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Magnetic susceptibility of polyaniline and polyaniline-polymethylmethacrylate blends

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While heat-capacity measurements on polyaniline yield density of states at the Fermi level $N(E_F)$ of the order of 3 states/(eV-2 rings), a value of 16 states/(eV-2 rings) was reportedly found from magnetic susceptibility measurements. We report here our magnetic susceptibility measurements, which, in agreement with the heat-capacity results, yield $N(E_F)$ of the order of 2 states/(eV-2 rings). These results firmly establish the metallic nature of charge carriers in the polyaniline family.

In an earlier paper,[1] we presented results of a low-temperature heat-capacity study of *p*-toluenesulfonic acid (PTSA) doped polyaniline (PANI), and PTSA doped polyaniline-polymethylmethacrylate (PANI-PMMA) blends. The specific heat (C), which is related to heat capacity (c) as $C=c/(m/M)$ with m and M being the sample mass and molecular mass, respectively, was represented at low temperatures by the expression $C=A+\gamma T+\beta T^3$ for PANI and by the expression $C=\gamma T+\beta T^3$ for PANI-PMMA blends; here, γT and βT^3 are the electronic and lattice contributions, respectively, to specific heat. Since γ is related to the density of states at the Fermi level as $N(E_F)$ (in states/eV mole) $=0.212\,\gamma$ (in mJ/mole K^2), analysis of the specific-heat data gave $N(E_F)$ values of 3 states/(eV 2-rings) for PANI, 17 states/(eV 2-rings) for PANI(40%)-PMMA(60%) (abbreviated as PP4060), and 17 states/(eV 2-rings) for PANI(33%)-PMMA(67%) (abbreviated as PP3367). These heat-capacity obtained values of $N(E_F)$ were then compared with those obtained by another group[2] from magnetic susceptibility (χ) measurements using the relation $N(E_F)=\chi_P/\mu_B^2$, where χ_P is Pauli susceptibility and μ_B is Bohr magneton. While the agreement between the $N(E_F)$ values obtained from these two different techniques was semiquantitative for PP4060 and PP3367, magnetic susceptibility obtained $N(E_F)$ for PANI was larger by a factor of 7 compared to its value from heat-capacity measurements.

The purpose of this paper is to present our own magnetic susceptibility measurements on PANI in view of the above difference between the heat capacity obtained (3 states/eV 2-rings) and the magnetic susceptibility determined (21 states/eV 2-rings) values of $N(E_F)$. It is significant to note that a typical value of $N(E_F)$ reported by various groups[3] on variously synthesized PANI is around 3 states/(eV 2-rings). Two samples of PANI from two different batches and one

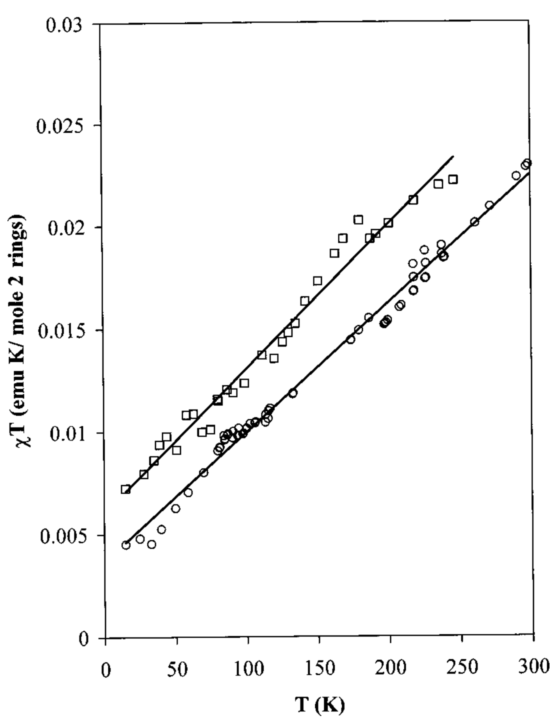

FIG. 1. χT as a function of T for two samples of polyaniline. Least-squares fit of the data is shown by lines passing through the data.

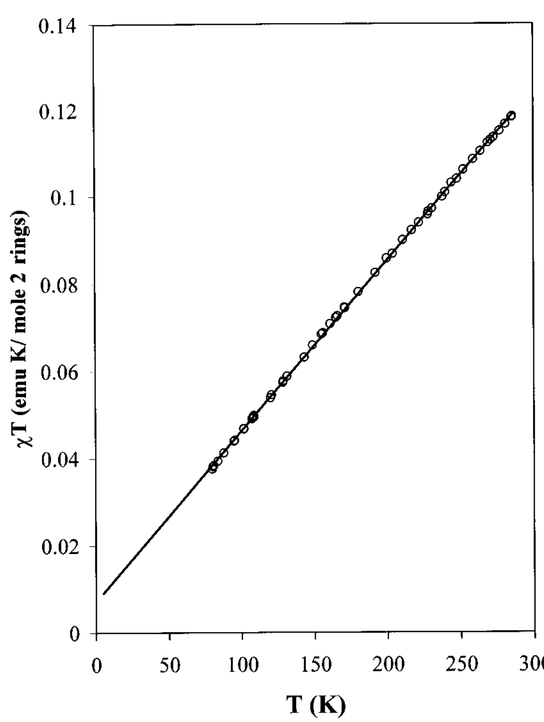

FIG. 2. χT as a function of T for the polyaniline-polymethylmethacrylate (40:60) blend. The line through the points shows least-squares fit of the data.

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sample of PP4060 were therefore obtained from Ormecon Chemie Gmbh & Co—the same supplier of earlier samples as well. χ measurements were made at 5 kG with a ''force'' magnetometer on PANI and PP4060 samples, which were pressed in the form of a parallel pipe and had a mass in the range 80–100 mg. The pressed samples were attached directly to a fine quartz fiber, whose other end connected to the electromicrobalance for force measurements; this arrangement avoided applying any addendum corrections due to the sample holder.

In the free-electron model, slope and intercept of the χT vs T plot yield Pauli susceptibility (χ_P) and Curie constant (C), respectively. Figure 1 therefore shows data in the χT vs T form for the two PANI samples. In obtaining this plot, the diamagnetic core susceptibility and the molecular mass were taken as -203.2×10^{-6} emu/(mole 2-rings) and 373, respectively. A simple analysis of the data shown in Fig. 1 gives the following values for χ_P and C: 66×10^{-6} emu/(mole 2-rings) and 0.0037 for sample 1, and 73×10^{-6} emu/(mole 2-rings) and 0.0059 for sample 2. $N(E_F)$ values obtained from these Pauli susceptibilities are 2.1 and 2.3 states/(eV 2-rings). These values are in disagreement with the one reported earlier,[2] but in agreement with the value obtained from our heat-capacity measurements.[1]

The χT vs T behavior for the PP4060 sample was also obtained which is shown in Fig. 2. Values of -319.4×10^{-6} emu/(mole 2-rings) and 653 were used, respectively, for the diamagnetic core susceptibility and the molecular mass in obtaining the shown behavior. The plot yields a χ_P of 420×10^{-6} emu/(mole 2-rings) or 14 states/(eV 2-rings) for $N(E_F)$, which is in agreement with the previously reported value.

In conclusion, the results from heat-capacity and magnetic susceptibility experiments, in spite of different sensitives of these techniques, have been shown to yield nearly the same corresponding values of $N(E_F)$ for polyaniline and polyaniline-methylmethacrylate systems. It therefore seems clear that theoretical models or experimental analyses which do not require the existence of free-electron spins or Pauli susceptibility may not correctly describe the underlying physics of systems in the polyaniline family.

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