Effects of pressure and magnetic field on the low-temperature conductivity of $FeCl_4^-$-doped polyacetylene: The influence of scattering by low-energy excitations

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The effects of hydrostatic pressure and magnetic field on the low-temperature conductivity of oriented polyacetylene doped with $FeCl_4^-$ up to a metallic state have been investigated. It was found that the conductivity at 10 kbar is greater than that at ambient pressure by a factor of 1.3. Application of pressure suppresses the resistivity minimum at 280 K and decreases the resistivity ratio $\rho_r = \rho(0.37\ \mathrm{K})/\rho(300\ \mathrm{K})$ from 2.4 down to 1.9. The temperature dependence of resistivity $\rho(T) \sim \ln T$ at temperatures below 1 K at ambient pressure and at 10 kbar, which remains almost unaltered by a magnetic field up to 14 T. The starting temperature of the logarithmic temperature dependence shifts by a magnetic field up to higher temperatures. Transverse magnetoresistance (MR) was found to be negative, linear, and almost temperature-independent at temperatures below 2 K. The low temperature $\rho(T)$ and MR behavior at $T<1$ K observed in heavily doped polyacetylene has been attributed to weak localization. We assumed that a dramatic increase of inelastic scattering due to low-energy vibrational excitation can ascribe the stronger temperature behavior of $\rho(T)$ and MR at $T>2$ K as a result of further suppression of weak localization due to a more effective dephasing effect. At higher temperature, the resistivity decrease is dominated by activation to additional conduction paths.

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I. INTRODUCTION

The electrical-conductivity of heavily doped polyacetylene (PA) has been studied intensively during recent years.[1,2] In particular, more attention has been paid to transport properties of $FeCl_4^-$-doped and ClO_4^--doped PA at very low temperatures.[3–5] These studies have resulted in revealing the ClO_4^--doped PA with a positive temperature coefficient of resistivity (TCR) over an entire temperature range 300–1.5 K.[6,7] From another side, the application of high hydrostatic pressure can increase and control the conductivity of anisotropic, highly doped conducting polymers with relatively weak interchain interactions by tuning the interchain overlap of the π electrons. The effects of high pressure and high magnetic fields on the temperature dependence of conductivity have been studied intensively for oriented PA films doped with iodine[8–10] and potassium,[11] as well as for some other highly doped polymers.[12–15]

In all the above cases, the pressure enhanced interchain interactions and thus induced a metallic or more pronounced metallic temperature dependence of the conductivity, whereas the magnetoresistance (MR) behavior was found to be more complicated, and the subtle interplay between positive and negative MR components has been found in particular for doped PA at low temperature.[9,10] The obtained results have been explained in the framework of various theoretical models, including the interaction-localization model[16] and the extended heterogeneous model.[17] However, an apparent disagreement of experimental data obtained for highly doped conjugated polymers with both of these theories has been found at temperatures below 1 K.[5,18,19] Thus an exact nature of charge-carrier transport in highly doped polymers at very low temperatures remains a subject of intensive discussion.

Recently, the influence of strong inelastic scattering by low-energy vibrational excitations on the low-temperature conductivity of highly doped conjugated polymers has been proposed.[19] These excitations related to an initial chemical structure of conducting polymers, and thus they could affect the charge-carrier transport at temperature ~ 1 K. The specific feature of these excitations is their great sensitivity to external pressure, as shown by the measurements of heat capacity, thermal conductivity, optical spectra, etc. (see, e.g., Ref. 20).

At present, there are no experimental data regarding the pressure-induced conductivity and MR in $FeCl_4^-$-doped PA at temperatures down to mK; moreover, there is no direct experimental evidence of the influence of hydrostatic pressure on the low-energy vibrational excitations in highly doped polymers at low temperature. These studies could provide additional information for further theoretical efforts in this area.

In this paper, we present the details of charge-carrier transport of oriented polyacetylene films doped with $FeCl_4^-$ up to a metallic state under hydrostatic pressure up to 14 kbar and at magnetic field up to 14 T at the temperature region down to 370 mK.

We have found that the conductivity of oriented polyacetylene doped with $FeCl_4^-$ at 10 kbar is greater than that at ambient pressure by a factor of 1.3. Application of pressure decreases the resistivity ratio $\rho_r = \rho(0.37\ \mathrm{K})/\rho(300\ \mathrm{K})$ from 2.4 down to 1.9. At $T < 1$ K, $\rho(T) \sim \ln T$ at ambient pressure and at 10 kbar. This behavior remains almost unaltered by a magnetic field up to 14 T. The MR was found to be negative, linear, and almost temperature-independent at temperatures below 2 K. We attributed the $\rho(T)$ and MR behavior observed in heavily doped polyacetylene at $T < 2$ K to a weak localization effect. At $T > 2$ K, the increase of inelastic scattering due to low-energy vibrational excitation can explain the further stronger temperature behavior of $\rho(T)$ and MR as a result of further suppression of weak localization due to a more effective dephasing effect. At higher temperature, the resistivity decrease is dominated by activation to additional conduction paths.

II. EXPERIMENT

The Naarmann-type polyacetylene films were synthesized by the modified Shirakawa method.[21] The samples were oriented by stretching with $l/l_0 = 3$–4. The $FeCl_4^-$ doping was done from solution by analogy with earlier publications.[6,7] The doping concentration was determined by measuring the weight of the sample after the films were doped to a saturation level equal to 7–8 wt. %.

The dc conductivity measurements were carried out using collinear four-probe geometry. High-pressure (up to 14 kbar) conductivity measurements were carried out in a self-clamped beryllium-copper pressure cell using a miniature sample holder with platinum contact wires. Electrical contacts to the sample were additionally improved by conductive carbon paint. Manganin wire was placed close to the sample to determine the pressure inside the pressure cell during experiment. The hydrostatic pressure-transmitting medium was Fluorinert^{TM}(3M Co). After pressurizing, the cell was clamped at room temperature and then cooled down to 370 mK in a ^{3}He cryostat system containing a superconducting magnet (0–14 T). Magnetic field was applied perpendicularly to the sample surface and current direction.

III. RESULTS

The typical pressure dependence of the normalized conductivity $\sigma(P)/\sigma(\text{ambient pressure})$ of $FeCl_4^-$-doped PA is shown in Fig. 1. The inset to Fig. 1 presents the pressure dependence of the conductivity for the sample, which has been studied at 10 kbar down to 370 mK. As can be seen from Fig. 1, the room-temperature conductivity increases by a factor of 1.3 (from 9100 S/cm up to 11 720 S/cm) with a pressure increase up to 10 kbar, while at higher pressure the conductivity decreases slightly.

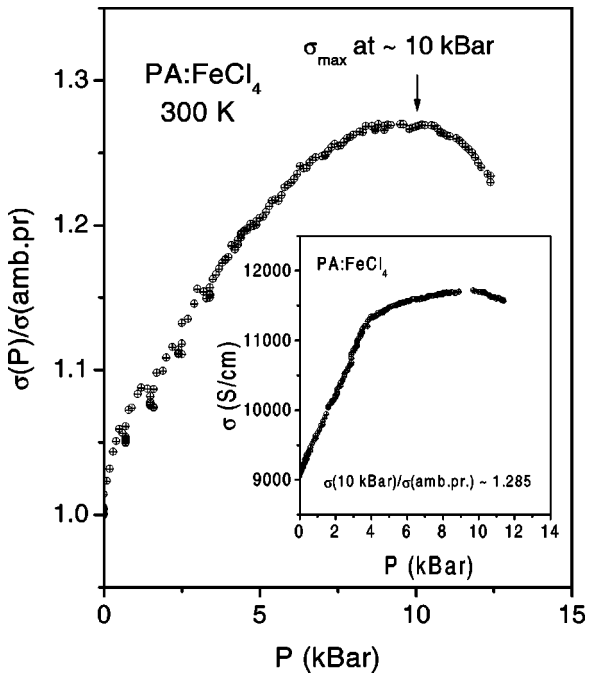

FIG. 1. Pressure dependence of the normalized conductivity $\sigma(P)/\sigma(\text{ambient pressure})$ of $FeCl_4^-$-doped PA at 300 K. The inset shows the pressure dependence of the conductivity for the PA sample, which has been studied at 10 kbar down to 370 mK.

The intrinsic nature of the conductivity maximum at high pressure in $FeCl_4^-$-doped PA was proved by an additional room-temperature pressure experiment with another highly doped conjugated polymer, poly(3,4-ethylenedioxythiophene) (PEDOT) (sample CF1 with a nearly metallic temperature dependence of the conductivity, described in detail at ambient pressure in Ref. 22). The conductivity of the PEDOT sample measured at the same conditions as the PA sample does not show any maximum at high pressure; the conductivity increases continuously with a pressure increase up to 14 kbar.

The typical temperature dependences of the normalized resistivity $\rho(T)/\rho(300\ \mathrm{K})$ of PA doped with $FeCl_4^-$ at ambient pressure and at 10 kbar are shown in Fig. 2. It is evident from Fig. 2 that application of hydrostatic pressure decreases the resistivity ratio $\rho_r = \rho(0.37\ \mathrm{K})/\rho(300\ \mathrm{K})$ from 2.4 (at ambient pressure) down to 1.9 (at 10 kbar). The resistivity minimum–a characteristic feature of highly doped PA–was observed at $T^\star \sim 280$ K for the PA sample at ambient pressure only. Application of pressure suppresses the resistivity minimum and leads to the negative TCR at temperatures from 300 K down to 370 mK in spite of the higher conductivity of the sample at 10 kbar with respect to it at ambient pressure (see the inset to Fig. 2). At temperatures below some specific temperature $T_0 \sim 1$ K, PA samples demonstrate $\rho(T) \sim \ln T$ dependence for both ambient pressure and $P = 10$ kbar, which remains almost unaltered by a magnetic field up to 14 T (Fig. 2 and Fig. 3). As can be seen from Fig. 3, at temperature below 4 K the $\rho(T)/\rho(300\ \mathrm{K})$ curves are being shifted as a whole by a magnetic field, according to negative MR. The slopes of $\rho(T)/\rho(300\ \mathrm{K})$ are almost parallel at zero magnetic field and at a magnetic

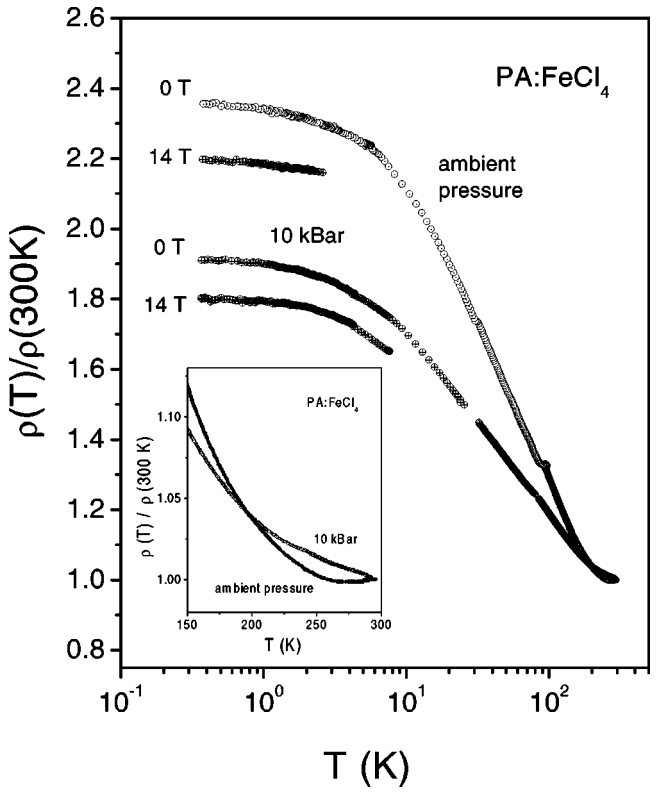

FIG. 2. Temperature dependence of the normalized resistivity $\rho(T)/\rho(300\ \mathrm{K})$ of $FeCl_4^-$-doped PA sample at ambient pressure and at 10 kbar, without magnetic field and at a magnetic field 14 T. The inset shows $\rho(T)/\rho(300\ \mathrm{K})$ vs T for the same sample at the temperature range 150–300 K.

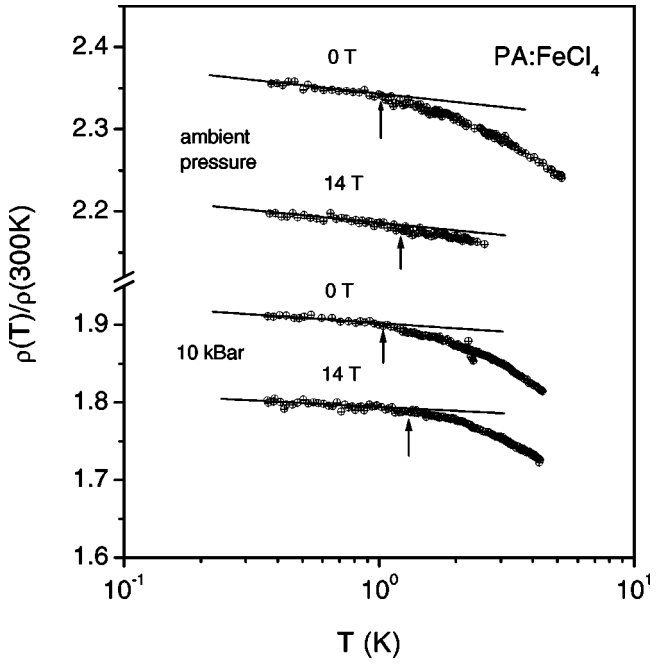

FIG. 3. $\rho(T)/\rho(300\ \mathrm{K})$ vs T for the same sample at the temperature range 0.37–4.0 K. Arrows indicate the starting temperature of $\rho(T)\sim\ln T$ dependence.

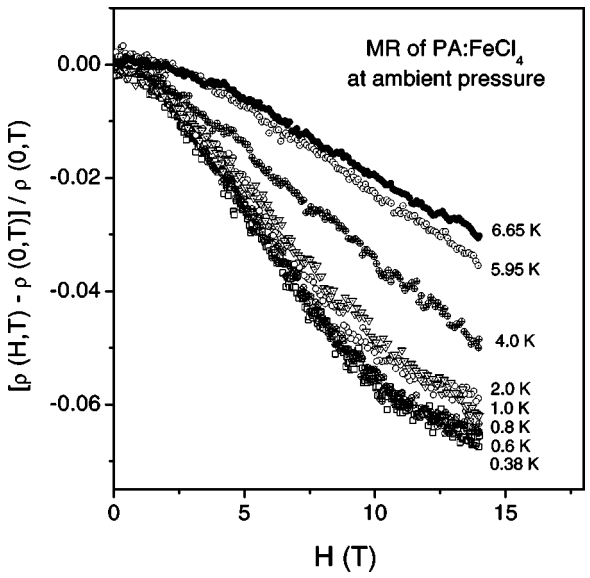

FIG. 4. Transverse magnetoresistance of $FeCl_4^-$-doped PA sample at ambient pressure at various temperatures.

field 14 T, whereas the resistivity ratio ρ_r decreases down to 2.2 (at ambient pressure) and 1.8 (at 10 kbar). The starting temperature of the $\rho(T)\sim\ln T$ dependence shifts slightly by a magnetic field up to higher temperatures (Fig. 3). Figures 4 and 5 show that both at ambient pressure and at 10 kbar the transverse MR of $FeCl_4^-$-doped PA is negative, almost linear, and in fact it is almost temperature-independent at $T<2$ K. As can be seen from Fig. 6, the magnitude of negative MR obtained at 14 T suppresses slightly (from 6.5% to 5.5% at 0.37 K) by application of hydrostatic pressure.

IV. DISCUSSION

The observed pressure dependence of the room-temperature conductivity is rather similar to that obtained in

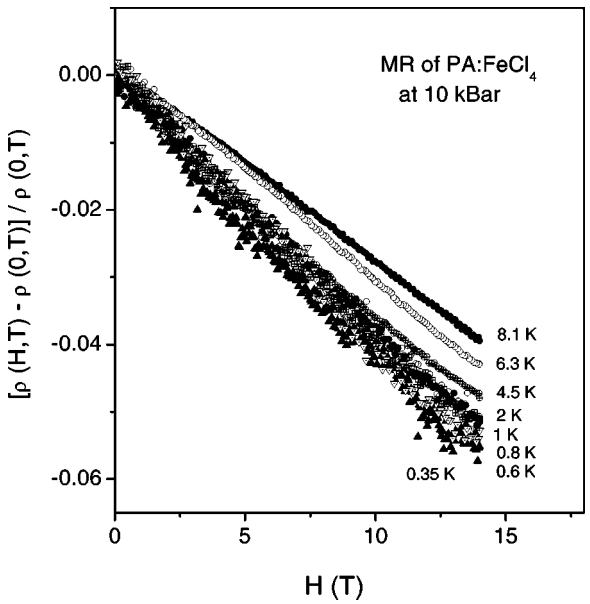

FIG. 5. Transverse magnetoresistance of $FeCl_4^-$-doped PA sample at 10 kbar at various temperatures.

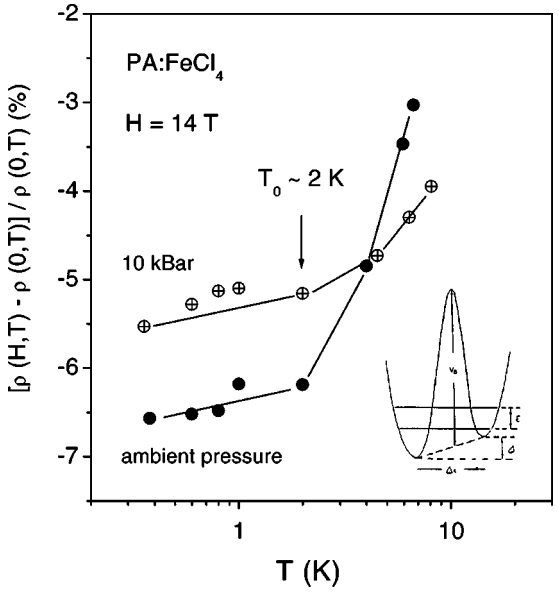

FIG. 6. Temperature dependence of the transverse magnetoresistance of $FeCl_4^-$-doped PA sample calculated at 14 T at ambient pressure and at 10 kbar. The inset shows the double well potential with *E*, the interlevel splitting, which can give rise to the two-level system at low temperature.

PA doped with some other dopants including iodine and alkali metals.[8–11] The initial increase in conductivity at pressure up to 10 kbar implies an enhancement of interchain interactions, while the following decrease could be connected to the creation of some structural defects at high hydrostatic pressure. The differences in the pressure dependences of the conductivity of PA and PEDOT could reflect the different response to the high pressure from fibril-like (PA) and partly globular-like (PEDOT) macromolecular polymeric structure.

The suppression of the resistivity minimum at $T^\star \sim 280$ K by a high hydrostatic pressure can be related to a significant spatial imhomogeneity, which is expected to exist in heavily doped polymers.[19,23] In this case, the mobility edge can be spatially dependent and thus the increase of the conductivity can be ascribed to additional conduction paths activated at the elevated temperature. Hydrostatic pressure affects this spatial inhomogeneity and enhances the strength of the interchain interactions and the total stiffness of the system. All of these factors result in a suppression of nonmonotonic resistivity behavior. This conclusion does not contradict with the modified heterogeneous model,[17] which successfully describes the behavior observed for $FeCl_4^-$-doped PA samples at the relatively high temperature range.[6] Thus we conclude that the transition to monotonous behavior observed for the $FeCl_4^-$-doped PA sample under high pressure results from a strong macroscopic disorder at high temperature.

Next we will focus our attention on the low-temperature ($T \leqslant 1$ K) region. The logarithmic temperature dependence of resistivity and the almost temperature-independent, linear, negative MR at temperatures from the mK region up to some K observed in $FeCl_4^-$-doped PA are rather similar to those observed earlier in iodine-doped PA and PF_6-doped polypyrrole.[18] At the same time, this $\rho(T)$ and MR behavior is similar to that in some metallic glasses, for example in $Fe_{1-x}B_x$ and some other alloys.[24,25] The behavior of $\rho(T)$ and MR at low temperature in metallic alloys has been explained in some different ways: in the framework of the anisotropic exchange interaction model,[24] by Coulomb-type interaction,[26] and by momentum dependence of conduction electron assisted tunneling using the symmetric two-level system model.[27] However, as can be seen from the temperature dependences of the resistivity of $FeCl_4^-$-doped PA, below 1 K, the magnetic field up to 14 T does not affect significantly the slope of the $\rho(T)$ dependence (Fig. 2 and Fig. 3). This indicates that the role of localized magnetic moments is not significant at this temperature region.

Following our approach presented in Ref. 19, we have ascribed the negative TCR with $\rho(T) \sim \ln T$ dependence observed in $FeCl_4^-$-doped PA at low temperatures to the dominant weak-localization contribution.[28] The anomalous $\rho(T)$ and MR behavior at $T < 1$ K seems to be similar to the one observed for ''saturation of dephasing time (τ_φ)'' found recently for crystalline semiconductors.[29] One has in mind that according to the temperature dependence of resistivity at $T < 1$ K, the dephasing time τ_φ is expected to be weakly temperature dependent, so the same is expected for MR. In the case of $FeCl_4^-$-doped PA, there is a correlation between the temperature behavior of resistivity and the temperature dependence of MR, so in this regard this behavior seems to be consistent with dominant weak-localization effects at this temperature region. The negative MR indicates that weak-localization effects with weak spin-orbit scattering dominate transport. The observed linear MR behavior can be considered as a composition of the $H^{1/2}$ law expected for weak localization at high magnetic field and some different mechanism (positive MR) for higher fields which imitates the linear law.

At the same time, taking into account the importance of weak localization, we would like to note that the observed $\rho(T)$ and MR features might also have a structural origin because at low temperatures the structure of polymers exhibits the properties typical for glasses.[19] The glassy properties become more pronounced in highly doped conjugated polymers, where the presence of dopant atoms leads to an increase of microscopic disorder and the weak bonding between the conjugated polymer chains supporting ''soft'' atomic potential configurations. At $T \sim 1$ K, various properties of glasses, e.g., specific heat and thermal conductivity, are mainly determined by the two-level systems (TLS) related to atoms and atomic groups, which move in double-well interatomic potentials with soft barriers[30–32] (see the inset to Fig. 6), whereas at higher temperatures ($1 \text{ K} < T < 10$ K), the main contribution comes from quasiharmonic soft localized modes (SLM).[32]

We have suggested that at $T > 1$ K, the further much stronger temperature behavior of resistivity in $FeCl_4^-$-doped PA can be associated with further suppression of weak localization due to more effective dephasing by low-energy vibrational excitations. Taking into account that highly doped

polymers are having a strong disorder, we assumed that asymmetric TLS are playing the main role in the low temperature inelastic electron scattering in $FeCl_4^-$-doped PA because any disorder would lift the TLS symmetry. According to the soft potential model,[32] the soft anharmonic oscillator potential at ambient pressure is

$$U(x)=E_0[\eta(x/a)^2+\xi(x/a)^3+(x/a)^4], \quad (1)$$

where E_0 is the binding energy of atomic scale in glasses ~ 10 eV, x is the displacement in terms of some generalized coordinate, a is the distance of the order of the interatomic spacing ~ 1 A, η and ξ are random parameters, and η reflects the effective elastic modulus while ξ reflects the potential asymmetry, $\eta,\xi<1$.

The energy scale for the low-energy excitations in glasses at ambient pressure is given by a characteristic energy $W=1\sim 10$ K. At $E<W$, the density of states (DOS) $P(w,r)$ is dominated by the TLS with $P(w,r)=P_0 r^{-1}(1-r)^{-1/2}=$const, where $r=\Delta_0/E$; $E=(\Delta^2+\Delta_0^2)^{1/2}$ is the TLS interlevel splitting; Δ is the atomic potential asymmetry; and Δ_0 is the tunneling matrix element. At $E<3W$, the excitations correspond to strongly anharmonic single-well and weak double-well effective potentials, and at $E>3W$, the excitations are dominated by the SLM–quasilocal harmonic vibrations with $P(w)\sim w^4$.

The effect of pressure (P_h) may be described by adding to $U(x)$ a term linear in P_h and x: $E_0 k P_h(x/a)$:[20]

$$U(x)=E_0[\eta(x/a)^2+\xi(x/a)^3+(x/a)^4+kP_h(x/a)]. \quad (2)$$

$k>0$ is the parameter related to the local compressibility, which for organic solids is $\sim 10^{-5}$ bar^{-1}. It was shown in Ref. 20 that at low $w\ll w_1=(2E_0/Ma_0^2)^{1/2}$, where M is the effective mass of SLM, an application of hydrostatic pressure gives

$$P(w)\sim w^4(1-\sigma_1 P_h), \quad (3)$$

where $\sigma_1=22\beta k/3$ (β is a positive parameter). So, an external pressure reduces the number of DOS without changing its w^4 power dependence. The $P(w)$ for the whole frequency range is[20]

$$P(w)\sim[w^4\Omega_P(1-\sigma_0 P_h)]/[w^6+\Omega_P^2(w^2-w_{0P}^2)^2], \quad (4)$$

where $w_{0P}^2\sim w_0^2(1+\alpha P_h)$, $\Omega_P\sim\Omega(1+\sigma_1 P/2-17\alpha P_h/16)$; $\sigma_0\sim(\sigma_1/2-15\alpha/16)$ and $\alpha=16\beta k w_1^2/9w_0^2$; w_0 and Ω are the characteristic frequency and width of the Lorentzian distribution function. As can be seen from the $P(w)$, an application of hydrostatic pressure P_h decreases significantly the number of the calculated DOS of SLM with possible conversion of SLM into TLS (at low temperature) and shifts the DOS to higher frequency.

This phenomenon may affect the inelastic scattering of electrons by TLS and SLM at low temperatures, leading (together with reduced interatomic spacing, enhanced stiffness of vibrating systems, and enhanced interchain interactions in PA fibril-like structure) to higher metallic conductivity of glassy and polymeric systems under high hydrostatic pressure. The details of scattering (the starting temperature of $\rho(T)\sim\ln T$ dependence, the absolute value of MR, etc.) can be different for different types of atoms (light or heavy) that move in the double-well potential, for symmetric and asymmetric double-well potentials, and for strong and weak coupling between atoms and electrons. In particular, the specific starting temperature of the logarithmic temperature dependence, $T_0\sim 1$ K, correlates well with the upper limit for applicability of the TLS model in metallic glasses. The shift of T_0 by pressure and by a magnetic field up to higher temperatures observed in $FeCl_4^-$-doped PA can be related to the influence of the pressure- and magnetic-field-induced conversion of SLM into TLS at low temperature predicted in Ref. 20.

TLS-related transport corresponds to microscopic disorder. We would like to note that macroscopic disorder could influence the transport properties at high temperature in heavily doped PA samples. This disorder leads to a relatively wide spatial scatter between the mobility edge E_C and the Fermi level E_F within the energy scale from some meV down to zero.[19,23] In this case, according to the percolation model, one can expect that a metallic cluster will be formed by regions where E_C is lower than E_F and the resistivity decrease can be ascribed to additional conduction paths activated at the elevated temperature.

V. CONCLUSIONS

We have studied the effects of hydrostatic pressure and magnetic field on the low-temperature conductivity of oriented $FeCl_4^-$-doped metallic polyacetylene. It was found that the conductivity at 10 kbar is greater than that at ambient pressure by a factor of 1.3. Pressure suppresses the resistivity minimum at 280 K and decreases the resistivity ratio $\rho_r=\rho(0.37\ \text{K})/\rho(300\ \text{K})$ from 2.4 down to 1.9. The temperature dependence of resistivity demonstrates the $\rho(T)\sim\ln T$ dependence at $T<1$ K, which remains almost unaltered by application of high pressure and a magnetic field up to 14 T. The transverse MR was found to be negative, almost linear, and almost temperature-independent at $T<2$ K at ambient pressure and at 10 kbar. We attributed the $\rho(T)$ and MR behavior observed at $T<1$ K in $FeCl_4^-$-doped polyacetylene to weak localization. The increase of inelastic scattering due to low-energy vibrational excitation can explain the further stronger temperature behavior of $\rho(T)$ and MR at $T>2$ K as a result of further suppression of weak localization due to a more effective dephasing effect. At higher temperature, the resistivity decrease is dominated by activation to additional conduction paths.

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