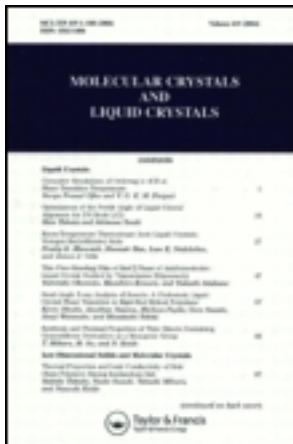


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Magnetoresistance of Metallic Polyacetylene: Variable Range Hopping in a Magnetic Field

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MAGNETORESISTANCE OF METALLIC POLYACETYLENE: VARIABLE RANGE HOPPING IN A MAGNETIC FIELD

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Abstract The magnetoresistance (MR) of heavily doped $(CH)_x$ has been measured between 4.2 K and 0.3 K in fields up to 3.4 Tesla. The data can be described by variable-range hopping (VRH) in a magnetic field and by postulating a second transport mechanism independent of the magnetic field. A model based on orbital shrinking of the localized wavefunction (positive MR) and on a Zeeman shift of the energy levels (negative MR) can be fit to the data and yields the Bohr radius and the binding energy of the localized states as well as the variation of the density of states at the Fermi level, which in most samples is found to be quadratic.

We have studied the dc conductivity of heavily doped polyacetylene (I^-_3, AsF_6^-) at temperatures between 4.2 K and 0.3 K and in magnetic fields up to 3.4 Tesla. From the MR in particular it is possible to obtain detailed information on the transport mechanism.

Our samples were polymerized by the Stuttgart group. Iodine doping was performed in Regensburg by exposing the samples to iodine vapor and consecutive vacuum pumping. The magnetic field was oriented parallel to the polyacetylene films and to the current.

Typical results are shown in Figs. 1 and 2 where the relative change of the resistance as a function of the magnetic field is displayed at various constant temperatures. At high temperatures (Fig. 1) the MR is negative but upon cool-down continuously changes to positive values (Fig. 2). At the lowest temperatures the positive MR is similar to our earlier data on less metallic samples¹, the differences being the two orders of magnitude smaller values

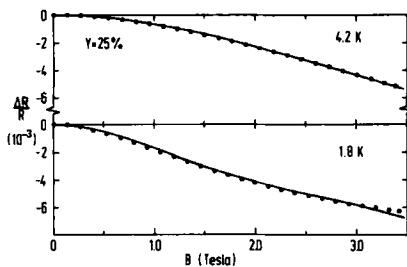


Fig. 1 Negative MR of $(\text{CHI}_{0.25})$ at high temperatures. The solid lines are fits of Eq.(1).

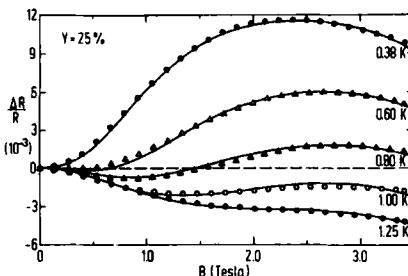


Fig. 2 Change from negative to positive MR at lower temperature (same sample as in Fig. 1). The solid lines are fits of Eq.(1).

and the maximum near 2.5 Tesla instead of a monotonic increase.

In accordance with the interpretation of our previous work we attribute the positive part of the MR to orbital shrinking² of the hole wavefunction resulting in a reduction of the conductivity by the factor $\exp(-B/B_0)^2$ in moderate fields or $\exp(-B/B_1)^{1/4}$ in high fields³. The characteristic fields B_0 and B_1 have particular temperature dependencies, which are determined by the variation of the density of states N at the Fermi level E_F (see Table I).

For intermediate fields we use an empirical interpolation formula $\exp(-f(B))$ where $f(B) = B^2/(B_0^2 + B_1^{1/4} B^{7/4})$. Hence the VRH conductivity in a magnetic field is given by

$$\sigma_{\text{VRH}}(B) = \sigma_{\text{VRH}}(0) \cdot \exp(-f(B)).$$

The negative part of the MR can be described by a Zeeman shift of the energy levels of the hole wavefunction, which leads to a spin dependent Bohr radius⁴. The overall effect is an increase of the conductivity. In the limit of a Zeeman shift being much smaller than the binding energy one finds⁴ the factor $\text{ch}(B/B_2)$, where the temperature dependence of the characteristic field B_2 again depends upon the density of states (see Table I).

Taking into account both contributions we write

$$\sigma_{VRH}(B) = \sigma_{VRH}(0) \cdot \exp(-f(B)) \cdot \text{ch}(B/B_2).$$

In addition, we postulate a second transport mechanism σ_2 being insensitive to a magnetic field, which in the limit of low temperatures and high fields shortens out the vanishing VRH and thus accounts for the saturation tendency of our data in this regime: $\sigma(B) = \sigma_{VRH}(B) + \sigma_2$. From the total conductivity we finally obtain for the MR

$$\frac{\Delta R}{R} = c \cdot (1 - \exp(-f(B)) \cdot \text{ch}(B/B_2)), \quad (1)$$

with $c = \sigma_{VRH}(0) / (\sigma_{VRH}(0) + \sigma_2)$.

The solid lines in Figs. 1 and 2 are fits of equation (1) to the data. Fitting parameters are c , the temperature independent coefficients of the characteristic fields B_i ($i = 0, 1, 2$), and the power law which determines their temperature dependence.

The results for two of our samples (8% I_3^- and 11% AsF_6^-) are compiled in Table I together with a listing of theoretical predictions for purely constant and quadratic density of states N .

Table I Temperature dependence of the characteristic fields

Characteristic fields	Theory $N = \text{const.}$	Theory $N = N_0 (E - E_F)^2$	Experiment AsF_6^-	Experiment I_3^-
B_0	$T^{3/8}$	$T^{3/4}$	$T^{0.47}$	$T^{0.65}$
B_1	$T^{2/3}$	T^2	$T^{0.84}$	$T^{1.73}$
B_2	$T^{1/4}$	$T^{1/2}$	$T^{0.31}$	$T^{0.43}$

For the iodine doped sample we find the following values at 1 K:

$B_0 = 1.1$ Tesla, $B_1 = 1.0 \cdot 10^{-3}$ Tesla, and $B_2 = 0.69$ Tesla.

A comparison between theory and experiment in Table I shows that for the iodine doped sample the temperature dependences of

the B_i ($i = 0, 1, 2$) indicate a parabolic density of states at the Fermi level as observed earlier¹ in contrast to the AsF_6^- doped sample, where N seems to be nearly constant. At present it is unclear whether the differences in the density of states are caused by the different dopants or by some other properties of the various samples.

From the coefficients of the three characteristic fields one can in principle derive the following three microscopic quantities: 1. The Bohr radius a of the hole, 2. its binding energy ΔE , and 3. the factor N_0 of the density of states. This, however, requires the knowledge of numerical factors which theories of VRH leave uncertain to within an order of magnitude. Therefore, we obtain only estimates as follows (for the iodine doped sample): $a \approx 100 \text{ \AA}$, $\Delta E \approx 2 \text{ K}$, $N_0 \approx 10^{26} (\text{eV cm})^{-3}$. These values do not appear to be unreasonable.

In summary, the MR of heavily doped polyacetylene can be described by 3D variable-range hopping of localized states with spin. Further work is in progress to determine the influence of the dopant concentration on the properties of the hole. Even more interesting would be an understanding of the nature of the σ_2 conduction mechanism.

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