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Direct Determination of Chemically Induced Doping Concentrations in (6,5)- Single-Wall Carbon Nanotubes by EPR

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Control over doping of semiconducting Single-Wall Carbon Nanotubes (SWNTs) by (electro-) chemical methods and intrinsic impurities are considered to be crucial for future applications of SWNTs in carbon-based electronics and photonics. Previous indirect all-optical techniques hypothesize charge carrier localization in semiconducting (6,5)-SWNTs following redox chemical AuCl3 doping, which is indicated by spectral and dynamical changes of the S1 exciton band [1].
Here, we present direct quantitative investigations of chemical impurities by means of electron paramagnetic resonance (EPR). For the as-produced (6,5)-SWNT samples we detected very low signal intensities stemming from two separate radical-like, isolated spin contributions that we assign to defects or residual impurities and which may be associated with what is sometimes referred to as intrinsic p-doping. Their concentration indicates exceptional nanotube quality with only few paramagnetic species per µm nanotube. Upon addition of the doping agent (AuCl3)2 we observe a linear increase of the EPR signal intensity which we ascribe to localized p-type impurities on the nanotube. However, at higher doping concentrations, the observed signal saturates, before decreasing again and finally vanishing at the highest AuCl3 concentrations. This seemingly puzzling result can be understood in terms of the formation of EPR silent, diamagnetic bi-radicals which start to appear at moderate doping concentrations. We model this behaviour with a stochastic formation of impurity sites in a 1D lattice, yielding good agreement with experimental data. In conclusion, we can confirm charge carrier confinement at low impurity concentrations in SWNTs with potentially far reaching implications for future device applications.

Summary

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