Electron-electron interaction in percolative network of polymer - amorphous carbon composites

S. Shekhar, V. Prasad and S. V. Subramanyam

Department of Physics, Indian Institute of Science, Bangalore, 560 012 India.

Abstract

The polymer-amorphous carbon composites show a negative magnetoconductance which varies as $B^2$ at low fields which changes to $B^{1/2}$ at sufficiently high fields. The magnetoconductance gives the evidence of electron-electron interaction in composites whose conductivity follows thermal fluctuation induced tunneling and falls in the critical regime.

Key words: Magnetoresistance; Electron-electron interaction; Transport properties

PACS: 72.80.Tm; 72.20.Mf; 72.15.Gd

1 Introduction

The effect of weak localization (WL) and electron-electron interaction in two dimensional as well as in three dimensional systems are well studied and documented [1,2]. These two effects individually substantially alter the transport and magnetotransport behavior of the disordered systems. The weak localization effect has been observed in wide range of disordered materials including metallic glasses, granular metals, semiconductors and amorphous alloys [4–6]. In many of the systems such as copper films, ion implanted polymers, conducting polymers and disordered carbon electron-electron interaction effect have been realized [11–13,21]. It is very natural to expect the WL and e-e effects simultaneously affect the transport properties. Their combined effects have been reflected in many disordered systems [7,15,16]. The effect of WL and e-e interaction are observed in percolating network, too [7,8]. In this letter we are reporting the result of magnetoconductance extensively carried out on a percolative network of polymer-carbon composites in the temperature range 1.3K to 50K.
2 Theoretical Background

The weak localization effect is often associated with spin-orbit scattering and in case it is significant it changes MC drastically. In three dimensional system the MC due to combined effect of weak localization along with spin-orbit contribution is given \cite{3,4} as:

\[
\Delta \sigma = [\sigma(B) - \sigma(0)] = \alpha \frac{e^2}{2\pi^2\hbar} \frac{eB}{\hbar} \frac{2}{2} \frac{3}{2} f_3 \left( \frac{B}{B_{so}} \right) - \left( \frac{1}{2} + \beta f_3 \left( \frac{B}{B_i} \right) \right) \tag{1}
\]

where \(B_i = \hbar/4eD\tau_i\) and \(B_{so} = \hbar/4eD(\tau_i^{-1} + 2\tau_{so}^{-1})\); D, \(\tau_i\) and \(\tau_{so}\) are electron diffusion coefficient, inelastic collision induced relaxation rate and spin-orbit scattering relaxation rate respectively. The function \(f_3(x)\) is given by

\[
f_3(x) = \begin{cases} 
\frac{x^{3/2}}{0.065}, & x \ll 1 \\
0.605, & x \gg 1
\end{cases}
\tag{2}
\]

The term \(\beta\) (known as Maki-Thompson contribution) in eq.1 is the contribution from the quenching of superconducting fluctuation due to electron pairing in Cooper channel and effective at temperatures far away from \(T_c\). In non superconducting materials this effect can be safely neglected. The MC depends on the relative strength of inelastic relaxation rate \(\tau_i^{-1}\) and spin orbit scattering relaxation rate \(\tau_{so}^{-1}\). When there is very weak or no spin orbit scattering (i.e. \(\tau_{so}^{-1} \ll \tau_i^{-1}\)) the eq.1 reduces to the case of pure weak localization effect and one gets positive magnetoconductance which is due to dephasing of back scattered charge carriers. In opposite limit when spin-orbit scattering is stronger \((\tau_{so}^{-1} \gg \tau_i^{-1} \Rightarrow B_{so} \gg B_i)\) eqn.1 gives

\[
\Delta \sigma \simeq -\alpha \frac{e^2}{192\pi^2\hbar} \left( \frac{e}{\hbar} \right)^{1/2} \frac{B^2}{B_i^{3/2}} \quad (B \ll B_i) \tag{3}
\]

\[
\Delta \sigma \simeq -\frac{0.605e^2}{4\pi^2\hbar} \left( \frac{eB}{\hbar} \right)^{1/2} \quad (B_i \ll B \ll B_{so}) \tag{4}
\]

\[
\Delta \sigma \simeq +\frac{0.605e^2}{2\pi^2\hbar} \left( \frac{eB}{\hbar} \right)^{1/2} \quad (B \gg B_{so}) \tag{5}
\]

The electron-electron interaction effects too, produce positive MR and the type of dependence mentioned above. The exact dependence of MC on field in electron electron interaction is written as

\[
\Delta \sigma = -\frac{e^2F}{4\pi^2\hbar} \left( \frac{kT}{2\hbar D} \right)^{1/2} g_3 \left( \frac{g\mu_B B}{kT} \right) \tag{6}
\]
where F is the interaction constant and the function $g_3$ in low field and high field limit is given by

$$g_3(h) \simeq 0.084h^2 \quad (h \ll 1) \quad (7)$$

$$g_3(h) \simeq \sqrt{h} - 1.3 \quad (h \gg 1) \quad (8)$$

Eq. 7 and 8 define the limit on the MC dependence as $B^2$ or $B^{1/2}$.

3 Experimental

The composites have been prepared by mixing amorphous carbon (a-C) flake in different proportion to Poly(vinyl chloride) with the help of magnetic stirrer [9]. The composites are disordered in nature. The composites filled with higher concentration of a-C are more conducting. The details have been discussed in reference [9]. The magnetoconductance has been measured by conventional (dc) four probe technique inside a commercial bath type cryostat (Janis) with superconducting magnet with maximum attainable field of 12T. The magnetoconductance of the composites having a-C content higher than percolation threshold (conducting samples) have been measured at different temperatures varying from 1.3K-50K. The fluctuation in temperature was less than 0.002K below 10K whereas above 10K the fluctuation was 0.01K. The value of MC does not vary much from sample to sample but the higher resistive samples show slightly lower magnetoresistance (Higher MC). The change in the direction of field has no effect on the magnetotransport properties. Above 25K a negligibly small magnetoresistance (MR) is observed.

4 Results and Discussion

The temperature dependence of resistivity for disordered metals at low temperatures due to electron-electron interaction-weak localization (ee-WL) model follows [10,12]

$$\sigma(T) = \sigma_0 + \alpha T^{1/2} + \beta T^{p/2} \quad (9)$$

The first term is a constant related to residual resistivity at $T=0$, the second term arises due to electron-electron interaction and third term takes care of weak localization effect on transport properties. The interaction effect is dominant at low temperatures and WL is effective relatively at higher temperatures. But electron-electron interaction and WL model are applicable to
Fig. 1. Low temperature normalized magnetoconductance \( \frac{\sigma(B) - \sigma(0)}{\sigma(0)} \times 100 \) of polymer composites disordered metals only. In those materials \( \rho_{1.3}/\rho_{300} \) is slightly greater than 1. In polymer composites the given ratio is nearly 2 for 33% of loading and increases gradually to 6 for 6.4% which is more towards the critical regime (near to metal insulator boundary) of insulating side. Moreover fits of eq.9 were very unphysical. The ratio \( \rho_{1.3}/\rho_{300} \) suggests that the samples are in critical regime. In the critical regime the conductivity follows [14]

\[
\sigma(T) \sim \frac{e^2 p_F}{\hbar^2} \left( \frac{T}{E_F} \right)^\beta
\]

where \( P_F \) is the Fermi momentum and \( e \) is the electronic charge. The \( \beta \) lies in the range \( 1/3 < \beta < 1 \). Again the fit was found to be extremely poor for all the samples for the specified value of \( \beta \).

The composites follow thermal fluctuation induced tunneling (Sheng’s model [17])

\[
\rho = \rho_0 \exp\left[\frac{T_1}{(T_0 + T)}\right]
\]

at temperatures below 50K-60K [9]. This theory takes care of large size of the particles which forms a junction with insulating material. In fact in the cases
Fig. 2. Normalized Magnetoconductance ($\frac{\sigma(B)-\sigma(0)}{\sigma(0)}\%$) of polymer composites in temperature range 10K-25K.

Table 1
Magnetic field dependence range of composites. The variation in the range is ±0.1 from sample to sample.

<table>
<thead>
<tr>
<th></th>
<th>1.3K</th>
<th>2.5K</th>
<th>4.2K</th>
<th>6K</th>
<th>8K</th>
<th>&gt;10K</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B^2$</td>
<td>$&lt; 1.4T$</td>
<td>$&lt; 3.1T$</td>
<td>$&lt; 4.8T$</td>
<td>$&lt; 7.4T$</td>
<td>$&lt; 10.9T$</td>
<td>12T and high</td>
</tr>
<tr>
<td>$B^{1/2}$</td>
<td>$&gt; 2.35T$</td>
<td>$&gt; 3.3T$</td>
<td>$&gt; 5.8T$</td>
<td>$&gt; 8.1T$</td>
<td>$\gg 11T$</td>
<td>$\gg 12T$</td>
</tr>
</tbody>
</table>

where filler particle size is larger experimental results follow this conduction mechanism [18,19]. Here $T_1$ and $T_0$ are energy required for charge carriers to tunnel and low temperature, temperature independent resistivity respectively[17]. Near to 250K the resistivity of composites exhibit a minimum (as clearly seen in fig.3). The difference in thermal expansion coefficient of PVC and a-C are responsible for this behavior. As temperature increases the matrix expands and the separation between a-C particles increase resulting in increase of resistance at higher temperature. Not much have been studied about the effect of magnetic field on the Sheng’s model. In this letter we have investigated the effect of magnetic field on Sheng’s conduction mechanism.

$\Delta \sigma$ varies as $B^2$ at lower fields. As the temperature increases the $B^2$ dependence extends to a larger field. Above 10K in entire 12T range $B^2$ dependence is observed. At sufficiently low temperature and high field $\Delta \sigma$ makes a tran-
Fig. 3. Plot shows temperature dependence of a-C composites in temperature range 1.3K-300K. In the temperature range 1.3K-50K the composites follow thermal fluctuation induced tunneling conduction mechanism [9].

Fig. 4. Magnetoconductance \( MC = \frac{\sigma(B) - \sigma(0)}{\sigma(0)} \) of composites in both positive and negative field. No appreciable change in MC is observed.

Transition from \( B^2 \) to \( B^{1/2} \) dependence as shown in the fig.5. The range of \( B^2 \) and \( B^{1/2} \) dependence are depicted in Table 1. This kind of behavior is observed in weak localization with spin-orbit interaction or electron-electron (e-e) interaction model. Neither PVC nor a-C contain heavy elements, so the effect of spin-orbit scattering is negligible. Moreover in weak localization model \( \Delta \rho / \rho \propto \rho \) so the higher resistive samples show higher MR (lower magnetoconductance) [20]. The behavior of magnetoconductance observed here is in well agreement with the electron-electron interaction model. Here the MR decreases as one goes to higher temperatures suggested by electron-electron interaction model. In e-e model the \( B^2 \) dependence should be observed in the limit given by eq.7 \( (g\mu_B B/\kappa T \ll 1) \). So at very low temperatures only in very low fields \( B^2 \).
dependence should be observed where as at high temperatures at very high field B^2 dependence should be observed. Similar kind of behavior is observed in composites. Above 10K in entire 12T range B^2 dependence is observed. In high field limit (gμBB/κT ≫ 1, i.e. at low temperature and high field) at low temperatures B^1/2 dependence has been noticed. So the e-e theory is precisely explaining the magnetoconductance behavior in the composites except for the fact that the low temperature transport does not exactly follow the T^1/2 dependence (e-e model). In critical regime the different mechanism for transport and magnetotransport has been observed earlier also [21]. It is worth to mention here that the a-C too (the filler), shows the e-e interaction effects both in terms of transport and magnetotransport properties [11]. But the MR values is much small compared to composites and moreover the MR of the system not necessarily depends on its constituents.

5 Conclusions

The negative magnetoconductance has been observed in polymer- amorphous carbon composites. At low fields the MC dependence on field is B^2 which changes to B^1/2 at higher fields. The range of low field and high field limits strongly depend on temperature. The MC behavior indicates the presence of electron-electron interaction effects. The transport properties of the composites at low temperatures are governed by thermal fluctuation induced tunneling (Sheng’s model). It is very interesting to observe the different mechanism
for transport properties and magnetotransport properties in critical regime. Besides that the effect of magnetic field in Sheng’s model has been studied extensively for which a theoretical model is not well established.

Acknowledgments: We would like to thank the DST, New Delhi for providing the National High Magnetic Field Facility where this work has been carried out. One of us (S.S.) expresses his gratitude to Madan Mithra for some useful discussions and providing several related references.

References


