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Eur. Phys. J. E **31**, 153–157 (2010)

DOI: 10.1140/epje/i2010-10555-9

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Received 4 August 2009

Published online: 1 March 2010 – © EDP Sciences / Società Italiana di Fisica / Springer-Verlag 2010

**Abstract.** We examine the magnetoresistance (MR) of conducting polymers with interest paid on the role of structural flexibility. Through Monte Carlo simulation and Green function method, we evaluate the electric transmission for a variety of polymer configurations. It is found that for a single polymer the transmission displays a complex oscillation and also a parity-dependent periodicity. For an ensemble of polymers the averaged transmission yields the nonlinear behavior of MR under varying magnetic fields. Interestingly, more flexible polymers are shown to achieve higher MR, depending on the population and the size of the loops.

Conducting polymers are materials electrically similar to inorganic semiconductors but flexible like rubber; the  $\pi$ -orbital overlap paves the way for electron conduction and the low-elastic modulus allows easy deformation to yield a variety of conformations. These intriguing properties have drawn much interest and researches have been done on their device applications and fundamental properties [1,2]. More recently, remarkable progress has been made in nano-scale sample fabrications, opening the field of molecular electronics. The idea is to take advantage of the self-assembly properties of molecules forming highly ordered nano-sized structures spontaneously. Carbon nanotube and DNA-based transistor can be mentioned as successful examples to realize the single-polymer electronics [3–5]. Also for other conductive polymers such as polyacetylene and polyaniline, a technique for isolating a polymer from the interpolymer attraction has been developing [6] to elevate the importance of research on polymer nanowires.

In theoretical models, conducting polymers can be viewed as one-dimensional tight-binding chains, where the structural flexibility should be carefully considered. For instance, polymer chains take coiled conformations, where the bending energy cost determines the orientational correlation length, the so-called persistence length, ranging from micrometers to nanometers [7–10]. When a polymer is longer than the persistence length, uncorrelated portions of a polymer can join at a link(s) to form a loop(s). Such crosslinks influence the electrical properties. On the one hand, it introduces off-diagonal disorder to cause electron localization, and on the other hand, it enhances the effective dimensions of the system. Recently, the resulting behaviors from the two competing effects are elaborated

to find phenomena of localization softening [11]. Also a polymer chain with crosslinks accommodates non-simply-connected paths, and therefore, quantum interference effects come into play, which should be more pronounced in the presence of magnetic fields [12–15].

The purpose of this work is to study the electric properties of conducting polymers in the presence of magnetic fields by explicitly considering the polymer flexibility. To this end, we generate polymer configurations via Monte Carlo simulation according to the wormlike chain model. The electric resistance of a given polymer configuration is evaluated by Green function method. We find that flexible polymers indeed have loops enclosing magnetic flux to yield conductance oscillations, as anticipated. For a single polymer, in a stretched state having a single loop, we find an intriguing parity effect in the oscillation harmonics. For a coiled state with multi-loops, the oscillation patterns become complex and their Fourier components have a long-range tail in a high “frequency” regime. Also we evaluate the conductance average for a collection of polymers, which is shown to nonlinearly increase with magnetic fields at weak fields, and become saturated at strong fields. Interestingly, the increment of conductance with respect to zero-field conductance is enhanced by the flexibility so that highly flexible polymers achieve giant magnetoresistance.

We start with a Hamiltonian describing noninteracting electrons moving along a single conducting polymer chain placed in a uniform magnetic field,

$$\mathcal{H}_e = \sum_i \epsilon_i c_i^\dagger c_i - \sum_{\langle i,j \rangle} (t_{ij} c_i^\dagger c_j + t_{ji} c_j^\dagger c_i), \quad (1)$$

where  $c_i^\dagger$  and  $c_i$  is a fermion creation and annihilation operator at site  $i$ , respectively. The on-site energy  $\epsilon_i$  can be

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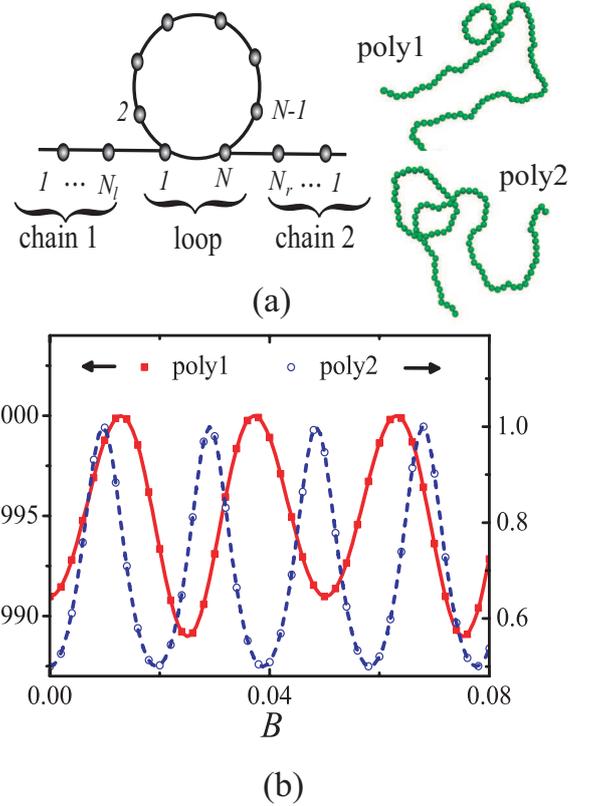
monomer dependent, but for simplicity we take uniform  $\epsilon_i$  and focus on the geometry-induced randomness in a coiled polymer. The second term represents electron hoppings between nearest-neighboring monomers. In the presence of magnetic flux, phase factors accompany the hopping integral as  $t_{ij} = |t_{ij}| \exp(iA_{ij})$ , where  $A_{ij} = (2\pi/\Phi_0) \int_i^j \mathcal{A} \cdot d\ell$  with  $\mathcal{A}$  and  $\Phi_0$  being the vector potential and the flux quantum, respectively. Although the hopping amplitude generally depends on the polymer geometry such as intermonomer distance and local orientations [16–18], we simply take uniform  $|t_{ij}| = t$  for  $i - j = \pm 1$  and  $|t_{ij}| = t_\perp$  for hoppings between two sites at a crosslink. Using  $t$  as an energy unit, we take  $t_\perp = 0.1$ , an eligible value for  $t_\perp$  presumably weaker than  $t$ . To consider the structural flexibility of polymers, we allocate the elastic energy for bent configurations by the wormlike chain model [19],

$$\mathcal{H}_s = \frac{\kappa}{2a^2} \sum_n (\mathbf{r}_n - \mathbf{r}_{n-1})^2. \quad (2)$$

Here we consider a polymer composed of  $N$  monomers with the position vector of the  $n$ -th monomer defined by  $\mathbf{R}_n$ , and  $\mathbf{r}_n = \mathbf{R}_{n+1} - \mathbf{R}_n$ . In particular, having an experiment on a substrate in mind, we consider polymers in two dimensions. Also, letting the intermonomer distance fixed as  $|\mathbf{r}_n| = a$ , nonadjacent pairs of monomers satisfying  $|\mathbf{R}_n - \mathbf{R}_{n' \neq n \pm 1}| \leq a/2$  are regarded to be in contact with each other to form a crosslink. How to define the link is in fact arbitrary and possibly changes the details of loop distributions. However, since we examine all the polymer configurations by varying flexibility, our results to be presented here would remain qualitatively intact. It is worthwhile to mention that eq. (2) yields  $\langle \mathbf{r}_i \cdot \mathbf{r}_j \rangle \sim e^{-ck_B T |i-j|/\kappa}$ , with  $c$  being an irrelevant constant, which indicates that  $\kappa/k_B T$  is a key parameter to determines the angle correlations, and hence the coiling property of polymer structures.

Given the structure Hamiltonian  $\mathcal{H}_s$  with the angular direction of each monomer taken as a dynamic variable, we perform the Monte Carlo (MC) simulation by using the Metropolis algorithm with the total number of monomers  $L = 100$ . Equilibrium polymer configurations are obtained after equilibration for  $10^6$  MC steps. Especially for the ensemble-averaged quantity, we collect configurations at every  $10^4$  MC steps at a given temperature  $T$  measured in units of  $\kappa/k_B$ . For a given polymer configuration, its electronic property can be examined by evaluating the dimensionless conductance or transmission  $g(E, B)$ , where  $E$  is the Fermi energy and  $B$  is the magnetic-field strength (in units of  $\Phi_0/a^2$ ). In the present work, the conductance  $g(E, B)$  is computed at  $E = 0$  for half-filled systems. According to Fisher-Lee relation, the conductance can be given in terms of Green function as  $g(E, B) = \text{Tr}\{G_L^r G_R^r G^a\}$ , with the retarded Green function  $G^r = [(E + i0^+)I - \mathcal{H}_e - \Sigma]^{-1}$ . The self-energy  $\Sigma$  comes from the coupling between the open ends of a polymer and metallic leads, and its imaginary part determines the coupling strength through a relation  $\Gamma = -2\text{Im}\Sigma$ .

We expect that at zero temperature the bending energy cost highly suppresses a curved configuration so that



**Fig. 1.** (Color online) (a) Schematic picture of a conducting polymer with a single loop (left) and configurations generated by the wormlike chain model at  $T = 0.2$  (right); (b) conductance *vs.* magnetic fields for poly1 and poly2, where the analytic calculation (the lines) are in a perfect agreement with numerical results (the points). The oscillation period for poly1 with area  $2\pi S = 125.2$  is shown to be  $1/S \approx 0.05$  while for poly2 with area  $2\pi S = 161.9$  the period is halved as  $1/2S \approx 0.02$ .

a polymer becomes a stiff wire with no loops. For this case, it is obvious that the applied magnetic flux yields null effects. On the other hand, as temperature increases, entropy plays a role to result in coiled configurations. It can be good to start with the simplest structure, that is, a chain with a single loop (see fig. 1). For this system, analytic results for  $g(E, B)$  can be obtained in the following. As shown in fig. 1(a) partitioning the single-loop polymer into chain 1 ( $c_1$ ), loop ( $\ell$ ), and chain 2 ( $c_2$ ) sectors, we write the system Green function to satisfy

$$\begin{pmatrix} G_{c_1} & G_{c_1,\ell} & G_{c_1,c_2} \\ G_{\ell,c_1} & G_\ell & G_{c_2,\ell} \\ G_{c_2,c_1} & G_{\ell,c_2} & G_{c_2} \end{pmatrix} = \begin{pmatrix} E - \mathcal{H}_{c_1} & -\mathcal{H}_{c_1,\ell} & 0 \\ -\mathcal{H}_{\ell,c_1} & E - \mathcal{H}_\ell & -\mathcal{H}_{c_2,\ell} \\ 0 & -\mathcal{H}_{\ell,c_2} & E - \mathcal{H}_{c_2} \end{pmatrix}^{-1}.$$

This is basically Löwdin's partitioning technique [20,21] to facilitate the Green function evaluation. It is easy to see that a loop Green function is given by  $G_\ell = [E - \tilde{H}]^{-1}$ , where  $\tilde{H} = \mathcal{H}_\ell + \Sigma_{c_1} + \Sigma_{c_2}$  with the coupling to the side chains counted by the self-energy corrections:  $\Sigma_{c_i} = \mathcal{H}_{\ell,c_i} \mathcal{G}_{c_i} \mathcal{H}_{c_i,\ell}$ , where  $\mathcal{G}_{c_i} = (E - \mathcal{H}_{c_i})^{-1}$ . Given the loop

Green function and the self-energies, we obtain transmission as

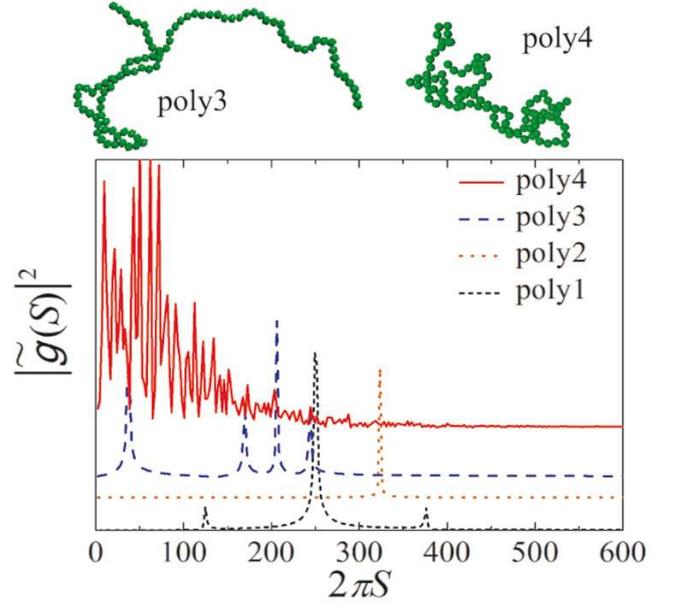
$$g(E, B) = 4 \text{Tr} \{ \text{Im}(\Sigma_{c_1}) G_\ell^{(r)} \text{Im}(\Sigma_{c_2}) G_\ell^{(a)} \}. \quad (3)$$

With the definition of a local coordinate as shown in fig. 1(a), we have  $(\Sigma_{c_1(c_2)})_{x,y} = t^2 (\mathcal{G}_{c_\ell(r)})_{N_\ell(r), N_\ell(r)} \delta_{x,y=1(N)} \equiv \Sigma_{1(2)} \delta_{x,y=1(N)}$  and hence, the transmission simply becomes  $g(E, B) = 4 \text{Im}[\Sigma_1] \text{Im}[\Sigma_2] |(G_\ell)_{1,N}|^2$ . Let us first evaluate the loop Green function with the effective Hamiltonian in a matrix representation:  $(\tilde{H})_{ij} = \Sigma_1 \delta_{i,j=1} + \Sigma_2 \delta_{i,j=N} + \delta_{i,i+1} + \delta_{i+1,i} + t_\perp e^{2\pi i f} \delta_{i=1,j=N} + t_\perp e^{-2\pi i f} \delta_{i=N,j=1}$ , where the total flux  $f$  enclosed by the loop is  $f = BS$ , with  $S$  being the loop area in units of  $a^2$ . Here we have transformed  $\mathcal{H}_e$  via  $\exp[i(2\pi/\Phi_0) \int^i \mathcal{A} \cdot d\ell] c_i = \tilde{c}_i$ , leaving the phase factors all transferred into the hopping terms between the sites at the link as  $t_\perp \exp[2\pi i f \mathcal{A} \cdot d\ell] \tilde{c}_1^\dagger \tilde{c}_N + \text{H.c.}$  It is straightforward to obtain

$$(G_\ell)_{1N} = \frac{1 + B(N, t_\perp) e^{2\pi i f}}{A(N, t_\perp, \Sigma) - 2e^{iN\pi} t_\perp \cos 2\pi f}, \quad (4)$$

where  $A(N, t_\perp, \Sigma) = D_N - D_{N-1}(\Sigma_r + \Sigma_\ell) + (\Sigma_r \Sigma_\ell - t_\perp^2) D_{N-2}$  and  $B(N, t_\perp) = t_\perp e^{i\pi N} D_{N-2}$ . Here  $D_N$  is the determinant of the  $(N \times N)$ -tridiagonal matrix with diagonal and off-diagonal components given by  $E$  and 1, respectively, which after some manipulation can be obtained as  $D_N = \sin(N+1)\theta / \sin \theta$  with  $2 \cos \theta = E$ . On the other hand, the chain self-energy can also be easily evaluated to give  $\Sigma_{2(1)} = S(N_{r(\ell)}, \theta, \Gamma) / S(N_{r(\ell)} + 1, \theta, \Gamma)$ , with  $S(n, \theta, \Gamma) = \sin n\theta - i\Gamma \sin(n-1)\theta$ , and  $N_{r(\ell)}$  being the number of monomers in the right (left) chain. These self-energies together with the Green function given by eq. (4) determine the transmission in eq. (3). The above consideration can be extended to the cases where loops are well separated along the polymer. For example when one more loop is located in the side chain 2, we can decompose the side chain into chain 2 ( $c_2$ ), loop 2 ( $\ell_2$ ), and chain 3 ( $c_3$ ), and yet the Hamiltonian can still be written in the tridiagonal form. For this case, the Green function of chain 2 in eq. (3) is modified as  $\mathcal{G}_2 = (E - \mathcal{H}_2 - \Sigma_{(\ell_2, c_3)})^{-1}$  due to the coupling to the additional loop and chain, where  $\Sigma_{(\ell_2, c_3)} = \mathcal{H}_{c_2, \ell_2} (E - \mathcal{H}_{\ell_2} - \mathcal{H}_{\ell_2, c_3} \mathcal{G}_{c_3} \mathcal{H}_{c_3, \ell_2})^{-1} \mathcal{H}_{\ell_2, c_2}$ , with  $\mathcal{G}_{c_3} = (E - \mathcal{H}_3)^{-1}$ . When the additional loop is introduced in chain 3, it is obvious that the Green function  $\mathcal{G}_{c_3}$  should be modified in the same way as  $\mathcal{G}_{c_2}$ . It appears that the procedure can be done for an arbitrary number of loops. However when many loops are present in the system, they can be entangled, not allowing the simple algebra. Albeit discarding the entanglement, in the presence of many loops numerical evaluation is more effective.

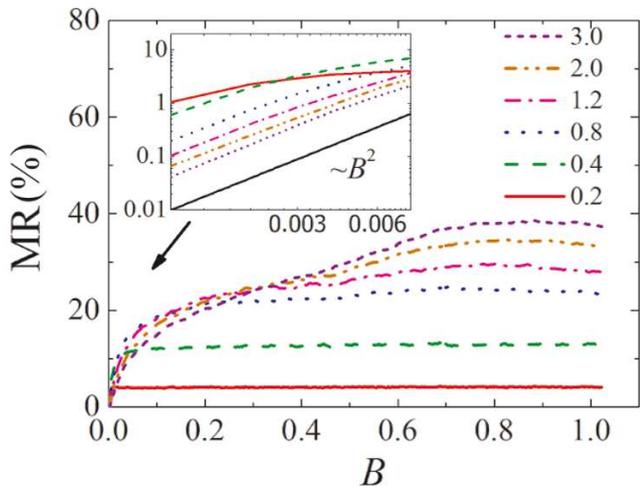
Figure 1(b) shows the resulting transmission  $g(B)$  that oscillates with the magnetic fields. This conductance oscillation is one of the well-known manifestations of the Aharonov-Bohm interference effect, and the oscillation period would be given by  $1/S$  for the lowest harmonics. We evaluate the loop area to find  $2\pi S = 125.2$  for poly1 and  $2\pi S = 161.9$  for poly2 and expect oscillation periods to be  $1/S \approx 0.05$  for poly1 and  $1/S \approx 0.04$  for poly2. We



**Fig. 2.** (Color online) Fourier amplitude  $|\tilde{g}(S)|^2$  with arbitrary offsets for each polymers, where the parity effect for poly1 and poly2 is clearly seen; peaks at integer multiples of  $2\pi S$  for poly1 and one peak only at  $4\pi S$  for poly2. Poly3 generated at  $T = 0.3$  has two loops with areas  $2\pi S_1 = 37.2$  and  $2\pi S_2 = 121.8$ , which determines the peak locations as  $2\pi S_1$ ,  $4\pi S_2 - 4\pi S_1$ ,  $4\pi S_2 - 2\pi S_1$ , and  $4\pi S_2$ . Poly4 at a higher temperature ( $T = 0.6$ ) has a large number of loops of small area to yield  $|\tilde{g}(S)|^2$  with larger weight at small  $S$  and a long-range tail.

find that this is the case only for poly1, but for poly2 period halving appears, as shown in fig. 1(b). Let us see how this interesting event of period halving happens in detail. For the fundamental periodicity  $1/S$  to be removed, we see that in eq. (4) the real part of  $A(N, t_\perp, \Sigma)$  and  $D_{N-2}$  should vanish. These can be equivalently stated as  $E = \text{Re}[\Sigma_1 + \Sigma_2]$ , which at the Fermi level ( $E = 0$ ) implies  $\text{Re}[\Sigma_{1,2}] = 0$ . This is indeed true, as we can see by inserting  $\theta = \pi/2$  into  $\Sigma_{1,2}$ . Meanwhile, another condition  $D_{N-2} = 0$  or  $\theta = n\pi/(N-1)$  with integers  $n$  can be met only when the loop is composed of an odd number of monomers. It is interesting to note that this is equivalent to a condition for the absence of interference effects. The phase difference  $\delta\phi$  between the paths along the upper and the lower perimeter of the loop is given by  $\delta\phi = (N-1)\theta - \theta$ , which for  $\theta = \pi/2$  becomes half-integer multiples of  $\pi$  for the odd-numbered loop. We check the loop parity of each polymers and confirm that the number of monomers in the loop is indeed odd for poly2 ( $N = 21$ ), and even for poly1 ( $N = 18$ ).

At higher temperatures, the polymer becomes more flexible and bend itself to accommodate more loops. For this case, the Fourier analysis must be effective to examine the conductance oscillation. In fig. 2, we present a Fourier amplitude  $|\tilde{g}(S)|^2$  with  $\tilde{g}(S) = \sum_B e^{2\pi i S B} g(B)$  for various configurations generated at different temperatures. Peaks in  $\tilde{g}(S)$  should be developed at multiples of inverse periods which are given by the loop area  $2\pi S$ .



**Fig. 3.** (Color online) MR at various temperatures (the curve labels denote the corresponding temperatures), which increases with  $B$  as shown in the inset with the line ( $B^2$ ) as a guide for the eye. As the temperature is increased, MR saturates slowly to reach giant MR.

For the single-loop polymer configurations generated at  $T = 0.2$ , fig. 2 clearly shows that for poly2 the peak develops at  $4\pi S$ , indicating the period halving, while periods  $1/S$ ,  $1/2S$ , and  $1/3S$  are found in poly1. At the higher temperature,  $T = 0.3$ , the configuration (poly3) has two loops with areas,  $2\pi S_1 = 37.2$  and  $2\pi S_2 = 121.8$ , yielding the multi-component harmonics. When the flexibility increases further, loops become abundant (see poly4), and the Fourier spectrum becomes complicated to have a long-range tail and larger weight at smaller  $S$ . This indicates that many crosslinks realize small-sized loops to make a major contribution to the conductance oscillation.

Let us now proceed to the case of an ensemble of polymers, for which the averaged transmission must be a meaningful quantity to examine. Especially we evaluate magnetoresistance defined as  $\text{MR} = (\langle g(B) \rangle - \langle g(0) \rangle) / \langle g(B) \rangle$ , where the angular bracket denotes an ensemble average of the polymer configurations generated at a given temperature. Figure 3 exhibits MR at various temperatures. In the weak-field region, MR nonlinearly increases with  $B$  as  $\text{MR} \sim B^2$  as shown in the inset of fig. 3 and saturates in the high-field regions. It can also be seen that increasing temperature slows down the saturation and enhances MR. Although, due to the system complexity a rigorous analytic proof is infeasible, we make a reasonable argument to explain the behaviors of MR qualitatively within the framework of the weak localization [22,23]. Since crosslinks give self-crossing trajectories, the averaged reflection probability can be written as  $\langle r(B) \rangle = \langle |\sum_n [A_n(B) + A_n^R(B)]|^2 \rangle$ , with counting contributions from possible paths and their time-reversed counterparts such as  $A_n(B) = A_n e^{-2\pi i B S_n}$  and  $A_n^R(B) = A_n e^{2\pi i B S_n}$ . Here,  $S_n$  is the area of the  $n$ -th loop (if any) in a given polymer configuration. Using an orthogonality between different  $n$ 's, we have the averaged

reflection probability reading

$$\begin{aligned} \langle r(0) \rangle - \langle r(B) \rangle &= 4 \left\langle \sum_n |A_n|^2 \sin^2(2\pi B S_n) \right\rangle \\ &= \langle g(B) \rangle - \langle g(0) \rangle \\ &\approx 4|A|^2 \|\sin^2(2\pi B S)\|, \end{aligned} \quad (5)$$

where the second line is the probability conservation, and in the last line we make an assumption that the operation of the summation over  $n$  and the ensemble average can be replaced with an average taken with respect to the probability distribution of loop areas  $P(S)$ , that is,  $\|X\| = N_c \int dS P(S) X(S)$ : If the variable  $X$  is the area itself, ensemble average  $\langle S \rangle$  should give the average loop area, and summation over the index  $n$  would be proportional to the number of crosslinks  $N_c$ , which is equivalently described also by  $\|X\|$ . Also, we assume that  $|A|^2$  is a slowly varying function of  $S$ . Although  $P(S)$  is unknown, we make a conjecture,  $P(S) = (4S/S_0^2) e^{-2S/S_0}$  with  $S_0$  being the mean loop area that decreases with increasing temperature in order to correspond to the understanding that, at higher temperature, the smaller loops become more prevalent. We then reach

$$\langle g(B) \rangle - \langle g(0) \rangle = 2N_c |A|^2 \left( 1 - \frac{1 - (2\pi S_0 B)^2}{(1 + (2\pi S_0 B)^2)^2} \right), \quad (6)$$

which increases with the magnetic fields, yielding  $\text{MR} \sim B^2$  for  $S_0 B \ll 1$ , and saturates when  $2\pi B S_0 \approx 1$  or when a unit flux quantum penetrates through a mean-area-sized loop. This explains the slowing down of the saturation of MR with increasing temperature. Note also that in eq. (6) MR is proportional to  $N_c$  increasing with temperature, which is in correspondence with the temperature dependence displayed in fig. 3.

In summary, we have studied MR for flexible conducting polymers. For the single-polymer realizations we have observed the conductance oscillations with periods determined by the area of the loops. Especially, for chains with a single loop the parity effect appears in the oscillation period; for even  $N_\ell$  the period is found to be  $1/S$ , while  $1/2S$  for odd  $N_\ell$ . For an ensemble of polymers, MR has been found to nonlinearly increase with magnetic fields, and eventually become saturated into higher values for more flexible wires to achieve the giant MR. To go beyond the simple picture adopted here, new physical ingredients are certainly necessary. For instance, at high temperature, not only the large-scale deformation considered here, but also a small-scale deformation such as ionic displacement would influence the system property. This can be examined by applying the Su-Schrieffer-Heeger model [24,25] to the coiled wire. Also, the effects of electrostatic interactions between electrons on the curved space must be nontrivial. These challenging questions would be answered by future theoretical and experimental studies.

This work was supported by the Korea Science and Engineering Foundation (Grant No. R01-2007-000-20084-0).

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