Theory of the Anomalous Specific Heat of Nickel and Copper-Nickel Alloys at Low Temperatures*

K. H. BENNEMANN†
Argonne National Laboratory, Argonne, Illinois
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The anomalously large specific heat observed at low temperatures for Ni and Cu–Ni alloys is shown to result from the electron-magnon interaction at temperatures below the Curie temperature, and from the electron-paramagnon interaction at temperatures above the Curie temperature.

I. INTRODUCTION

THE low-temperature specific heat of Ni [1] and Cu_xNi_{1-x} alloys[2] is considerably enhanced above the values which follow from band theory.[3] For instance, in the Cu_xNi_{1-x} alloys the magnetic saturation moment decreases with increasing Cu content indicating the filling up of the 3*d* band of Ni by the copper valence electrons. Assuming that any renormalization of the electron mass due to many-body effects is not changing much with the alloy composition, and neglecting for the moment the change in the electron density of states resulting from the Zeeman splitting of the Fermi surface, then consequently the specific-heat contribution which is linear in temperature and which is proportional to the electron density of states at the Fermi surface should decrease monotonically with increasing Cu concentrations until the specific-heat value appropriate for Cu with a filled 3*d* band is reached. However, contrary to this expectation, it was observed some years ago[2,4] that the specific-heat contribution which is linear in temperature is much larger for all Cu–Ni alloys than expected from band theory and exhibits an anomalous increase and a peak for alloys with about 55 at.% Cu just where the magnetic saturation moment and the Curie temperature become zero. Also, at Cu concentrations higher than 55 at.% the specific-heat coefficient γ decreases much slower than expected from band theory. Furthermore, dividing the specific heat C_v by the temperature, one finds an anomalous increase with decreasing temperature for alloys with about 55 at.% Cu which have small Curie temperatures and "reduced" Fermi temperatures, respectively.[2] This anomalous behavior of C_v/T has been interpreted by several authors[2,5] as resulting from fluctuations in the alloy composition leading to ferromagnetic clusters. However, this analysis cannot explain the dependence of the anomalous specific heat on the composition of the alloy; in particular it cannot explain the specific heat of the alloys with very high or very small Ni concentrations. Also, such an explanation of the specific heat of the Cu–Ni alloys in terms of ferromagnetic Ni clusters yields an unreasonable dependence of the Debye temperature on the Ni concentration.

In contrast to this attempt of explaining the experimental specific-heat results we will show that the anomalous behavior of the specific heat of Ni and the Cu_xNi_{1-x} alloys at low temperatures can be explained as resulting from the coupling between electrons and spin fluctuations, or more specifically, as resulting from the electron-magnon interaction in the case of the ferromagnets and from the electron-paramagnon interaction in the case of the "almost" ferromagnetic alloys. Such an explanation is strongly suggested by recent theoretical studies of the effect of electron-spin-fluctuation coupling on the dynamical properties of the electrons.[6] Some years ago it had been speculated by Phillips and Mattheiss[7] that electron-electron interactions might contribute significantly to the effective electron mass of Ni.

Taking into account the coupling between electrons and spin fluctuations one finds that at low temperatures the temperature dependence of the electronic specific heat is approximately given by $C_v=\gamma T+AT^n \ln T+BT\,|\,1-T/T_{\text{Curie}}\,|^{-1/2}$. It is shown that such an expression for the specific heat can explain the experimental results which have been a puzzle for many years. Notice that the exponent n occurring in the above expression for the specific heat depends sensitively on the energy-momentum distribution function of the spin fluctuations, e.g., of the magnons and paramagnons, respectively.

The spectral density function of the spin fluctuations

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† Permanent address: Physics Department, University of Rochester, Rochester, N.Y.
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specifies completely the nature of the spin fluctuations (their spatial extent lifetime, etc.) and hence is basic for any treatment of the coupling between electrons and spin fluctuations. It is remarkable that the theory of the coupling between electrons and spin fluctuations has the same form for spin fluctuations described by the itinerant electron model and for *s-d* or *s-f* exchange interaction involving localized spins and that only the spectral function of the spin excitations specifies the used spin model.

It is of great interest to study the change of the spectral density function around the ferromagnetic phase transition. Therefore, Cu_xNi_{1-x} alloys are a fine candidate for such a study since they form fcc solutions over the entire range of composition and since they become ferromagnetic below 55 at.% Cu.

Notice that the temperature dependence of the specific-heat term $T^n \ln T$ resulting from the coupling between the electrons and the spin fluctuations is scaled by the Curie temperature for the ferromagnets, and by the "reduced" Fermi temperature, which is a measure for the average spin excitation energy, for the "almost" ferromagnetic alloys. Consequently, the anomalous behavior of C_v/T at low temperatures should decrease and finally disappear with increasing Curie temperatures and "reduced" Fermi temperatures, respectively; e.g., on either side of $Cu_{55}Ni_{45}$ with respect to the composition scale.

When the temperature approaches the Curie temperature, then the magnons become more and more strongly damped by the increasing hybridization with the collective Stoner spin excitations, but they persist as so-called paramagnons for temperatures not too far above the Curie temperature. Notice that below the Curie temperature the Zeeman splitting of the Fermi surface tends to suppress the electron scattering involving emission and reabsorption of spin excitations.

All the results of the electron-spin-fluctuation interaction discussed so far are derived in Secs. II and IV. Section II presents a detailed derivation of the specific-heat contribution resulting from the electron-spin-fluctuation coupling. In Sec. III the spectral density function of the spin excitations is discussed in detail. In Sec. IV various approximate expressions are derived for the electronic specific-heat. In Sec. V the theory is used to calculate the specific-heat coefficient γ of Ni and the Cu-Ni alloys and C_v/T for several Cu–Ni alloys with about 55 at.% Cu.

II. THEORY

The interaction between electrons and spin fluctuations is mathematically treated as follows. The specific heat C_v is determined by

$$C_v = \partial E/\partial T, \tag{II1}$$

where the energy E of the electrons is approximately given by

$$E(T) = -T \sum_{n=-\infty,\alpha=\downarrow,\uparrow}^{\infty} \int \frac{d^3p}{(2\pi)^3} \mathrm{Tr}\{[\xi_p + \tfrac{1}{2}\Sigma_\alpha(p,\omega_n)] \times G_\alpha(p,\omega_n)\}, \qquad \omega_n = (2n+1)\pi T. \tag{II2}$$

Here, $\xi_p \equiv (p^2/2m) - \epsilon_F$, and the electronic self-energy Σ_α resulting from the interaction with transverse and longitudinal spin fluctuations is given by

$$\Sigma_\alpha(p,\omega) = \frac{1}{2} \sum_{\lambda;\nu=l,t} \int_{-\infty}^{\infty} \frac{dz}{2\pi} \int_{-\infty}^{\infty} \frac{dz'}{2\pi} \int \frac{d^3p'}{(2\pi)^3} A_{-\alpha}(p',z') \times B_\lambda{}^\nu(p-p',z) \frac{\tanh(z'/2T)}{\omega - z - z' + i\delta}. \tag{II3}$$

Here, $-\alpha$ refers to a spin direction which is opposite to α. The symbol $A_\alpha(p,\omega)$ denotes the spectral weight function of the electronic thermal Green's function $G_\alpha(p,\omega_n)$. Similarly $B_\lambda{}^\nu(q,\omega)$ is the spectral weight function of the spin fluctuations of kind λ. The subscripts l and t refer to longitudinal and transverse spin fluctuations, respectively. Equation (II2) is rewritten in the standard way as[8]

$$E(T) = -\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \tanh\frac{\omega}{2T} \int \frac{d^3p}{(2\pi)^3} \times \frac{1}{2} \sum_{\alpha=\uparrow,\downarrow} \mathrm{Im}\{[\xi_p + \tfrac{1}{2}\Sigma_\alpha(p,\omega)] G_\alpha(p,\omega)\}, \tag{II4}$$

where $G_\alpha(p,\omega)$ and $\Sigma_\alpha(p,\omega)$ are obtained from $G_\alpha(p,\omega_n)$ and $\Sigma_\alpha(p,\omega_n)$, respectively, by analytical continuation performed by replacing $i\omega_n$ by $\omega + i\delta$. Using

$$2\,\mathrm{Im}G_\alpha(p,\omega) = 2\pi\delta(\omega - \xi_p),$$
$$\mathrm{Re}\Sigma_\alpha(p,\omega) = -\mathrm{Re}\Sigma_\alpha(p,-\omega),$$

and assuming an isotropic Fermi surface and writing

$$\frac{dp^3}{(2\pi)^3} = \frac{N(0)}{2p_F{}^2} \frac{d\varphi}{2\pi} dq q d\xi_p,$$

where $q \equiv |p - p'|$ and $N(0)$ is the electronic density of states at the Fermi-surface, one obtains for $C_v(T)$ at low temperatures the expression[6]

$$C_v(T) = \gamma_0[1 + (4.5\pi^2/\sqrt{2})\,UN(0)\,(T_{\mathrm{Curie}}/\epsilon_F)^2 \times |1 - T/T_{\mathrm{Curie}}|^{-1/2} + \Omega(T)]T, \tag{II5}$$

with $\gamma_0 \equiv \frac{2}{3}\pi^2 N(0)$, and where the enhancement factor $\Omega(T)$ is given by

$$\Omega(T) = \frac{3}{8\pi^2} T^{-1} \int_0^{\infty} dx \frac{x}{\cosh^2 x} \int_{-\infty}^{\infty} \frac{dz}{2\pi} D(z) P \times \int_{-\infty}^{\infty} dy \frac{\tanh[x + (y/2T)]}{y + z}. \tag{II6}$$

[8] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinski, *Quantum Field Theory in Statistical Physics* (Prentice-Hall, Inc., Englewood Cliffs, N.J., 1963).

Here, P means principal value integration and the energy distribution function $D(z)$ of the spin fluctuations is given by

$$D(z)\equiv\frac{N(0)}{2p_F^2}\sum_{\lambda;\nu=l,t}\int_{q_1}^{q_2}dq\,qB_\lambda{}^\nu(q,z),$$

$$q_2\equiv p_{F\uparrow}+p_{F\downarrow},\qquad q_1\equiv p_{F\downarrow}-p_{F\uparrow}. \tag{II7}$$

$p_{F\sigma}$ is the Fermi momentum for electrons with spin σ. The momentum threshold q_1 results from the Zeeman splitting of the Fermi surface occurring in the ferromagnetic metals. Clearly, in the presence of a Zeeman splitting of the Fermi surface the electrons cannot absorb magnons with low energy if $v_Fq<2\eta$, where 2η is the Zeeman energy and where v_F is the Fermi velocity. Since one finds at low temperatures that

$$P\int_{-\infty}^{\infty}dy\,\frac{\tanh[t+(y/2T)]}{y+z}\simeq P\int_{-2Tt}^{2Tt}\frac{dy}{y+z}$$

$$=\ln\left|\frac{t+(z/2T)}{t-(z/2T)}\right|,$$

one obtains

$$\Omega(T)=\frac{3}{4\pi^2}T^{-1}\int_0^\infty dx\,\frac{x}{\cosh^2x}\int_0^\infty\frac{dz}{2\pi}D(z)$$

$$\times\ln\left|\frac{x+(z/2T)}{x+(z/2T)}\right|. \tag{II8}$$

A further explicit evaluation of the temperature dependence of $\Omega(T)$ requires an explicit expression for the spectral function $D(z)$ of the spin excitations. $D(z)$ which specifies the nature of the spin fluctuations is discussed in detail in the next section. Notice, that Eq. (II8) applies to electron spin-flip exchange scattering due to localized spins as well as to electron coupling to spin fluctuations in the d and f bands described by the itinerant-electron model. $D(z)$ describes the characteristic change in the lifetime of the spin fluctuations occurring upon the transition from the ferromagnetic state to the paramagnetic state. It can be easily shown by choosing various approximations for $D(z)$ (the Einstein, Debye, Lorentzian, or Ising spectral function, for example), that the temperature dependence of $\Omega(T)$ depends sensitively on $D(z)$, which in turn is essentially determined by $B_\lambda{}^\nu(q,z)$. Notice, in particular, that the q dependence of the electron-electron interaction potential $V_\lambda(q)$ affects sensitively the temperature-dependence of the specific heat at low temperatures.

Notice, that for very small temperatures such that $z/2T\gg1$, we might further evaluate $\Omega(T)$ by expanding

$$\ln\left|\frac{x+(z/2T)}{x-(z/2T)}\right|$$

in terms of $2Tx/z$. Using then the dispersion relation

$$E_\lambda{}^\nu(q,\omega)=\int_{-\infty}^{\infty}\frac{dz}{2\pi}B_\lambda{}^\nu(q,z)/(\omega-z),$$

one obtains approximately

$$\Omega(T)\simeq\tfrac{1}{4}[D_R(0)]+\cdots, \tag{II9}$$

with

$$D_R(0)\equiv-\sum_{\nu=l,t;m}\frac{N(0)}{2p_F^2}\int_{q_1}^{q_2}dq\,qE_{mm}{}^\nu(q,\omega). \tag{II10}$$

In the case of the itinerant-electron model and transverse spin fluctuations one obtains for $E_{mm}{}^t(q,\omega)$ the expression[6,9–12]

$$E_{mm}{}^t(q,\omega)=\sum_{m'}V_{mm'}(q)P_{m'}{}^t(q,\omega)t_{m'm}(q,\omega),$$

where

$$P_m{}^t(q,\omega)\equiv\int\frac{d^3k}{(2\pi)^3}\frac{f_{km\uparrow}-f_{k+qm\downarrow}}{\epsilon_\downarrow(k+q,m)-\epsilon_\uparrow(k,m)-\omega+i\delta}$$

is the electron polarization function of the band m [10]; $V_{mm'}$ denotes the effective interaction between electrons in the bands m and m', respectively; and the t matrix $t_{mm'}(q,\omega)$ for the multiple electron-hole scattering involving the electron bands m and m' is given within the "ladder" approximation by

$$t_{mm'}(q,\omega)=V_{mm'}(q)+\sum_{m''}V_{mm''}(q)P_{m''}{}^t(q,\omega)t_{m''m'}(q,\omega).$$

$f_{km\sigma}$ gives the Fermi distribution function. k, m, and σ specify the electronic states. In the case of the longitudinal spin fluctuations one finds[6]

$$E_{mm}{}^l(q,\omega)=(\tfrac{1}{2})^2\sum_{\sigma=\uparrow,\downarrow}V_{mm'}(q)P_{m'\sigma}{}^l(q,\omega)\Gamma_{m'm\sigma}{}^l(q,\omega),$$

with

$$P_{m\sigma}{}^l(q,\omega)\equiv\int\frac{d^3k}{(2\pi)^3}\frac{f_{km\sigma}-f_{k+qm\sigma}}{\epsilon(k+q,m)-\epsilon(k,m)-\omega+i\delta},$$

and

$$\Gamma_{mm'\sigma}{}^l(q,\omega)=V_{mm'}(q)+\sum_{m''}V_{mm''}(q)P_{m''-\sigma}{}^l(q,\omega)V_{m''m'}(q)+\sum_{m'',m'''}V_{mm''}(q)P_{m''-\sigma}{}^l(q,\omega)V_{m''m'''}(q)P_{m'''\sigma}{}^l(q,\omega)\Gamma_{m'''m'\sigma}{}^l(q,\omega).$$

[9] N. F. Berk, Ph.D. thesis, University of Pennsylvania, 1966 (unpublished).
[10] H. Ehrenreich and M. H. Cohen, Phys. Rev. **115**, 786 (1959).
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[12] L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin, Inc., New York, 1962).

Notice that $\epsilon(k, m)$ does not include the exchange self energy. It is evident from these expressions that in the paramagnetic state $E_{mm}{}^{l}(q,\omega)=\frac{1}{2}E_{mm}{}^{t}(q,\omega)$. This relationship holds also approximately for ferromagnets close to the ferromagnetic phase transition. Using now for the t matrix the approximate solution[11]

$$t_{mm'}=t_m\delta_{mm'},$$

with

$$t_m(q,\omega)=\frac{U(q)-J(q)}{1-[U(q)-J(q)]P(q,\omega)}+\frac{J(q)}{\{1-[U(q)-2J(q)]P(q,\omega)\}\{1-[U(q)-J(q)]P(q,\omega)\}},$$

one finds

$$D_R(0)=-\frac{3}{2}\frac{N(0)}{2p_F{}^2}\int_{q_1}^{q_2}dq\,qP(q,0)\left\{\frac{1}{3}\frac{[U(q)+2J(q)]^2}{1-[U(q)+2J(q)]P(q,0)}+\frac{2}{3}\frac{[U(q)-J(q)]^2}{1-[U(q)-J(q)]P(q,0)}\right\}. \quad \text{(II11)}$$

U denotes the effective direct Coulomb interaction between the electrons. J denotes the intra-atomic exchange coupling to which Hund's rule refers. Approximating the Lindhard function $P(q,0)$ by $N(0)[1+\frac{1}{3}(q/2p_F)^2]+\cdots$ and performing the integrations one obtains

$$D_R(0)=\frac{9}{2}\left\{\frac{2}{3}\ln\left|\frac{1-[U-J]P(q_2,0)}{1-[U-J]P(q_1,0)}\right|+\frac{1}{3}\ln\left|\frac{1-[U+2J]P(q_2,0)}{1-[U+2J]P(q_1,0)}\right|+U[P(q_2,0)-P(q_1,0)]+\cdots\right\}. \quad \text{(II12)}$$

For simplicity we have here assumed that U and J are q-independent.

Assuming now that $N(0)U$ changes linearly with the concentration c of Ni, and putting for simplicity $J=0$, and also neglecting the Zeeman splitting of the Fermi surface occurring for the ferromagnetic alloys, one finds for the alloys with $N(0)U\simeq 1$ that

$$D_R(0)\propto \ln|c-c_0|^{-1}, \quad \text{(II13)}$$

where c_0 denotes the concentration of Ni at which the alloys become ferromagnetic, e.g., for which $N(0)U=1$. For the ferromagnetic Cu-Ni alloys, Eq. (II12) can also be rewritten approximately as ($J=0$)

$$D_R(0)\simeq\tfrac{3}{2}\ln|\omega(q_2)/\omega(q_1)|\simeq 3\ln(q_2/q_1). \quad \text{(II14)}$$

Notice, that the magnon energy $\omega(q_2)$ is of the order of the Curie temperature and that $\omega(q_1)$ is of the order of MU, where M denotes the magnetization per atom.

It follows from Eq. (II12) that the specific-heat coefficient γ diverges at the ferromagnetic instability. The decrease in γ occurring below the ferromagnetic instability for increasing Ni concentrations results from the increasing Zeeman splitting of the Fermi surface, which tends to suppress the electron spin-flip scattering by the spin fluctuations.

III. SPECTRAL FUNCTION OF THE SPIN FLUCTUATIONS

The spectral weight function $B_\lambda{}^t(q,z)$ for the transverse spin-fluctuation mode λ can in general be written as[12]

$$B_\lambda{}^t(q,z)=\frac{\Gamma_\lambda(q,z)\,\mathrm{sgn}z}{K_\lambda{}^2(q,z)+[\Gamma_\lambda(q,z)/2]^2}, \quad \text{(III1)}$$

where $K_\lambda(q,z)=0$ yields the dispersion relation for the spin-excitation mode λ. The quantity $\Gamma_\lambda{}^{-1}$ measures the lifetime of the spin excitation. In the case of the transverse spin fluctuations described by the itinerant-electron model it is[6] $B_\lambda{}^t(q,z)=2\,\mathrm{Im}t_\lambda(q,z)$, where t_λ denotes the t matrix for the electron-hole scattering which causes the spin fluctuation of kind λ. One obtains then $K_\lambda(q,z)\equiv V_\lambda(q)[1-V_\lambda(q)\mathrm{Re}P_\lambda(q,z)]$ and $\Gamma_\lambda(q,z)\equiv V_\lambda{}^2(q)\,\mathrm{Im}P_\lambda(q,z)$. An expression for the electron-spin polarization function $P_\lambda(q,z)$ has been given in Sec. II. V_λ is an effective potential including screening and residual-electron correlations. Below the Curie temperature $K_\lambda(q,z)$ and $\Gamma_\lambda(q,z)$ can be expanded in terms of z/MV_λ and q/MV_λ, where $M\equiv(N_\uparrow-N_\downarrow)/N_A$ gives the magnetization per atom. Here, N_A denotes the total number of lattice atoms, and N_σ is the number of electrons with spin σ. One finds now for $K_\lambda(q,z)$ the approximate expression

$$K_\lambda(q,z)=M^{-1}[z-z_\lambda(q)], \quad \text{(III2)}$$

with

$$z_\lambda(q)=A_\lambda q^2+\cdots,\qquad A_\lambda\equiv\tfrac{1}{3}[N(0)V_\lambda](MV_\lambda/2p_F{}^2), \quad \text{(III3)}$$

and

$$\Gamma_\lambda(q,z)=C_\lambda(z/q)+\cdots, \quad \text{(III4)}$$

with

$$C_\lambda\equiv(\pi/2)N(0)|V_\lambda|^2(p_F/\epsilon_F),\quad \text{if } |z|<V_Fq,$$
$$C_\lambda\equiv 0,\quad \text{if } |z|>V_Fq. \quad \text{(III5)}$$

In the case where the spin excitations have a long lifetime, one obtains from these expressions

$$B_\lambda{}^t(q,z)=2\pi M|V_\lambda|^2\delta[z-z_\lambda(q)]. \quad \text{(III6)}$$

Notice, that in the case of localized spins M becomes $2\langle S_z\rangle$, where $\langle S_z\rangle$ is the expectation value of the localized spin in the direction of the magnetization. Substituting Eq. (III6) into Eq. (II6), and using $z_\lambda(q)=A_\lambda q^2$, one obtains

$$D(z)=2\pi\sum_\lambda D_{0\lambda}\int_{z_\lambda(q_1)}^{z_\lambda(q_2)} dz_\lambda(q)\,g_\lambda^2(q)\,\delta(z-z_\lambda(q)) +D_l(z),$$

with

$$D_{0\lambda}\equiv\frac{N(0)\langle V_\lambda(q)\rangle_q}{2\omega_\lambda(q_2)}M\langle V_\lambda(q)\rangle_q\left(\frac{q_2}{p_F}\right)^2,$$

and

$$g_\lambda^2(q)\equiv\left|\frac{V_\lambda(q)}{\langle V_\lambda(q)\rangle_q}\right|^2.$$

$D_l(z)$ results from the longitudinal spin fluctuations and is proportional to $\exp[-MV_\lambda/T]$. Hence, for $MV_\lambda\gg T$, $D_l(z)$ is assumed to be negligible.

When one approaches the ferromagnetic instability with increasing Cu concentration, or when one approaches the Curie temperature the magnons hybridize strongly with the collective Stoner spin excitations and therefore cease to be good, long-lived quasiparticles. With increasing Γ_λ the spin waves become more and more Stoner-like excitations.[6] For temperatures close to the Curie temperature and for alloys with $N(0)V_\lambda$ close to 1 the spectral function $B_\lambda^\nu(q,z)$ is still strongly peaked for $q/p_F\ll1$ and $\omega/q\ll1$. One finds then

$$K_\lambda(q,z)\simeq|V_\lambda(q)|^2\left\{F_\lambda+H_\lambda\left[\left(\frac{q}{p_F}\right)^2-3\left(\frac{z/\epsilon_F}{q/p_F}\right)^2\right]\right\},\qquad\text{(III7)}$$

with

$$F_\lambda\equiv[1-N(0)V_\lambda]/V_\lambda N(0)^2\underset{T\to T_{\text{Curie}}}{\propto}(T-T_{\text{Curie}}),\qquad\text{(III8)}$$

and

$$H_\lambda\equiv V_\lambda P(0,0)/12N(0).\qquad\text{(III9)}$$

Here, $P(0,0)$ is the Pauli spin susceptibility. From these expressions one finds approximately[6]

$$B_\lambda^t(q,z)\simeq B_{0\lambda}(z/q),$$
$$B_\lambda^l(q,z)\simeq\tfrac{1}{2}B_\lambda^t(q,z),\qquad\text{(III10)}$$

with

$$B_{0\lambda}\equiv2\pi|V_\lambda|^2\frac{p_F}{\omega_\lambda(p_F)}\frac{N(0)/\pi}{1-N(0)V_\lambda},\quad\text{if}\quad 0\le|z|\le|z_\lambda(q)|,\qquad\text{(III11)}$$

$$B_{0\lambda}\equiv0,\quad\text{if}\quad z_\lambda(q)<z,$$

and

$$z_\lambda(q)\equiv\frac{4}{\pi}\frac{1-N(0)V_\lambda}{N(0)V_\lambda}\left(\frac{\epsilon_F}{p_F}\right)q,\quad\text{if}\quad q_1<q<q_2,$$
$$z_\lambda(q)\equiv0,\quad\text{if}\quad q_2<q.\qquad\text{(III12)}$$

One has for the ferromagnets $q_1\simeq2\eta/v_F$ and $q_2=\min(p_{F\uparrow}+p_{F\downarrow},q_c)$ q_c is a Debye-like cutoff for the magnon momenta. For the paramagnetic metals q_1 is zero and q_2 is kept as a disposable cutoff parameter.

If $B_\lambda^\nu(q,z)$ is not peaked with respect to q and z, but a very smooth function, then it is obvious from Eq. (II6) that $\Omega(T)$ becomes negligible.

In order to obtain the explicit dependence of $D(z)$ on z, the q dependence of $g_\lambda(q)$ has to be known. Taking into account intraband coupling and interband exchange coupling, for example, one obtains[10] for the three resulting transverse modes specified by $\lambda=1$, 2, and 3

$$V_1(q)\equiv U+2J,$$

and

$$V_2(q)=V_3(q)\equiv U-J,$$

where U denotes the effective intraband interaction potential and J the effective interband exchange interaction potential. $J(q)$ will in general be q dependent. Assuming that $J(q)=J_0q$, using for $B_\lambda^t(q,z)$ the expression given by Eq. (III6), and neglecting $B_\lambda^l(q,z)$, then one finds approximately

$$D(z)=\tfrac{2}{3}\pi D_{01}\left\{1+2\frac{D_{02}}{D_{01}}+\mu_1z^{1/2}+\mu_2z+\cdots\right\},\quad\text{if}\quad z(q_1)<z<z(q_2),\qquad\text{(III13)}$$

and

$$D(z)=0,\qquad\text{otherwise.}\qquad\text{(III14)}$$

Here, the constants μ_1, and μ_2 are given by

$$\mu_1\equiv4\frac{J_0}{A_1^{1/2}}\left(\frac{1}{U+2J_0}-\frac{D_{02}}{D_{01}}\left(\frac{A_1}{A_2}\right)^{1/2}\frac{1}{U-J_0}\right)$$

and

$$\mu_2\equiv4\frac{J_0^2}{A_1}\left[\frac{1}{(U+2J_0)^2}+\frac{D_{02}}{2D_{01}}\frac{A_1}{A_2}\frac{1}{(U-J_0)^2}\right].$$

This suffices to demonstrate that the q dependence of $V_\lambda(q)$ is sensitively reflected in $D(z)$, and thus also in the temperature dependence of $\Omega(T)$.

IV. APPROXIMATE EXPRESSIONS FOR THE SPECIFIC HEAT AT LOW TEMPERATURES

Using now the expressions obtained in the previous section for $B_\lambda^\nu(q,z)$ and $D(z)$, respectively, one derives the following approximate expressions for $C_v(T)$.

Substituting Eq. (III13) into Eq. (II8) one finds straightforwardly that

$$\Omega(T)=\frac{D_{01}}{2\pi^2}\int_0^\infty dx\,\frac{x^2}{\cosh^2x}\left\{\left(1+2\frac{D_{02}}{D_{01}}\right)[\Phi(x,\omega_1(q_2))-\Phi(x,\omega_1(q_1))]+\mu_1[R(x,\omega_2(q_2))-R(x,\omega_2(q_1))]+\mu_2[P(x,\omega_2(q_2))-P(x,\omega_2(q_1))]+O\exp(-MV_\lambda/T)\right\},\qquad\text{(IV1)}$$

where

$$\Phi(x,\omega)\equiv\frac{\omega}{2Tx}\ln\left|\frac{x+(\omega/2T)}{x-(\omega/2T)}\right|+\ln\left|x^2-\left(\frac{\omega}{2T}\right)^2\right|, \tag{IV2}$$

$$R(x,\omega)\equiv\tfrac{8}{3}\omega^{1/2}+\tfrac{2}{3}\left(\frac{\omega}{2T}\right)^{3/2}\ln\left|\frac{x+(\omega/2T)}{x-(\omega/2T)}\right|\frac{(2T)^{1/2}}{x}$$
$$-\tfrac{4}{3}(2Tx)^{1/2}\tan^{-1}\left(\frac{\omega}{2Tx}\right)^{1/2}-\tfrac{2}{3}(2Tx)^{1/2}\ln\left|\frac{x^{1/2}+(\omega/2T)^{1/2}}{x^{1/2}-(\omega/2T)^{1/2}}\right|, \tag{IV3}$$

and

$$P(x,\omega)\equiv\omega+T\frac{(\omega/2T)^2-x^2}{x}\ln\left|\frac{x+(\omega/2T)}{x-(\omega/2T)}\right|. \tag{IV4}$$

Using now these expressions one obtains for the temperature range $0<T<\omega_\lambda(q_1)\simeq 2\eta/V_F$ for $\Omega(T)$ the approximate expression

$$\Omega(T)\simeq\tfrac{1}{12}D_{01}\Bigg\{\left(1+2\frac{D_{02}}{D_{01}}\right)\left[\ln\frac{\omega_1(q_2)}{\omega_1(q_1)}-\tfrac{7}{30}\pi^2\left(1-\left(\frac{\omega_1(q_2)}{\omega_1(q_1)}\right)^2\right)\left(\frac{T}{\omega_1(q_2)}\right)^2\right]$$
$$+\mu_1[\omega_2(q_2)]^{1/2}\left[2\left(1-\left(\frac{\omega_2(q_1)}{\omega_2(q_2)}\right)^{1/2}\right)-1.2\frac{18}{\pi^2}\left(1-\left(\frac{\omega_2(q_2)}{\omega_2(q_1)}\right)^{1/2}\right)\frac{T}{\omega_2(q_2)}+\frac{8}{\sqrt{2}\pi^2}(1-2^{-5/2})\Gamma(\tfrac{9}{2})\zeta(\tfrac{7}{2})\right.$$
$$\left.\times\left(1-\frac{\omega_2(q_2)}{\omega_2(q_1)}\right)\left(\frac{T}{\omega_2(q_2)}\right)^{3/2}\right]+\mu_2\omega_2(q_2)\left[1-\frac{\omega_2(q_1)}{\omega_2(q_2)}-\tfrac{7}{15}\pi^2\left(1-\left(\frac{\omega_2(q_2)}{\omega_2(q_1)}\right)\right)\left(\frac{T}{\omega_2(q_2)}\right)^2\right]+\cdots\Bigg\}. \tag{IV5}$$

Here, Γ and ζ denote the Γ and the Riemann ζ function, respectively. For temperatures such as $\omega_\lambda(q_1)<T<\omega_\lambda(q_2)$, one finds that the enhancement factor $\Omega(T)$ is approximately given by

$$\Omega(T)\simeq\tfrac{1}{12}D_{01}\Bigg\{\left(1+2\frac{D_{02}}{D_{01}}\right)\left[1+\frac{12}{\pi^2}\beta+\ln\frac{\omega_1(q_2)}{2T}-\tfrac{7}{30}\pi^2\left(\frac{T}{\omega_1(q_2)}\right)^2-\tfrac{3}{2}\pi^2\left(\frac{\omega_1(q_1)}{T}\right)^2\right]$$
$$+\mu_1[\omega_2(q_2)]^{1/2}\left[2\left(1-\frac{1}{3}\left(\frac{\omega_2(q_1)}{\omega_2(q_2)}\right)^{1/2}\right)-\left(\frac{\omega_2(q_1)}{\omega_2(q_2)}\right)^{1/2}\left(\frac{\omega_2(q_1)}{T}\right)^2+\frac{4\ln 2}{3\pi^2}\left(\frac{\omega_2(q_1)}{\omega_2(q_2)}\right)^{1/2}\frac{\omega_2(q_1)}{T}\right.$$
$$+\frac{4}{\pi^2}(1-2^{-1/2})\Gamma(\tfrac{3}{2})\zeta(\tfrac{1}{2})\left(\frac{\omega_2(q_1)}{\omega_2(q_2)}\right)^{1/2}\left(\frac{\omega_2(q_1)}{T}\right)^{1/2}+\frac{2-2^{-1/2}}{\gamma^2}\Gamma(\tfrac{7}{2})\zeta(\tfrac{5}{2})\left(\frac{T}{\omega_2(q_2)}\right)^{1/2}$$
$$\left.-\frac{3.6}{\pi^2}6\frac{T}{\omega_2(q_2)}+\frac{4}{\pi^2}(1-2^{-5/2})\Gamma(\tfrac{9}{2})\zeta(\tfrac{7}{2})\left(\frac{T}{\omega_2(q_2)}\right)^{3/2}\right]+\mu_2\omega_2(q_2)$$
$$\times\left[1-\tfrac{7}{15}\pi^2\left(\frac{T}{\omega_2(q_2)}\right)^2-\frac{1}{\pi^2}\frac{\omega_2(q_1)}{\omega_2(q_2)}\left(\frac{\omega_2(q_1)}{T}\right)^2+\cdots\right]+\cdots\Bigg\}, \tag{IV6}$$

where

$$\beta\equiv-\int_0^\infty dx\, x\ln x/\cosh^2 x\simeq 0.66.$$

These results demonstrate clearly that the q dependence of $g_\lambda^2(q)$ affects sensitively the temperature dependence of the specific heat.

These approximate expressions cannot be used for the precursor ferromagnetism in the "almost" ferromagnetic metals and also not for the ferromagnets very close to the Curie temperature, since it becomes then very important to take into account the longitudinal spin fluctuations and the damping of the magnons. In order to take approximately into account the damping of the spin excitations around the ferromagnetic phase transition, we use for $B_\lambda''(q,z)$ the approximate expressions given by Eq. (III10) and also assume for simplicity that V_λ is q independent.

Substituting then Eq. (III10) into Eq. (II6) one obtains

$$\Omega(T)\simeq\frac{2}{\pi^4}\sum_{\lambda=1}^{3}\frac{N(0)V_\lambda}{\omega_\lambda^2(p_F)}\frac{\epsilon_F}{\omega_\lambda(p_F)}T\int_0^\infty dx\,\frac{x}{\cosh^2 x}\{F[x,\omega_\lambda(q_2)]-F[x,\omega_\lambda(q_1)]\},\tag{IV7}$$

with

$$F(x,\omega)\equiv\frac{3}{2}\int_0^\omega dz\int_0^{z/2T}dy\,y\ln\left|\frac{x+y}{x-y}\right|.\tag{IV8}$$

The integrations can be performed straightforwardly and one obtains

$$F(x,\omega)=\frac{3}{2}\left\{\tfrac{2}{3}x\left(\frac{\omega}{2T}\right)^2+\left[\frac{1}{6}\left(\frac{\omega}{2T}\right)^2-\frac{x^2}{2}\right]\ln\left|\frac{x+(\omega/2T)}{x-(\omega/2T)}\right|\frac{\omega}{2T}-\tfrac{1}{3}x^3\left[\ln\left|x^2-\left(\frac{\omega}{2T}\right)^2\right|\right]2T\right\}.\tag{IV9}$$

Further evaluation gives for the temperature range $\omega_\lambda(q_1)<T<\omega_\lambda(q_2)$ for $\Omega(T)$ the approximate expression

$$\Omega(T)\simeq\tfrac{1}{8}\pi\sum_{\lambda=1}^{3}\frac{[\mathrm{N}(0)V_\lambda]^2}{1-\mathrm{N}(0)V_\lambda}\left\{\frac{1}{6}\left[\frac{\omega_\lambda(q_2)}{\omega_\lambda(q_1)}\right]^2+\left[\frac{8S}{\pi^2}-\tfrac{14}{45}\pi^2-\tfrac{7}{30}\pi^2\ln\frac{\omega_\lambda(q_2)}{2T}\right]\left[\frac{T}{\omega_\lambda(p_F)}\right]^2-\tfrac{1}{8}\pi^2\left[\frac{\omega_2(q_1)}{\omega_\lambda(p_F)}\right]^2\left[\frac{\omega_\lambda(q_1)}{T}\right]^2+\cdots\right\},\tag{IV10}$$

where

$$S\equiv\int_0^\infty dx\,x^4\ln x/\cosh^2 x.$$

One finds for ferromagnets at temperatures close to the Curie temperature T_c the expression

$$\Omega(T)\simeq\tfrac{1}{8}\pi\sum_{\lambda=1}^{3}\frac{[N(0)V_\lambda]^2}{1-N(0)V_\lambda}\left\{\frac{1}{6}\frac{\omega_\lambda^2(q_2)-\omega_\lambda^2(q_1)}{\omega_\lambda^2(p_F)}-\tfrac{7}{30}\pi^2\left[\ln\frac{\omega_\lambda(q_2)}{2T}\right]\left[\frac{T}{\omega_\lambda(p_F)}\right]^2+\cdots\right\},\tag{IV11}$$

if $0<T<\omega_\lambda(q_1)$.

Notice that for the ferromagnets a better approximation for $B_\lambda{}^l(q,z)$ than given by Eq. (III10) is given by[6]

$$B_\lambda{}^l(q,z)=\frac{1}{2}\frac{\Gamma_\lambda(q,z)}{[K_\lambda(q,z)+\Lambda_\lambda]^2+(\frac{1}{2}[\Gamma_\lambda(q,z)])^2},$$

with

$$\Lambda_\lambda\equiv-V_\lambda\left(\frac{\partial}{\partial T}P_\lambda(0,0)\bigg|_{T=T_c}\right)(T-T_c)\left\{1-\frac{F(1)F(3)}{F^2(2)}\right\},$$

and

$$F(\nu)\equiv\int_0^\infty dx\,x^{1/2}\frac{d^\nu f(x)}{dx^\nu}.$$

V. NUMERICAL RESULTS FOR Ni AND Cu-Ni ALLOYS

The theory for the interaction between electrons and spin excitations is now used to calculate γ for Ni and all Cu–Ni alloys and C_v/T for some Cu–Ni alloys with about 57 at.% Cu. Since the specific heat has not been measured at sufficiently small temperatures γ cannot be determined directly from the experimental results.[2,4] According to the theory outlined in the previous sections $\gamma_{\exp}$ is determined by fitting the experimental results to $C_v/T=\gamma+pT^2+sT^n\ln T+lT\,|\,1-T/T_{\mathrm{Curie}}\,|^{-1/2}$, where

$$p\simeq(464.3/T_{\mathrm{Debye}}{}^3)[\mathrm{cal/mole}(^\circ\mathrm{K})^4].$$

It is $n=0$ for $T\ll T_{\mathrm{Curie}}$, and $n=2$ if $T\sim T_{\mathrm{Curie}}$. For the paramagnetic alloys γ is theoretically determined by using for γ the expression given by Eq. (II9) and assuming that $N(0)V_\lambda$ changes linearly with the Ni concentration like $N(0)V_\lambda=[N(0)V_\lambda]_{\mathrm{Cu}}(1+\Lambda C_{\mathrm{Ni}})$. The parameter Λ is determined by fitting the theory to $\gamma_{\exp}$ for Ni concentrations smaller than 10 at.% Ni. For the ferromagnetic alloys γ is calculated by taking into account the cutoff $q_1\simeq2\eta/V_F=(\eta p_F/\epsilon_F)\propto(C_{\mathrm{Ni}}-C_0)$, where $C_0=43$ at.% Ni denotes the (critical) smallest Ni-concentration for which the CuNi alloys are ferromagnetic. Putting $J=0$, one has approximately for the ferromagnetic alloys

$$\gamma\cong\gamma_0\{1+0.75\ln[\omega(q_2)/\omega(q_1)]\}.$$

Here,

$$\omega(q_1)\propto(C_{Ni}-C_0)$$

and

$$\omega(q_2)/\omega(q_1)=(q_2/q_1)^2\simeq(R\epsilon_F/\eta)^2,$$

where

$$R\equiv q_2/p_F.$$

The reduction factor R can be estimated by noticing that $\omega(q_2)\sim T_{\text{Curie}}$. Also, notice that $\omega(q_1)\propto\eta\propto M$.

In the absence of any band calculations for the Cu-Ni alloys, $\gamma_0\equiv\frac{2}{3}\pi^2N(0)$ has been obtained by interpolation from band theory results for Ni [13] and Cu.[14] Due to the Zeeman splitting of the Fermi surface, γ_0 decreases initially above C_0 with increasing Ni concentrations. The obtained results for the theoretically determined γ and for γ_{exp} are shown in Fig. 1.

Using $C_v/T=\gamma+pT^2+A(T/T_c)\ln(T/T_c)$, with $A\simeq(1/4\pi)(7\pi^2/30)[N(0)V_\lambda]^2/[1-N(0)V_\lambda]$, the specific heat of several alloys with about 55 at.% Cu is calculated. The characteristic temperature T_c is the Curie temperature T_c for the ferromagnets and the "reduced" Fermi temperature T_F for the "almost" ferromagnetic metals. For T_{Curie} the observed values are taken and T_F is calculated from $N(0)V_\lambda$. The results obtained are shown in Fig. 2.

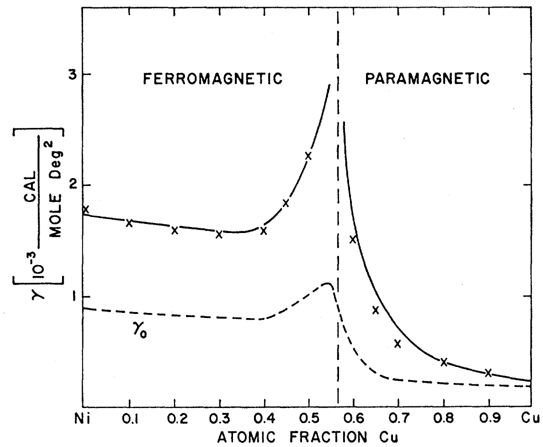

FIG. 1. The coefficient γ of the low-temperature specific-heat contribution, which is linear in temperature, is shown. The continuous curve is obtained by fitting $C_v/T=\gamma+pT^2+AT^n\ln T$ to the experimental specific-heat results. pT^2 is the lattice contribution. It is $p=464.3T_{\text{Debye}}^{-3}[\text{cal/mole}(°\text{K})^4]$. n is taken to be zero for $T\ll T_{\text{Curie}}$, and to be two for $T\sim T_{\text{Curie}}$ and for all paramagnetic alloys. The crosses are the theoretical results calculated by $\gamma=\gamma_0\{1+1.12\ln|1/[1-N(0)U]|\}$, assuming $N(0)U=(1+\Lambda C_{\text{Ni}})[N(0)U]_{\text{Cu}}$, and using $[N(0)U]_{\text{Cu}}=0.30$, $C_0=43$ at.% Ni, and $\Lambda=\{1-[N(0)U]_{\text{Cu}}\}/[N(0)U]_{\text{Cu}}C_0$. In the case of the ferromagnetic alloys γ is calculated by $\gamma=\gamma_0[1+1.5\ln(q_2/q_1)]$. Here, we take $q_2/q_1\equiv F(C_{\text{Ni}}-C_0)$ and determine the function $F(C_{\text{Ni}})$ by fitting to the experimental results for the ferromagnetic alloys with less than 60 at.% Ni. γ_0 is estimated using the rigid-band model, and using for the ferromagnetic Ni, $N_{\text{Ni}}(0)=0.80$ states/(eV) atom and $N_{\text{Cu}}(0)=0.16$ states/(eV) atom for copper.

VI. CONCLUSION

It has been shown that the coupling between the electrons and spin fluctuations (magnons) has a strong effect on the electronic specific heat. In view of the fact that an isotropic Fermi surface has been assumed, a q-independent interaction V_λ, and that any detailed band-structure effects have been neglected, the agreement between theory and experiment is surprisingly good. It is very likely, that the anomalous behavior of the specific heat observed for Ti–Fe and Fe–V alloys,[15] and other alloys, Fe, etc., which is similar to the one observed for the Cu-Ni alloys, can also be explained as resulting from the interaction between the electrons and magnons (spin fluctuations).

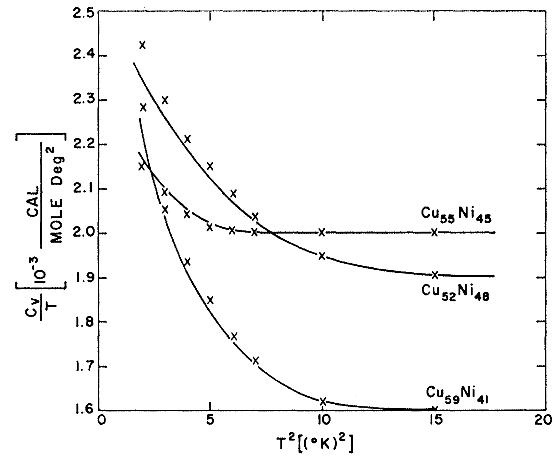

FIG. 2. Specific heat of several Cu-Ni alloys with small Curie temperature or small "reduced" Fermi temperature, respectively. The continuous curve gives the experimental results. The crosses are the theoretical results, obtained by fitting to the experimental results at temperatures above 3°K and using $T_{\text{Curie}}=18°$K for $Cu_{55}Ni_{45}$, $T_{\text{Curie}}=50°$K for $Cu_{52}Ni_{48}$, and T_c(=reduced Fermi temperature) =26°K for $Cu_{59}Ni_{41}$.

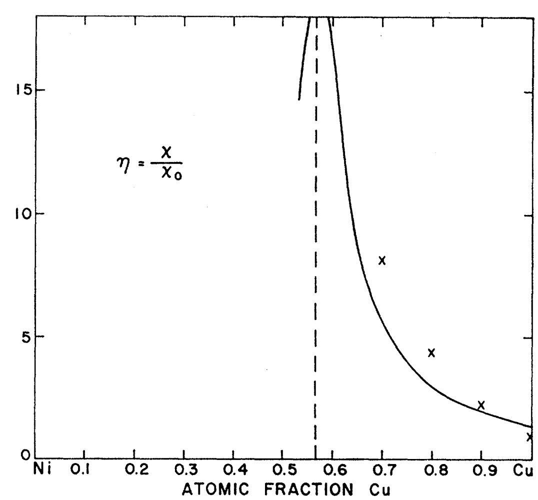

FIG. 3. The enhancement factor $g=1/[1-N(0)U]$ of the Pauli spin susceptibility is calculated for the Cu-Ni alloys taking for $N(0)U$ the values used in determining γ. The crosses represent the experimental results.

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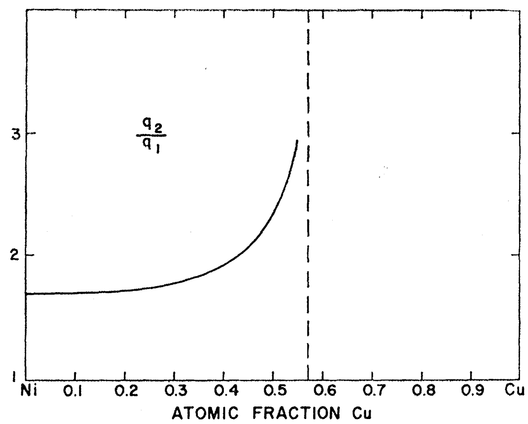

FIG. 4. The function $q_2/q_1 \equiv F(C_{\rm Ni})$ is determined by fitting $\gamma_{\rm exp}$ for the ferromagnetic alloys with less than 60 at.% Ni. Using $\epsilon_F^{\rm Ni}=8.0$ eV and $\eta=0.3$ eV one finds $q_1^{\rm Ni}\simeq 0.06p_F$.

For a further improvement of the analysis presented here, it would be very useful to measure the specific heat at temperatures well below 1°K. Thus it is possible to determine γ directly from experiment and to find out experimentally, whether the coupling of the electrons to the magnons (spin fluctuations) gives rise to a contribution $\ln T$, T^{-1}, or $T^2 \ln T$ to C_v/T. Notice, that the first two contributions to C_v/T diverge at $T\rightarrow 0$ while the third contribution goes to zero at $T=0$. Also, a band calculation should be performed determining $N(0)$ for all Cu–Ni alloys.

The contribution to the effective electron mass resulting from the electron-phonon coupling is likely to be much smaller than the contribution resulting from the electron coupling to the magnons (spin fluctuations). Notice, however, that the contribution to the effective electron mass due to the electron-phonon coupling should also reflect the possible peak in $N(0)$ or γ_0, respectively, occurring in the ferromagnets which is due to the Zeeman splitting of the Fermi surface.

A more accurate determination of q_2/q_1, is desirable, since γ seems to depend sensitively on q_2/q_1.

It is very likely that electron spin-orbit coupling has a strong effect on the coupling between electrons and spin excitations. It would be very interesting to study this effect, for example, for the Cu–Ni alloys using impurities, which give rise to strong spin-orbit scattering, for Pd, Pt, etc., or by a comparative study of the electron-spin fluctuation coupling in alloys differing mainly with respect spin-orbit coupling.

According to the theory of the coupling between the electrons and the spin fluctuations the transverse static electron spin susceptibility should approximately be given by $\chi_{\rm el}{}^t = P(0,\ 0)/[1-N(0)V_\lambda]$, as has indeed been observed.[16] Putting $V_\lambda = U$, and taking for $N(0)U$ the values used in determining γ, one finds reasonable agreement with the existing experimental results. This might indicate that J/U is small and that the s-d hybridization is not changing much with alloy composition. However, a small ratio J/U might be in conflict with other experimental results. See Fig. 3.

The electrical resistivity and electronic thermal conductivity, for example, of the Cu–Ni alloys should also exhibit an anomalous temperature dependence at low temperatures due to the interaction between the electrons and the (spin fluctuations) magnons. See Fig. 4.

Finally, it is interesting to point out that Beck and co-workers observed[17] in the ternary alloys Cu–Ni–Al with about 10 at.% Al that γ increases immediately upon adding Cu to Ni. This behavior, which is puzzling in the light of band theory, can be explained as follows. Since Al tends to decrease the magnetic saturation moment, the peak in γ observed for the Cu–Ni alloys should shift to smaller Cu concentrations in the ternary alloys and occurs for 10 at.% Al already at very small Cu concentrations.

ACKNOWLEDGMENTS

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