Organic magnetoresistance based on hopping theory*

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(Received 26 December 2013; revised manuscript received 5 March 2014; published online 23 July 2014)

For the organic magnetoresistance (OMAR) effect, we suggest a spin-related hopping of carriers (polarons) based on Marcus theory. The mobility of polarons is calculated with the master equation (ME) and then the magnetoresistance (MR) is obtained. The theoretical results are consistent with the experimental observation. Especially, the sign inversion of the MR under different driving bias voltages found in the experiment is predicted. Besides, the effects of molecule disorder, hyperfine interaction (HFI), polaron localization, and temperature on the MR are investigated.

Keywords: hopping, organic magnetoresistance, master equation, hyperfine interaction

PACS: 73.50.–h, 72.20.Ee, 75.47.–m **DOI:** 10.1088/1674-1056/23/9/097306

1. Introduction

Organic spintronics have generated considerable interest in recent years[1,2] due to the long spin lifetimes of organic semiconductors (OSCs) as well as the flexibility, low cost, and chemical tunability of organic devices.[3–5] In the last few years, magnetic field-modulated resistance of organic devices, named organic magnetoresistance (OMAR), has been widely studied.[6,7] Usually, an OMAR value around 0–15% has been observed in a number of organic materials at room temperature under a low magnetic field (in the scale of mT). OMAR behaviors display universal line shapes that can be fitted by empirical Lorentzian $B^2/(B^2+B_0^2)$, non-Lorentzian $[B/(|B|+B_0)]^2$ or their combination.[8,9] Some are also fitted with power law B^n, $f_1/B^2+f_2/B^4$, or $d_1B^2+d_2B^4$ (in these formulae, B is the applied magnetic field, B_0 and the rest of the symbols are fitting coefficients).[10,11] Both positive and negative OMAR have been reported in the OSCs, exhibiting a transition between negative and positive depending on the voltage and the concrete samples.[9,12–15] For example, Mermer *et al.* reported the MR of a polyfluorene device under different bias voltages. They found that the MR shows a sign inversion from negative to positive as the voltage increases.[8] Besides, Kang *et al.* reported the sign change in OMAR of an Alq_3 film with the morphological change from amorphous to crystalline state upon annealing.[15] The investigation of the sign change could provide vital clues for further unraveling the origin of OMAR. Some explanations for the sign change have been reported,[12,14,16–18] and generally it has been thought that different signs of OMAR correspond to differences in the microscopic mechanism under different device operating conditions. For example, Desai *et al.* described the changes in the MR are a result of the reduction in the triplet exciton concentration and the reduced role of free carrier trapping at triplet states.[17] Bloom *et al.* assigned the sign inversion behavior to a transition from a single-carrier regime to a double-carrier regime.[18] Up to now, the underlying mechanism of OMAR is still under debate. Most of the mechanisms that have been proposed to explain OMAR are involved with the spin dependent transport of charged spin carriers (polarons).[19–21] It is usually suggested that the external magnetic field together with the hyperfine interaction (HFI) of hydrogen nuclei act on the spin of polarons and affect their transport among molecules.[22,23] As a point of view, the mobility-related theory has been widely supported. For example, Ding *et al.* showed that the organic magnetic-field effect (OMFE) has a close relationship with the carrier mobility.[24] In addition, Veeraraghavan *et al.* measured the magnetoresistance (MR) in polyfluorene devices and found that the magnetic field effect acts on the carrier mobility rather than carrier density.[25] Li *et al.* studied the velocity of polaron based on a quantum quasi-band model and gave a rational explanation for MR found in small molecule devices.[26] Based on percolative hopping transport, Harman and Flatté provided a description of OMAR for positionally disordered organic materials. They analytically obtained the MR and the results agree with some measurements.[27] As most organic materials have an amorphous structure, transport takes place mainly by hopping of localized carriers between sites (molecules) instead of the band transport. The localized carriers are spin-sensitive to the external magnetic field and the hyperfine field.[21] In this paper, we try to consider spin dependent polaron transport with the master equation (ME).[28–30] The spin orientation is determined by the external magnetic field as well as the HFI. As an approach, we suppose that a polaron keeps its spin orientation during hopping. The model details are described in Section 2. Results and discussion are given in Section 3. Finally, in Section 4, a conclusion is made.

*Project supported by the National Basic Research Program of China (Grant No. 2010CB923402), the National Natural Science Foundation of China (Grant Nos. 11174181 and 21161160445), and the Program of Introducing Talents of Discipline to Universities, China (Grant No. B13029).

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2. Model and method

We consider a localized polaron at site i, and its spin is determined by the external magnetic field B and the internal effective hyperfine field $B_{\mathrm{hpf},i}$. The Hamiltonian of the system can be written as

$$\begin{aligned}\hat{H}_i &= -g\mu_{\mathrm{B}}\hat{\boldsymbol{s}}\cdot\boldsymbol{B} - g\mu_{\mathrm{B}}\hat{\boldsymbol{s}}\cdot\boldsymbol{B}_{\mathrm{hpf},i} \\ &= -g\mu_{\mathrm{B}}B\hat{s}_z - g\mu_{\mathrm{B}}B_{\mathrm{hpf},i}\left(\hat{s}_z\cos\theta_i + \hat{s}_x\sin\theta_i\right), \end{aligned} \quad (1)$$

where g is the electronic g-factor, μ_{B} is the Bohr magneton, $\hat{s}$ is the electronic spin operator, $\hat{s}_z$ and $\hat{s}_x$ are the z and x components of the electronic spin, respectively, and θ_i is the angle between the magnetic field $\boldsymbol{B}$ (along the z axis) and the local hyperfine field $\boldsymbol{B}_{\mathrm{hpf},i}$. The ground state of the polaron is solved to be $\phi_{\mathrm{G}} = \begin{pmatrix} Z_\uparrow \\ Z_\downarrow \end{pmatrix}$, so we get the probability of spin up

$$p_i^{\uparrow} = \frac{|Z_\uparrow|^2}{|Z_\uparrow|^2 + |Z_\downarrow|^2}$$

and the probability of spin down

$$p_i^{\downarrow} = 1 - p_i^{\uparrow} = \frac{|Z_\downarrow|^2}{|Z_\uparrow|^2 + |Z_\downarrow|^2}.$$

Equation (1) can be written as

$$\hat{H}_i = -\frac{1}{2}\begin{pmatrix} g\mu_{\mathrm{B}}B + g\mu_{\mathrm{B}}B_{\mathrm{hpf},i}\cos\theta_i & g\mu_{\mathrm{B}}B_{\mathrm{hpf},i}\sin\theta_i \\ g\mu_{\mathrm{B}}B_{\mathrm{hpf},i}\sin\theta_i & -g\mu_{\mathrm{B}}B - g\mu_{\mathrm{B}}B_{\mathrm{hpf},i}\cos\theta_i \end{pmatrix} = -\begin{pmatrix} A_i & \delta_i \\ \delta_i & -A_i \end{pmatrix}. \quad (2)$$

Here, $\delta_i = \frac{1}{2}g\mu_{\mathrm{B}}B_{\mathrm{hpf},i}\sin\theta_i$. It can be seen from the formula that the spin of site i is the mixed state of spin up and spin down states due to the existence of δ_i. Then the results of the probabilities p_i^{s} are given as

$$\begin{cases} p_i^{\uparrow} = \dfrac{1}{2}\dfrac{\left(A_i + \sqrt{A_i^2+\delta_i^2}\right)^2}{A_i^2+\delta_i^2 + A_i\sqrt{A_i^2+\delta_i^2}}, \\ p_i^{\downarrow} = \dfrac{1}{2}\dfrac{\delta_i^2}{A_i^2+\delta_i^2 + A_i\sqrt{A_i^2+\delta_i^2}}. \end{cases} \quad (3)$$

We suppose that a polaron keeps its spin orientation during hopping under a low carrier density. In our work, to describe the transport of the system, we need to know the hopping rate of a polaron. The charge hopping rate ω_{ij} is described by the Marcus formula, which can be written as

$$\omega_{ij} = \frac{t_{ij}^2}{\hbar}\left[\frac{\pi}{k_{\mathrm{B}}T\lambda}\right]^{1/2}\exp\left[-\frac{(\lambda+\varepsilon_j-\varepsilon_i)^2}{4k_{\mathrm{B}}T\lambda}\right], \quad (4)$$

where $\hbar$ is the reduced Planck constant, $t_{ij} = t_0\exp(-2\gamma R_{ij})$ is the transfer integral between molecules i and j, t_0 is a constant, $R_{ij} = \left|\boldsymbol{R}_j - \boldsymbol{R}_i\right|$ is the distance between the two molecules, γ is the inverse localization length of the localized wave functions, k_{B} is the Boltzmann constant, π is the circumference ratio, T is the temperature, and ε_i is the on-site energy of site i. The energy differences in Eq. (4) are supposed to contain a contribution $-eER_{ij,x}$ due to an electric field E in the x direction (e is the charge of the carriers), λ is the reorganization energy responding to the charge moving from site i to j.[31]

To determine the mobility of organic devices, the model of hopping between different sites has been investigated by the ME method.[29,30] The ME approach has an advantage: it guarantees the steady state solution. When it comes to the spin-related system, the ME can be written as

$$\frac{\mathrm{d}n_{is}}{\mathrm{d}t} = \sum_{j\neq i}\left[-\omega_{ij}n_{is}(1-n_{js}) + \omega_{ji}n_{js}(1-n_{is})\right], \quad (5)$$

where n_{is} is the occupancy number of polarons at site i with spin s. In the steady state $\mathrm{d}n_{is}/\mathrm{d}t = 0$, equation (5) is solved through an iteration procedure. Firstly, we set the energy levels $\{\varepsilon_{is}\}$ from a Gaussian distribution of width σ. The hyperfine field is set as $B_{\mathrm{hyp},i} = B_{\mathrm{hyp}}$, and the angles $\{\theta_i\}$ obey a uniform distribution. The polaron density of the system is $n = N_{\mathrm{T}}/M = \sum_s N_s/M$, where N_{T} and N_s are the total polaron population of the system and the total polaron population of spin s, respectively, and M is the total number of the sites. During the calculation, $N_s = N_{\mathrm{T}}\sum_i p_i^{\mathrm{s}}/M$. The iteration is carried out till the difference of $\left|(n_{is}^{k+1} - n_{is}^{k})/n_{is}^{k}\right|$ between two successive calculations is smaller than a given small value for each site i, where k is the iteration number.

When the stable distribution n_{is} is obtained, the mobility of the route of spin s is given by

$$\mu_s(B) = \frac{1}{N_s E}\sum_{i,j}\omega_{ij}n_{is}(1-n_{js})R_{ij,x}. \quad (6)$$

While the total mobility of the system is

$$\mu_{\mathrm{T}}(B) = \sum_s N_s\mu_s(B)/N_{\mathrm{T}}. \quad (7)$$

Therefore,

$$\mathrm{MR} = \frac{\mu_{\mathrm{T}}(0) - \mu_{\mathrm{T}}(B)}{\mu_{\mathrm{T}}(B)}.$$

In calculation, we take a regular cubic molecular lattice with the total number of sites $M = 50\times50\times50$. The molecule nearest neighbor distance (lattice constant) $a = 1$ nm. The hopping takes place to a maximum distance of $d_{\max} = \sqrt{3}a$

and we apply periodic boundary conditions. The reorganization energy is $\lambda = 0.1$ eV. The simulation is carried out under a low carrier density $n = 0.01$ and the temperature is set as $T = 300$ K. The mobility is averaged over twenty samples to reduce statistical errors as much as possible.

3. Results and discussion

OMAR is found to appear complex from a large number of experimental measurements on organic devices. It is associated with the concrete organic materials and the polarity of the device, as well as the driving voltage and temperature.

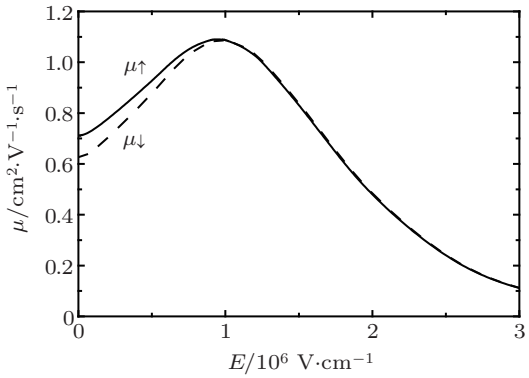


Fig. 1. Dependence of polaron mobility on the strength of the driving electric field under magnetic field $B = 30$ mT. The parameters are set as $\sigma = 0.05$ eV, $t_0 = 0.1$ eV, $B_{\mathrm{hyp}} = 10$ mT, and $\gamma = 1$ nm^{-1}.

The electric field dependence of mobility is important to understand the transport property of an organic device. Time-of-flight mobility measurements showed that the field dependence in many OSCs approximates the Poole–Frenkel form i.e., $\mu = \mu_0 \exp(\gamma_0 \sqrt{E})$, where μ_0 and γ_0 are the constants.[32] As shown in Fig. 1, the mobility shows a similar Poole–Frenkel behavior in a low intensity electric field. While in a strong electric field when $\left|eER_{ij,x}\right|$ are comparable or larger than the reorganization energy, the mobility decreases with an increase of the electric field. This performance is described as Marcus inverted region behavior. In our work, the reorganization energy is $\lambda = 0.1$ eV. From the relation of $\lambda = |eE_\lambda a|$, we conclude the electric field is close to $E_\lambda = 1 \times 10^6$ V/cm. It can be seen from the figure that the mobilities $\mu_\uparrow$ and $\mu_\downarrow$ both have a peak value near E_λ.

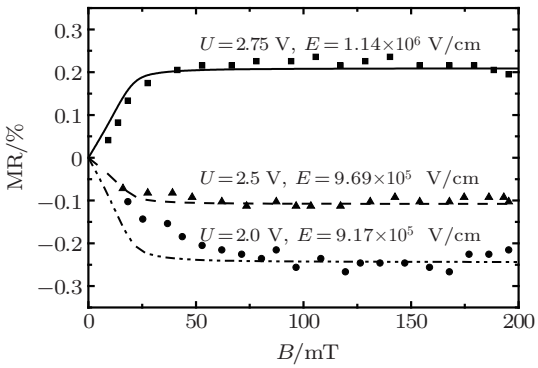


Fig. 2. Dependence of MR on B under different electric fields. The experimental data (squares, triangles, and circles) under different driving voltages are also shown for comparison.[14] The parameters are set as $\sigma = 0.05$ eV, $t_0 = 0.1$ eV, $B_{\mathrm{hyp}} = 10$ mT, and $\gamma = 1$ nm^{-1}.

The effects of the electric field on the MR are calculated by the ME. As shown in Fig. 2, the MR is negative at a low electric field. As the electric field increases, the MR's absolute value drops. Finally, the MR changes its sign at a higher electric field. For the electric field is related to the bias voltage, the experimental data (squares, triangles, and circles) under different driving voltages are shown for comparison.[14] It is found that the theoretical simulation coincides well with the experimental data. Under these parameters, our results show that the sign of OMAR changes from negative to positive as the bias increases. As is shown in Fig. 1, the mobility reaches a peak value near $E_\lambda = 1 \times 10^6$ V/cm. The electric field corresponding to the sign change of OMAR is also close to E_λ, so we deduce the mobility's behavior results in the sign inversion.

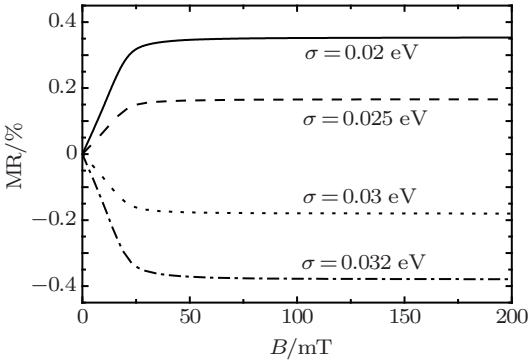


Fig. 3. Dependence of MR on B under different energetic disorder widths σ. The parameters are set as $E = 1 \times 10^5$ V/cm, $t_0 = 0.1$ eV, $B_{\mathrm{hyp}} = 10$ mT, and $\gamma = 1$ nm^{-1}.

For amorphous organic materials, the parameter σ is the description of the disorder extent of the system. The system will be more disordered when σ becomes larger, and the mobility of OSCs increases as σ decreases. As shown in Fig. 3, the MR's sign changes from negative to positive as σ decreases. We set $\Delta G = \varepsilon_j - \varepsilon_i - eER_{ij,x}$ from Eq. (4). Thus σ works together with the electric field to influence the energy differences. When the electric field is constant, the energetic disorder width σ can also change the MR's sign. Recently, Kang *et al*. investigated OMAR of an Alq_3 film with the morphological change from amorphous to crystalline state upon annealing. They found that the sign of the MR changes from negative to positive when the structure of the system changes from disorder to order.[15] Our results show the same sign inversion.

The indispensable role of HFI from the hydrogen nuclear spins is proposed in the mechanism of understanding on OMAR nearly at the time when strong OMAR is found. For instance, Nguyen *et al*. compared the magneto–electroluminescence response in organic light-emitting diodes

based on π-conjugated polymers made of protonated and deuterated hydrogen (the latter has a weaker HFI strength).[33] They reported that the device based on the D-polymers shows substantially narrower magneto–electroluminescence response, which demonstrates that the HFI does indeed have a crucial role in OMFE. Qin *et al.* studied OMFE from a phenomenological theory and found that OMFE behavior is sensitive to HFI.[34] Especially, they obtained that the saturated magnetoconductance value is independent of HFI, which is consistent with the experimental observation (see Fig. 3 in Ref. [34]). In our work, the strong HFI reduces the influences of the magnetic field. As shown in Fig. 4, OMAR changes when we change the strength of HFI. For a strong HFI, it is found that a large magnetic field is needed for the MR's value to saturate. In addition, we note that the saturated MR value is independent of HFI. This result is in good agreement with the experimental measurement.[33]

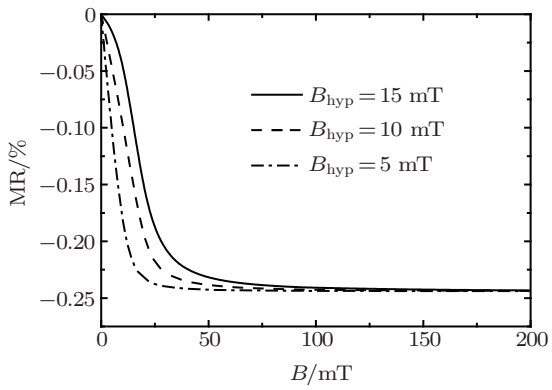


Fig. 4. Dependence of MR on B under different hyperfine fields $B_{\rm hyp}$. The parameters are set as $\sigma = 0.05$ eV, $E = 9.17 \times 10^5$ V/cm, $t_0 = 0.1$ eV, and $\gamma = 1$ nm^{-1}.

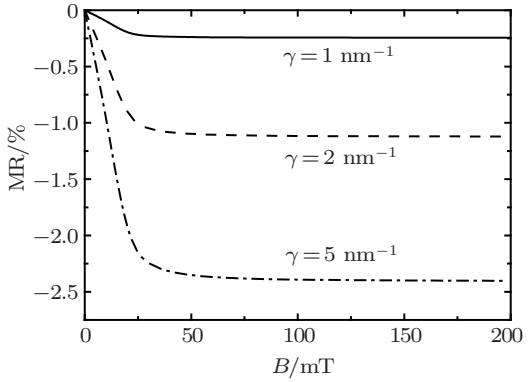


Fig. 5. Dependence of MR on B under different localization factors γ. The parameters are set as $\sigma = 0.05$ eV, $E = 9.17 \times 10^5$ V/cm, $t_0 = 0.1$ eV, and $B_{\rm hyp} = 10$ mT.

In normal inorganic semiconductors, the carriers are usually extended electrons or holes, which are extended over the whole crystal lattice. While in OSCs, the charged carriers are localized polarons due to the strong electron-lattice interaction. The difference in carrier property results in the different transport characteristics. In the present model, the localization of a polaron is reflected by parameter γ which is the inverse localization length of the wave functions. A small γ means a large polaron and so a large hopping integral between molecules. We can see from Eq. (4) that the mobility decreases exponentially with the localization of the polaron. As shown in Fig. 5, the MR value becomes larger for a stronger localized polaron. It indicates that, in organic materials, the MR should be much more apparent than in their inorganic counterparts.

The temperature dependence of the MR on the magnetic field is shown in Fig. 6. In our simulation, the MR value is negative at a low temperature under these parameters. When the temperature becomes high, the MR's absolute value decreases. Finally, the MR value becomes positive at a higher temperature. We set $\hat{\sigma}=\sigma/k_{\rm B}T$ and we can see that the temperature also influences the system's disorder property. As the temperature increases, the value of $\hat{\sigma}$ drops. Thus the system's disorder strength decreases. Therefore, the results in Fig. 6 are consistent with those in Fig. 3, which shows the MR dependence of energetic disorder widths. Besides, the temperature's influence on spin relaxation is also important,[35] and we will investigate the relative effect on the MR in future.

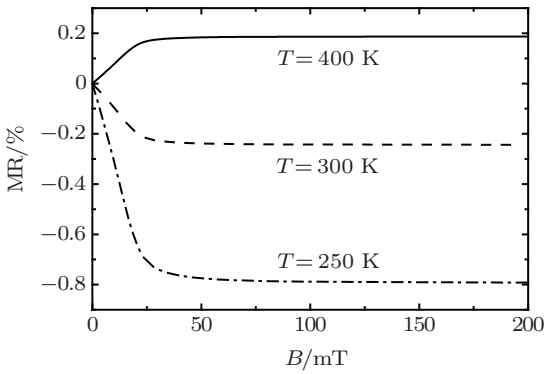


Fig. 6. Dependence of MR on B under different temperatures. The parameters are set as $\sigma = 0.05$ eV, $E = 9.17 \times 10^5$ V/cm, $t_0 = 0.1$ eV, $B_{\rm hyp} = 10$ mT, and $\gamma = 1$ nm^{-1}.

4. Conclusion

We use the ME to calculate the mobility of the system. Under a low carrier density, the polaron is assumed to hop with its spin orientation unchanged. By choosing suitable parameters, the theoretical simulation is well consistent with the experimental observation. It is found that OMAR is dependent upon the material properties as well as the operating conditions. Our simulation shows that the sign of OMAR changes from negative to positive as the electric field increases. Besides, the sign inversion of MR also happens when energetic disorder parameter decreases. MR will reach its saturation slower for a stronger HFI, which reveals the importance of HFI in OMAR. As the carriers in OSCs are localized polarons, it is found that the MR value becomes larger for a stronger localized polaron. It explains why an apparent MR is obtained

in an OSC rather than in its inorganic counterpart. Besides, in our simulation, the MR's temperature dependence is consistent with that of the energetic disorder widths dependence.

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Phys. Rev. B **74**, 045213 – Published 26 July 2006

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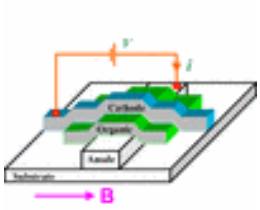
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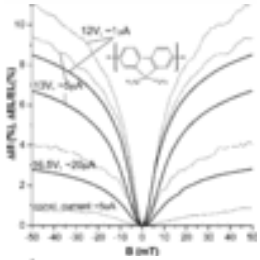
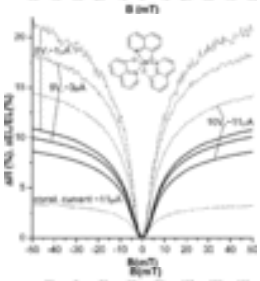
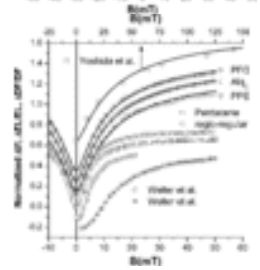
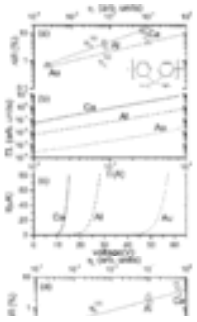
Abstract
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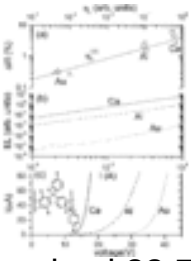
Abstract

We explore the possibility that hyperfine interaction causes the recently discovered organic magnetoresistance (OMAR) effect. We deduce a simple fitting formula from the hyperfine Hamiltonian that relates the saturation field of the OMAR traces to the hyperfine coupling constant. We compare the fitting results to literature values for this parameter. Furthermore, we apply an excitonic pair mechanism model based on hyperfine interaction, previously suggested by others to explain various magnetic-field effects in organics, to the OMAR data. Whereas this model can explain a few key aspects of the experimental data, we uncover several fundamental

contradictions as well. By varying the injection efficiency for minority carriers in the devices, we show experimentally that OMAR is only weakly dependent on the ratio between excitons formed and carriers injected, likely excluding any excitonic effect as the origin of OMAR.
DOI: http://dx.doi.org/10.1103/PhysRevB.74.045213

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- Received 22 February 2006
- Revised 9 May 2006
- Published 26 July 2006

©

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