B mechanism for fringe-field organic magnetoresistance

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Fringe fields emanating from magnetic domain structures can give rise to magnetoresistance in organic semiconductors. In this article, we explain these magnetic-field effects in terms of a $\Delta B$ mechanism. This mechanism describes how variations in magnetic-field strength between two polaron hopping sites can induce a difference in precessional motion of the polaron spins, leading to mixing of their spin states. In order to experimentally explore the fringe-field effects, polymer thin-film devices on top of a rough in-plane magnetized cobalt layer are investigated. The cobalt layer can be described by a distribution of out-of-plane magnetic anisotropies, most likely induced by thickness variations in the cobalt. With a magnetic field perpendicular to the cobalt layer, fringe fields are created because some domains are magnetized out of plane whereas the magnetization of other domains remains approximately in plane. By varying the distance between the polymer layer and the cobalt layer, we find that the magnetoresistance arising from these fringe fields reduces with the gradient in the fringe fields, in agreement with the $\Delta B$ mechanism.

I. INTRODUCTION

Many organic semiconducting devices exhibit intrinsic magnetic-field effects (MFEs) in their conductive or luminescent properties at low magnetic fields [1–7]. This makes organic semiconductors interesting for magnetic-field sensing applications [8]. Local hyperfine fields are involved in the origin of these magnetic-field effects [5]. These hyperfine fields can cause mixing, or intersystem crossing, of spin states. The suppression of this spin mixing by an applied magnetic field is believed to be responsible for the magnetic-field effects [9,10]. Unfortunately, relying on hyperfine fields only offers very little control over the amount of spin mixing. They are determined by the organic material itself and vary only little between different materials. However, there are also other sources of spin mixing. In particular, the (inhomogeneous) fringe fields emanating from magnetic domains or nanoparticles could play a role similar to that of hyperfine fields [11]. Such fringe fields depend on the properties of the magnetic material, which can be controlled, for example, by patterning.

Recently, this idea was applied to organic semiconductors by Wang et al. [12] In their work the magnetoconductance (MC) in an organic device on top of a ferromagnetic layer was shown to depend on the magnetized state of the layer. In the presence of fringe fields emanating from the magnetic domains in this layer, the magnetoconductance was found to decrease significantly. Control experiments showed that the observed effect was not related to spin injection. An explanation based on mixing of spin states of a spin pair on adjacent sites prior to their reaction (“two-site model”) by differently oriented magnetic fields at these sites was dismissed by the authors because of the large correlation length of the fringe fields, resulting in almost completely aligned fields at adjacent sites. On the other hand, Harmon et al. recently suggested the fringe-field gradients might cause additional spin mixing [13]. These gradients are on the order of a few millitesla per nanometer. As mentioned by the authors, assuming that the polarons are separated by just a few nanometers, this means the difference in magnetic-field strength felt by the polarons due to the fringe fields is comparable or even larger than the size of the hyperfine fields.

In this paper, we will show that a straightforward two-site model can explain the enhanced spin mixing due to the fringe-field gradients, even at large applied magnetic fields. A small difference in magnetic field $\Delta B$ at the two sites can lead to a difference in precession frequency of the two spins and hence to mixing between their spin states. Because this mechanism is similar to the so-called $\Delta g$ mechanism, we coin it the $\Delta B$ mechanism. We investigate devices with a magnetic layer causing fringe fields in the active organic layer. By increasing the distance between the magnetic and the active layers the magnetoresistance caused by the fringe fields decreases. The reduction of this fringe-field magnetoresistance is found to correlate well with the calculated distance dependence of the fringe-field gradients emanating from the magnetic layer.

II. $\Delta B$ MECHANISM

In a two-site model, magnetic-field effects, such as magnetoresistance are generally described by the alignment of the effective magnetic fields at the two sites when an external magnetic field is applied [10,14]. When these sites are occupied by two spin-1/2 polarons subject to random hyperfine fields, the spin mixing between the singlet $S$ and $T_0$ triplet with the $T_-$ and $T_+$ triplet states is suppressed due to this alignment. However, even with perfectly aligned magnetic fields spin mixing can still take place between the $S$ and the $T_0$ states. A reaction between the polarons can take place by hopping of one of the polarons to the site of the other. When the hopping rate is much slower than the hyperfine precession frequency and the singlet and $T_0$ triplet are exactly degenerate, complete mixing takes place. When either of these two criteria is violated, the amount of spin mixing becomes dependent on the difference in precession frequency.

To understand why this is the case, consider an electron or hole polaron in a magnetic field $B$. The spin of the polaron will precess about the magnetic field with a frequency given
Here we consider an electron and a hole that are about to form an exciton, but the obtained results are also valid for two electron or hole polarons forming a bipolaron. Experimental results suggest that magnetic-field-dependent singlet and triplet exciton formation is responsible for the large magnetic-field effects observed in several organic light-emitting devices (OLEDs) [6,16]. An increase in the fraction of formed singlet excitons will be accompanied by a decrease in the fraction of formed triplet excitons on application of a magnetic field [10]. The long-living triplet excitons influence the polaron transport, which results in the observed magnetoresistance [2,3,6]. Therefore, we assume that the magnetic-field-dependent singlet fraction \( \chi^S \) is a measure for the magnetoresistance in the OLEDs studied in this paper.

We assume that the g-factor difference between the polarons is negligible and that the effective magnetic fields at the two sites are perfectly aligned (here chosen in the \( z \) direction). In experimental situations this latter assumption is valid as long as the difference in angle between the effective magnetic fields at neighboring polaron sites is small. We argue that this situation is valid for the experimental work described in the remainder of this paper because large external magnetic fields are used to induce fringe fields, which effectively reduce any (significant) difference in angles induced by the fringe fields. We found through numerical calculations that small differences in angles between the magnetic fields at neighboring sites lead to negligible spin mixing. In this section, we therefore assume perfect alignment in order to focus on an intuitive and analytical derivation. To continue, the spin Hamiltonian of the polaron system described above is given by

\[
H = g \mu_B B e \chi_z / \hbar + g \mu_B B_h \chi_z / \hbar,
\]

in which \( S_{z, \text{eb}} \) is the spin operator in the \( z \) direction for the electron (hole). The steady-state fraction of singlet excitons \( \chi^S \) that is formed from polaron pairs can be derived using a stochastic Liouville equation [10,14]. We investigate polaron pairs that are formed with a rate \( k_u \) and from which singlet and triplet excitons are created with rates \( k_S \) and \( k_T \), respectively. Because large aligned magnetic fields are assumed, mixing with \( T_S \) and \( T_T \) is suppressed due to the large Zeeman interaction. Therefore, these states can be neglected in the derivation. The steady-state density operator is found by demanding time independence of the stochastic Liouville equation,

\[
0 = \frac{\partial \rho}{\partial t} = -i \hbar [H, \rho] - \frac{1}{2} \{\Lambda, \rho\} + \Gamma,
\]

where \( \Lambda = \Sigma \chi^S \chi^T / \Lambda \) and \( \Gamma = k_u / 4 \), where \( I \) is the identity operator. As formation rates, we take \( k_T = k_{\text{hop}} / \gamma \) and \( k_S = k_{\text{hop}} \). Here \( \gamma \) is taken as the ratio between the singlet and the triplet exciton formation rates, which have to be different to obtain magnetic-field-dependent exciton fractions [10]. From the obtained density matrix one can straightforwardly derive the singlet excitation fraction \( \chi^S \), which is given by

\[
\chi^S = \frac{\gamma}{1 + \gamma} \left( 2 - \frac{\gamma - 1}{(g \mu_B B \gamma / k_{\text{hop}})^2 + \gamma} \right).
\]

Figure 1(b) shows how \( \chi^S \) evolves with the relative hopping rate \( r \) for \( B = 10 B_{\text{ho}} \) (solid black curve) in which \( r \) is defined by

\[
\omega_B = g \mu_B B / \hbar,
\]

in which \( g \) is the \( g \) factor of the polaron, \( \mu_B \) is the Bohr magneton, and \( \hbar \) is the Planck constant. A difference in precession frequency between two polarons at adjacent sites can to linear order be written as

\[
\Delta \omega_B = \frac{\mu_B}{\hbar} (B \Delta g + g \Delta B).
\]

The first term \( B \Delta g \) describes how a difference in \( g \) factor between the two polarons \( \Delta g \) causes a difference in precession frequency proportional to the applied magnetic field. The resulting dephasing of the polaron pair’s spin configuration leads to an increase in spin mixing between \( S \) and \( T_0 \), partially compensating for the loss in spin mixing due to the alignment of the effective magnetic fields at the two sites [14]. In the literature, this is referred to as the \( \Delta g \) mechanism, which has been used to explain magnetic-field effects observed at large applied fields [6,15]. The second term \( g \Delta B \) could arise due to fringe fields, where \( \Delta B \) is the difference in magnetic field at the two sites. Similar to the \( \Delta g \) mechanism, a difference in precession frequency due to \( \Delta B \) should also result in additional spin mixing. Figure 1(a) illustrates the spin dephasing of two polarons at adjacent sites due to a difference in magnetic field \( \Delta B \). In analogy to the \( \Delta g \) mechanism, we call this the \( \Delta B \) mechanism.

It is possible to derive an analytical expression for the \( \Delta B \) mechanism and its corresponding magnetic-field effect. Here we consider an electron and a hole that are about
as $r = k_{\text{hop}}/\omega_{\text{hf}}$ with the hyperfine frequency $\omega_{\text{hf}} = g\mu_B B_{\text{hf}}/\hbar$ at a hyperfine field $B_{\text{hf}}$. Because $\gamma$ is generally considered to be close to unity [10], we have linearized $\chi_S$ with respect to $(\gamma - 1)$ in order to show the most general result. The statistical singlet fraction is 1/4, but spin mixing changes it to $\gamma/2(1 + \gamma)$ at slow hopping ($r \ll 1$). When the hopping rate increases, the exciton formation rate becomes faster than the precession frequency, thereby quenching the spin mixing. The transition occurs around $r = \Delta B \sqrt{\gamma}/B_{\text{hf}}$.

In the absence of fringe fields, the hyperfine fields are still capable of causing mixing between the $S$ and $T_0$ states. With a large external magnetic field $B_z$, $\Delta B$ is given by the difference in the $z$ component of the hyperfine fields. $\Delta B$ can then be described with a Gaussian distribution with standard deviation $2B_{\text{hf}}$. Averaging Eq. (5) over the hyperfine fields is performed analytically (not shown here). As can be seen in Fig. 1(b), the transition from the slow-hopping to the fast-hopping result is slightly less abrupt than with a fixed $\Delta B$. Note that in reality the gradient in the fringe fields experienced by all the polarons in a device may also have a certain distribution.

In the transition from slow to fast hopping, the spin mixing is gradually quenched. When polarons form excitons with intermediate-hopping rates, we find that the amount of spin mixing and thus the singlet exciton fraction depends on the strength of $\Delta B$. By controlling the fringe fields, e.g., with domains in a magnetic layer, the singlet and triplet exciton fractions can be influenced and hence the magnetoresistance.

The above describes the main aspects of the $\Delta B$ mechanism, but other ingredients can be added. For example, incomplete spin mixing arises at slow-hopping rates due to spin-spin interactions in the form of dipolar coupling or exchange interaction because this lead to an energy difference between $S$ and $T_0$. Dipolar coupling generally dominates the exchange interaction at longer distances [17]. Therefore, we will here consider how dipolar coupling influences the $\Delta B$ mechanism. For the case that the two polarons are separated from each other in the $z$ direction (assuming polaron transport takes place along the magnetic field), the following term has to be added to the Hamiltonian:

$$H_{\text{dip}} = -2g\mu_B DS_zc_zS_z/b,$$

in which the dipolar coupling strength is given by $D = \mu_0\xi g\mu_B/(4\pi R^3)$. For a typical separation distance $R$ of 1.5 nm [18] and a hyperfine field strength of 1 mT [5], the dipolar coupling strength would be equivalent to $D = 0.5B_{\text{hf}}$, but the exact coupling strength is not critical for the present paper. Similar to the derivation above, it is possible to find a density matrix from the steady-state solution of Eq. (4). The result for $\chi_S$ with dipolar coupling is also shown in Fig. 1(b) (dashed lines). Essentially, the spin-spin interactions suppress the spin mixing and shift the singlet fraction towards the statistical ratio of 1/4. The amount of suppression depends on the relative strength $D/\Delta B$, which explains why the effect is best observed for the calculation with the hyperfine fields. With the addition of a larger $\Delta B$ due to the fringe fields, spin mixing can thus also be increased at slow-hopping rates.

The singlet and triplet fractions and other quantities that depend on them, such as the current, are magnetic-field dependent. The singlet fractions derived above are the saturated values at large magnetic fields. As we have shown, these values depend on the size of $\Delta B$, which can be increased by “turning on” fringe fields. One can define a relative magnetic-field effect caused by the fringe fields as $(\chi_{S,\text{fringe}} - \chi_{S,\text{fringe}})/\chi_{S,\text{fringe}}$. This MFE is shown in Fig. 1(c) as a function of the relative hopping rate. The size of this MFE depends on the hopping rate, spin-spin interactions, and the dynamics of the spin-dependent processes (the latter is included in $\gamma$ for the case of exciton formation).

### III. FRINGE-FIELD MAGNETORESISTANCE

In order to investigate the influence of fringe fields on the magnetoresistance, we fabricated devices with a layer stack similar to OLEDs, only with a magnetic cobalt layer inserted close to the organic layer. In contrast to previous work [12,19,20], we use a single (rough) cobalt layer with an in-plane magnetization due to shape anisotropy. The cobalt layer can be placed on a seed layer underneath the functional device or on top of the device. The experiments in this paper will focus on devices with a cobalt layer on top. By applying a magnetic field in plane or out of plane the fringe fields can be turned on and off, respectively, making this a convenient system to study the magnetoresistance arising from fringe fields. A superconducting quantum interference device (SQUID) was used to characterize the magnetic properties of the cobalt layers, and atomic force microscopy (AFM) was used to investigate their roughness.

The results of the magnetization and roughness are shown in Figs. 2(a) and 2(b) for a cobalt layer on top of an aluminum seed layer. We note that we found similar results for cobalt layers on top of an organic layer. The organic layer may also induce height variations in the aluminum and cobalt layers which do not result in significant local variations in shape anisotropy. Therefore, we will here focus on the magnetic characterization and thickness variations in the cobalt (and aluminum) layer only. Figure 2(a) shows that when the magnetic field is applied in plane we find an abrupt switch with a coercive field of 1.2 mT as expected from the shape anisotropy. When the magnetic field is applied out of plane, the magnetization slowly reaches saturation at more than 1.5 T. For an ideal in-plane magnetized layer the magnetization should follow a linear function according to Stoner-Wohlfarth theory [21]. However, we find two regimes; at fields roughly below 200 mT the magnetization changes more rapidly than at higher fields. The existence of these two regimes can be explained from the roughness of the layer, shown in Fig. 2(b), leading to different local out-of-plane shape anisotropies. In some regions the layer is thicker than in other regions, and in these regions the magnetization may be pulled out of plane more easily. The fact that there are two distinct regimes in the magnetization is indicative of two separate distributions of shape anisotropies, corresponding to the thick and thin regions in the film. This means that at a magnetic field around the transition between these two regimes, some parts of the cobalt layer are completely magnetized out of plane, whereas the other parts hardly are magnetized. The implications for the fringe fields emanating from the cobalt layer are as follows: For in-plane applied magnetic fields all domains are magnetized in the same direction, resulting in a minimum of fringe fields emanating from the cobalt layer. With a sufficiently large
relative MC at 500 mT for varying aluminum spacer layer thickness. The red line is a fit with a \( 1/(d + d_0)^2 \) function where we found \( d_0 = 15 \) nm for the case shown.

out-of-plane applied magnetic field, the different magnetic domains will induce additional fringe fields, which should affect the magnetoresistance.

To investigate the effect of such a cobalt layer on the magnetoresistance, typical devices were fabricated. The devices consist of glass substrates with prepatterned indium tin oxide and 40 nm of spin-coated poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) as the bottom electrode. The active layer is composed of a phenyl-substituted poly(p-phenylene vinylene) (PPV) copolymer also known as “superyellow” PPV, which is known to exhibit large magnetic-field effects \([4]\). The top contact consisting of LiF (1 nm)/Al (10–100 nm) was thermally evaporated inside a high-vacuum system in another glovebox. On top of the LiF/Al layer we thermally evaporated 15 nm of cobalt, which was capped with 100 nm of Al. The first aluminum layer acts both as an electrical contact and as a spacer layer between the cobalt and the organic layer. All measurements were performed inside a nitrogen-filled glovebox where the sample is placed between the poles of an electromagnet. The current density \( J \) is measured with a Keithley 2400 series source meter. The MC is defined as \( \Delta J(B) - \Delta J(0) \)/\( J(0) \).

Figure 2(c) shows the MC for a magnetic field applied parallel and perpendicular to the plane of the device. These two orientations result in distinctly different responses. For a parallel orientation we find a monotonously increasing MC, which is generally found in the literature for standard OLED devices \([1,2,4,22]\). This corresponds to the case where all domains are magnetized in plane, minimizing the fringe fields. For a perpendicular magnetic field the MC first follows this trend, but then deviates from it and eventually even decreases again. This deviation occurs at the transition between the two regimes in the magnetization measurement in Fig. 2(a) where domains with different magnetizations are present leading to the creation of additional fringe fields. Wang et al. also found a reduction in the MC when magnetic domains in the magnetic layer are created. This reduction is indicative of enhanced spin mixing due to the presence of fringe fields as can be explained with the \( \Delta B \) mechanism. At larger fields we expect the perpendicular MC to return to the value of the parallel MC because the entire cobalt layer will then be magnetized in the same direction, leading to vanishing fringe fields. We forego the verification of this interesting feature as measuring at such large fields would require a complete redesign of our setup and because we believe it is beyond the scope of the current paper.

The same measurements were performed with varying aluminum spacer thicknesses. Figure 2(d) shows the relative difference in MC at 500 mT (\( \delta MC = (MC_\parallel - MC_\perp)/MC_\parallel \)) as a function of the spacer thickness. The \( \delta MC \) is normalized with respect to the regular (hyperfine) MC given by \( MC_\parallel \) to account for device variations in the MC. As the organic layer is further separated from the magnetic layer, the fringe fields in the organic layer will reduce. Accordingly, the relative MC reduces with increasing spacer thickness as can be observed in Fig. 2(d). We find that this reduction can be described with a \((d + d_0)^{-2}\) function, where \( d \) is the spacer thickness and \( d_0 \) is an offset distance. Note that the relative MC will not completely approach zero at large \( d \) because of a small intrinsic angle dependence with a \( \delta MC \) on the order of 1% \([22]\). In the next section, we will show that the gradients in the fringe fields are also expected to follow a similar dependence as a function of the distance away from the magnetic layer. The \( d^{-2} \) dependence can readily be understood intuitively. For a magnetic dipole, the magnetic field reduces with \( 1/R^3 \), where \( R \) is the distance from the dipole. After performing a surface integral, the magnetic field depends on distance \( d \) to the plane as \( 1/d \) and its gradient as \( 1/d^2 \). The offset distance could arise because the relevant polaron pairs are not positioned at \( d = 0 \) but some distance away from the plane into the organic layer. Therefore, \( d_0 \) might be related to the location of the recombination region. We will show in the next section that \( d_0 \) can also be related to the magnetic domain structure. Finally, we note that our results show that with a hybrid device the weak intrinsic angle dependence of organic magnetoresistance can be enhanced by the insertion of magnetic materials with angle-dependent fringe fields.

IV. FRINGE-FIELD MAGNETORESISTANCE CALCULATIONS

In order to investigate how the fringe-field magnetoresistance depends on the distance to the magnetic layer, we use the \( \Delta B \) mechanism and a straightforward model of the magnetic layer. Magnetic fields emanating from a magnetic layer can easily be calculated with a discrete dipole approximation where the magnetic layer is divided into (small) segments that are treated as single magnetic dipole moments. The magnetic fields are then calculated by summing over all
The average gradient \( \sigma_z \) in the fringe fields as a function of the distance \( z \) above the magnetic layer increases as \( z \) approaches \( z_0 \) and then saturates at small \( z \). Therefore, in order to be in the linear regime, one needs either small \( \Delta B \) or a large hopping rate [see \( \hbar_{\text{hop}} \) in Eq. (5)]. We numerically simulated the magnetic-field effect, defined as the relative change in singlet fraction, corresponding to the fringe-field distributions calculated in this section. For this purpose we used a two-site model similar to the analytical calculations, including the \( T_+ \) and \( T_- \) states. All magnetic-field orientations were allowed, i.e., we did not make the assumption of aligned magnetic fields at the sites. Figure 4 shows the results as a function of the height above the magnetic layer for different hopping rates. Taking the perpendicular MFE, \( \chi \) is \( (\chi_{\text{fringe},B} - \chi_{\text{fringe},0})/\chi_{\text{fringe},0} \) and the parallel MFE, \( \chi \) is \( (\chi_{\text{fringe},B} - \chi_{\text{fringe},0})/\chi_{\text{fringe},0} \) then the relative magnetic-field effect is \( \Delta \text{MFE} = (\chi_{\text{fringe},B} - \chi_{\text{fringe},0})/\chi_{\text{fringe},0} \), which corresponds to the experimental definition used in Fig. 2(d). For lower hopping rates, the \( \Delta \text{MFE} \) does not follow the \( 1/z^2 \) dependence of the fringe-field gradient. Instead it reaches only a saturated value at small \( z \), approximate \( 1/(z + z_0)^2 \) dependence, where \( z \) is the distance to the plane (which is related to the aluminum thickness \( d \) in the experiments). Different orientations of the domains do not significantly influence the \( z \) dependence, except for the total amplitude. Interestingly, we also find an offset \( z_0 \) is required to obtain a good fit, similar to the experimental results.

This offset is related to the size of the domains and requires some explanation. Due to the nature of the dipole, the magnetic field is zero above an infinite magnetic film with a single magnetization. Close to a finite plane it is approximately constant. The \( 1/z \) dependence of the magnetic field (and thus a \( 1/z^2 \) gradient) only arises above a change in magnetization, i.e., at the edge of two domains. By taking the average over many domains in our calculations, the \( 1/(z + z_0)^2 \) dependence is found for the gradients. For smaller domain sizes, so on average more domain edges, we find that the offset \( z_0 \) decreases, thereby approaching the expected \( 1/z \) dependence. More specifically, the constant \( z_0 \) scales with the typical domain size, provided that the probing height scales equivalently.

These results indicate that the magnetoresistance caused by the magnetic layer in our experiments is proportional to the size of the fringe-field gradients. This implies that we are in the linear regime of the \( \Delta B \) mechanism. For larger \( \Delta B \) the singlet fraction in Eq. (5), and hence the magnetic-field effect, will saturate at fixed \( r \). Therefore, in order to be in the linear regime, one needs either small \( \Delta B \) or a large hopping rate [see \( \hbar_{\text{hop}} \) in Eq. (5)]. We numerically simulated the magnetic-field effect, defined as the relative change in singlet fraction, corresponding to the fringe-field distributions calculated in this section. For this purpose we used a two-site model similar to the analytical calculations, including the \( T_+ \) and \( T_- \) states. All magnetic-field orientations were allowed, i.e., we did not make the assumption of aligned magnetic fields at the sites. Figure 4 shows the results as a function of the height above the magnetic layer for different hopping rates. Taking the perpendicular MFE, \( \chi \) is \( (\chi_{\text{fringe},B} - \chi_{\text{fringe},0})/\chi_{\text{fringe},0} \) and the parallel MFE, \( \chi \) is \( (\chi_{\text{fringe},B} - \chi_{\text{fringe},0})/\chi_{\text{fringe},0} \) then the relative magnetic-field effect is \( \Delta \text{MFE} = (\chi_{\text{fringe},B} - \chi_{\text{fringe},0})/\chi_{\text{fringe},0} \), which corresponds to the experimental definition used in Fig. 2(d). For lower hopping rates, the \( \Delta \text{MFE} \) does not follow the \( 1/z^2 \) dependence of the fringe-field gradient. Instead it reaches only a saturated value at small \( z \), approximate \( 1/(z + z_0)^2 \) dependence, where \( z \) is the distance to the plane (which is related to the aluminum thickness \( d \) in the experiments). Different orientations of the domains do not significantly influence the \( z \) dependence, except for the total amplitude. Interestingly, we also find an offset \( z_0 \) is required to obtain a good fit, similar to the experimental results.

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i.e., at large $\Delta B$. For increasing hopping rates this does not happen, and the distance dependence seems to approach the $1/z^2$ dependence.

**Roughness**

So far, we considered the roughness in the magnetic layer as a source of spatially fluctuating magnetic anisotropy, leading to the formation of magnetic domains and corresponding fringe fields. What we did not consider is that such roughness might on itself induce fringe fields independent of the magnetic domain structure. Our silent assumptions have been twofold. Either these roughness-induced fringe fields are negligible at distances above the minimum spacer layer thickness of 10 nm used in the experiments, or otherwise, these fringe fields are independent of the magnetization direction in which case they function as a constant source of spin mixing which divides out when considering $\delta MC$.

To investigate these assumptions, we performed magnetic dipole calculations on magnetic films with thickness and height variations. For comparison, all parameters are kept similar to those used in the calculations shown in Fig. 3. The roughness can be implemented in the calculations either as thickness variations in the magnetic film, which we propose are responsible for variations in magnetic anisotropy, or as height variations induced by roughness in the underlying polymer and aluminum layers. Additionally, the roughness can be considered to consist of sharp local variations or smooth transitions. The measured AFM topology shown Fig. 2(b) suggests the latter situation is most likely for the layers used in the experiments, but we will investigate both cases here.

Figure 5(a) shows calculations of the average fringe-field gradients as a function of distance above the layer with either discrete steps or sinusoidal variations in the film thickness (both the same total volume of magnetic material) and for both cases with either in-plane or out-of-plane magnetization direction. These results clearly show that sharp features induce stronger gradients than smooth features. The sharp features also show fringe-field gradients that depend strongly on the magnetization direction.

Figure 5(b) shows the average fringe-field gradients induced by height variations, e.g., caused by roughness in the underlying layers. The reader should be aware that a continuously flat magnetic film induces no gradients, therefore, the gradients induced by height variations decrease rapidly with distance. Nevertheless, sharp features can induce significant gradients close to the magnetic film.

Given the fact that a relatively smooth roughness produces small gradients independent of the magnetization direction, we can consider the gradients induced by the roughness as an angle-independent contribution to the experimental results described in this paper. Our interpretation thus remains valid: The large anisotropic fringe-field effects in the MC are most likely caused by differences in magnetic anisotropy, leading to different magnetic domains under an applied magnetic field.

**V. CONCLUSION**

We have presented a mechanism, coined the $\Delta B$ mechanism, for a new type of magnetoresistance in organic semiconductors that is based on magnetic-field gradients. Magnetic-field gradients can be created with fringe fields emanating from a magnetic layer. We created such gradients by magnetizing a rough cobalt thin film out of plane, which was placed in proximity to a conductive organic layer. Our experimental results combined with calculations of the magnetic fields above such a magnetic layer show that the resulting magnetic-field effects are proportional to the gradients in the fringe fields. We conclude that fringe fields can be used to control magnetic-field effects in organic semiconductors and that these effects can be explained in a natural way with the $\Delta B$ mechanism.

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