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LOW-TEMPERATURE PROPERTIES OF NEARLY FERROMAGNETIC FERMI LIQUIDS

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Effective-mass corrections due to emission and reabsorption of persistent spin fluctuations are shown to be large in Pd and to have a temperature dependence which fits qualitatively that of the low-temperature specific heat of He^3.

A characteristic feature of two strongly interacting Fermi liquids, namely the d-band holes in metallic palladium, and liquid He^3, is the considerable enhancement of the observed static paramagnetic susceptibility at low temperatures over the Pauli susceptibility deduced from the observed value of the low-temperature specific heat using noninteracting Fermi-gas theory. The work of Slater and of Stoner[1] on the magnetic properties of fermion systems shows that the physical origin of this enhancement can be represented, within the framework of a molecular field or random phase approximation (RPA), as resulting directly from a semiphenomenological short-range repulsion between fermions. We take this to be

$$H_{\text{int}} = I\int d^3x\, n_\uparrow(\vec{x})n_\downarrow(\vec{x}')\delta(\vec{x}-\vec{x}'), \tag{1}$$

where $n_\uparrow(\vec{x}) = \psi_\uparrow^\dagger(\vec{x})\psi_\uparrow(\vec{x})$ is the spin-up number density.

In this Letter we show that there is a class of corrections to RPA which become increasingly important as the exchange enhancement is increased, and we discuss some of their consequences. These corrections are the contributions to the one-particle self-energy, and hence effective mass, arising from the emission and reabsorption of persistent spin fluctuations (or critically damped spin waves) analagous to the phonon corrections to the effective mass resulting from electron-phonon interaction.[2] These are taken into account by summation of the particle-hole t-matrix contribution to the self-energy. The propagator for the spin fluctuations, which results from this summation, is defined by

$$\chi^{-+}(\vec{x}-\vec{x}', t-t') = +i\langle T\{\sigma^-(\vec{x},t)\sigma^+(\vec{x}',t')\}\rangle, \tag{2}$$

where $\sigma^+(\vec{x}) = \psi_\uparrow^\dagger(\vec{x})\psi_\downarrow(\vec{x})$. This has been discussed by Izuyama, Kim, and Kubo[3] who show that the Fourier transform of (2), $\chi(\vec{k}, k_0)$, is given in RPA in terms of $\chi^0(\vec{k}, k_0)$ for the noninteracting Fermi gas by

$$\chi^{-+}(\vec{k}, k_0) = \frac{\chi^0(\vec{k}, k_0)}{1 - I\chi^0(\vec{k}, k_0)}. \tag{3}$$

The persistence of the spin fluctuations represented by χ is now seen by looking at χ in the region of small $|\vec{k}|/p_F$ when χ^0 may be expanded in powers of k/p_F to give

$$\chi^{-+}(\vec{k}, k_0) = \frac{N(0)}{K_0^2 + (\bar{I}/12)\bar{k}^2 - i(\pi/4)\bar{I}|k_0|/\bar{k}} \tag{4}$$

for $\bar{k}_0 = k_0/\epsilon_F < 2\bar{k} = 2|\vec{k}|/p_F \ll 1$. For $\bar{k}_0 \gtrsim 2\bar{k}$ the imaginary term in the denominator goes to zero. $N(0)$ is the density of states, $mp_F/2\pi^2$, at the Fermi level; $\bar{I} = IN(0)$; and $K_0^2 = (1-\bar{I})$ is the inverse of the Stoner enhancement factor. For paramagnets for which $K_0^2 \ll 1$, this function is strongly peaked at $k_0 \cong \epsilon_F K_0^2 \bar{k}$ so that although the spin fluctuations are strongly damped, their characteristic excitation energy is reduced by the factor K_0^2 by the effect of the interactions, i.e., they tend to persist for very long times as $K_0^2 \to 0$ at which point the Stoner criterion for transition to the ferromagnetic state is satisfied.

We now consider the following correction to the ground-state energy based on the use of the RPA formula (3) for the particle-hole correlation to evaluate $\langle H_{\text{int}}\rangle$, which may then be integrated with respect to the coupling constant to give

$$\Delta E = I\langle n_\uparrow\rangle\langle n_\downarrow\rangle - i\int_C \frac{dk_0}{2\pi}\sum_{\vec{k}}\{\ln[1-I\chi^0(\vec{k},k_0)] + I\chi^0(\vec{k},k_0)\}, \qquad (5)$$

where the contour is closed in the upper $\frac{1}{2}$ plane. Here

$$\chi^0(\vec{k},k_0) = \sum_{\vec{p}}\left\{\frac{f_{\vec{p}\uparrow}(1-f_{\vec{p}+\vec{k}\downarrow})}{\epsilon_{\vec{p}+\vec{k}\downarrow}-\epsilon_{\vec{p}\uparrow}-k_0-i\delta} - \frac{f_{\vec{p}+\vec{k}\downarrow}(1-f_{\vec{p}\uparrow})}{\epsilon_{\vec{p}+\vec{k}\downarrow}-\epsilon_{\vec{p}\uparrow}-k_0+i\delta}\right\}, \qquad (6)$$

where $f_{\vec{p}}$ are the Fermi functions and $\epsilon_{\vec{p}} = p^2/2m - \epsilon_F$. At finite temperatures (5) provides an approximation to the free energy, on using the temperature-dependent Fermi functions. The one-particle self-energy, $\Sigma(\vec{p}, p_0)$, resulting from the spin fluctuations may be derived from the diagram in Fig. 1 to be

$$\Sigma_\uparrow(\vec{p},p_0) = -iI^2\int[d^4k/(2\pi)^4] \times G_\downarrow{}^0(\vec{p}+\vec{k},p_0+k_0)\chi^{-+}(\vec{k},k_0). \qquad (7)$$

The same expression results, on the energy shell $\epsilon_{\vec{p}} = p_0$, by functional differentiation of ΔE with respect to $f_{\vec{p}\uparrow}$. The integral (7) may be evaluated by using the spectral representation

$$\chi^{-+}(\vec{k},k_0) = -\int_0^\infty \frac{d\Omega}{\pi}\operatorname{Im}\chi^{-+}(\vec{k},\Omega)D^0(\Omega,k_0), \qquad (8)$$

where $D^0(\Omega,k_0) = 2\Omega/(k_0^2-\Omega^2+i\delta)$. Introducing a cutoff momentum $\bar{p}_1$, with $\bar{p}_1 = p_1/p_F$, as an arbitrary parameter which gives some measure of the inadequacy of (4) at large k and also of the range of the forces in (1), (7) may be evaluated at the Fermi level, leading to

$$\operatorname{Re}\Sigma(p_F,p_0) = \frac{I^2m}{p_F}\int_0^{p_1}\frac{kdk}{(2\pi)^2}\int_0^\infty\frac{d\Omega}{\pi}\operatorname{Im}\chi^{-+}(\vec{k},\Omega) \times\int_{-\infty}^{\infty}d\epsilon'\frac{\partial f'}{\partial\epsilon'}\ln\left|\frac{p_0-\epsilon'+\Omega}{p_0-\epsilon'-\Omega}\right|. \qquad (9)$$

Using the approximation (4) this leads to an effective mass correction at $T=0$ of

$$\frac{m^*}{m} - 1 = -\lim_{p_0\to 0}\frac{\partial}{\partial p_0}\Sigma(p_F,p_0) = 3\bar{I}\ln\left(1+\frac{\bar{p}_1{}^2\bar{I}}{12K_0{}^2}\right) \qquad (10)$$

which diverges as $K_0{}^2 \to 0$. In fact (10) may be seen to be very closely related to the critical scattering discussed by Izuyama, Kim, and Kubo[3] and this divergence represents the physical effect of the long range of the spin fluctuations on the one-particle spectrum as the ferromagnetic instability is approached. In order to apply these results to experiment, we will determine $K_0{}^2$ in terms of the measured susceptibility. For this purpose we need to calculate both the susceptibility and the low-temperature specific heat consistently within the proposed approximation scheme. To consider the effects of the effective mass corrections (10) on the measured susceptibility, we consider the dependence of ΔE, Eq. (5), on a local field h expressed in units such that $\epsilon_{p\uparrow} = \epsilon_p - h$, $\epsilon_{p\downarrow} = \epsilon_p + h$. The magnetization $M = -\partial E/\partial h$ at finite h is calculated by differentiating (5) as

$$(1-\bar{I})M = 2N(0)h - iI^2\int\frac{d^4k}{(2\pi)^4}\frac{\partial x^0}{\partial h}(k)\chi^{-+}(k). \qquad (11)$$

The first term on the right-hand side gives the unperturbed static susceptibility in the present units. We now show the second term to be identically zero to the order in which the effective mass (10) was calculated.[4] This results from a cancellation in $\partial x^0/\partial h$: The derivative of the first term in Eq. (6) is

$$\sum(\epsilon_\downarrow{}'-\epsilon_\uparrow-i\delta)^{-1}\frac{\partial}{\partial h}(f_\uparrow(1-f_\downarrow{}')) + \sum f_\uparrow(1-f_\downarrow{}')\frac{\partial}{\partial h} \times[(\epsilon_\downarrow{}'-\epsilon_\uparrow-k_0+i\delta)^{-1}].$$

On replacing $\partial/\partial h$ by $[\partial/\partial\epsilon_\downarrow{}' - \partial/\partial\epsilon_\uparrow]$ in the second term, performing a partial integration on ϵ and ϵ', and neglecting end-point contributions, these two terms cancel identically. The same cancellation also occurs in the second term of (6). This result indicates that the Stoner enhancement factor usually calculated for Pd from the observed specific heat should really be calculated using the bare band mass of the d holes for the unenhanced Pauli susceptibility. This is rather difficult to calculate with

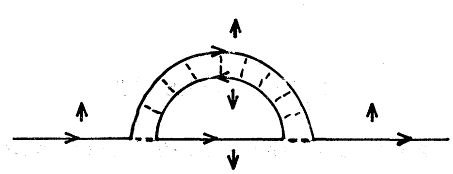

FIG. 1. Spin-fluctuation contribution to fermion self-energy.

any precision owing to uncertainty in the cutoff momentum $\bar{p}_1$ in Eq. (10). However, the recent estimate of Freeman, Furdyna, and Dimmock,[5] based on an augmented-plane-wave calculation of $m^*/m \cong 2$, enables us to apply formula (10) to estimate this cutoff. The exchange enhancement in Pd, calculated on the basis of the observed specific heat, is[6] of order 6.7; using $m^*/m = 2$ leads to an estimate of $1/K_0^2 \cong 13$ leading to a cutoff of order $\bar{p}_1 = 0.6$.[7] The existence of a large mass enhancement has also been proposed in nickel.[8] It seems likely that corrections similar to those proposed here might also apply in that case if suitably modified for the ferromagnetic state.

We now consider He^3. Data on the low-temperature susceptibility of He^3 given by Wheatley[9] lead to a value of $K_0^2 = \frac{1}{9}$ at 0.28 atm and $K_0^2 = 1/21$ near the melting pressure (27 atm). If we take an extreme point of view, and assume that all the observed specific-heat mass[10] arises due to the mechanism proposed here, these can be fitted to formula (10) using cutoff values $\bar{p}_1 = 1.3$ at 0.28 atm and $\bar{p}_1 = 1.6$ at 27 atm. These estimates may now be used to discuss the temperature dependence of the low-temperature specific heat in He^3. From (5) we have the shift in entropy given as

$$\Delta S = -\frac{\partial}{\partial T}\Delta E = -2\sum_{\vec{p}} \frac{\partial f_{\vec{p}}}{\partial T}\Sigma(\vec{p}, \epsilon_{\vec{p}}). \qquad (12)$$

This is evaluated using the approximation (4) and constant density of states via Eq. (9) to give a leading contribution at low temperatures of

$$\frac{C_v}{C_v^{\,0}} = \frac{m^*}{m} + \frac{6}{5}\pi^2 \frac{\bar{I}^2}{K_0^{\,2}}\left(\frac{T}{\bar{T}_F}\right)^2 \left\{\ln\left(\frac{T}{\bar{T}_F}\right) - \left(\tfrac{1}{3} + \ln\bar{p}_1\right)\right\} + O\left(\left[\frac{T}{\bar{T}_F}\right]^4\right), \qquad (13)$$

where

$$K_B \bar{T}_F = 4\epsilon_F K_0^{\,2}/\pi\bar{I}$$

and

$$C_v^{\,0} = \tfrac{2}{3}\pi^2 N(0) K_B^{\,2} T.$$

In the calculation, the $\bar{k}^2$ in the denominator of (4) has been neglected in the second term as the contributions come mainly from the low-k region of the integral; we also neglect the temperature dependence of χ. Use of the estimates of K_0^2 given above, in formula (13), leads to a variation in the low-temperature specific heat which is much faster than that observed.[10] However, a variation of order of that observed may be obtained by choice of somewhat lower values, $1/K_0^2 = 7$ at 0.28 atm and $1/K_0^2 = 11$ at 27 atm. This renormalization of K_0^2 may indicate breakdown of the approximation used above to show that the susceptibility is not renormalized. Using $T_F = 5.0$ and 6.2°K at 0.28 and 27 atm, we then find $\bar{T}_F = 1.3$ and 0.78°K, respectively. The resulting variation of the specific heat is plotted in Fig. 2. The qualitative fit rapidly becomes bad above 40 mdeg. There may be a number of contributions to this: use of constant density of states, neglect of the temperature dependence of the spin propagator, and use of the small-k approximation of Eq. (4). The temperature dependence of (13) is to be contrasted with that from acoustic phonons which will also contribute to mass enhancement and will lead to a similar $T^2 \ln T$ variation in C_v/T. However, the scale on which this variation occurs will be determined by some effective Debye temperature which may be expected to be of order or larger than T_F in contrast to the characteristic temperature $\bar{T}_F$ for the spin fluctuations which becomes considerably reduced as $K_0^2 \to 0$.[11] Our main conclusion is, therefore, that it is the magnetic fluctuations which have sufficiently low ex-

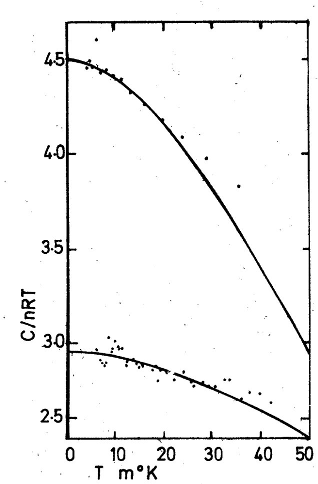

FIG. 2. (C_v/NRT) for He^3. Full curves are given by Eq. (13). Use of the cutoff parameter fits to observed value at $T = 0$, but does not affect significantly the predicted temperature dependence. Points are data of the Illinois group.[10]

citation energies to account qualitatively for the low-temperature variation of the specific heat in liquid He^3.

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[11]A similar temperature dependence may be expected for the specific heat of Pd for which $\overline{T}_F$ would be of order 200°K. This variation combines with a similar variation due to phonons.

DIATOMIC FERROELECTRICS

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Experimental measurements of the phonon dispersion relation for SnTe suggest that the phase transition of GeTe and SnTe/GeTe alloys is of a displacive character, similar to that of the perovskite ferroelectrics. They therefore provide the first examples both of diatomic ferroelectrics and of "ferroelectric semiconductors," with a narrow band gap.

These measurements are part of a program to study the phonon dispersion relations of diatomic crystals having high dielectric constants by means of the method of inelastic neutron scattering.[1] SnTe has the sodium chloride structure, and as-grown crystals are slightly nonstoichiometric with a relatively high-carrier concentration[2] which prevents any direct measurement of the static dielectric constant, $\epsilon(0)$. The value of the high-frequency dielectric constant is $\epsilon(\infty) = 42 \pm 4$.[3]

Figure 1 shows the phonon frequencies $\nu_j(\vec{q})$ for the optic branches, for wave vectors in the [001] direction, at a number of temperatures. The transverse optic (TO) branch is very temperature dependent, in contrast to the weak temperature dependence of the other [001] branches of the dispersion relation. Figure 2 shows the square of the frequency of the TO mode at small wave vector $\vec{q}$, as a function of temperature. The bars indicate uncertainty mainly due to the difficulty of correcting these particular measurements for the effect of the finite resolution of the triple-axis spectrometer.

It is evident from Fig. 1 that as $\vec{q}$ approaches zero the frequency of the longitudinal optic (LO) mode falls sharply. A similar effect has been found in PbTe[4] and in PbS,[5] although it is much less marked in these materials. This effect can be understood in terms of the screening of the LO mode by carriers in the conduc-