exchange interaction, whereas $P(T/\Theta)$ gives the temperature dependence.

First, let us carry out a numerical calculation of L by assuming the parabolic electronic density of states of Eq. (3.16) which is shown in Fig. 2 for different values of $\bar{V}$ and ϵ_F/W. The result is given in Fig. 4. Depending upon the values of $\bar{V}$ and ϵ_F/W, L can be either positive or negative and its magnitude can easily be larger than ~ 10. Note that in order to make $|L|\sim 10$, $\bar{V}$ is required to be close to unity. Also note that for $\bar{V}>1$, L becomes always negative: The electron-phonon interaction effect on the spin susceptibility is destructive ($J_{\rm ph}<0$) if the exchange interaction between electrons is strong enough to satisfy the Stoner condition for ferromagnetism ($\bar{V}>1$).

We calculated L also for a Gaussian form of electronic density of states. The result is essentially similar to that of Fig. 4.

D. Temperature dependence of the phonon contribution to spin susceptibility: A new mechanism of Curie-Weiss law

In Eqs. (3.19') and (3.21'), the temperature dependence of the spin susceptibility is determined by that of the function $P(T/\Theta)$ introduced in Eq. (2.20). The following limiting behavior can be easily checked:

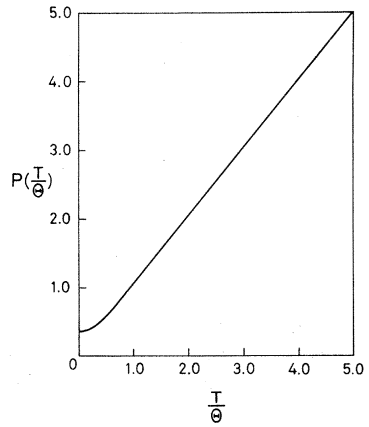

FIG. 5. Temperature dependence in $P(T/\Theta)$ defined by Eq. (2.20).

$$P\left[\frac{T}{\Theta}\right]=\begin{cases}\frac{3}{8} & \text{for } T\ll\Theta\\ \dfrac{T}{\Theta} & \text{for } T\gg\Theta\ .\end{cases}$$

The most interesting point was to see at about what temperature the high-temperature behavior of $P(T/\Theta)$ would start. The result of our numerical calculation on $P(T/\Theta)$ is given in Fig. 5.

Quite surprisingly, the linear T dependence already begins near $T/\Theta\simeq 0.2$. Thus, for $T/\Theta\gtrsim 0.2$ we may set

$$P\left[\frac{T}{\Theta}\right]=\frac{T}{\Theta}\ . \tag{3.27}$$

With the simplification of Eq. (3.27), Eq. (3.19′) is rewritten as

$$\chi=\frac{2\mu_B^2 N(0)}{(1-\bar{V})-L(k_BT/W)}\ , \tag{3.28}$$

where we restored μ_B^2. Let us discuss the cases of $L<0$ and $L>0$ separately.

1. $L<0$: *Destructive phonon effect*

Depending on whether $\bar{V}$ is larger or smaller than unity the susceptibility behaves differently.

(a) $\bar{V}>1$:

$$\chi=\frac{C}{T-T_c}\ , \tag{3.29}$$

$$T_c=(\bar{V}-1)W/k_B\,|L|\ , \tag{3.30}$$

$$C=2\mu_B^2 N(0)W/k_B\,|L|\ . \tag{3.31}$$

(b) $\bar{V}<1$:

$$\chi=\frac{C}{T+T_0}\ , \tag{3.32}$$

$$T_0=(1-\bar{V})W/k_B\,|L|\ . \tag{3.33}$$

In Eq. (3.32), C is the same as in Eq. (3.31).

Case (a) gives the Curie-Weiss-law susceptibility. Our new Curie-Weiss susceptibility has the following convenient features:

(i) As can be seen from the result of Fig. 5 the Curie-Weiss-type temperature dependence is better realized for higher temperatures and valid up to quite low temperatures of $T/\Theta\simeq 0.2$.

(ii) The Curie temperature can be much lower than that which is given by the Stoner theory: Suppose $\bar{V}=1.05$, $W=3$ eV, and $L=-5$. Then Eq. (3.30) gives $T_c\simeq 3\times 10^2$ K. The corresponding value in the Stoner theory is T_c (Stoner) $\simeq(\bar{V}-1)^{1/2}W/k_B\simeq 7\times 10^3$ K.

As for case (b), where the exchange interaction $\bar{V}$ is not strong enough to produce ferromagnetism, note that actually we quite often observe spin susceptibility of the form of Eq. (3.32).[34] An example, for instance, might be Pd for $T>100$ K.

Observation of the paramagnetic spin susceptibility of the form of Eq. (3.32) is usually associated with an antiferromagnetic tendency. According to our phonon mechanism, however, such temperature dependence of spin susceptibility can be produced if (i) $\bar{V}<1$ and (ii) ϵ_F is located at or near the peak of the electronic density of states to make $L<0$ (see Fig. 4), quite independently of anitferromagnetism.

2. $L>0$: *Constructive phonon effect*

According to the results of Fig. 4, in order to have a positive L, we require (i) $\bar{V}<1$ and (ii) that the Fermi surface should not be at or very near the peak of the density of states; we do not need to consider the situation of $\bar{V}>1$ in the case of $L>0$. Thus we obtain the susceptibility in the following form:

$$\chi=\frac{C_a}{T_a-T}\ , \tag{3.34}$$

$$T_a=(1-\bar{V})\frac{W}{k_BL}\ , \tag{3.35}$$

$$C_a=2\mu_B^2 N(0)\frac{W}{k_BL}\ . \tag{3.36}$$

Note that observation of an increasing spin sus-

ceptibility with increasing temperature is rather common among transition metals[34]: Typical examples are Ti ($T<1200$ K), Hf, Zr ($T<1000$ K), Rh, and so on. Although such observations have been attributed entirely to the effects of the electronic structure, our result seems to offer an alternative way of understanding them.

What would happen if T approaches T_a? It is a question to be further pursued. If T is as high as ϵ_F/k_B or T_a, Eq. (3.34) should be drastically modified.

IV. PHONON EFFECT ON MAGNETIZATION BELOW T_C

According to Eq. (2.3), in order to estimate the size of the possible electron-phonon interaction effect on magnetization we must know how the phonon frequency or the sound velocity would change under magnetization or external magnetic field in the temperature region $T<T_C$. We pursue this problem in Sec. IV A. Then with the result of Sec. IV A we treat the problem of the phonon effect on magnetization perturbationally in Sec. IV B. The result of such a perturbational estimation of phonon effect turns out to be quite large and diverges in some cases. Thus in Sec. IV C we reexamine the problem now nonperturbationally.

A. Magnetization and/or magnetic field dependence of the sound velocity for $T<T_C$

The same equation (2.12) as we used in Sec. III A is the basis of our discussion. The difference here is that now the system is in the ferromagnetic state; the system has spontaneous magnetization without the external magnetic field. What we are going to study is how the sound velocity would change if we further magnetize the system by an external magnetic field.

Exactly as in Sec. III A we are required to know how the $q\rightarrow 0$ limit of the Lindhard function would change with the additional spin splitting in the ferromagnetic state; we can use the same equation (3.3). The difference, however, is the η dependence of the shift in the chemical potential; in the ferromagnetic state, in place of Eq. (3.6), we obtain

$$\Delta\mu=-\frac{F_+(0)-F_-(0)}{F_+(0)+F_-(0)}\eta\ . \tag{4.1}$$

Correspondingly, in place of Eq. (3.7), we obtain

$$F_\pm(0;\eta)\cong F_\pm(0)\mp F'_\pm(0)\frac{2F_\mp(0)}{F_+(0)+F_-(0)}\eta\ . \tag{4.2}$$

Note that 2η represents an additional spin splitting of the bands over the spontaneous Stoner magnetization.

Since we know that in the ferromagnetic state, unlike in the paramagnetic state, the η dependence in the sound velocity starts from the linear term, we retain only terms up to first order in η. Putting Eq. (4.2) into Eq. (2.12), with the approximation of Eq. (3.8), we obtain the following result:

$$\frac{\Delta s(\eta)}{s(0)}=A(\epsilon_F,\xi,M)\frac{\mu_B H}{W}\ , \tag{4.3}$$

where

$$A(\epsilon_F,\xi,M)=[s_0/s(0)]^2 Y(\epsilon_F,M)\chi_{\rm HF}(M)\ , \tag{4.4}$$

$$Y(\epsilon_F,M)=-\tfrac{1}{2}N(0)W\left[\frac{N'_+(0)/N_+(0)}{[1-\tilde{V}N_+(0)]^2}-\frac{N'_-(0)/N_-(0)}{[1-\tilde{V}N_-(0)]^2}\right]\left[\frac{N_+(0)}{1-\tilde{V}N_+(0)}+\frac{N_-(0)}{1-\tilde{V}N_-(0)}\right]^{-2}\ , \tag{4.5}$$

and $\chi_{\rm HF}(M)$ is the so-called high-field susceptibility (with $\mu_B^2=1$),

$$\chi_{\rm HF}(M)=\frac{4N_+(0)N_-(0)}{N_+(0)+N_-(0)-2\tilde{V}N_+(0)N_-(0)}\ . \tag{4.6}$$

The magnetic field dependence of the sound velocity has been measured in a number of ferromagnetic metals below T_C, and the result is quite varied.[32] In the case of pure Ni, the size of the field dependence is very small, implying $O(|A|)\simeq 1$ in Eq. (4.3). On the other hand, in the Fe-Ni alloys that show the Invar properties, a magnetic field of less than 10 kG produced a relative increase of $\sim 1\%$ in the sound velocity; with $W\simeq 1$ eV and $\mu_B H\simeq 10^{-4}$ eV this implies $A\simeq 10^2$. To see whether such results can be explained by our result of this section, we carried out a numerical calculation of A for the parabolic density of states of Eq. (3.16). The result is given in Fig. 6.

In Fig. 6, the real lines show the result of our numerical calculation on $A(\epsilon_F,\xi,M)$ for different occupations ϵ_F/W of the band as the function of

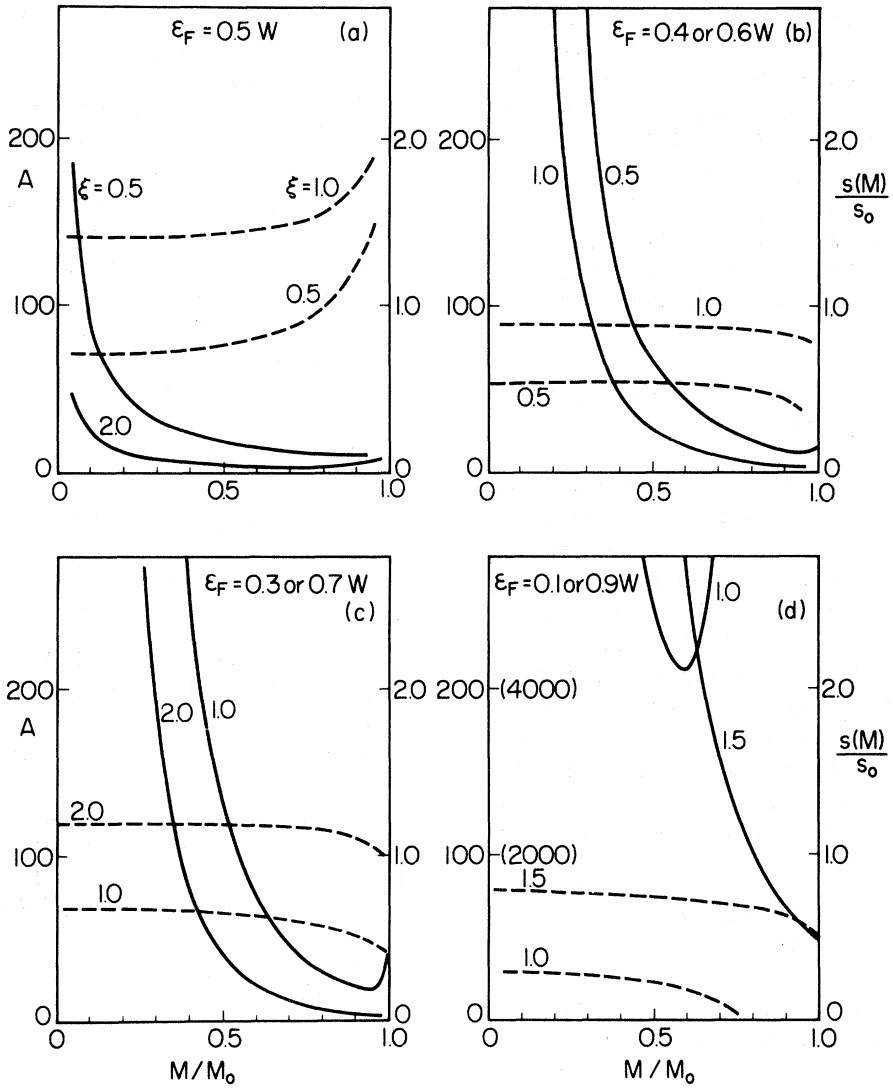

FIG. 6. Magnetization (M) dependence of A defined by Eq. (4.4) for different occupations ϵ_F/W of the band shown in Fig. 2 and for different values of ξ is shown by the real lines. Note that in (d) a different scale shown with parentheses is used for the ordinate for the case of $\xi=1$. The broken lines are for the magnetization dependence of the sound velocity without the external magnetic field.

the parameter ξ and the magnetization M, normalized by the maximum possible magnetization M_0; note that ϵ_F is the Fermi energy measured from the bottom of the band in the paramagnetic state without spin splitting and that M_0 are different for different ϵ_F/W. With a given ϵ_F in the density of states, values of different magnetization M are realized by changing the values of $\bar{V}$. For reference, the sound velocity $s(0)/s_0$ without the external magnetic field is also given by broken lines; to emphasize that $s(0)$ is the sound velocity under the "spontaneous" magnetization M we have $s(M)/s_0$ in place of $s(0)/s_0$ in Fig. 6. Note that with the density of states of Eq. (3.16) both $A(\epsilon_F,\xi,M)$ and $s(M)/s_0=s(0)/s_0$ are the same for $\epsilon_F/W=x$ and $1-x$.

The result of Fig. 6 shows that the magnitude of A can be indeed as large as 10^2-10^3, with positive sign, just as it was required to account for the experimental observations on Fe-Ni and Fe-Pt alloys. Depending upon the values of ϵ_F/W and ξ, however, the detailed behavior of A can be quite different; if the Fermi energy in the paramagnetic state is at or near the maximum of the density of states ($\epsilon_F/W\simeq 0.5$) the magnitude of A is small; for the same value of ϵ_F/W, A becomes smaller for

a larger value of ξ. Remember that in pure Ni the observed magnetic field effect on the sound velocity is negligibly small compared with that of Fe-Ni and Fe-Pt alloys. Within our present model calculation, such a behavior of Ni may be associated with case (a) with $\xi=2$ or larger in Fig. 6. In this respect, it is interesting to note that in Ni the sound velocity $s(M)$ [$=s(0)$] was observed to increase with increasing magnetization[32] in accordance with such behavior of $s(M)$ in Fig. 6; in Fe-Ni and Fe-Pt $s(M)$ decreases with increasing magnetization.

How the sound velocity is affected by the external magnetic field below T_C of of an itinerant-electron ferromagnet is closely related to how the magnetization of the ferromagnet can be affected by the electron-phonon interaction, as we will see in Sec. IV B. Note that our success here in accounting for the observed magnetic field dependence of sound velocity implies the validity of our discussion on the effect of the electron-phonon interaction on magnetization to be given in Secs. IV B and IV C.

B. Phonon effect on magnetization: Perturbational approach

Let us first assume that the phonon effect on magnetization is small and take a perturbational approach. The major part of the magnetization M_e is obtained from the condition

$$\left.\frac{\partial F_e(M)}{\partial M}\right|_{M=M_e}=0 . \tag{4.7}$$

The effect of the electron-phonon interaction is expected slightly to shift the position of the free-energy minimum from M_e to $M=M_e+M_{\rm ph}$. Under the assumption of $|M_{\rm ph}| \ll M_e$, the electron part of the free energy may be expanded as

$$F_e(M)=F_e(M_e)+\frac{1}{2}\left.\frac{\partial^2 F_e(M)}{\partial M^2}\right|_{M_e}(M-M_e)^2+\cdots . \tag{4.8}$$

We retain only the first two terms in the expansion. Then putting Eq. (4.8) into Eq. (2.3) we obtain the phonon contribution to magnetization as

$$M_{\rm ph}\cong -\left.\left[\frac{\partial F_{\rm ph}(M)}{\partial M}\Big/\frac{\partial^2 F_e(M)}{\partial M^2}\right]\right|_{M=M_e} . \tag{4.9}$$

Note that the denominator of the right-hand side of Eq. (4.9) gives the inverse of the high-field susceptibility given by Eq. (4.6):

$$\left.\frac{\partial^2 F_e(M)}{\partial M^2}\right|_{M_e}=1/\chi_{\rm HF}(M_e) .$$

[See Eq. (2.4).] Then Eq. (4.9) can be rewritten in the following form:

$$M_{\rm ph}=\chi_{\rm HF}(M_e)H_{\rm ph} , \tag{4.9'}$$

where the effective magnetic field due to the electron-phonon interaction $H_{\rm ph}$ is defined quite naturally as

$$H_{\rm ph}(M_e)=-\left.\frac{\partial F_{\rm ph}(M)}{\partial M}\right|_{M=M_e} . \tag{4.10}$$

While in the calculation of the magnetic field dependence of sound velocity in Sec. IV A η was related to the change ΔM in magnetization as

$$\eta=H+\tfrac{1}{2}\tilde{V}\Delta M=\frac{N_+(0)+N_-(0)}{4N_+(0)N_-(0)}\chi_{\rm HF}H , \tag{4.11}$$

in the present calculation of Eq. (4.10), as is seen from Eq. (4.2), we should use the following relation:

$$\eta=\frac{N_+(0)+N_-(0)}{4N_+(0)N_-(0)}\Delta M . \tag{4.11'}$$

The difference between Eqs. (4.11) and (4.11') exactly corresponds to that between Eqs. (2.16) and (2.16') of the paramagnetic state; the former change in magnetization is in the thermal equilibrium, but the latter change in magnetization is a variational one.

If we note the relation between Eqs. (4.11) and (4.11') the calculation of Eq. (4.10) can be related to that of Sec. IV A. Corresponding to Eq. (4.3) we obtain

$$\frac{\Delta s(\eta)}{s(0)}=\frac{A}{\chi_{\rm HF}}\frac{\Delta M}{W} , \tag{4.12}$$

where A is given in Eqs. (4.3)–(4.5). From Eqs. (2.18) and (4.12) it is straightforward to obtain

$$H_{\rm ph}=-N\frac{A}{\chi_{\rm HF}}P\left[\frac{T}{\Theta}\right]\frac{\hbar\omega_D(M_e)}{W} , \tag{4.10'}$$

where $\omega_D(M_e)=s(M_e)q_m$, q_m being the Debye cutoff wave number. Putting Eq. (4.10') into Eq. (4.9') we arrive at the final result: If we define a

quantity B as

$$M_{\rm ph}/N\mu_B = B(\hbar\omega_D/W) , \tag{4.13}$$

corresponding to A of Eq. (4.3), it is given as

$$B = -P\left[\frac{T}{\Theta}\right]A , \tag{4.14}$$

$P(T/\Theta)$ being discussed in detail in Sec. III.

The result of Eq. (4.14) shows that the sign of B is opposite to that of A and that the magnitude of B is of the same order as that of A and increases proportionally to the temperature. The fact that A can be easily $\sim 10^2$ with positive sign implies, therefore, that B can be as large as 10^2 with negative sign. If we note that $\hbar\omega_D/W \simeq 10^{-2}$ in Eq. (4.13), $B = -10^2$ implies that the effect of the electron-phonon interaction is to destroy the magnetization by as much as $1\mu_B$ per atom. Such large effect of the electron-phonon interaction on magnetization was never before anticipated.

As we discussed in the preceding subsection, experimentally, A is found to be positive and as large as $\sim 10^2$ in some Fe-Ni and Fe-Pt alloys. Such experimental observations on A strongly suggest that in those alloys B would be negative and as large as $\sim 10^2$.

Clearly the result of Fig. 6, indicates that the effect of the electron-phonon interaction is much too large to justify our present perturbational treatment. We carry out a nonperturbational treatment in Sec. IV C.

C. Phonon effect on magnetization: Nonperturbational approach

In this section we calculate numerically how $E_e(M)$ and $F_{\rm ph}(M)$ change with M; the equilibrium magnetization is determined from the minimum of the total free energy. In order to make the discussion simple let us consider the situation of the zero temperature. Then the electron part of the energy in the mean-field approximation $E_e(M)$ is given as

$$E_e(M) = \sum_{\sigma=\pm}\left[\int_0^{\epsilon_{F\sigma}} \epsilon N(\epsilon)d\epsilon - \frac{\tilde{V}}{2}n_\sigma^2\right] , \tag{4.15}$$

where $\epsilon_{F_\pm}(M)$ is the Fermi energy of the $\pm$ spin electrons measured from the bottom of each band. Correspondingly, the energy of phonons is given from Eq. (2.18) as

$$E_{\rm ph}(M) = \tfrac{3}{8}N\hbar s_0 q_m[s(M)/s_0]$$
$$= a(NW)s(M)/s_0 , \tag{4.16}$$

where we note $P(T/\Theta) = 3/8$ for $T = 0$, $N = q_m^3/3\pi^2$, and $a \simeq \hbar\omega_D/W \simeq 10^{-2}$.

Suppose the minimum of $E_e(M)$ alone is located at $M = M_e$. The problem is how far the location of the energy minimum would shift from M_e if we take $E_{\rm ph}(M)$ into consideration. At first glance, since

$$E_{\rm ph}(M)/NW \simeq O(10^{-2}) ,$$

whereas

$$E_e(M)/NW \simeq O(1) ,$$

the electron-phonon interaction effect may appear negligible within an error of $\sim 1\%$. Such simple reasoning is not warranted, as our numerical result in Sec. IV B shows. Here note that generally the variation of $E_e(M)$ becomes small in the neighborhood of the stationary point of $M = M_e$; near M_e the size of variation in $E_e(M)$ may not necessarily be much larger than that of $E_{\rm ph}(M)$.

In carrying out a numerical calculation of $E_e(M)$ and $E_{\rm ph}(M)$ we use the parabolic electronic density of states of Eq. (3.16). As for a in Eq. (4.16), we assume $a = 10^{-2}$. The result of our numerical calculation is given in Figs. 7–9. In Fig. 7 we show how $E_e(M)$, $E_{\rm ph}(M)$ change with M/M_0 for the case of $\epsilon_F/W = 0.5$. All three energies E_e, $E_{\rm ph}$ and $E_e + E_{\rm ph}$ are plotted with the same energy scale in the unit of NW, although from different origins. As anticipated, near the stationary point of $E_e(M)$ the size of the M dependence of $E_{\rm ph}(M)$ can be as large as that of $E_e(M)$; the location of the minimum of $E_e(M) + E_{\rm ph}(M)$ shifts appreciably from that of $E_e(M)$ alone. A typical situation is shown in the case (i) of Fig. 7; by the phonon effect the magnetization is reduced as much as by $\sim 30\%$, from $M/M_0 = 0.35$ to ~ 0.25. In case (ii) of the smaller exchange interaction $\bar{V} = \tilde{V}N(0) = 1.006$, the Stoner magnetization of $M/M_0 \simeq 0.2$ is totally destroyed.

The case of $\epsilon_F/W = 0.3$ shown in Fig. 8 is qualitatively different from the above two situations of Fig. 7; in Fig. 8 the magnetization dependence of sound velocity or $E_{\rm ph}(M)$ becomes singular at a certain value of M and there the total energy of the system becomes minimum. Such singular behavior is due to the fact that at zero temperature the following situation can be produced in Eq. (2.12),

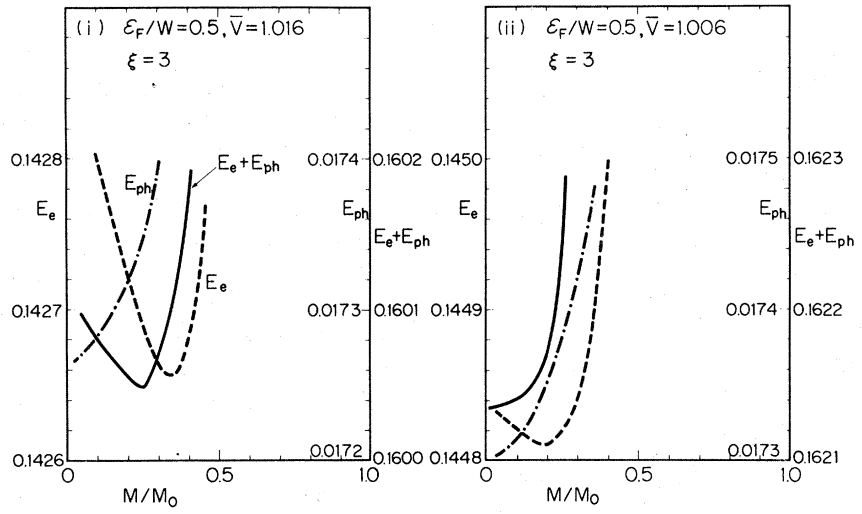

FIG. 7. Magnetization dependence of $E_e(M)$, $E_{ph}(M)$, and $E_e(M)+E_{ph}(M)$ for the case of $\epsilon_F/W=0.5$ in the electronic density of states of Fig. 2. All the energies are measured in the same unit of NW, although from different origins.

$$\tilde{F}_+(0)+\tilde{F}_-(0) = \frac{N_+(0)}{1-\tilde{V}N_+(0)}+\frac{N_-(0)}{1-\tilde{V}N_-(0)}\cong 0\,, \qquad (4.17)$$

for a certain value of M. If the situation of Eq. (4.17) holds, however, we are not justified to use Eq. (2.12); we must treat the problem more dynamically, abandoning the adiabatic approximation for the electron response[28] (see the Appendix). With such an improved treatment the singular behavior of $E_{ph}(M)$ would be rounded off; we should not take too literally the detailed behavior of $E_{ph}(M)$ in Fig. 8.

In Fig. 9 we show how differently the magnetization would be modified for two different occupa-

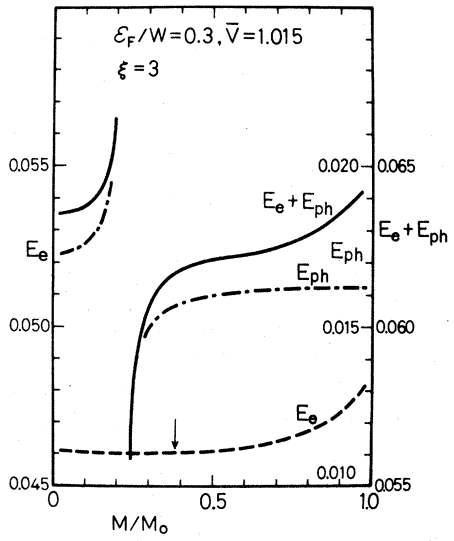

FIG. 8. Magnetization dependence of $E_e(M)$, $E_{ph}(M)$, and $E_e(M)+E_{ph}(M)$ for the case of $\epsilon_F/W=0.3$, as in Fig. 7. The arrow shows the minimum of $E_e(M)$.

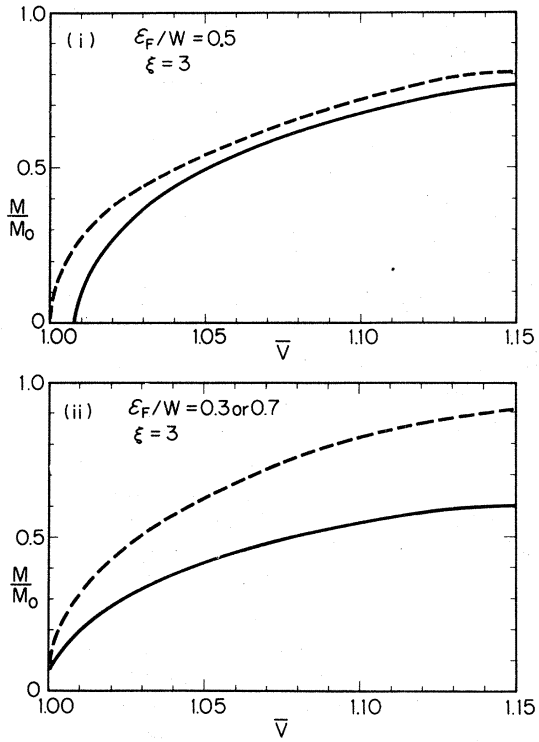

FIG. 9. Equilibrium magnetization with (solid lines) and without (broken lines) the phonon effect as a function of $\bar{V}$, for two different locations of the Fermi energy in the density of states of Fig. 2.

tions of the electron band. Note that with the density of states of Eq. (3.16) the cases of $\epsilon_F/W=x$ and $1-x$ are equivalent.

We carried out similar calculations for more different values of $\epsilon_F/W=0.1(0.9)$, 0.2(0.8), and 0.4(0.6) with the same density of states, and the result is found to be qualitatively similar to that of Figs. 8 and 9 (ii).

A point to note in our result of Fig. 9 is that the manner in which $M_{\rm ph}$ changes with $\bar{V}$ is quite different for different locations of ϵ_F in the density of states; while in Fig. 9 (ii) the phonon effect is generally large and increases with increasing $\bar{V}$, in Fig. 9 (i) the phonon effect is generally small and decreases with increasing $\bar{V}$. With this result let us see how we can understand the drastic difference between the magnetization behaviors of Fe-Ni alloys and Ni (or Fe). In a very approximate view, these two systems have nearly the same form of the density of states but different locations of ϵ_F.

It is well known that in Fe-Ni alloys the temperature dependence of magnetization is anomalous; with increasing temperature the magnetization decreases much faster than, say, in pure Ni. Since in these alloys $\bar{V}$ is considered rather large, such magnetization behavior implies that these systems belong to the case of Fig. 9 (ii); ϵ_F is not located at or very near the maximum of $N(\epsilon)$.

In Ni the phonon effect on magnetization appears much smaller. Since $\bar{V}$ is not considered to be small in Ni, it should belong to the case of Fig. 9 (i); ϵ_F is at or very near the maximum of the density of states.

Remember that earlier we reached exactly the same conclusion on the difference in the electronic structures of Fe-Ni and pure Ni from the magnetization dependence of sound velocity and from the external magnetic field dependence of sound velocity both above and below T_C.

Finally let us note that we set $P(T/\Theta)=\frac{3}{8}$ by assuming $T=0$ in this section. In our numerical calculation the variation of magnetization is realized by changing the value of the effective exchange interaction $\bar{V}$ alone. In reality, the variation of magnetization is driven by temperature; smaller M/M_0 implies higher temperatures. As shown in Fig. 5, $P(T/\Theta)$ increases nearly linear with temperatures. Thus the actual phonon effect on magnetization should be enhanced by the temperature-dependent factor $P(T/\Theta)/\frac{3}{8}$ from the result of Fig. 9; the thermal enhancement is larger for smaller M/M_0, or, as the temperature approaches T_C, from below.

V. CONCLUDING REMARKS

In this paper we pointed out the fundamental importance of considering the role of the electron-phonon interaction in understanding itinerant-electron ferromagnetism. The Curie-Weiss behavior of spin susceptibility for $T>T_C$ and the anomalous temperature dependence of magnetization for $T<T_C$, widely observed in itinerant electron ferromagnets, are shown to be understood from the effect of the electron-phonon interaction. To test such drastic conclusions concerning the phonon effect on magnetism, with the same model and approach we discussed the effect of the magnetic field on sound velocity, which is very closely related to the former effect; unlike the former effect, the latter effect can be measured directly. Our theory was shown to be able to account for the varied observations concerning the latter effect.

Prior to the present work we extensively studied the opposite problem of how the magnetic properties of a metal, either ferromagnetic or not, would affect the phonon properties of the metal. There, too, our model and approach were proved to be quite useful. In addition to what we already mentioned in this paper, we could give an explanation[35] why in Ni the peak in the ultrasonic attenuation is observed not at T_C but at slightly below T_C,[36] for instance. With the present work, we now have a base to discuss comprehensively how the magnetic properties and the phonon properties are related to each other in a metal.

An interesting and challenging problem for our theory is the so-called Invar problem. Some itinerant-electron ferromagnets called Invars,[2] such as Fe-Ni alloys and Fe-Pt alloys, for instance, are quite different from Ni both in magnetic and elastic properties. In this paper we showed how the various aspects of such differences between Fe-Ni alloys and Ni can be systematically understood simply by assuming differences in the locations of the Fermi energy in the electronic density of states, together with the difference in the value of the parameter ξ. According to our analysis, in Ni the Fermi energy in the paramagnetic state is located at or very near the peak in the density of states and the value of ξ is signficantly larger than unity. Correspondingly, in Fe-Ni Invar alloys, the Fermi energy in the paramagnetic state is not expected to be located near the peak in the density of states and the value of ξ is not expected to be larger than unity.

Our discussion throughout this paper is based on a simplest possible model; as for the electron-

phonon interaction, we used essentially the jellium model extended with the introduction of the parameter ξ; as for the electronic density of states, we used the simple form of Eq. (3.16). Owing to the simplicity of our model, we were not required to introduce parameters of nontransparent nature.

We do not claim that we have considered all of the important aspects of the electron-phonon interaction effect in this paper. An important effect of the electron-phonon interaction in a metal is to modify the electronic density of states near the Fermi surface of the metal. What would such an effect be in a itinerant-electron ferromagnet? We discussed this problem both for $T > T_C$ (Ref. 37) and $T < T_C$ (Ref. 38) elsewhere. There are many other aspects to be discussed further.

In going beyond the Stoner theory there are many other mechanisms to be considered besides the electron-phonon interaction. As mentioned already, recently various mechanisms[4-17] have been proposed to understand the Curie-Weiss-law behavior of spin susceptibility together with its low Curie temperature. Since all of those other mechanisms, such as spin-fluctuation effect, consist in going beyond the approximation of Eq. (3.17) in treating the electronic part of the free energy, they are compatible with our phonon mechanism. Which mechanism is more important than others would depend upon materials and may be determined by asking which one would give a lower T_C. In this respect it is important to note that in our phonon mechanism, for given values of $\bar{V}$ and W (or ϵ_F), $T_C \propto 1/|L|$, and according to the result of Fig. 4, $|L|$ can readily be larger than 10 if ϵ_F is not located too close to the peak of the electronic density of states. If $|L| > 10$ it is quite likely that the dominant mechanism to make the spin susceptibility of an itinerant-electron ferromagnet Curie-Weiss-type is the effect of the electron-phonon interaction.

In concluding, let us note that our discussion in this paper is a perturbational one. For $T > T_C$, for instance, we started from the finding that the phonon frequency of a metal is closely related to the spin susceptibility $\chi_0(q)$ of the metal, $\omega_q = \omega_q(\chi_0)$. Then we proceeded to find that the spin susceptibility of the metal is modified by the effect of phonons; the spin susceptibility χ including the effect of the electron-phonon interaction is drastically different from χ_0 used in calculating phonon frequency. Our theory at the present stage is not fully self-consistent; there remain many things to be done.

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APPENDIX: PHONON FREQUENCY IN A FERROMAGNETIC METAL: DERIVATION OF EQ. (2.5)

Since the entire discussion of the present paper is based on Eq. (2.5) let us briefly describe how it is obtained. With the familiar forms of Hamiltonians for phonons and electron-phonon interaction[29]

$$H_{\rm ph} = \frac{1}{2} \sum_q (P_q^\dagger P_q + \Omega_q^2 Q_q^\dagger Q_q) , \tag{A1}$$

$$H_{e\text{-ph}} = \sum_q g(q) Q_q n(-q) , \tag{A2}$$

where Q_q and P_q are, respectively, the normal coordinate and corresponding momentum of a phonon with the bare frequency Ω_q and

$$n(q) = \sum_{k,\sigma} a_{k,\sigma}^\dagger a_{k+q,\sigma} = n_+(q) + n_-(q)$$

is the Fourier component of electron-density operator, $a_{k,\sigma}^\dagger$ being the creation operator of an electron at the state with momentum $\hbar k$ and spin σ, we obtain

$$\frac{\partial^2}{\partial t^2} Q_q + \Omega_q^2 Q_q = -g(-q) n(q) . \tag{A3}$$

If we use a linear-response approximation on $n(q)$ by replacing it by its thermal average $\langle n(q) \rangle$, viz.,

$$n(q) \cong \langle n(q) \rangle = -g(q) Q_q \chi_e(q, \omega_q)$$

noting that $Q_q \propto \exp(-i\omega_q t)$, we obtain the equation to determine the modified phonon frequency ω_q,

$$\omega_q^2 = \Omega_q^2 - |g(q)|^2 \chi_e(q, \omega_q) , \tag{A4}$$

where $g(q)g(-q) = |g(q)|^2$. The electron-density susceptibility $\chi_e(q,\omega)$ is defined with respect to the external-charge potential

$$U(q) \exp(iqr - i\omega t) ,$$

which gives rise to the following form of perturbing Hamiltonian:

$$\mathcal{H}_e' = -eU(q) e^{-i\omega t} n(-q) , \tag{A5}$$

as

$$\chi_e(q,\omega)=\frac{\langle n_+(q,\omega)\rangle+\langle n_-(q,\omega)\rangle}{eU(q)}\ , \tag{A6}$$

where we set

$$\langle n_\pm(q)\rangle=\langle n_\pm(q,\omega)\rangle\exp(-i\omega t)\ .$$

We use the following Hamiltonian for the electrons[29]:

$$\mathscr{H}_0+\mathscr{H}_c=\sum_{k,\sigma}\epsilon_k a^\dagger_{k\sigma}a_{k\sigma}+\frac{1}{2}{\sum_{\substack{k,k',\kappa\\ \sigma,\sigma'}}}' V(\kappa)a^\dagger_{k\sigma}a^\dagger_{k'\sigma'}a_{k'-\kappa,\sigma'}a_{k+\kappa,\sigma}\ , \tag{A7}$$

where $V(\kappa)=4\pi e^2/\kappa^2$ is the Coulomb repulsion and the prime on the summation indicates the exclusion of $\kappa=0$ from the sum. Then with the mean-field approximation on the Coulomb interaction between electrons $\mathscr{H}_c$, the electron-density response is obtained from the following self-consistent equation:

$$\langle n_\pm(q,\omega)\rangle=F_\pm(q,\omega)eU(q)-F_\pm(q,\omega)V(q)[\langle n_+(q,\omega)\rangle+\langle n_-(q,\omega)\rangle]+F_\pm(q,\omega)\tilde{V}(q)\langle n_\pm(q,\omega)\rangle\ , \tag{A8}$$

where

$$F_\sigma(q,\omega)=\sum_k\frac{f(\epsilon_{k,\sigma})-f(\epsilon_{k+q,\sigma})}{\epsilon_{k+q,\sigma}-\epsilon_{k,\sigma}+\hbar\omega} \tag{A9}$$

is the dynamical Lindhard response function. The effective exchange interaction $\tilde{V}(q)$ is introduced by the approximation[39]

$$\begin{aligned}\mathscr{H}_c&\cong{\sum_{k,k',\kappa,\sigma,\sigma'}}' V(\kappa)\langle a^\dagger_{k'\sigma'}a_{k'-\kappa,\sigma'}\rangle a^\dagger_{k\sigma}a_{k+\kappa,\sigma}-{\sum_{k,k',\kappa,\sigma,\sigma'}}' V(\kappa)\langle a^\dagger_{k'\sigma'}a_{k+\kappa,\sigma}\rangle a^\dagger_{k\sigma}a_{k'-\kappa,\sigma'}\\
&\cong V(-q)(\langle n_+(q)\rangle+\langle n_-(q)\rangle)\sum_{k,\sigma}a^\dagger_{k\sigma}a_{k-q,\sigma}-\sum_{k,\kappa,\sigma'}V(\kappa)\langle a^\dagger_{k+\kappa,\sigma}a_{k+\kappa,\sigma}\rangle a^\dagger_{k\sigma}a_{k\sigma}\\
&\quad-\sum_{k',k,\sigma}V(k'-k+q)\langle a^\dagger_{k'\sigma}a_{k'+q,\sigma}\rangle a^\dagger_{k\sigma}a_{k-q,\sigma}\\
&\equiv V(-q)(\langle n_+(q)\rangle+\langle n_-(q)\rangle)\sum_{k,\sigma}a^\dagger_{k\sigma}a_{k-q,\sigma}-\sum_{k\sigma}[\tilde{V}(0)n_\sigma]a^\dagger_{k\sigma}a_{k\sigma}\\
&\quad-\sum_{k\sigma}(\tilde{V}(q)\langle n_\sigma(q)\rangle)a^\dagger_{k\sigma}a_{k-q,\sigma}\ ,\end{aligned} \tag{A10}$$

by noting that under the external field with wave vector q the only nonvanishing thermal averages concerning electron density are $\langle n_\sigma(q)\rangle$ and $\langle n_\sigma(0)\rangle=n_\sigma$. The second term on the right-hand side of the last equation represents the exchange self-energy and it can be incorporated into the one-particle energy of an electron as

$$\epsilon_{k\sigma}=\epsilon_k-\tilde{V}n_\sigma\ , \tag{A11}$$

where we set $\tilde{V}(0)=\tilde{V}$; the one-particle energy appearing in Eq. (A9), for instance, is of this sense.

If we notice that $F_\pm(q,\omega)$ is the response function of noninteracting electrons, the meaning of Eq. (A8) becomes clear. On the right-hand side of Eq. (A8) the first term represents the noninteracting electron response to the external potential $U(q)$; if there were no interaction between electrons [$V(q)=\tilde{V}(q)=0$] we would have only this term. The second and third terms represent, respectively, the effects of the Coulomb repulsion and exchange interaction between electrons; the second term is of the form of a noninteracting electron response to the Coulomb potential due to the induced electron-density polarization, and similarly, the third term is of the form of a noninteracting electron response to the exchange potential. Note that in Eq. (A8) the exchange term tends to enhance the electron-density polarization whereas the Coulomb term tends to suppress the polarization. Solving Eq. (A8) for $\langle n_\sigma(q,\omega)\rangle$ and putting them into Eq. (A6) we obtain the electron-density susceptibility,

$$\chi_e(q,\omega)=\frac{\tilde{F}_+(q,\omega)+\tilde{F}_-(q,\omega)}{1+V(q)[\tilde{F}_+(q,\omega)+\tilde{F}_-(q,\omega)]}\ , \tag{A12}$$

with

$$\tilde{F}_\pm(q,\omega)=F_\pm(q,\omega)/[1-\tilde{V}(q)F_\pm(q,\omega)]\ . \tag{A13}$$

The final equation to determine the modified phonon frequency ω_q is obtained by putting Eq. (A12) into Eq. (A4). In Eq. (2.5), noting that $\hbar\omega_q \ll \epsilon_F$, we approximated the dynamical electron-density susceptibility $\chi_e(q,\omega_q)$ by a static one $\chi_e(q,0)$.

Note that the electron spin susceptibility of an interacting electron gas can be treated exactly on the same footing as above. The self-consistent equation for the electron-density response to a magnetic field

$$H(q)\exp(iqr - i\omega t)$$

in the direction of negative z axis (note that we assumed $n_+ > n_-$) is obtained simply by replacing the first term on the right-hand side of Eq. (A8) by

$$\pm F_\pm(q,\omega)\mu_B H(q) .$$

Then the spin susceptibility defined by

$$\chi_m(q,\omega) = \frac{\langle n_+(q,\omega)\rangle - \langle n_-(q,\omega)\rangle}{\mu_B H(q)} \tag{A14}$$

is obtained as

$$\chi_m(q,\omega) = \frac{\tilde{F}_+(q,\omega)+\tilde{F}_-(q,\omega)+4V(q)\tilde{F}_+(q,\omega)\tilde{F}_-(q,\omega)}{1+V(q)[\tilde{F}_+(q,\omega)+\tilde{F}_-(q,\omega)]} . \tag{A15}$$

The static paramagnetic spin susceptibility [Eq. (2.9)] and the high-field spin susceptibility below T_C [Eq. (4.6)] are obtained from Eq. (A15).

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