Thermodynamic approach to obtaining a highly spin-polarized strongly correlated Fermi liquid in solid-state systems

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We present a thermodynamic approach to obtain a highly polarized heavy Fermi liquid in solid-state systems. It is similar to that proposed by Castaing and Nozières [J. Phys. (Paris) **40**, 257 (1979)] for polarizing liquid ^{3}He. The mechanism applies to a class of materials where local moments coexist with a Fermi liquid at a first-order transition. *Fast melting* of the magnetic field aligned local moment phase, due to a change of pressure or magnetic field, yields a highly polarized Fermi liquid state. This fast melting implies that the relaxation time of the spin polarization is long compared to the melting time of the local moment phase. We discuss the conditions for attaining it in the materials considered.

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Systems of spin polarized electrons have been extensively studied due to their scientific interest and many possible applications. An area of spintronics has emerged associated with these fluids and their use in devices.[1] Although huge magnetic fields are required to polarize the conduction electron in metals, these fields naturally arise in ferromagnetic metals. In particular, in *strong* ferromagnets the Fermi level may lie entirely on the minority spin band, the majority band being completely full. These systems are in their totality based on transition metals and the polarized spins occupy electronic *d* states. From the scientific point of view spin polarized Fermi liquids are extremely interesting as they present unusual properties.[2,3] The study of polarized liquid ^{3}He is an important area of research and many techniques have been used to produce these systems. In particular, a thermodynamic approach[2] turned out to be quite successful. In this method the local moments of solid ^{3}He are polarized by an external magnetic field and subsequently the solid is melted giving rise to a highly polarized Fermi liquid. The lifetime of this state is sufficient to allow for the study of its properties.

In a series of recent of papers[4,5] we pointed out a similarity between the solid state compound $YbInCu_4$ and its alloys with the extreme quantum system ^{3}He. We have shown the existence of a Pomeranchuk effect in the former materials that can be used to produce cooling.[5] The basic feature that these systems share with ^{3}He is a first order transition where a local moment phase coexists with a strongly correlated Fermi liquid.[6] In the solid-state materials this is a valence, or volume transition and in ^{3}He the melting transition.

If we neglect the sign of the relative volume change at the valence transition T_V, this list of materials can be enlarged to contain, metallic Ce (Refs. 5 and 7) and possibly $Eu(Pd_{1-x}Au_x)_2Si_2$ (Refs. 8 and 9) and $EuNi_2(Si_{1-x}Ge_x)_2$ (Ref. 10) for selected values of the concentration x. These systems have at high temperatures, above T_V, a phase with well-defined local moments as indicated by the Curie-Weiss behavior of their magnetic susceptibility. On the low temperature side, for $T \ll T_V$, they can be described as moderatly heavy fermions with enhanced Pauli susceptibility, power law behavior of the resistivity (with a T^2 regime in some range), and linear temperature-dependent specific heat. At the first order valence transition T_V, the two phases local moment and Fermi liquid coexist.

Let us concentrate on $YbInCu_4$ as a paradigm. Above $T_V \approx 42$ K the effective magnetic moments of $YbInCu_4$ obtained from the Curie-Weiss susceptibility present the full value $\mu_{\text{eff}}=4.54\ \mu_B$ of an $Yb^{3+}(4f^{13}, J=7/2, g_J=8/7)$ ion.[11] At T_V the valence change of Yb is only 0.1 as derived from thermal expansion and x-ray absorption.[12] The relative volume change at the transition is $\Delta V/V=0.5\%$. The low temperature phase is a non-magnetic, renormalized Fermi liquid with an enhanced Pauli-like susceptibility χ_P and an equally enhanced coefficient of the linear term of the specific heat $\gamma=50$ mJ/Mol K. Again, in common with ^{3}He, this material has a first order transition where a renormalized Fermi liquid phase coexists with a phase of nearly independent local moments (the solid phase in ^{3}He).

In this paper, we extend further the analogy between these solid-state Pomeranchuk materials and ^{3}He. We consider using the thermodynamic approach of Castaing and Nozières[2] for polarizing ^{3}He, to produce a highly polarized Fermi liquid state in the former systems. This would allow us to obtain a class of polarized electrons, with heavy masses, essentially *f* character and still unexplored.

Returning to the system $YbInCu_4$, the valence or volume transition at T_V is highly sensitive to pressure and magnetic field.[11] A magnetic field applied on the local moment phase above T_V has two main effects. It polarizes the local moments and shifts the volume, or valence transition to lower temperatures. That this shift is always to lower temperatures, independently of the sign of the relative volume change, can be seen from the magnetic Clausius-Clapeyron equation[5]

$$\left(\frac{dT_V}{dH}\right)_{H_V} = \frac{-(M_{\text{LM}} - M_{\text{FL}})_{H_V}}{(S_{\text{LM}} - S_{\text{FL}})_{H_V}}. \tag{1}$$

This is always negative, since the entropy and magnetization of the local moment high temperature phase are larger than those of the Fermi liquid.

We can now envisage the mechanism for polarizing the correlated Fermi liquid. Imagine, for example, a

sample of $YbInCu_4$ in thermodynamic equilibrium with a heat reservoir at a temperature T_0 which is lower than $T_V(H=0)[T_0<T_V(H=0)]$. The thermodynamic equilibrium state at this point of the phase diagram $(T=T_0,H=0)$ is the Fermi liquid state. Next, we apply an external magnetic field H_0 that reduces the valence transition temperature to exactly T_0, i.e., $T_V(H_0)=T_0$. At this point a local moment phase, highly polarized by the external magnetic field H_0, is in coexistence with the Fermi liquid phase at the instability temperature. If the magnetic field is now reduced and the thermodynamic equilibrium state of the system at T_0 is again the Fermi liquid, the polarized local moment phase will *melt*. The resulting Fermi liquid in a time scale shorter than the relaxation time of the magnetization must keep the original, large polarization, of the local moment phase.

Similar results can be accomplished changing adiabatically the external pressure P on the system, but keeping the magnetic field constant, to vary $T_V(P,H)$. If pressure has to be increased or relieved will depend on the sign of ΔV at the transition. In the cases of ^{3}He and $YbInCu_4$ pressure has to be released to melt the local moment phase. The resulting strongly polarized heavy Fermi liquid is a still unexplored state of matter which may reveal interesting properties.

Let us consider in more detail the polarization process through a change of the external magnetic field. The total normalized magnetization at the coexistence line is given by: $m=(1-x_{\mathrm{LM}})m_{\mathrm{FL}}+x_{\mathrm{LM}}m_{\mathrm{LM}}$, where m_{LM} and m_{FL} are the magnetization of the local moment and Fermi liquid phases, normalized by their fully polarized values, respectively. A similar expression applies for the entropy of the system. The equation for m can be rewritten as, $(m/m_{\mathrm{LM}})=(1-x_{\mathrm{LM}})(\chi_{\mathrm{FL}}/\chi_{\mathrm{LM}})+x_{\mathrm{LM}}$, where χ_i is the magnetic susceptibility of the ith phase. When the field is reduced, heating of the sample will occur during the local moment-Fermi liquid conversion. This is the reverse of the Pomeranchuk effect.[4,5] The decrease of the magnetization, starting from the polarized local moment phase at T_0, due to the *magnetic decompression* can be related to the temperature variation in the sample by

$$\frac{\Delta m}{m_{\mathrm{LM}}} \approx \frac{C_P(T_0)(1-\chi_{\mathrm{FL}}/\chi_{\mathrm{LM}})}{T_0 R \ln(2J+1)}\Delta T,$$

where C_P is the specific per mole of the sample and $\Delta T=T-T_0$. Notice that after the conversion process is complete, temperature will lower again since the sample is in contact with the heat reservoir.[13] In deriving the above equation, notice that the entropy $S=x_{\mathrm{LM}}S_{\mathrm{LM}}+(1-x_{\mathrm{LM}})S_{\mathrm{FL}}$, where $S_{\mathrm{LM}}=R\ln(2J+1)$ and $S_{\mathrm{FL}}=\gamma T(S_{\mathrm{LM}}\gg S_{\mathrm{FL}})$ are the entropies of the local moment and Fermi liquid phases, respectively.

A basic question for obtaining the highly polarized Fermi liquid state concerns the competition between two time scales, namely, the lifetime of the polarized state and the melting time of the local moment phase. What we want to achieve is a condition of *fast melting*, such that, the former is much longer than the latter.

At the coexistence temperature $T_V(H_0)$, the local moment and Fermi liquid phases can be seen as a two component AB mixture both with spins up and down.[2] The free energy, which depends on the total magnetization M induced by the external field H is invariant under exchange of $A\uparrow(A\downarrow)$ and $B\uparrow(B\downarrow)$ particles. The spin dependent chemical potentials of the two phases, $\mu_A^{\uparrow}=\mu_B^{\uparrow}=\mu_{\uparrow}$ and $\mu_A^{\downarrow}=\mu_B^{\downarrow}=\mu_{\downarrow}$. On a time scale shorter than the relaxation time of the magnetization the effective magnetic field acting on the mixture is $H_{\mathrm{eff}}=\mu_{\uparrow}-\mu_{\downarrow}$. Then, for sufficiently *fast melting* of the local moment phase, such that, the magnetization M is conserved the effective magnetic field on the Fermi liquid is given by $H_{\mathrm{eff}}=(C_{\mathrm{LM}}/C_{\mathrm{FL}})(T_{\mathrm{FL}}/T)H$. This can attain large values yielding strongly polarized Fermi liquid states. We used that the magnetization of the Fermi liquid is $M_{\mathrm{FL}}=\chi_{\mathrm{FL}}H=(C_{\mathrm{FL}}/T_{\mathrm{FL}})H$ and that of the local moment phase, $M_{\mathrm{LM}}=\chi_{\mathrm{LM}}H=(C_{\mathrm{LM}}/T)H$. The Curie constant $C_{\mathrm{LM}}=Np^2\mu_B^2/3k_B$ in the local moment phase is very close to the value obtained from the magnetic configuration of Table I.[14] For the Pauli-like susceptibility of the Fermi liquid χ_{FL}, we can take $T_{\mathrm{FL}}=T_V$, but C_{FL} cannot be obtained from the nonmagnetic configuration in Table I since of course this vanishes.

TABLE I. Relevant parameters that determine the magnetic properties of the systems discussed in the text in their local moment phase. Notice that $p=g\sqrt{J(J+1)}$.

	L	S	J	g	p
Eu^{3+}	3	3	0	0	0
Eu^{2+}	0	7/2	7/2	2	7.94
Ce^{4+}	0	0	0		0
Ce^{3+}	3	1/2	5/2	6/7	2.54
Yb^{3+}	3	1/2	7/2	8/7	4.54
Yb^{2+}	0	0	0		0

In ^{3}He, at the temperatures of the polarization experiments, the time scale of the relaxation of the magnetization is of the order of minutes.[15] In the solid-state Pomeranchuk materials, due to their metallic nature, magnetic relaxation should be faster, even if $T_V(H)$ is reduced to the temperatures of the ^{3}He experiments. An estimate of the relaxation processes in the unstable valence materials can be obtained from electron spin resonance (ESR). The most investigated of the compounds discussed here is $YbInCu_4$ for which at least two ESR studies have been published.[16,17] However, the resonance line of Yb in this cubic compound could not be observed, even in the local moment phase for $T>T_V$. In order to get an ESR signal, the material is doped with a very small concentration of Gd which is used as a probe. Below T_V, the relaxation of the electromagnetic radiation absorbed by the Gd ions occurs through the electrons of the highly correlated Fermi liquid state as indicated by the observed Korringa-like linewidth.[17] Above T_V, in the local moment phase, relaxation is not so efficient (see Fig. 1). If the Yb ions in the cubic environment relax as the Gd impurities, we may expect to find long relaxation times in the local moment phase at low temperatures. An extrapolation of the temperature dependent Gd linewidth shown in Fig. 1 for temperatures below T_V yield an infinite lifetime at finite temperatures. Since we can reduce T_V through the application of large magnetic fields, which at the same time polarize the local moments, this al-

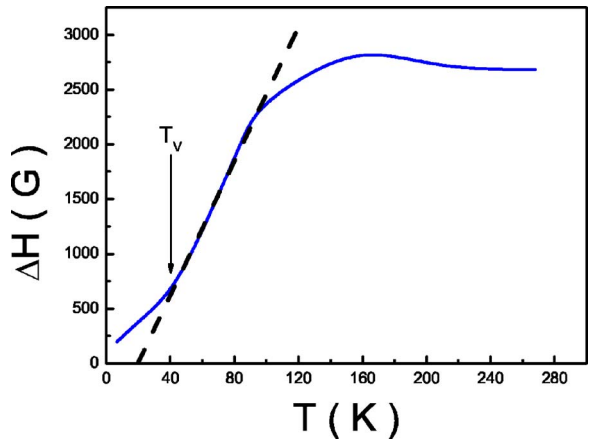

FIG. 1. (Color online) Linewidth of Gd ions diluted in $YbInCu_4$ as a function of temperature (adapted from Ref. 16) Below T_V the linewidth is Korringa-like, due to electrons in a narrow band (Ref. 17). The extrapolation of the linewidth in the local moment phase ($T > T_V$) to low temperatures vanishes at a finite T.

lows us to obtain long relaxation times for the local moment phase.

Relaxation of the magnetization in the renormalized Fermi liquid phase is harder to estimate. The local Gd probes in this phase relax through a Korringa mechanism but then this is different from the relaxation of the renormalized Fermi liquid itself. Recently, the Yb resonance has been observed in a heavy fermion material with an anisotropic structure.[18] Above the coherence temperature[4] where the system behaves as local moments, the relaxation of the Yb ions is through phonons and is quite inefficient. The lifetime of the polarization is directly related to the inverse linewidth and extrapolates to an infinite value at finite temperatures. However, below T_{coh} the linewidth increases linearly with temperature suggesting a Korringa mechanism which has been attributed to some remaining Yb^{3+} moments relaxing through the correlated Fermi liquid. The Fermi liquid may relax through electron-phonon and electron-electron scattering. The latter relaxation time increases as T^{-2} when T decreases.[19] It should involve anisotropic exchange or dipolar interactions which are non-spin conserving. No doubt this fast Fermi liquid relaxation is the most restrictive factor for the observation of the proposed effect, which may require the use of techniques to measure fast relaxation.[20]

Let us now briefly discuss the other mechanism competing with the relaxation of the magnetization, namely, the melting of the local moment phase. The nature and mechanisms of this *melting* process is in itself a very interesting problem. It is clearly connected to elastic processes as the transformation of the local moment phase into the renormalized Fermi liquid occurs through an expansion of the lattice.[14] These processes slow down with decreasing temperature. On the other hand the effect of the applied magnetic field is to facilitate the melting process as it reduces the melting temperature T_V.

During fast melting, equal numbers of spin up and spin down ions leave a bubble of local moment phase to the liquid. This will give rise to an excess of magnetization on the surface of the bubble (as long as spin diffusion is inefficient

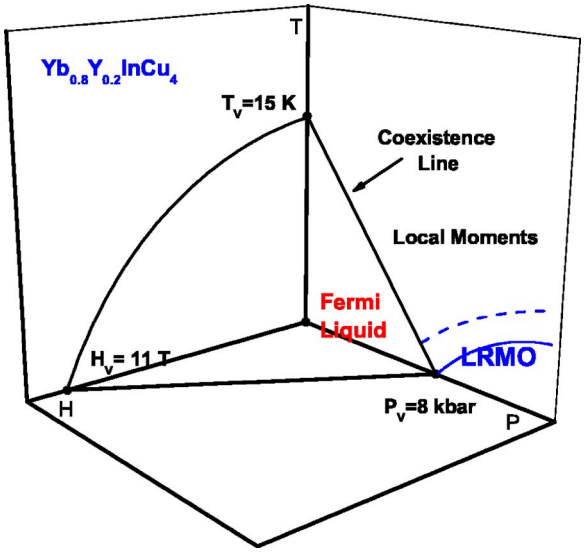

FIG. 2. (Color online) Schematic phase diagram of $Yb_{0.8}Y_{0.2}InCu_4$ as a function of temperature, pressure, and magnetic field (see Ref. 22. The two frontiers of the long range magnetic ordered (LRMO) phase, dashed and full lines, represent two different possibilities not yet distinguished by the experiments. In both cases, the zero temperature magnetic instability occurs from the Fermi liquid phase (Ref. 22).

in redistributing the magnetization). As a consequence the effective magnetic field on the surface increases and further enhances the melting process. In this type of *dendritic* melting the liquid penetrates on the local moment regions giving rise to further small bubbles and so on. This type of process stops when a minimum size is reached where diffusion occurs sufficiently fast to homogenize the magnetization in the bubble.[21]

The systems $YbInCu_4$ and the Eu compounds mentioned above can be brought to the proximity of a magnetic instability either by pressure or doping.[22] In the case of $YbInCu_4$ the long range ordered magnetic phase is a weak ferromagnetic phase which can be attained for sufficiently high pressures.[22] A schematic phase diagram is shown in Fig. 2. The experiments are not yet sufficiently detailed to determine whether the magnetic frontier rises from a $T=0$ quantum critical point[23] or starts at finite temperatures at the line $T_V(P)$ of first order instabilities.[23,24] However in both cases shown in Fig. 2, the zero temperature ferromagnetic instability[23,24] is always from the Fermi liquid phase. This phase diagram strongly suggests that to obtain a polarized Fermi liquid starting from the local moment phase it is convenient to work close to the magnetic instability as this will surely enhance the lifetime of the magnetic polarization.

In this paper, we have proposed a mechanism for obtaining a highly spin polarized strongly correlated Fermi liquid. This can be accomplished in a class of solid-state materials and represents a still unexplored state of matter whose properties may be very interesting to investigate. These are unstable f-shells systems that present a first order transition from a magnetic configuration to a renormalized Fermi liquid state. The essential condition to observe this effect is that of *fast melting*. This implies that the *melting* of the local

moment phase polarized by the external magnetic field occurs faster than the relaxation time of the magnetization. We have made a brief discussion of the nature of both the relaxation and melting processes. If experiments are carried out at sufficiently low temperatures, which imply using large fields, fast melting can be achieved. Large magnetic fields reduce T_V increasing the relaxation time of the magnetization, facilitate melting, and produce high polarizations. We pointed out that the *melting process* through which the local moment phase transforms in the highly correlated polarized Fermi liquid state is an interesting problem on its own. For observing the solid-state Pomeranchuk effect and obtaining highly polarized Fermi liquids, the rare earth materials are convenient due to their large effective moments which couples strongly to the external field. Also the large entropy associated with these moments makes them useful when used for cooling purposes. However, rather similar effects should be possible to find in other materials which exhibit first order transitions and the nature of the electrons involved in these transitions changes discontinuously from localized to itinerant. For example, near Mott transitions.[25] We hope these ideas can encourage further experimental work in these systems.

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[1] G. A. Prinz, Science **282**, 1660 (1998); S. A. Wolf, *ibid.* **294**, 1488 (2001); I. Zutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

[2] B. Castaing and P. Nozières, J. Phys. (Paris) **40**, 257 (1979).

[3] C. R. Sanchez-Castro, K. S. Bedell, and S. A. J. Wiegers, Phys. Rev. B **40**, 437 (1989); B. Hébral, A. S. Greenberg, M. T. Béal-Monod, M. Papoular, G. Frossati, H. Godfrin, and D. Thoulouze, Phys. Rev. Lett. **46**, 42 (1981).

[4] M. A. Continentino and A. S. Ferreira, Phys. Rev. B **69**, 233104 (2004); Solid State Commun. **131**, 195 (2004).

[5] M. A. Continentino and A. S. Ferreira, Cryogenics **45**, 331 (2005); M. A. Continentino, A. S. Ferreira, P. G. Pagliuso, C. Rettori, and J. L. Sarrao, Physica B **359&361**, 744 (2005).

[6] J. K. Freericks and Z. Vlatic, Phys. Status Solidi B **236**, 265 (2003); V. Zlatic and J. K. Freericks, Acta Phys. Pol. B **32**, 3253 (2001).

[7] B. Coqblin and A. Blandin, Adv. Phys. **17**, 281 (1968).

[8] C. U. Segre, M. Croft, J. A. Hodges, V. Murgai, L. C. Gupta, and R. D. Parks, Phys. Rev. Lett. **49**, 1947 (1982).

[9] M. M. Abd-Elmeguid, Ch. Sauer, and W. Zinn, Phys. Rev. Lett. **55**, 2467 (1985); M. M. Abd-Elmeguid, H. Micklitz, and G. Kaindl, Phys. Rev. B **23**, 75 (1981).

[10] E. M. Levin, B. S. Kuzhel, O. I. Bodak, B. D. Belan, and I. N. Stets, Phys. Status Solidi B **161**, 783 (1990); G. Wortmann, I. Nowik, B. Perscheid, G. Kaindl, and I. Felner, Phys. Rev. B **43**, 5261 (1991); H. Wada, A. Nakamura, A. Mitsuda, M. Shiga, T. Tanaka, H. Mitamura, and T. Goto, J. Phys.: Condens. Matter **9**, 7913 (1997); H. Wada, M. F. Hundley, R. Movshovich, and J. D. Thompson, Phys. Rev. B **59**, 1141 (1999).

[11] W. Zhang, N. Sato, K. Yoshimura, A. Mitsuda, T. Goto, and K. Kosuge, Phys. Rev. B **66**, 024112 (2002).

[12] I. Felner, I. Nowik, D. Vaknin, Ulrike Potzel, J. Moser, G. M. Kalvius, G. Wortmann, G. Schmiester, G. Hilscher, E. Gratz, C. Schmitzer, N. Pillmayr, K. G. Prasad, H. de Waard, and H. Pinto, Phys. Rev. B **35**, 6956 (1987).

[13] A. Dutta and C. N. Archie, Phys. Rev. Lett. **55**, 2949 (1985).

[14] J. L. Sarrao, Physica B **259–261**, 128 (1999) and references therein.

[15] M. Chapellier, G. Frossati, and F. B. Rasmussen, Phys. Rev. Lett. **42**, 904 (1979).

[16] T. S. Altshuler, M. S. Bresler, M. Schlott, B. Elschner, and E. Gratz, Z. Phys. B: Condens. Matter **99**, 57 (1995).

[17] C. Rettori, S. B. Oseroff, D. Rao, P. G. Pagliuso, G. E. Barberis, J. Sarrao, Z. Fisk, and M. Hundley, Phys. Rev. B **55**, 1016 (1997).

[18] J. Sichelschmidt, V. A. Ivanshin, J. Ferstl, C. Geibel, and F. Steglich, Phys. Rev. Lett. **91**, 156401 (2003); see also K. Ishida, K. Okamoto, Y. Kawasaki, Y. Kitaoka, O. Trovarelli, C. Geibel, and F. Steglich, *ibid.* **89**, 107202 (2002).

[19] D. Shaltiel, D. Ioshpe, A. Grayevsky, V. Zevin, and J. L. Smith, Phys. Rev. B **36**, 4090 (1987); F. Beuneu and P. Monod, Phys. Rev. B **18**, 2422 (1978).

[20] A. Y. Elezzabi, M. R. Freeman, and M. Johnson, Phys. Rev. Lett. **77**, 3220 (1996); A. Vaterlaus, T. Beutler, and F. Meier, *ibid.* **67**, 3314 (1991).

[21] G. Bonfait, L. Puech, A. S. Greenberg, G. Eska, B. Castaing, and D. Thoulouze, Phys. Rev. Lett. **53**, 1092 (1984).

[22] A. Mitsuda, T. Goto, K. Yoshimura, W. Zhang, N. Sato, K. Kosuge, and H. Wada, Phys. Rev. Lett. **88**, 137204 (2002).

[23] See M. A. Continentino, *Quantum Scaling in Many-Body Systems* (World Scientific, Singapore, 2001), Chap. 12.

[24] M. A. Continentino and A. S. Ferreira, Physica A **339**, 461 (2004).

[25] J. Spalek, A. Datta, and J. M. Honig, Phys. Rev. Lett. **59**, 728 (1987).