Thermoelectric power during magnetic-field-induced localization in degenerately doped n-type Ge

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Thermoelectric power, resistivity, and Hall-effect measurements are presented of the magnetic-field-induced metal-insulator transition in degenerately doped n-type Ge. At low temperatures and high magnetic fields, this system changes from an isotropic but "dirty" metal to an isotropic insulator. The resistivity tensor is indicative of a pure mobility transition with no appreciable loss of charge carriers, consistent with Mott-type variable-range hopping. The thermoelectric power of these samples, however, increases with increasing field and decreasing temperature, possibly indicating that the density of states vanishes as the system passes into the insulating state.

Impurity conduction and the metal-insulator transition in doped semiconductors has been of interest for over 40 years.[1–5] In recent years magnetic-field-induced metal-insulator transitions in degenerately doped semiconductors have become of interest due to suggestions that collective phenomena are playing an important role.[5,6] Traditionally, magnetic freeze-out[7] was used to describe the behavior of heavily doped semiconductors in large magnetic fields. In recent years new ideas have been formulated to describe these systems. Depending on the strength of various interaction energies, Wigner crystallization,[8] charge-density waves,[9] spin-density waves,[10] and valley waves[11] have also been proposed as possible ground states of degenerate semiconductors in large magnetic fields.

Experiments on compound semiconductors have interpreted these magnetic-field-induced metal-insulator transitions in terms of both collective phenomena and magnetic freezeout. Experiments on InSb are usually interpreted as evidence for magnetic freeze-out.[12,13] The case for $Hg_xCd_{1-x}Te$ is a matter of some controversy with magnetic freeze-out,[13] Wigner crystallization,[14,15] and "correlated viscous liquid"[16] states proposed as the ground state in a magnetic field. Experiments on InAs have been used as evidence for a proposed "Hall insulator" state,[17] in which the Hall effect remains metallic while the resistivity undergoes a metal-insulator transition.

Elemental semiconductors have also been recently studied in large fields. Magnetic tuning experiments in Si (Ref. 18) have been performed to probe the behavior in high fields while in the metallic state. Experiments on Ge (Refs. 19–21) have succeeded in driving samples insulating without an apparent change in the Hall coefficient, similar to the InAs studies mentioned above. While in this later study the anomalous behavior of the Hall coefficient could also be interpreted as evidence for collective phenomena such as strongly correlated hopping, it is not required to explain the experimental data in Refs. 19–21.

In this paper thermopower, resistivity, and Hall measurements are presented which may indicate the opening of an energy gap in degenerately doped n-type Ge as the magnetic field tunes the system into the high-field insulating state reported in Refs. 19–21.

While resistance (and magnetoresistance) measurements probe the density and mobility of the carriers, the thermoelectric power (S) probes their energy distribution. This is why the thermopower can sometimes be viewed as the "entropy per carrier" for a system. The third law of thermodynamics requires, for all systems, that the product of the thermopower and the conductivity must go to zero as the temperature goes to zero ($\sigma S \rightarrow 0$ as $T \rightarrow 0$).[22,23] A metal with finite conductivity at $T=0$ must have $S \rightarrow 0$ as $T \rightarrow 0$. For the insulating case where $\sigma \rightarrow 0$ as $T \rightarrow 0$, we may have S finite or diverging less rapidly than $1/\sigma$ (as for intrinsic semiconductors which have a minimum carrier energy) or $S \rightarrow 0$ (as for Mott-type variable-range hopping). If the energy distribution of the carriers falls to zero width about the Fermi energy as $T \rightarrow 0$ we will have $S \rightarrow 0$. Such is the usual case for metals as well as for noninteracting (Mott-type) variable-range hopping. If S increases as $T \rightarrow 0$ there is evidence for the existence of a minimum excitation energy (energy gap). There have been simple calculations suggesting that a soft gap, as can be produced by the presence of strong Coulomb interactions, can also lead to a nonzero thermopower at zero temperature.[24,25] It should be noted, however, that strong spin-flip scattering, as in Kondo alloys, can produce an increasing S with decreasing T over a limited range above the Kondo temperature. Eventually $S \rightarrow 0$ as $T \rightarrow 0$ for these systems.[23]

We find that as degenerately doped n-type Ge samples are driven into the insulating state by the application of a large magnetic field at low temperature, the thermopower increases considerably although the Hall coefficient changes are very small. Furthermore, over the temperature and magnetic-field range covered, the increase of the thermopower with decreasing temperature gets larger as the sample is driven farther into the insulating state. The thermopower measurements possibly indicate that the density of states at the Fermi level vanishes as the samples pass into this high-field insulating state.

The samples used in this study were cut from uncompensated Czochralski-grown Ge degenerately doped with Sb donors, obtained from Eagle-Picher. Careful checks of dopant homogeneity were performed as described in Refs. 19–21. The samples were cut with a diamond wire saw into long bars, lapped, and etched in 3:1 (by volume) concentrated HNO_3 to HF. Electrical contacts were made using a Sb-saturated Sn solder comprised of 80% Sn and 20% Sb. This produced typical contact resistances of 0.5–3 Ω at low temperatures. The final sample sizes were approximately $1.1\times3.3\times24$ mm^3. Two contacts were placed on the ends of the bars to be used as current injection contacts during resistivity and Hall measurements and to provide thermal and electrical contact during the thermopower measurements. Three small voltage contacts were placed along opposite sides of the samples to permit virtual-contact Hall measurements,[26,27] as well as four-terminal resistance measurements.

The experiments were performed on a temperature-controlled copper sample holder in an evacuated can and submerged in liquid helium. The sample arrangement, a modified version of the apparatus of Geballe *et al.*,[28] is shown in Fig. 1. The sample was mounted so that one end was silver epoxied to a single-crystal quartz heat sink. The remainder of the sample was cantilevered into the vacuum. A small thin single-crystal sapphire slide with a heater attached was then silver epoxied onto the opposite end of the sample. Au wires were then Sn-Sb soldered to the previously applied Sn-Sb contacts and a 25-μm Chromel-Constantan differential thermocouple was attached using silver epoxy to monitor the temperature difference between the sapphire gradient heater platform and the quartz sample heat sink. The relatively small (compared to the sample presented here) magnetic field dependence of the Au wires and the magnetic field dependence of the Chromel-Constantan were extrapolated using the information given in Refs. 29 and 30. The accuracy of this method for measuring the absolute thermopower, for these samples under these conditions, is $\sim$10%, ±5 μV/K and is practical above $\sim$1 K.

Careful checks were performed with helium exchange gas in the can as well as with the can evacuated to determine that the samples were heat sunk adequately to insure there were no temperature gradients between the sample and the copper platform.

Resistivity and virtual-contact Hall-effect measurements were made using conventional low-noise ac ($\sim$25 Hz) lock-in amplifier techniques as in Refs. 19–21. Thermopower measurements were performed by measuring (with a nanovoltmeter) the change in the voltage response of the sample as a measured temperature gradient ($\sim$0.1 to $\sim$0.3 K along the sample as determined by the differential thermocouple) is imposed periodically upon it. It should be noted that the period with which the gradient was switched on and off was several times the thermal time constant of the system thus allowing the sample to reach a steady state before a voltage measurement was taken. As a further check on the small field dependence of the differential thermocouple, the power into the gradient heater was kept constant for each on cycle, independent of field. The samples were first mounted with the current and temperature gradient perpendicular to the applied magnetic field. After the transverse resistance, the thermopower and Hall effect were measured the samples were remounted parallel to the field to allow longitudinal measurements of resistivity and thermopower.

In Fig. 2 we show logarithmic plots of the transverse and longitudinal magnetoresistivity at various temperatures of a sample of donor density $N_D\approx2.2\times10^{17}$ cm^{-3} as determined by room-temperature resistivity measurements. Low-field Hall measurements indicated that the carrier density was $n\approx1.7\times10^{17}$ cm^{-3} at 4.2 K, assuming a Hall factor of 1. The magnetoresistance changes from a weak field and temperature-dependent metallic regime below some characteristic field to an activated hopping regime at high fields. The data in Fig. 1 show clear evidence of a metal-insulator transition consistent with the data of Refs. 19–21.

Figure 3 shows the measured Hall coefficient of the

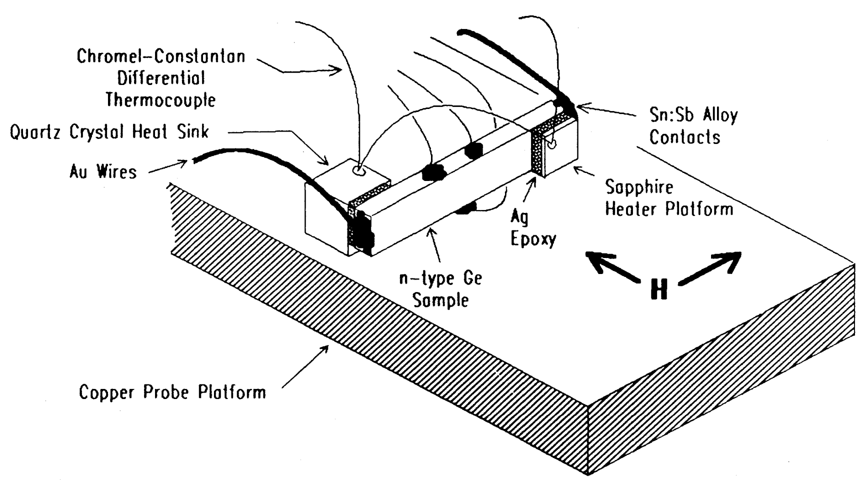

FIG. 1. Sample arrangement on copper holder. Sample was cantilevered on a step of a holder with heater placed on far end. All leads to samples and thermocouples were heat sunk to avoid additional gradients from the room-temperature leads.

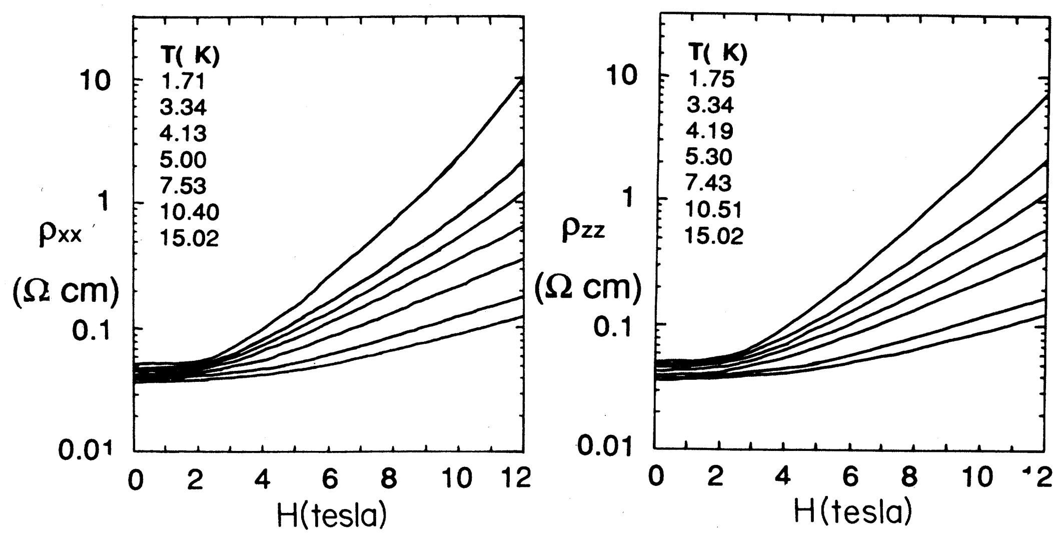

FIG. 2. Transverse and longitudinal magnetoresistance for a sample with $N_D \approx 2.0 \times 10^{17}$ cm^{-3}.

sample in Fig. 2 at two temperatures in fields up to 15 T. The inset shows the transverse magnetoresistivity of the sample over the same field range and at the same temperatures. As pointed out in previous publications,[19–21] the striking feature of the Hall coefficient in this system is the absence of the strong metal-insulator transition. These data may imply that the apparent carrier concentration changes relatively little across this metal-insulator transition, indicating that this system undergoes a pure mobility transition into a hopping regime. We note, however, that there is no consensus on calculations of the Hall resistance for hopping systems.[2–4,31–33]

These resistivity and Hall-effect studies are primarily presented to show that the samples in this study have the same behavior as the samples presented in Refs. 19–21 where only resistivity and Hall effect were performed. We have observed this behavior in all *homogeneous n*-type Sb-doped Ge samples in this dopant range.

The absolute transverse thermoelectric power of the sample from Figs. 2 and 3 is shown as a function of magnetic field for various temperatures in Fig. 4. Checks of the thermopower longitudinal to the field indicates the thermopower was isotropic to within the accuracy of the measurement. The zero-field data are consistent with

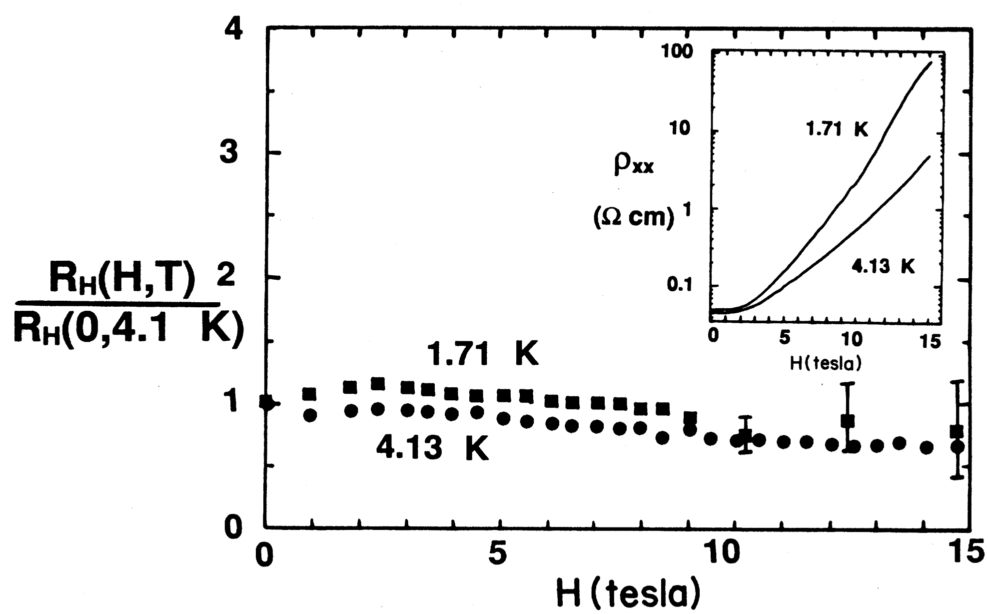

FIG. 3. Hall coefficient R_H normalized to the value at $H = 0$ and $T = 4.1$ K, at the temperatures indicated, for the sample shown in Fig. 2. Inset shows the transverse resistivity at the same temperatures.

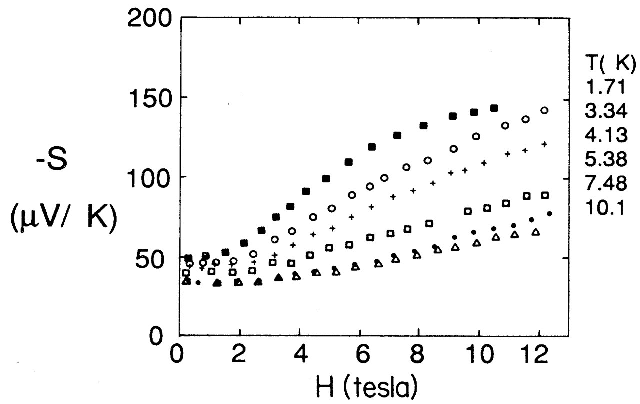

FIG. 4. Thermopower as a function of magnetic field for the sample shown in Figs. 2 and 3.

published zero-field thermopower data over the same temperature range (down to ~5 K) for Sb-doped Ge of this dopant concentration, given the slight geometry dependence (due to the large phonon mean free path) of the phonon drag peak at ~55 K for Ge in this dopant range.[28,34] Except for the study presented in this paper, to our knowledge the thermopower of germanium at these dopant levels has not been measured below ~5 K.[34]

In Fig. 5 we show the data of Fig. 4 plotted versus $1/T$ for various magnetic fields. As can be seen in Fig. 5, the thermopower at a given field increases strongly with decreasing temperature. At the lower temperatures measured, the rate of change slows down, perhaps indicating that the thermopower will saturate below a few degrees. This behavior appears inconsistent with a simple heating effect. dc measurements made nearly simultaneously with the thermopower (within a few milliseconds) give resistivity results identical to Fig. 1, which are similar to the results of Refs. 19–21. Considering the temperature sensitivity of the resistivity, any uncontrolled sample heating of the size required to straighten the curves in Fig. 5 would be quite pronounced in the resistivity. Since

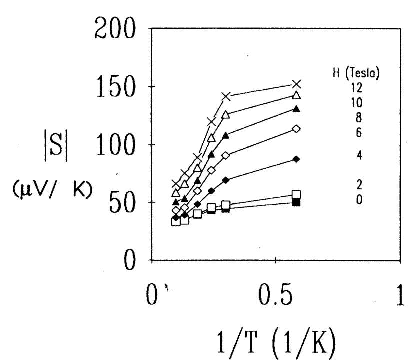

FIG. 5. Data of Fig. 4 plotted vs $1/T$ at constant magnetic field. Thermopower for a semiconducting system increases as $1/T$.

thermopower is a zero constant measurement, no external power is placed into the sample except the controlled amounts supplied by the gradient heater. We checked the response of the samples to different size-temperature gradients. Such checks give results consistent with Fig. 5 independent of the size of the gradient, provided the shift in the average sample temperature was taken into account. If the Fermi level was in a energy gap (of finite width) in the density of states, the thermopower would display a $1/T$ dependence for $k_B T \ll E_{\rm gap}$,[23] where k_B is Boltzmann's constant. Such is the observed behavior for intrinsic semiconductors. There are several possible models for magnetic-field-induced metal-insulator transitions, some involving collective phenomena, others are single-particle models.

Charge-density waves have energy gaps at the Fermi level.[35,36] Wigner crystallization and valley waves, which can often be treated as three-dimensional charge-density wave states,[5,6] as well as spin-density waves which can be treated as two charge-density waves 180° out of phase,[37] would also have a gap at the Fermi level. All of these gaps are of finite width, and hence at low enough temperature should give rise to semiconducting behavior. This is observed in conventional charge and spin-density wave systems such as the organic charge-transfer salts of the tetramethyltetraselenafulvalene family.[38] Thermopower in traditional charge and spin-density wave systems with fully gaped Fermi surfaces, may show increases at higher temperatures but typically start to display clear $1/T$ behavior when $k_B T < \sim 5\%$ of $E_{\rm gap}$.[38,39] If the assumption were made that a finite width gap is appearing in this degenerately doped Ge system, the lowest temperature resistivity data if Fig. 1 would crudely indicate a gap of ~5 K at 12 T. Using this value, one would not expect to see unambiguous $1/T$ behavior in the 12 T thermopower unless $T < \sim 200$ mK. One should note however, that measurements of resistivity and Hall measurements on a similar sample, presented in Ref. 20, show no evidence for a finite width gap down to temperatures of 30 mK and at magnetic fields up to 7 T.

Strong spin-flip scattering from local moments can produce an increasing thermopower in zero field as, for example, in Kondo alloys.[23] The presence of a symmetry-breaking field such as a magnetic field should tend to suppress such scattering mechanisms. Of course, such an enhancement would decrease as the temperature decreased below a characteristic temperature as the spins begin to order, eventually resulting in $S \to 0$ as $T \to 0$. Experiments on spin susceptibility in Si:P seem to be inconsistent with the presence of such a scattering mechanism.[40–42] It is interesting to note that recent resistivity and Hall-effect measurements from magnetic tuning experiments[43] on Si:P and Si:As on the metallic side of the transition appear to be similar to the results in the Ge:Sb samples presented in this paper and in Refs. 19–21. The implications of the Si:P susceptibility studies might, therefore, apply to Ge:Sb as well.

So called "single-particle" hopping models fall into two general categories. Standard hopping theories such as Mott-type variable-range hopping[2–4,44] and Miller-Abrahams hopping[2–4,45] ignore the role of interactions.

These models allow the energy distribution of the carriers to fall to zero as $T \rightarrow 0$ and hence result in $S \rightarrow 0$ as $T \rightarrow 0$.[2-4]

Hopping models which do not ignore interactions display quite a different behavior. Efros *et al.*[2-4,46] considered the problem of electron-electron interactions in a system which would otherwise display Mott-type variable-range hopping. A result of this model is that a hop from an occupied state below the Fermi level (E_F) to an unoccupied state above E_F is possibly only if the resulting electron and hole have enough energy to resist recombination. This criteria results in a depression of the density of states (from the noninteracting case) about the Fermi level with the density of states exactly zero at E_F. The calculations of the thermopower in this case indicate this system follows the Mott result at higher temperatures but goes to a nonzero constant as $T \rightarrow 0$.[24,25]

The other major interacting hopping model which may apply to this system is the Hubbard model.[2-4,47-50] The thermopower for this model has been calculated only in the narrow-band limit. Depending on the band filling and interaction energies, a narrow-band Hubbard system may display a variety of thermopower behavior in a manner which is very sensitive to the band filling.[50] The data presented in this paper as a function of fields do display some similarity to the calculations of Ref. 50 (plotted in Fig. 5 of Ref. 50), at a band filling near 0.6 for fixed temperature, if the magnetic field is tuning the near-neighbor interaction energy. However, these calculations are valid only in the limit of zero bandwidth and $T \rightarrow \infty$,[46-49] and imply a gap comparable in size to the near-neighbor Coulomb interaction energy.

We should note that there is one other property of thermopower which applies to all models. The thermopower of any system with electron-hole symmetry is identically zero, and so the above remarks assume only one carrier is dominant.

One can estimate the size of the Coulomb interactions with some rather simple assumptions. The bulk dielectric constant (κ) of Ge is 16, and the Coulomb interactions should be of the Yukawa form:[2-4]

$$V(r) = [e^2/(\kappa r)]\exp(-r/\lambda) ,$$

where λ is the Thomas-Fermi screening length.[2-4] For a system with N_D donor density, the density of states can be assumed to be on the order of N_D/E_F for $\hbar\omega_c \leq E_F$ and on the order of $N_D/\hbar\omega_c$, when $\hbar\omega_c > E_F$, where ω_c is the cyclotron frequency of a carrier. We can take the average spacing of the localized states which form in the high-field state to be approximately the same as the average impurity spacing, a reasonable assumption since the sample maintains charge neutrality on a macroscopic scale. This gives the on-site Coulomb interaction in zero field to be of the order of ~ 79 K, and the near-neighbor interaction energy ~ 15 K and next near neighbor ~ 1 K. As the field increases, the bandwidth changes very little but the decrease in screening increases the on-site energy to ~ 120 K, near-neighbor to ~ 30 K, and next-neighbor energy to ~ 5 K at 12 T. While these estimates are crude, they are probably indicative of the magnitude of the Coulomb interaction energies in this system.[2-4] A simple tight-binding estimate of the bandwidth of the impurity band in this sample gives ~ 100 K, clearly not a narrow-band limit, considering that $k_B T = \sim 1 - \sim 10$ K and the interaction energies estimated above. It is interesting to note that there is little change in the resistivity and thermopower by 12 T for temperatures on the order of the near-neighbor interaction energy, ~ 10 K.

Clearly, Coulomb interactions must play an important role in the insulating state, especially at low temperatures. While the data in Fig. 5 do not specifically rule out the various density-wave states, the most likely candidate of the hopping models mentioned above may be the Efros model. In the Efros model the carriers behave as "electronic polarons."[46] While, to our knowledge the Hall effect has not been specifically calculated for this model, in traditional polaron systems the Hall mobility is activated with temperature, and the density of polarons does not need to change.[2-4]

Recent scaling theories which include both single-particle "localization" terms and the presence of disordered Coulomb interactions address the behavior of the conductivity near the metal-insulator transition in strong magnetic fields. These theories indicate that at the field-induced metal-insulator transition, the quasiparticle density of states remains nonzero, while the quasiparticle diffusion constant vanishes.[51] Unfortunately, however, there appear to be no corresponding calculations for either the Hall effect or thermoelectric effects and hence comparison with the data presented in this paper is not possible.

In conclusion, we have measured the resistivity, Hall effect, and thermoelectric power of degenerately doped n-type Ge in the presence of large magnetic fields. The resistivity and Hall-effect data are consistent with previous reports in Ge:Sb of magnetic-field-induced localization, in which the Hall coefficient changes relatively little, while the magnetoresistance increases sharply. The result of this study is the thermopower in the insulating regime, for constant magnetic field, increases with decreasing temperature. If strong spin-flip scattering is present, this trend in the thermopower will reverse at lower temperatures and the thermopower should decrease. Unless the observed trend changes at lower temperatures, then the third law of thermodynamics indicates the density of states at the Fermi level vanishes creating a gap, probably a "soft" Coulomb gap due to the increase in the strength of the Coulomb interactions as the screening decreases.

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