Thermodynamics of metal-insulator systems: The two-fluid model in the presence of a magnetic field

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The two-fluid model for metal-insulator systems such as Si:P, involving localized and delocalized electrons, is extended to allow for magnetic field effects. Specific-heat and spin susceptibility expressions are given for the extended model and the predictions are compared with available experimental results. The agreement is satisfactory but further measurements of, for example, the Wilson ratio are needed in order to provide a more stringent test of the model. [S0163-1829(96)02124-8]

For metal-insulator (MI) systems, such as heavily doped semiconductors, there are important differences in character between transport properties and thermodynamic properties in the vicinity of the MI transition. The transport properties, in general, exhibit critical behavior near the critical concentration n_c, while the thermodynamic properties vary smoothly across the transition even at very low temperatures. A recent review[1] gives a comprehensive account of developments related to the Anderson-Mott transition, with emphasis on the transport phenomena and an effective field theory approach.

Paalanen *et al.*[2,3] have developed a phenomenological two-fluid model for MI systems, which allows for both localized and delocalized moments in systems close to the MI transition. Localized moments can play an important role in determining properties such as the magnetic susceptibility and the specific heat. These moments are also believed to play a dominant role in the nuclear relaxation processes near the transition.[4,5] The delocalized electrons, on the other hand, determine the measured NMR Knight shift[6,7] and the transport properties.[8]

While the two-fluid model is phenomenological, some progress has been made in placing it on a theoretical basis using an effective Hubbard model Hamiltonian.[9] Bhatt and Fisher,[10] on the basis of a positionally disordered Anderson-Hubbard model, have argued that the low-temperature thermodynamics of the disordered metallic phase is dominated by spin excitations in regions of low impurity concentration where the Kondo temperature is extremely low. The basic ideas are related to those of Bhatt and Lee[11] (BL) on the insulating side of the transition. Lakner *et al.*[12] have recently put forward a theoretical model that allows for a distribution of Kondo temperatures and that gives a quantitative description of the concentration of local moments. Related ideas have been put forward by Dobrosavljević *et al.*[13,14]

Local moments are important in other highly correlated electron systems including heavy fermions such as UPd_2Al_3 and $CeCu_{2.2}Si_2$, which exhibit local moment antiferromagnetism and superconductivity at low temperatures.[15] Magnetic order and superconductivity are competing temperature-dependent effects.[16,17]

The application of a magnetic field can produce changes in the behavior of the MI systems such as the universality class in which a given system falls. Marked changes in the behavior of thermodynamic properties such as the specific heat,[18] which develops Schottky-type peaks,[19] may also be brought about in this way.

This paper is concerned with generalizing the two-fluid model of Paalanen *et al.*[2,3] to allow for magnetic field effects. The dominant contribution to such effects is believed to be due to the localized moments and this is the basis for extending the model.

The thermodynamic two-fluid predictions for $B=0$ are expressed in the following equations for the specific-heat coefficient γ and the magnetic susceptibility χ:

$$\frac{\gamma}{\gamma_0}=\frac{m^*}{m_0^*}+\left(\frac{T}{T_0}\right)^{-\alpha}, \tag{1}$$

$$\frac{\chi}{\chi_0}=\frac{m^*}{m_0^*}+\beta_0\left(\frac{T}{T_0}\right)^{-\alpha}. \tag{2}$$

γ_0 and χ_0 are Fermi-liquid values, with m^* the Fermi-liquid effective mass and $m_0^*=0.34m_0$ the Si conduction-band mass.[2] T_0 is a parameter that includes the fraction of localized moments and β_0 is a constant. The exponent α is discussed below.

The local moment contributions to γ and χ contained in the second terms in Eqs. (1) and (2) are obtained using ideas put forward by BL to explain the low-field ($B\to 0$) behavior of χ for MI systems on the insulating side of the transition. The exchange coupling between the randomly positioned local moments is assumed to follow the distribution

$$P(J)dJ\simeq\kappa J^{-\alpha}dJ, \tag{3}$$

where the exponent $\alpha\sim 0.62$ for Si:P and other similar systems[3] for $n\simeq n_c$. κ is a normalization constant. A maximum cutoff value J_0 may be assumed for the distribution[2] but this is sufficiently high that for many purposes integrals containing J_0 as an upper limit may be extended to infinity.

The local moment contribution to Eqs. (1) and (2) may be obtained readily. For $B=0$, the exchange Hamiltonian H_e is of dominant importance for the local moments, with $H_e=\Sigma_{i<j}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j$. J_{ij} is the exchange coupling between spins i and j. The partition function for a pair of localized spins with exchange coupling J may be written as $Z=1+3e^{-\beta J}$, where $\beta=1/k_BT$. Using the distribution func-

tion given in Eq. (3), one can then obtain the localized contributions shown in Eqs. (1) and (2). The parameter T_0 is then defined as

$$\left(\frac{1}{T_0}\right)^{-\alpha} = \frac{1}{\pi^2}\left(\kappa\int_0^\infty dx\,\frac{3x^{2-\alpha}e^{-x}}{(1+3e^{-x})^2}\right)k_B^{1-\alpha}T_f\left(\frac{n_e}{n}\right),$$

where $x=\beta J$ and T_F is the Fermi temperature; n_e and n are the localized and itinerant electron concentrations, respectively. Numerical evaluation gives $\beta_0 \simeq 10.3$, for $\alpha=0.62$, in close agreement with the value given by Paalanen *et al.*[2]

When a magnetic field B is applied to the system, a Zeeman term must be included in the Hamiltonian for the local moment subsystem. The partition function for a spin pair is then $Z=1+e^{-\beta J}+e^{-\beta(J+g\mu_B B)}+e^{-\beta(J-g\mu_B B)}$, where μ_B is the Bohr magneton and g is the g factor. Again using the distribution function in Eq. (3), the modified two-fluid model equations for $B\neq 0$ may be written as

$$\frac{\gamma}{\gamma_0} = \frac{m^*}{m_0^*} + \delta(y)\left(\frac{T}{T_0}\right)^{-\alpha}, \tag{4}$$

$$\frac{\chi}{\chi_0} = \frac{m^*}{m_0^*} + \varepsilon(y)\left(\frac{T}{T_0}\right)^{-\alpha}, \tag{5}$$

where the factors $\delta(y)$ and $\varepsilon(y)$, using $y=g\mu_B B/k_B T$, are given by

$$\delta(y) = \frac{I_\delta(y)}{I_\delta(0)}, \quad \varepsilon(y) = \beta_0\frac{I_\varepsilon(y)}{I_\varepsilon(0)}, \tag{6}$$

with the integrals $I_\delta(y)$ and $I_\varepsilon(y)$, which have to be evaluated numerically, defined as

$$I_\delta(y) = \int_0^\infty dx\left(x^2\frac{\partial^2 \ln Z}{\partial x^2} + 2xy\frac{\partial^2 \ln Z}{\partial x \partial y} + y^2\frac{\partial^2 \ln Z}{\partial y^2}\right)x^{-\alpha}, \tag{7}$$

$$I_\varepsilon(y) = \int_0^\infty dx\left(\frac{1}{2y}\frac{\partial \ln Z}{\partial y}\right)x^{-\alpha}. \tag{8}$$

The behavior of the coefficients δ and ε is shown as a function of $1/y$ in Fig. 1. The specific-heat coefficient δ shows a peak for $y \simeq 2.6$, which leads to the Schottky-type peak observed in the specific heat. Sarachik *et al.*[20] have previously obtained a similar expression for χ for the localized moments in a magnetic field.

Equations (4) and (5) reduce to Eqs. (1) and (2) in the $B=0$ limit. It is to be expected that the extended two-fluid model equations will be of some value in accounting for the effects of magnetic fields.

In making comparisons of the two-fluid model predictions with experiment, one is faced with choosing the parameters α and T_0. It is possible that T_0 (Ref. 21) and, to a lesser extent, α will depend on the field B. We allow for a slight field dependence of T_0, due to an enhanced localization of spins, but, for simplicity, fix α for a given n/n_c value.

As a test of the model Paalanen *et al.*[2] have plotted experimental values of the Wilson ratio $(\chi/\chi_0)/(\gamma/\gamma_0)$ versus T for three samples on which they made low-field heat capacity and susceptibility measurements. The $B=0$, two-fluid model predictions give good one-parameter fits to the experimental results. The present extended two-fluid model provides Wilson ratio values as a function of temperature for nonzero magnetic fields. Figure 2 gives plots of this kind. The minimum feature corresponds to the maximum in the plot of $\delta(y)$ shown in Fig. 1. Unfortunately, no sufficiently complete measurements of the field dependence of the specific heat and magnetic susceptibility for a given sample near the MI transition appear to have been made. It is therefore not possible for us to make a comparison of experimentally determined Wilson ratio values for nonzero magnetic fields with the extended two-fluid model predictions. Measurements that permit such a comparison to be made would be of great interest.

The asymptotic $T\to 0$ forms of the specific heat and susceptibility can be directly inferred from the asymptotic forms of $\delta(y)$ and $\varepsilon(y)$, as shown in Fig. 1, using Eqs. (4) and (5). At low temperatures (T[mK]$\lesssim 100B$[T]) the model predicts that the total electronic specific heat will be linear in T. Accurate verification of this prediction is made difficult by the presence at low temperatures of what is believed to be a nuclear contribution.[18] The susceptibility tends to a constant

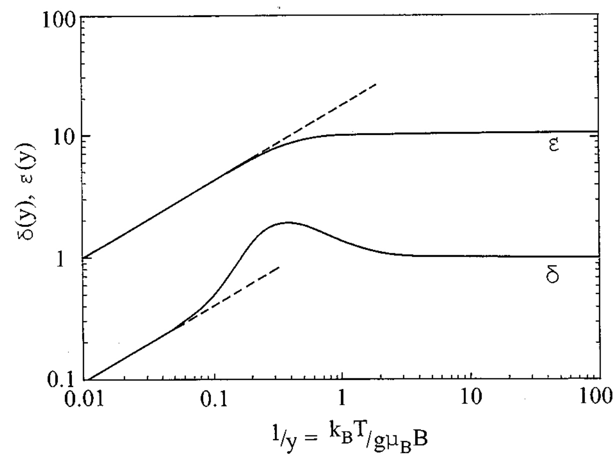

FIG. 1. The coefficients δ and ε, as defined in Eqs. (6), vs $k_B T/g\mu_B B$. The dashed lines show the low-temperature asymptotic behavior where $\delta,\varepsilon \sim y^{-\alpha}$.

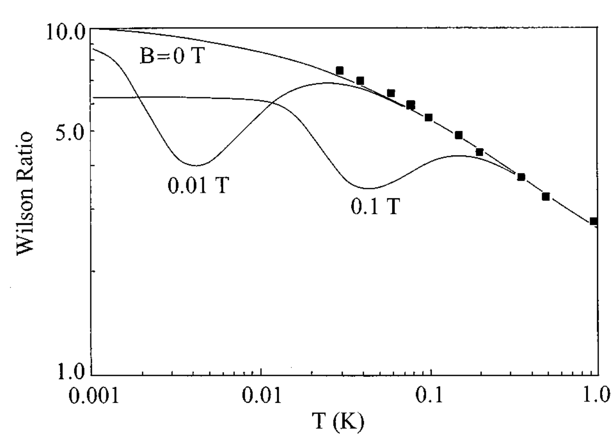

FIG. 2. Two-fluid model predictions of the Wilson ratio vs temperature for $n/n_c=1.09$. The zero-field experimental points shown are from Ref. 2.

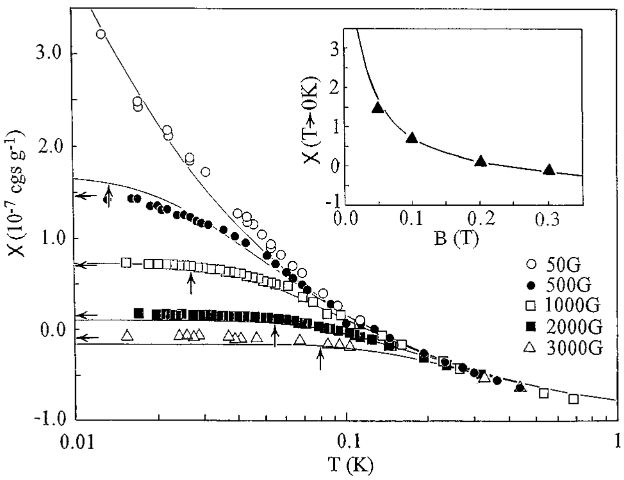

FIG. 3. Fits of Eq. (10) to the susceptibility (Ref. 22) for various magnetic fields, for $n=3.3\times10^{18}$ cm^{-3} ($n/n_c=1.1$ according to Ref. 22). Values used were $\chi^*=-1.1$ and $\rho=0.44$. The inset shows the asymptotic low-temperature behavior (indicated by the horizontal arrows) as a function of the applied field. The vertical arrows show the predicted temperatures where the susceptibility should start leveling off.

value, in this model, as $T\to0$. For $T[\mathrm{K}]\lesssim0.27B[\mathrm{T}]$ the localized contribution to the susceptibility has the field-dependent form

$$\chi=\left[\frac{\kappa\mu_0(g\mu_B)^{2-\alpha}}{2(1-\alpha)}n_e\right]B^{-\alpha}. \tag{9}$$

From Eq. (5) the total susceptibility χ can be written as

$$\chi=\chi^*+\rho I_\varepsilon(y)T^{-\alpha}, \tag{10}$$

where χ^* now also includes the constant diamagnetic Si host susceptibility, and ρ is a constant. Matsunage and Ootuka[22] have measured the magnetic susceptibility for a Si:P sample very close to the MI transition as a function of temperature and applied magnetic field. Their results are shown in Fig. 3. The fitted curves are obtained using Eq. (10) above, with $\alpha=0.6$. The constant $\chi^*=-1.1\times10^{-7}$ cgs/g is consistent with estimates of the diamagnetic Si susceptibility[23] and the Pauli-Landau susceptibility of itinerant electrons. ρ is treated as a parameter, depending on several unknowns. The inset in Fig. 3 shows the field dependence of the asymptotic low-temperature behavior, which can be read for four values of B. The fitted curve is based on Eq. (9). For the lowest field (50 G), the model predicts that convergence will only occur for temperatures around 1 mK.

Lakner and Löhneysen[12,18] have measured the specific heat for a number of MI systems in fields up to 6 T. We have used Eq. (4) to make comparisons of theory with their results for Si:P. In zero magnetic field, the total specific heat may be written as a function of temperature in the form

$$C(T)=\sigma T^{1-\alpha}+\gamma_i T+\beta T^3, \tag{11}$$

where σ is a constant, involving the parameter T_0, which is proportional to n_e. $\gamma_i=\gamma_0 m^*/m_0^*$ is the itinerant electron specific-heat coefficient and β is the phonon coefficient. We have used the Debye temperature of 660 K for silicon[18] to determine β. This reduces the number of fitting parameters. σ and γ_i for a particular specimen may be determined by fitting the zero-field data. We take $n_c=3.52\times10^{18}$ cm^{-3}.[24] σ, which gives a measure of the localized moment contribution, increases with n and then decreases smoothly through the transition. γ_i increases from a value close to zero for $n<n_c$ to finite values fairly abruptly at the transition. For $n/n_c>1$, γ_i tends to the value expected for the multivalley system,[12,25] with an effective mass m^* close to the band effective mass.[2]

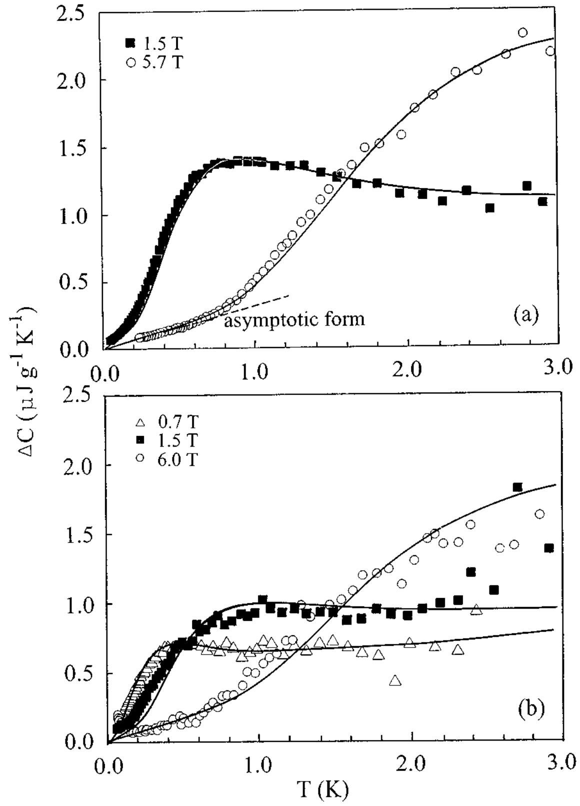

FIG. 4. Fits of Eq. (12) to the experimental ΔC values in various magnetic fields (Ref. 12) for (a) $n/n_c=0.45$ and (b) $n/n_c=1.02$. For the insulating side (a), using $\alpha=0.7$, we find $(\sigma,\gamma_i)=$ (0.64,0) and (0.70,0) for $B=1.5$ and 5.7 T, respectively. For the metallic sample (b), using $\alpha=0.63$, we find $(\sigma,\gamma_i)=$ (0.49,1.04), (0.55,0.92), and (0.63,0.94) for $B=0.7$, 1.5, and 6 T, respectively.

For the heat capacity data $C(B,T)$ obtained with an applied magnetic field present, it is convenient to introduce the excess specific heat ΔC, defined as $C(B,T)=\Delta C+\gamma_i T+\beta T^3$. This quantity may be compared with the theoretical prediction based on Eq. (4):

$$\Delta C=\sigma\delta(y)T^{1-\alpha}. \tag{12}$$

Figures 4(a) and 4(b) show plots of experimental ΔC values, obtained from the data of Lakner *et al.*,[12] for $n/n_c=0.45$ and 1.02. The theoretical predictions of the extended two-fluid model are shown as the fitted curves. σ and γ_i values are given in the captions for each figure. Note the absence of an itinerant contribution for $n/n_c<1$. While there may be a slight field dependence of σ obtained from the fits, it is not possible to draw any firm conclusions on field-induced localization effects.[21]

Nuclear moment contributions to the specific heat have been ignored but it is clear that at the lowest temperatures and for high fields these effects are becoming important. A nuclear Schottky term can be added to Eq. (11) to account for the upturn in ΔC observed below 100 mK.

The entropy of the system can be directly obtained by using the specific-heat expression in Eq. (4). At sufficiently low temperatures the nuclear spin entropy contribution will dominate, as has been pointed out by Lakner and Löhneysen.[18]

In summary, the two-fluid model[2] has been generalized to include the effects of an applied magnetic field. The predictions of the modified equations are compared with the limited number of measurements of thermodynamic response functions made on MI systems in the presence of a magnetic field. The agreement of the present model with both the experimental ΔC and χ values is gratifying. Further experiments are needed to test the model in greater detail.

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