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Anomalous organic magnetoresistance from competing carrier-spin-dependent interactions with localized electronic and nuclear spins

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Transport of carriers through disordered electronic energy landscapes occurs via hopping or tunneling through various sites, and can enhance the effects of carrier spin dynamics on the transport. When incoherent hopping preserves the spin orientation of carriers, the magnetic-field-dependent correlations between pairs of spins influence the charge conductivity of the material. Examples of these phenomena have been identified in hopping transport in organic semiconductors and colloidal quantum dots, as well as tunneling through oxide barriers in complex oxide devices, among other materials. The resulting room-temperature magnetic field effects on the conductivity or electroluminescence require external fields of only a few milliTesla. These magnetic field effects can be dramatically modified by changes in the local spin environment. Recent theoretical and experimental work has identified a regime for low-field magnetoresistance in organic semiconductors in which the spin-relaxing effects of localized nuclear spins and electronic spins interfere1. The regime is studied experimentally by the controlled addition of localized electronic spins, through the addition of a stable free radical (galvinoxyl) to a material (MEH-PPV) that exhibits substantial room-temperature magnetoresistance (20initially suppressed by the doping, as the localized electronic spin mixes one of the two spins whose correlation controls the transport. At intermediate doping, when one spin is fully decohered but the other is not, there is a regime where the magnetoresistance is insensitive to the doping level. For much greater doping concentrations the magnetoresistance is fully suppressed as both spins that control the charge conductivity of the material are mixed. The behavior is described within a theoretical model describing the effect of carrier spin dynamics on the current. Generalizations to amorphous and other disordered crystalline semiconductors will also be described. This work was supported by DOE and an ARO MURI and was done in collaboration with N. J. Harmon, K. Sahin-Tiras, Y. Wang and M. Wohlgenannt.