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Competing Mechanisms in Organic Magnetoresistance

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A surprisingly large "organic magnetoresistance" (OMAR) has been found in both polymers and small molecule organic semiconductors at relatively small applied magnetic fields ($\sim$ 5 mT) and at room temperature. Unlike spin-injection devices, where the occurrence of a finite *spin polarization* of the current is essential for measuring a finite magnetoresistance, OMAR is generally considered to be due to *spin correlations* between spin carrying particles in the organic material. Although the microscopic mechanisms of hyperfine field induced spin mixing are relatively well understood, it is still intensively debated which particles are involved and how they can affect the current in such a drastic manner. In this presentation recent developments and new insights as to the underlying physics are discussed. Quantitative models will be introduced, based on different pairs of particles and mechanisms, and giving rise to effects at a variety of field scales. It will be discussed how specific device physics causes a non-trivial relation between microscopic spin-dependent reactions and macroscopic device behaviour. Finally, it will be shown how comprehensive studies on especially engineered organic systems, including polymer-fullerene blends and molecular doping, can be used to pinpoint the relevance of different mechanisms in the complementary regimes. The experimentally observed linewidth, sign and amplitude of both "high-field" (>100 mT) and "low-field" ($\sim$ 5 mT) effects, as well as their bias voltage dependence display very pronounced features as a function of fullerene doping. They provide unique fingerprints for which mechanism is of relevance. After careful analysis, this allows for identification of three earlier proposed mechanisms, involving exciton-charge, electron-hole and bipolaron (polarons of like charge) reactions. Present activities are aiming at using this insight for tailoring OMAR response by design.