# Magnetic-Field Modulated Carrier Hopping in Organic Semiconductors: An Entanglement Perspective


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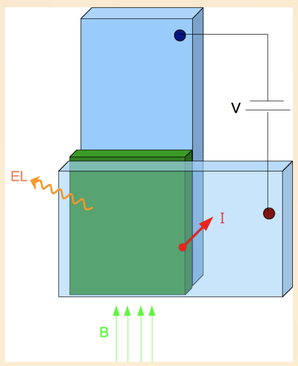
**For spintronics purposes, we studied the influence of the charge carrier's spin on its intermolecular hopping rate in amorphous organic semiconductors (OSC), beyond the common framework based on some "spin-blocking" rules. It is proved that the this hopping rate is directly related to the entanglement between the carrier's spin and its local environment. This enables the modulation of this rate by a magnetic field *B*. Based on a model of hyperfine interaction, we calculated the *B*-dependence of hopping rate , which exhibits a negative component at small *B* and a saturation component at large *B*. Both the overall behavior and its isotope effect agree well with experiment [1].**

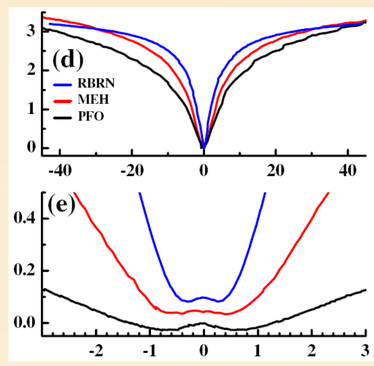

## I. Background: Organic Spintronics

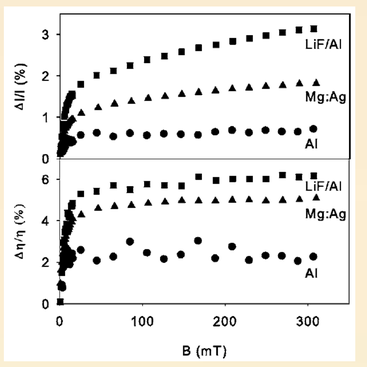
**Magneto-conductance (MC) [2] and Magneto-electroluminescence (MEL) [3]**

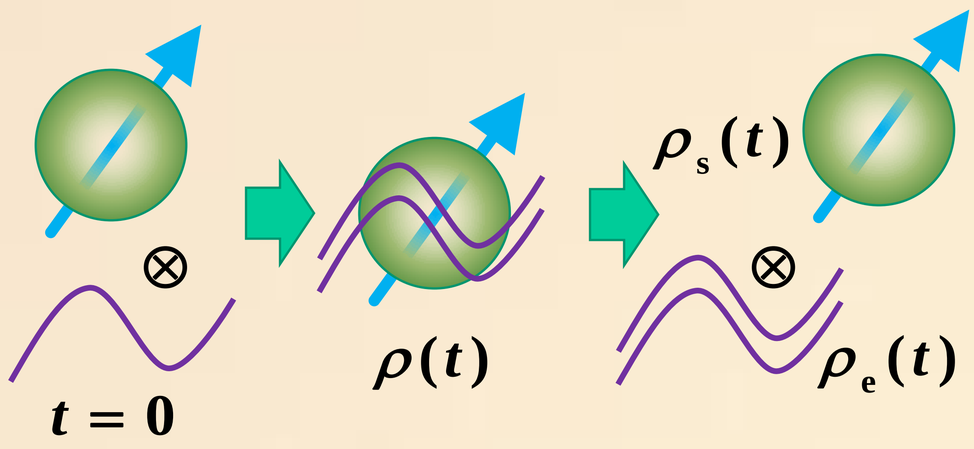
- **Sizable effect from the thin film devices of various organic materials;**
- **Robust in a wide temperature range from 4K to room temperature;**
- **Similar lineshape with a characteristic *B* of the order of milli-Tesla.**

**The universality calls for a general theory.**




## II. Physical Process

- **Charge transport in amorphous OSC is by incoherent hopping of carriers among molecules. However, the dynamics of the carrier's spin is thought to be coherent due to weak interaction.**
- **For a carrier hopping onto a specific molecule at time *t*=0, the interaction between its spin (s) and the local environment (e) brings them into entangled states at time t, denoted as $\rho(t)$.**
- **The incoherent hopping terminates this correlation, and the density matrix of the final state takes a direct-product form $\rho_f(t) = \rho_s(t) \otimes \rho_e(t)$, with $\rho_{s(e)}(t) = \mathrm{tr}_{e(s)}\rho(t)$**
- **The hopping rate is determined by Fermi golden rule: $\nu(t) = \nu_0 \mathrm{tr}\{\rho(t)\rho_f(t)\} = \nu_0\eta(t;B)$, in which $\nu_0$ is for contributions from other factors independent of the carrier's spin.**

## III. Main Results

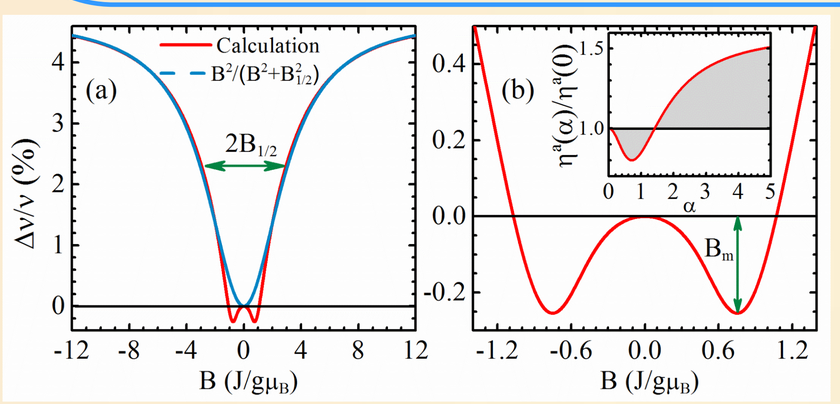
- **We take a model of hyperfine interaction as an example for quantitative results, which is:**

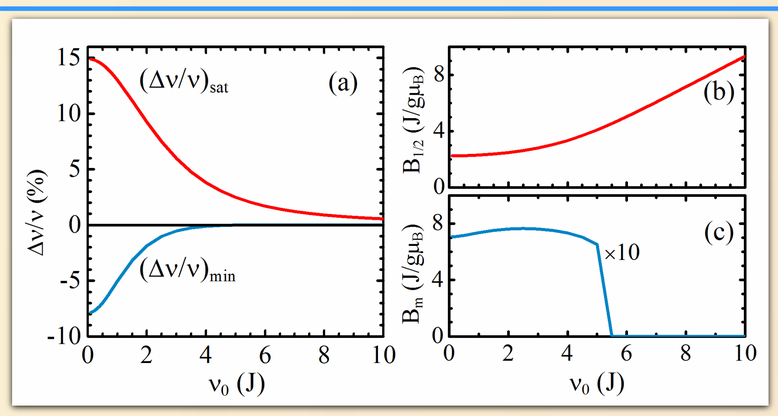
$$H = \sum_i J_i \mathbf{I}_i \cdot \mathbf{s} + g\mu_B B s_z .$$

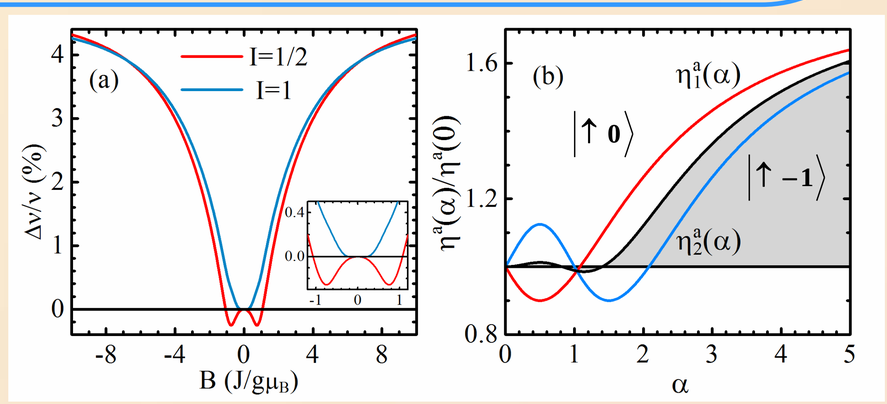
- **The initial state of the composite system is not entangled, and the *t*-dependent entanglement will make the hopping rate varying with time. We treat the hopping process as Markovian, and the inverse of the average waiting time can be seen as an effective hopping rate, which is:**

$$\nu = \langle t_w \rangle^{-1} = \left[ \int_0^\infty \exp\left( -\nu_0 \int_0^t \eta(\tau;B)\mathrm{d}\tau \right) \mathrm{d}t \right]^{-1} .$$

- **The following results are based on the example of a single spin *I*=1/2 as the local environment.**
- **There is a Lorentzian-like saturation at large *B*, together with a negative component at small *B*.**
- **The origin of the negative component can be understood by the *t*-averaged $\eta(t;B)$ for the initial state $|\uparrow\downarrow\rangle$. Here, $\alpha = g\mu_B B / J$.**
- **The magnitude of the variation of the hopping rate with *B* decreases with $\nu_0$, which is roughly linear to electric field in the device. This follows the same trend with experimental results [3].**
- **The half-width at half-maximum $B_{1/2}$ also increases, which is observed by experiment [1].**
- **A single spin *I*=1 as local environment is chosen for deuterons. The negative component is nearly smeared out, while the saturation remains.**
- **The difference can be understood by the result of two initial states: $|\uparrow 0\rangle$ as example 1 and $|\uparrow -1\rangle$ as example 2. In the second case the energy splitting first decrease and then increases with *B*.**




## IV. Conclusion

**We prove that in the hopping regime of OSC, the destroy of quantum entanglement between the carrier's spin and its local environment by the incoherent hopping will influence the hopping rate. A magnetic-field dependent hopping rate emerges from this process. The results based on a model of hyperfine interaction agrees qualitatively with experimental results. The detailed relation with experimental observables calls for future work.**


[1] T. D. Nguyen, G. Hukic-Markosian, F. J. Wang, L. Wojcik, X. G. Li, E. Ehrenfreund, and Z. V. Vardeny Nature Mater. **9**, 345 (2010).
[2] T. D. Nguyen, B. R. Gautam, E. Ehrenfreund, and Z. V. Vardeny Phys. Rev. Lett. **105**, 166804 (2010).
[3] P. Desai, P. Shakya, T. Kreouzis, W. P. Gillin, N. A. Morley, and M. R. J. Gibbs Phys. Rev. B **75**, 094423 (2007).