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Kinetic Properties of Current Carriers in GICs and Low Density Carbon Materials

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The Hall effect, Shubnikov-de Haas effect (SdH), and electrical conductivity have been studied in the temperature range 0.3–4.2 K in magnetic fields up to 8 T in acceptor type graphite intercalated compounds (GICs) of low stages and in low density carbon materials (graphite foils), produced by the thermal destruction of GICs. The parameters of the energy spectrum of carriers were determined. For graphite foils (GFs) the negative magnetoresistance and logarithmic dependence of the electrical conductivity on temperature was found at low temperatures. These data testify to weak localization beyond the diffusion limit.

Keywords: intercalation compounds of graphite; graphite foils; electronic properties; the energy spectrum; weak localization

INTRODUCTION

GICs have a very high electrical conductivity under normal conditions ^[1]. The electrical conductivity and physical properties of GICs depend on intercalate species, stage number, synthesis method etc. The study of the energy spectrum of GICs is important for explaining of the nature of their high electrical conductivity and the search of some new GICs. Compounds produced on the base of singlecrystalline graphite are quite perfect singlecrystals, that allows to observe the oscillations of transverse magnetoresistance (SdH effect). The extremal cross-sections of Fermi surface, effective masses of carriers and other

parameters of the energy spectrum are evaluated from SdH effect. Taking into account the Hall effect data in the frame of the band model ^[2] the concentration of carriers for these compounds was determined in the paper ^[3].

EXPERIMENTAL

GICs were produced on the basis of HOPG by the vapor phase method in a two-zone ampoule or by the liquid phase method. The SdH and Hall effects at liquid helium temperatures have been studied in the temperature range 0.3–4.2 K in magnetic fields up to 8 T. The electrical conductivity of GICs was measured by a four-contacts method and by a contactless inductive method using a frequency of $\approx 10^5$ Hz.

For producing GFs the GICs samples were hydrolyzed, and dried at a temperature of $\approx 110^\circ$ C. Then the samples were sharply heated at a temperature $\geq 950^\circ$ C and were rolled without a binder between cylinders. Finally GFs with different densities from 0.8 to 1.2 g/cm³ were produced.

RESULTS AND DISCUSSION

Values of the electrical conductivity in the basal plane of GICs (σ_a) at room temperature, the hole concentrations (p) calculated from quantum oscillations in the transverse magnetoresistance and Hall effect, and the mean free path of carriers (Λ), calculated from the electrical conductivity and quantum oscillations are presented in table 1.

TABLE 1. The characteristics of carriers for acceptor type GICs, N-stage number

Compound	N	$\sigma_a, 10^7$ $\text{Ohm}^{-1} \cdot \text{m}^{-1}$	$p, 10^{26} \text{ m}^{-3}$	$\Lambda, 10^3 \text{ E}$
$\text{C}_{16.5 \pm 0.5} \text{ICl}_{1.07 \pm 0.03}$	2	2.8 ± 0.6	2.7	3.7
$\text{C}_{24.8 \pm 0.5} \text{ICl}_{1.06 \pm 0.05}$	3	4.4 ± 0.6	3.7	5.9
$\text{C}_{32.8 \pm 0.5} \text{ICl}_{1.06 \pm 0.05}$	4	1.3 ± 0.3	0.7	3.8
$\text{C}_{27.5 \pm 0.5} \text{ICl}_{3.0 \pm 0.1}$	2	2.5 ± 0.3	2.8	3.5
$\text{C}_{9.5 \pm 0.2} \text{AlCl}_3 \text{Br}_{0.6 \pm 0.05}$	1	2.7 ± 0.2	3.6	3.1
$\text{C}_{9.8 \pm 0.1} \text{CuCl}_{2.05 \pm 0.02}$	2	1.2 ± 0.1	1.3	2.7

In fig. 1 the dependence of the oscillatory part of transverse magnetoresistance $\tilde{\rho}$ on magnetic field for some GICs is shown.

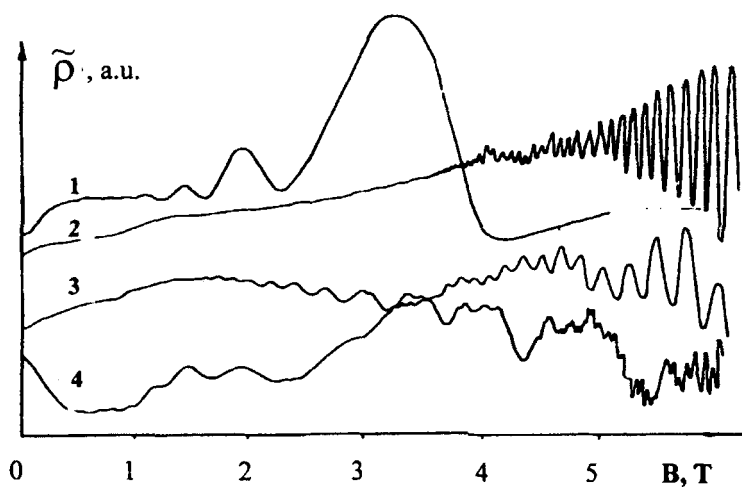


FIGURE 1. Dependence of oscillatory part of transverse magnetoresistance $\tilde{\rho}$ on magnetic field B: 1 – graphite; 2 – $\text{C}_{16.5} \text{ICl}_{1.1}$; 3 – $\text{C}_{24.8} \text{ICl}_{1.1}$; 4 – $\text{C}_{32.8} \text{ICl}_{1.1}$.

Galvanomagnetic and SdH effect measurements give a possibility to obtain the

dependence of the electrical conductivity σ_a not only on stoichiometric composition (stage number N) but also on carrier concentration p . We found that the value of σ_a increases with p . The value of σ_a for GICs increases about 5–20 times as compared with pristine graphite and becomes comparable with the electrical conductivity of normal metals. For example, in the compound $C_{24.8}ICl_{1.06}$ the value of σ_a is about 80 % of the electrical conductivity of copper under normal conditions. When the temperature decreases, the electrical resistance ρ of the GICs decreases linearly with temperature above 70 K. For temperatures below 70 K $\rho(T)$ obeys a power law $\rho \sim T^\alpha$, where α is about 3–4. It should be mentioned that in the general case the resistance of GICs in the basal plane measured by a four-contact method (ρ_a^K) is higher than (ρ_a^N) measured on the same samples with contactless inductive method. The main contribution to the resistance in DC measurements is due to macrodefects of block quasi-single crystals of GICs and the regions where the current flows are along the trigonal axes. Due to this reason the correct value of ρ_a^K may be obtained only on samples with a perfect structure. The coincidence of ρ_a^K with ρ_a^N on the same sample may be used as correctness criteria of such measurements.

The magnetotransport properties of a series of GFs have been studied at low temperatures ($0.4 \text{ K} < T < 4.2 \text{ K}$) and in magnetic fields up to 8 T. All foils show the main features of weak localization: a logarithmic dependence of resistance on temperature ($T < 2.5 \text{ K}$) and negative magnetoresistance in low magnetic fields ($B < 0.5 \text{ T}$). The negative magnetoresistance has been explained by the theory of quantum corrections to the conductivity for the 2D case [4]. The data were analyzed within the model of Wittmann and Schmid for weak localization beyond the diffusion limit [5].

In fig. 2 the dependence of the magnetoresistance of GFs on magnetic field at different temperatures is presented.

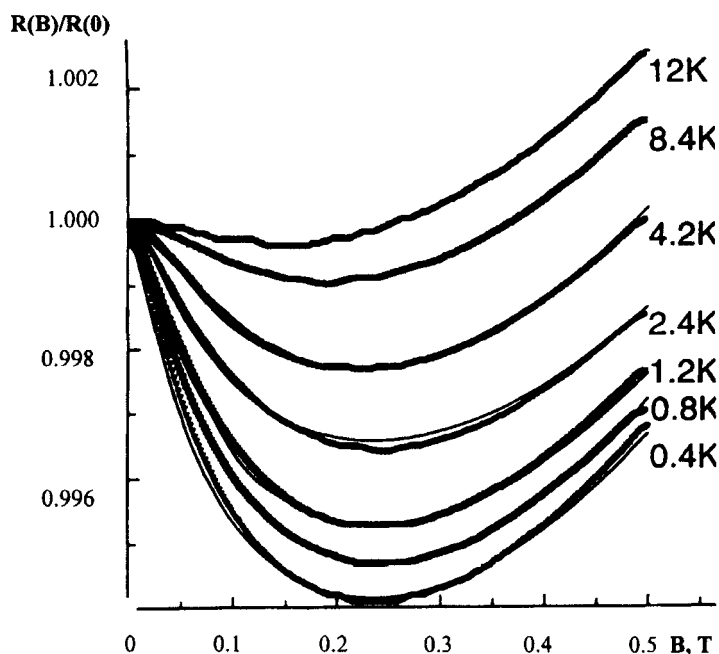


FIGURE 2. Dependence of the magnetoresistance of GFs on magnetic field at different temperatures.

The effect of structural differences in the foils on the negative magnetoresistance was investigated by varying the density and the temperature at which the samples were heat treated. The negative magnetoresistance did not change significantly with the density, indicating that the intergrain scattering process plays a minor role. X-ray analysis of the exfoliated graphite samples revealed that the number of turbostratic layers is small. The structural disorder induced by the turbostratic layers is the most likely cause of the weak localization. This is established by annealing the exfoliated graphite foils at $T=2800^{\circ}\text{C}$: the negative magnetoresistance becomes significantly smaller, while the disorder in the stacking sequence decreases.

CONCLUSIONS

It is established that the high electrical conductivity of acceptor type GICs at room temperature is caused not only by an increase the carrier density in the 2D carbon layers as compared with pristine graphite, but also by the reconstruction of the phonon spectra of GICs. That leads to the relation $\Lambda_{\text{GIC}} > \Lambda_{\text{graph}}$, i. e., the energy spectrum of GICs becomes more two-dimensional. The negative magnetoresistance in low magnetic fields and a logarithmic dependence of the resistance on temperature for GFs were found. The negative magnetoresistance is explained by the theory of quantum corrections to the conductivity for the 2D case. The analysis of the magnetoresistance yields the relaxation time of the phase of the wave function of the electron and its temperature dependence. The weak localization is attributed to disorder in the stacking sequence of the graphene layers.

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