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行政院國家科學委員會專題研究計畫成果報告

鍺合金系列的非有序導致量子相變研究

Disorder tuned quantum phase transition in Ge-based alloys

計畫編號：NSC 90-2112-M-009-033

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主持人：許世英 國立交通大學電子物理系

一、中文摘要

我們研究一系列不同程度（從弱無序跨越到強無序區域）無序三維樣品的量子干涉效應相關的磁電阻與穿隧電子能階密度；主要系統有二種：銅鍺合金和錫鍺合金。我們利用錫鍺系統作為無序導致的超導性破壞的研究。我們製作一系列的錫鍺樣品，藉由控制熱蒸鍍的熱瓦數來造成其成長結構的不同，因此改變無序程度與相關的超導特質。針對超導態的似電子能階密度、溫度相關的臨界磁場、磁電阻作一系列分析，瞭解無序程度造成的相對物理性質影響，尤其是靠近相變的鄰近區域的行為變化。銅鍺系統則用以作為無序導致的金屬性破壞的研究，我們製作一系列的銅鍺樣品，藉由在燒製時控制鍺與銅的相對莫耳比例來製造所需的無序程度樣品，針對電子波函數受到無序的位障導致的似電子能階密度、溫度相關的電阻率、磁電阻作一系列分析，瞭解無序程度造成的相對影響，尤其是靠近相變的鄰近區域的行為變化。

關鍵詞：量子干涉效應、無序程度、弱-強無序轉變、超導-絕緣態轉變。

Abstract

We have studied the quantum interference effect induced magnetoresistivity and electronic tunneling density of states on thick disordered samples spanning from the weakly and strongly localized regimes. Two systems, CuGe and SnGe alloy systems, have been fabricated in this study. The measurements in SnGe system allowed us to understand the mechanisms of degradation of superconductivity by the presence of

disorder. The thermal evaporation power affected the sample's micro-structure and hence, resulted in different superconducting properties due to different degree of disorder. We have performed a series of systematical analyses in quasi-particle tunneling density of states, temperature dependent critical field, magento-transport. A special attention is on the evolution of those physical properties through the phase change. The measurements in CuGe system allowed us to understand the mechanisms of degradation of metallic behaviors by the presence of disorder. Degree of disorder of the sample can be easily obtained by adjusting the relative molar concentration of Cu to Ge. We have performed a series of systematical analyses in tunneling density of states, temperature dependent electro- and mageto- transports. A special attention is on the evolution of those physical properties through the phase change.

Keywords: quantum interference effects, weak-to-strong localization transition, superconductor-insulator transition, disorder.

二、緣由與目的

Disordered samples have a long history as systems in which new and interesting phenomena can be studied and uncovered. We performed careful experiments on Ge-based disordered samples to elucidate the physical effects behind the profound and sometimes spectacular changes that occur in the strongly disordered limit, $k_F\ell \sim 1$. In this regime, superconductors turn into insulators, single electron system extended states become localized, and the quantum hall states become insulating. Experiments on our samples provide important insights into the mechanisms driving the superconductor-to insulator transition and bring to light a simple scaling dependence of temperature conductance of strongly disordered samples.

The 3-year project entitled “disorder tuned quantum phase transition in Ge-based alloys” supported fully by NSC has resulted in four published and some on-going manuscripts. Six Master students have performed the bulk of it. Through this work we obtained a better quantitative and qualitative understanding of the electronic properties of samples ranging from weakly disordered regime, through transition, to the strongly localized regime.

Studies of magento-transport and tunneling density of states on a series of thick disordered $\text{Cu}_x\text{Ge}_{1-x}$ samples have revealed the fact that backscattering interference effect dominates and disorder enhanced coulomb interaction effect is weak in weakly disordered Cu-rich samples ($x > 0.4$). Magento-transport can be ascribed by the weak anti-localization theory. Scattering rates $1/\tau_{s0}$ and $1/\tau_{in}$ are enhanced by disorder.[1]

Near the weak-to-strong localization transition, coulomb interaction effect becomes more important and a coulomb cusp starts to appear near the Fermi energy at low temperatures. [2,3] As sample becomes much more disorder, the electro-transport demonstrate variable range hopping behavior and magneto-transport can be attributed to quantum interference effects of forward hopping trajectories.

Studies of SnGe systems suggest that the SI transition is brought to occur when intergrain Josephson coupling becomes so weak and intergrain charging energies so strong that quantum fluctuations in phase of order parameter prevent the establishment of long range phase coherence.

三、實驗結果

(I) Weak-to-strong localization transition

3 dimensional $\text{Cu}_x\text{Ge}_{1-x}$ alloys

For Cu-rich samples in the weakly disordered regime, magneto-transport can be described by the weak anti-localization theory and reveals that both spin-orbit scattering rate and inelastic scattering rate are enhanced by disorder. Fits to the weak anti-localization theory for normalized magnetoresistivities, $\Delta\rho(B)/\rho^2(0)$, of samples in the weakly disordered regime ($0.85 > x > 0.4$) allow us to obtain $1/\tau_{\text{so}}$ and $1/\tau_{\text{in}}$. [1] We plot $1/\tau_{\text{so}}$ and $1/\tau_{\text{in}}$ versus the resistivity ρ for numerous samples. As shown, $1/\tau_{\text{so}}$ is bigger than $1/\tau_{\text{in}}$ confirming the anti-localization behaviors. $1/\tau_{\text{so}}$ increases rapidly with increasing resistivity in less disordered samples and seems saturate in slightly more disordered samples. $1/\tau_{\text{in}}$ increases with increasing resistivity, more or less linearly.

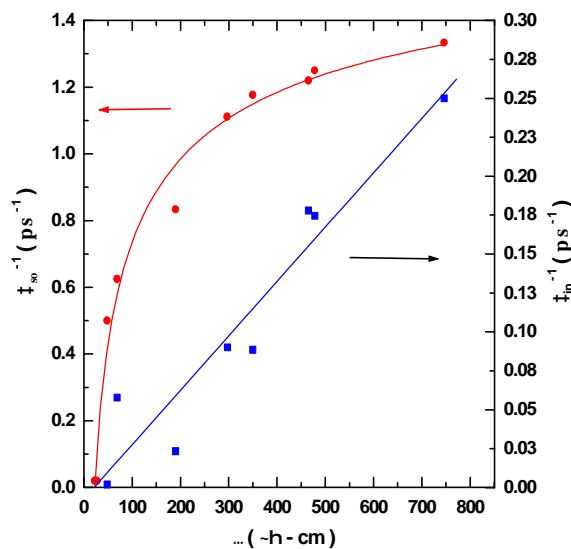


Fig.1. Both spin-orbit scattering rates $1/\tau_{\text{so}}$ ()

and inelastic scattering rates $1/\tau_{in}$ () versus resistivity for numerous CuGe samples at T=5K. The lines are guides to eye.

As x decreases more, magnetoresistivities start to deviate from expectation of the weak anti-localization theory. As shown in figure 2, amplitude, $\Delta\rho(B)/\rho(0)$, at 1.5K increases with increasing disorder in the weakly localized region ($x>0.26$), turns over near the weak-to-strong localization transition, and becomes negative in the strongly localized regime ($x<0.22$).

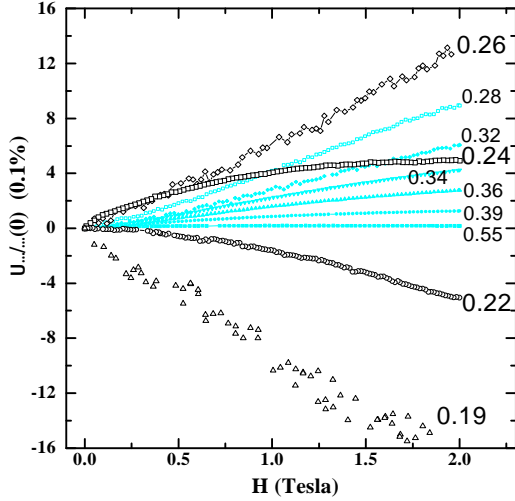


Fig.2. Evolution of $\Delta\rho(B)/\rho(0)$ versus applied magnetic field for numerous Cu_xGe_{1-x} samples ($0.19 < x < 0.6$) at 1.5K.

It has been known that magnetoresistivity in a disordered system is dominated by quantum interference effects and therefore, depends sensitively on whether the electronic states involved in the transport are extended or localized.[4] In the strongly localized regime, conduction electrons are spatially localized and hop from intermediate state to the other. Quantum interference effects of forward hopping trajectories dominate magneto- transport behaviors. Based on the idea, Nguyen et al. predicted the negative MR for strongly localized system.[5] Our data that negative MR and size of MR increases with decreasing temperature is in consistent qualitatively with theoretical prediction.

Electro-transport in the absence of magnetic field for samples in this strongly localization regime follows the expected hopping form, $\rho(T) \propto \exp(1/T^{1/2})$. Figure 3 demonstrates this behavior that can be attributed to the variable range hopping mechanism with the strong electron-electron interaction proposed by Efros and Shklovskii.

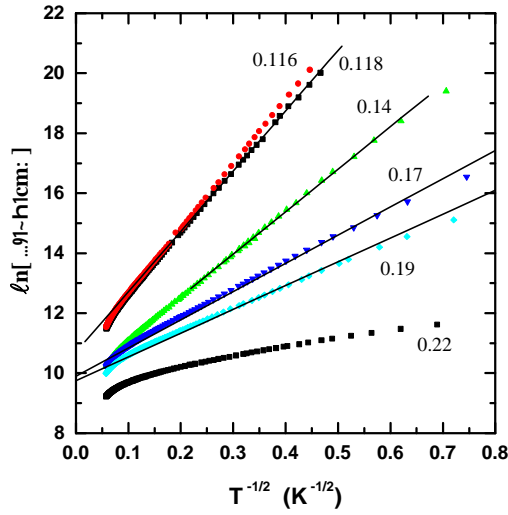


Fig.3. Semi-logarithmic plot of $\ln(\rho(T))$ versus $T^{-1/2}$ for numerous $\text{Cu}_x\text{Ge}_{1-x}$ samples ($0.11 < x < 0.2$) in the absence of magnetic field.

They thought that this Coulomb interaction effect could cause a parabolic depletion of density of states near the Fermi energy. People refer to this opening of density of states as a “soft gap”. Our data fit very well to their model for a very wide temperature range. The slope of linear fit gives information of the characteristic energy $k_B T_0$ relating to strength of Coulomb interaction and corresponding localization length ξ . As the disorder increases, $k_B T_0$ increases and ξ decreases rapidly confirming this hopping properties.[6]

We have performed tunneling measurements in these samples to obtain their tunneling density of states. A Al/ AlO_y strip had been previously deposited serving as counter-electrode and barrier, respectively, for the tunneling junction, Al(20nm)/ $\text{AlO}_y/\text{Cu}_x\text{Ge}_{1-x}$. [7] Since junction conductance $G(V,T)$ is proportional to $N(E)$ of disordered samples and the careful normalization to $G(V,T)$ reveals that the change of $N(E)$ due to the disorder effects.

In figure 4 we plot the normalized junction conductance versus applied bias voltage for some samples in strongly localized regime. $V=0$ refers to the Fermi energy. As shown in figure, clear cusps are present near the Fermi energy for all samples indicating disorder enhanced Coulomb interaction are present. The phenomena are consistent with zero-field electro-transport and magneto-transport.

No singularity is observed at E_F in agreement with earlier data on numerous systems. Thermal effects smear the singularity. For $eV > k_B T$, this effect disappears and $N(E)$ remains temperature independent. The data show that the anomaly grows with increasing disorder. For very strongly localized samples, $N(E)$ have a nearly parabolic

energy dependence as expected by Altshuler and Aronov.[8] The cusp width grows with increasing disorder, the characteristic energy $k_B T$. However, samples with less disorder deviates from this expected behavior and demonstrates a logarithmic dependence of energy instead. This mechanism for this evolution of $N(E)$ is still unknown.

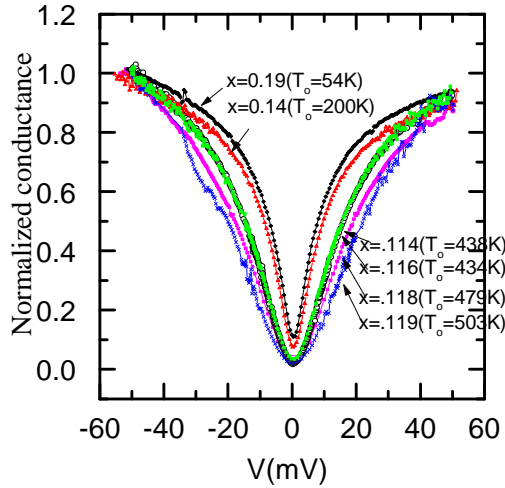


Fig.4 Normalized junction conductance versus applied bias voltage for Cu_xGe_{1-x} samples in the strongly localization regime at $T=5K$.

(II) Superconductor-to-insulator transition

3 dimensional SnGe alloys

We have fabricated a series of SnGe samples. The degree of disorder is sensitive to its micro-structure that can be slightly adjusted by controlling the thermal evaporation power. Systematical analyses in quasi-particle tunneling density of states, temperature dependent critical field, magento-transport allow us to understand the degradation of superconductivity by the presence of disorder.

Figure 5 shows evolution of $\rho(T)$ for numerous SnGe samples span SI transition. This type of evolution has been reported for many different materials and can be qualitatively described in terms of a disordered Josephson junction matrix model.[9]

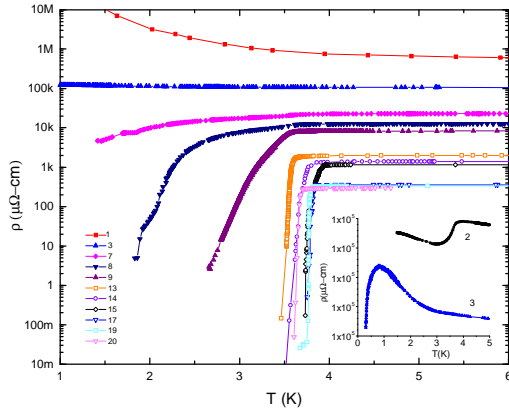


Fig.5 Semi-logarithmic plot of resistivity as a function of temperature for a series of SnGe samples.

At low resistivity sample ($\rho < 1000 \mu\Omega\text{cm}$), the intergrain Josephson coupling energy are large enough that order parameter on the individual grain can lock phases and the sample superconduct. The transition of $\rho(T)$ is sharp. Increases in ρ weaken the the intergrain coupling and make order parameter phase more susceptible to thermal and quantum fluctuations. This leads to a broad transition to superconducting state. ρ drops slowly with decreasing temperature. Eventually ρ becomes great enough that average intergrain coupling is so weak that disorder enables phase fluctuations to prevent the establishment of long range phase coherence. $\rho(T)$ shows only quasireentrant behavior and insulates at $T=0$ (See sample#2 and #3 in the inset). For much more disordered sample such as #1, quasireentrant behavior disappears and ρ increases exponentially with decreasing temperature due to the localization of Cooper pairs.

For sample with less disorder, the superconducting properties are similar to that bulk Sn. The tunneling density of states of sample #20 is plotted in figure 6. Data can be well described by BCS theory. As disorder is increased, the quasi-particle superconducting density of states is still BCS-like. In figure 7 we show tunneling densities of states of four SnGe samples with its transport properties span from weakly disorder to strongly disorder at $T=1.5\text{K}$. It shows the evolution of tunneling density of state with the increase of disorder, sample's resistivity. Symbols are data and the solid line represents BCS density of states with order parameter, $\Delta=0.58 \text{ meV}$, and temperature at 1.5K

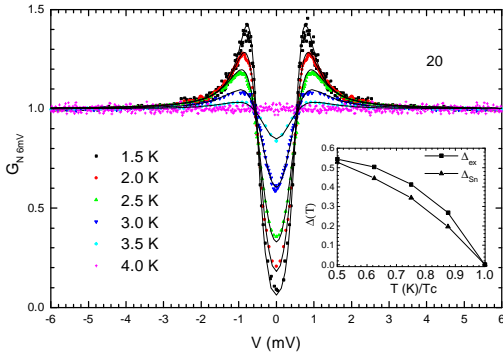


Fig.6 normalized tunneling conductance of Al/AlO_x/SnGe(#20) at different temperatures. Lines are least square fits to BCS density of states. In the inset: amplitude of order parameter obtained from fit to SnGe sample#20 (○) and bulk Sn (□), respectively.

From figure 6 and 7, we can confirm the transport picture of intergrain Josephson junction matrix model. The superconducting properties are robust in each grain even for those strongly disordered samples with hopping transport. A bulk ρ_0 can be obtained in insulating regime of granular SnGe samples. A broaden form of quasiparticle density of states was first observed on granular Sn by White et al. It was suggested that the quasiparticle lifetime effects in granular films modified the superconducting density of states to be

$$N_s = \text{Re} \left\{ \frac{E - i\tilde{\Gamma}}{[(E - i\tilde{\Gamma})^2 - \Delta^2]^{1/2}} \right\}$$

where $\tilde{\Gamma}$ represents the broadening energy near $\tilde{\Delta}$. In their report, $\tilde{\Gamma}$ increases with increasing disorder and eventually becomes comparable to energy gap at the superconductor-insulator transition. This broadening was also observed in 3D granular Al film and strong-coupled superconductor, Pb_{0.9}Bi_{0.1}.

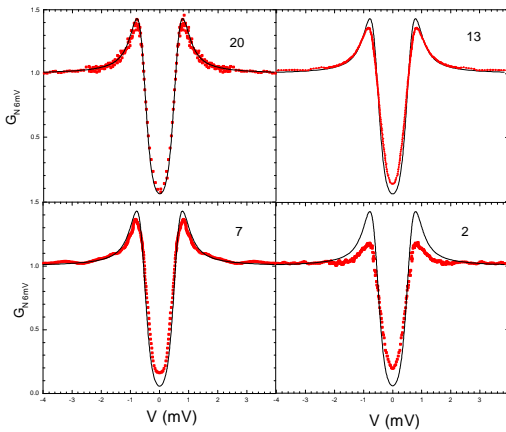


Fig.7, Junction conductance versus bias voltage for sample #20, #13, #7, and #2 at T=1.5K. Lines are BCS fits with $\rho_0=0.58\text{meV}$ for bulk Sn..

We have also performed H_{c2} measurements to understand the influence of

micro-structure to superconducting properties. Discussion above gives us the idea that high resistivity samples are made of isolated grains that have robust bulk superconducting properties. The broad resistive transitions in this regime come from weak Josephson coupling between grains. How does magnetic field affect the broad tail of temperature dependent resistive transition near the SI transition. Figure 8a and 8b demonstrate that the detailed response of $\rho(T)$ depends on magnitude of applied field. Fig.8a is for sample #5 (more disorder) and Fig.8b is for sample #9 (less disorder), respectively. As $H > 6$ Tesla, the superconducting disappear for both samples. The result indicates that the magnetic field tuned SI transition in both samples is driven by pair breaking effects of magnetic field. This pair breaking effects reduce the amplitude of order parameter and the transition temperature of the individual grains of the sample.

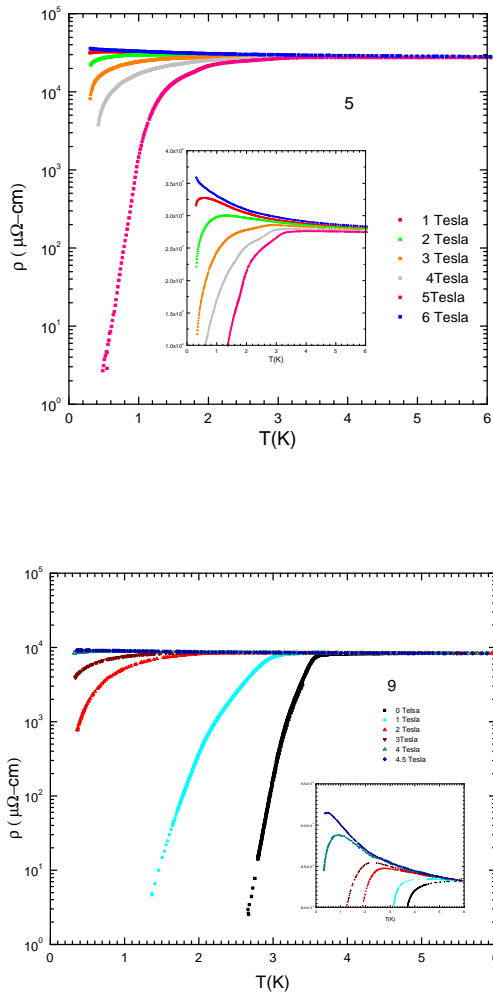


Figure 8. Temperature dependent resistive transitions for sample #5 (a) and sample #9 (b), at different magnetic fields. Inset show the close look around superconducting turn around regime.

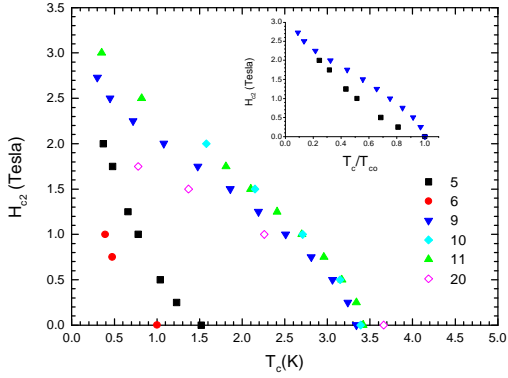


Figure 9. Upper critical field as a function of temperature for numerous SnGe samples.

Figure 9 shows the results for $H_{c2}(T)$ for numerous SnGe samples span from weak to strong disorder. We plot H_{c2} as a function of T/T_{c0} since the zero transition temperature T_{c0} at which resistivity drop to $\rho_N/2$ at zero field depends slightly on ρ_N . Samples near the SI transition show different behaviors of $H_{c2}(T)$ from that of samples in the weakly disordered regime.

3 dimensional CuGe alloys

Moreover, the superconducting properties of CuGe were observed. Earlier studies of electrical transport and tunneling measurements show that weak-to-strong localization transition occurs around $x \sim 0.2$. At much lower temperatures, we found some weakly disordered CuGe samples demonstrate superconducting behaviors.[10] Figure 10 shows the evolution of temperature dependent resistivities for four CuGe samples with $0.38 \leq x \leq 0.67$. The resistive transitions are very sharp for all samples indicating the superconducting of each sample is very robust. We associate this drop with mean-field transition of a superconductor, T_{c0} . For samples with higher concentration of Cu, $x > 0.7$, no superconducting behavior above 0.28K was observed. $T_{c0} = 0.38\text{K}$ for $x = 0.67$ and then, decreases with increasing ρ (decreasing x).

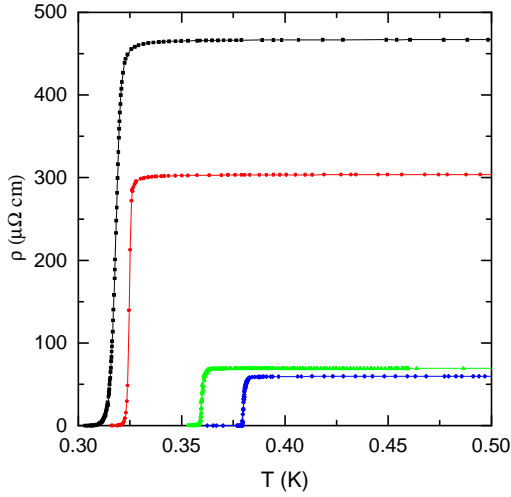


Figure 10. Resistivity ρ versus temperature T for four $\text{Cu}_x\text{Ge}_{1-x}$ samples with $x=0.38$ (), $x=0.45$ (), $x=0.64$ (), and $x=0.67$ ().

For a type II superconductor, upper critical field $H_{c2}(T)$ is linear in temperature near the zero field transition temperature T_{c0} . We plot $H_{c2}(T)$ of these four samples in figure 11. All data seem to fall on lines except in the regime very close to zero field for the highest disordered sample. The slope gives the information of diffusion constant,

$$D = \frac{4k_B}{\delta e} \frac{1}{dH_{c2}/dT}$$

The slight deviation from linear relation near the zero magnetic field for the highest disordered sample may be due to the magnetic field enhanced superconductivity that was observed in Au/Ge films by Seguchi et al.

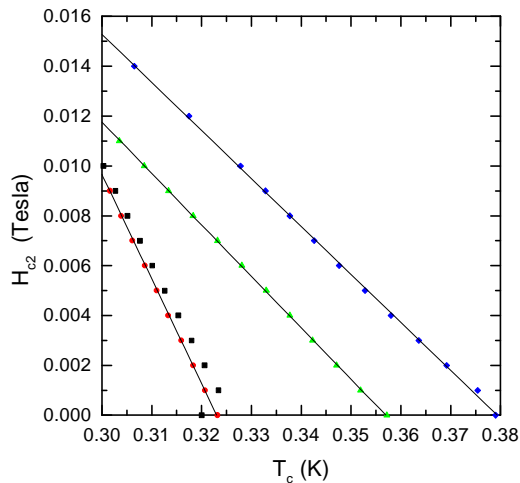


Figure 11. Critical field H_{c2} versus T_c for four $\text{Cu}_x\text{Ge}_{1-x}$ samples. The symbols are experimental results

and lines are linear fits to data.

Superconducting properties were observed in weakly disordered CuGe samples. Disorder enhanced electron-electron interaction effects certainly degrade the superconducting properties. However the anomaly in tunneling density of state for samples in this regime is small and therefore, the combination of spin-orbital scattering and weak localization may be responsible for such superconducting behaviors. That may answer why T_c is higher in both AuGe and AuSb systems than that in our CuGe samples. As disorder increases, enhanced Coulomb interaction grows at the Ge rich samples ($x > 0.7$). The interaction effects certainly break the Cooper pair leading a degradation of superconducting properties in it. [3,4]

四、結論

From the studies of $\text{Cu}_x\text{Ge}_{1-x}$ systems, we know that disorder induced localization and enhanced electron-electron interactions have important influences on transport properties and tunneling density of states. Our data show that the magnetoconductance is positive for samples in strongly localized regime consistent with theoretical predictions of Meir et al. Samples in this regime, electro-transport in the absence of magnetic field is attributed the variable range mechanism with disorder enhanced electron-electron interactions. A clear Coulomb cusp is also observed in these samples. We believe that the disorder enhanced electron-electron interaction effect play a dominant role in the weak-to-strong transition. Samples in weak disorder regime show weak anti-localization behaviors. Scattering rates $1/\tau_{so}$ and $1/\tau_{in}$ increase with increasing disorder.

From the studies of SnGe systems, we know that the granular structure limits the effect of disorder and therefore, superconducting properties of grains such as T_c and Δ are barely changed. The superconductivity of grains is robust. Our results also show that disorder weakens the Josephson couplings between grains by destroying the long range phase coherence, and the coupling strength is decreased. Phase slipping between grains results in a long tail in its temperature dependent resistivities and broadens its quasiparticle tunneling density of states below T_c . Transport properties and tunneling density of states of these granular SnGe samples at low temperatures show that disorder enhanced electron-electron interactions dominates the nature of conduction electrons.

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