Spin-polarized electron tunneling across a disordered insulator

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It is shown that the presence of disorder within an insulator has a dramatic effect on the mechanism and the spin polarization of tunneling in ferromagnet-insulator-metal tunnel junctions. We have calculated the conductance of the tunnel junction within a quantum-mechanical treatment of the electronic transport. The spin-polarized band structure of the ferromagnet was approximated by exchange-split tight-binding bands and the disorder was represented by a randomness in on-site atomic energies of the insulator. We demonstrate that for each realization of the disorder the conductance displays numerous resonances, which are determined by multiple scattering processes. The distribution of the conductance with respect to different random configurations is extremely broad, covering many orders of magnitude. The dominant contribution to the tunneling current comes from a few random configurations of disorder which provide highly conducting resonant electronic channels. We find that the spin polarization P of the tunneling current is determined not only by the intrinsic properties of the ferromagnet alone, but in a regime of relatively high disorder which is typical for experiments, the spin polarization decreases with increasing disorder and the thickness of the insulator. This behavior can be explained qualitatively in terms of quasi-one-dimensional tunnelling through an effective potential barrier, the height of which decreases with increasing disorder and insulator thickness. At high disorder the tunneling magnetoresistance calculated directly by modelling the parallel and antiparallel alignments of the ferromagnets agrees with that predicted by Julliere's formula $2P^2/(1+P^2)$.
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Recent advances in tunneling magnetoresistance (TMR) (Refs. 1 and 2) have demonstrated that thin-film tunnel junctions, in which two ferromagnets are separated by a thin insulating layer, are very promising from the point of view of applications as magnetic sensors and as magnetic random-access memory elements. The actual magnitude of TMR which is important for the performance of future devices is determined by the spin polarization of the tunneling current and was found to be in agreement with Julliere's formula.[3] The spin-polarization values were measured for numerous ferromagnetic metals in experiments on tunneling to superconductors across an alumina spacer.[4]

Theoretical formulations of the TMR problem are usually based on models which assume perfect systems.[5–8] It is known, however, that the presence of localized electronic states within the gap of the insulator due to impurities and defects leads to resonant tunneling (e.g., Ref. 9). Resonant tunneling has a much slower decrease of the transmission coefficient with increasing barrier thickness in comparison to the direct tunneling and, therefore, dominates for sufficiently thick barriers. In amorphous oxides which are usually used in TMR experiments,[1,2,10] the structural disorder can easily generate intermediate states in the gap of the oxide. The influence of this disorder on the spin polarization of tunneling and TMR is the subject of the present work.

We consider tunneling between two semiinfinite perfect metals through an insulator layer within a single-band tight-binding model. The metals and the insulator have a simple cubic geometry of lattice parameter a and (001) orientation of atomic layers. The band structure of the system is shown schematically in Fig. 1. The parameters of the model are chosen as follows. The hopping integrals are nonzero only between nearest neighbors and are set equal to 1 both for the metals and the insulator. All energies are measured in units of the hopping integral relative to the Fermi energy which lies at zero. In order to simulate the exchange splitting of the spin bands in the left ferromagnetic metal, we set the on-site atomic energies of the ''up-spin'' and ''down-spin'' elec-

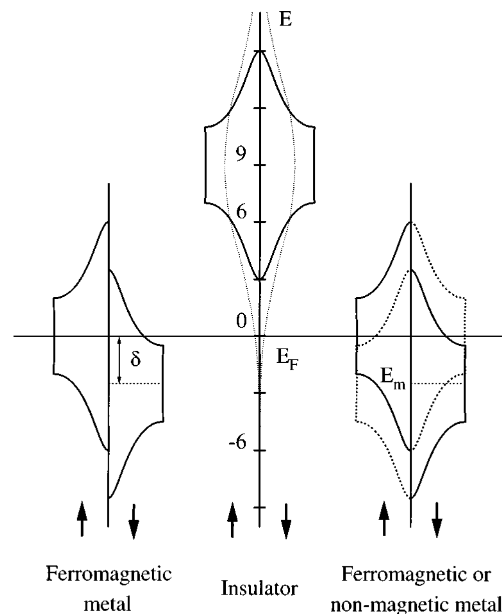

FIG. 1. Schematic representation of the band structure of the system. Up-spin and down-spin bands of ferromagnetic metals are split by the exchange potential δ. The dashed line shows the position of the bands for the case of antiparallel alignment. E_m determines the position of on-site atomic energy of the right metal when it is considered to be nonmagnetic. The dotted line represents broadening of the density of states in the insulator due to disorder.

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trons equal to 0 and $-\delta$, respectively. The right metal was assumed either nonmagnetic with on-site atomic energies E_m equal to 0 or $-\delta$ or ferromagnetic with the same parameters as the left metal. The insulator bands are spin independent and shifted from the Fermi energy by 9 energy units. Since the half width of the bands is equal to 6, this position of the on-site atomic energy for the insulator corresponds to a band gap for the insulator equal to 6, provided that the Fermi energy lies in the middle of the band gap. Bulk disorder within the insulator layer takes the form of random variation of the on-site atomic energies, with a uniform distribution with a mean of 9 and a standard deviation of γ which was varied in our calculations from 0 to 8.

For calculations of the conductance we use the Kubo formula within the real space representation,[11] in which the scattering of electrons by the disorder is treated exactly. The details of the technique may be found, e.g., in Refs. 8, 11, and 12. In order to extend the size of the system to infinity in the direction perpendicular to the current, we consider a unit cell in the transverse direction and impose periodic boundary conditions. In general, the results of the calculations depend on the size of the unit cell, because of the correlations in the scattering potential due to the imposed transverse periodicity. Increasing the size of the unit cell, however, makes the computations very expensive. We have, therefore, fixed the transverse cross section of the unit cell to be four by four atoms in all our calculations.

Since the results of the calculations are extremely sensitive to the particular realization of disorder, the conductance has to be averaged over different disorder configurations. In order to achieve an acceptable accuracy in the spin polarization of the tunneling P, the averaging over more than $N = 10^4$ configurations has to be performed. For example, in the case when the thickness of the insulator $L=10a$ and $\gamma = 6$, $N=5\times10^4$ gives an *absolute* error in P of 0.02. Note that the procedure of configurational averaging relates directly to the assumed periodicity of the disorder. Increasing the number of random configurations N effectively corresponds to increasing the area of the tunnel junction S, which has noncorrelated disorder, so that $S=NS_0$, where S_0 is the area of the unit cell.

Figure 2 shows the mean conductance $\langle G\rangle$ per unit area as a function of the electron energy for a disorder insulator with $\gamma=6$ and various values of N. The results presented in this figure and Figs. 3–6 below are obtained for the tunneling of the up-spin electrons of the ferromagnet to the nonmagnetic metal with $E_m=0$. As seen from the dashed curve in Fig. 2, for each realization of disorder the conductance displays numerous resonances, which are determined by multiple scattering processes. Some of the resonances overlap, producing energy regions where the conductance is high. With increasing N the conductance averages out, eventually giving in a relatively smooth profile curve (the solid line in Fig. 2). We see that in this case the conductance *gradually* increases with energy, reflecting a higher density of tunnel-assisted electronic states, when moving towards the gravity center of the insulator band. We note that this increase in the conductance has a different origin compared to that in a perfect insulator, where one would expect a *strong* exponential enhancement of the conductance due to the decrease in the barrier height.

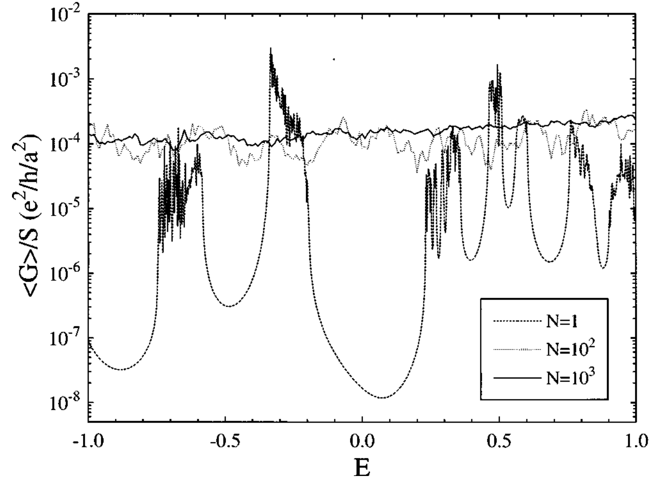

FIG. 2. Normalized conductance $\langle G\rangle/S$ per spin averaged over a different number of random configurations of disorder N versus electron energy for $\gamma=6$ and $L=10$.

Figure 3(a) shows the distribution of the conductance with respect to different random configurations of disorder. The distribution for a particular value of disorder γ is extremely broad, covering many orders of magnitude, and it becomes broader with increasing disorder. As can be seen from the vertical lines in Fig. 3(a), at higher disorder the position of the mean conductance $\langle G\rangle$ is shifted far away from the distribution maximum. This reflects the fact that the dominant contribution to the tunneling current comes from a few random configurations which provide a high conductance. This becomes obvious in Fig. 3(b), where the relative contribution of the conductances to the mean conductance is shown. By comparing Figs. 3(a) and 3(b), we see that the maximum contribution to $\langle G\rangle$ lies in the tails of the conductance dis-

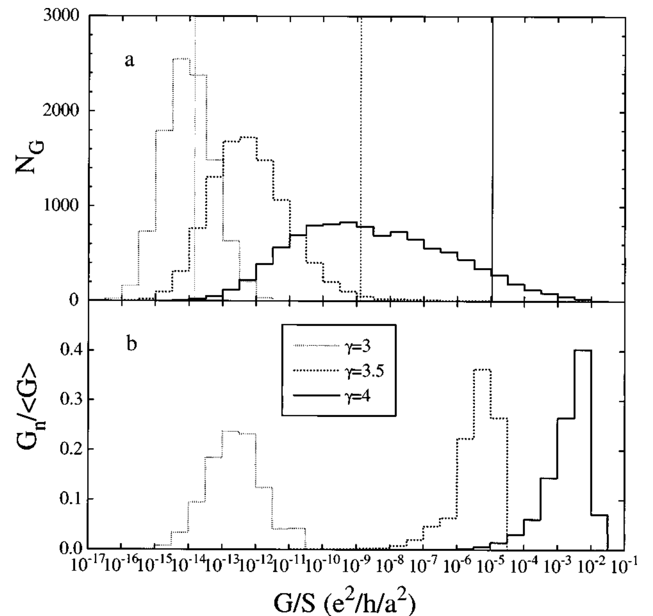

FIG. 3. Normalized conductance distribution for different configurations of disorder (a) and relative contribution of conductances to the mean conductance $\langle G\rangle$ (b) for $L=10$, $N=10^4$, and various values of γ. Vertical lines denote the position of the mean conductances $\langle G\rangle$.

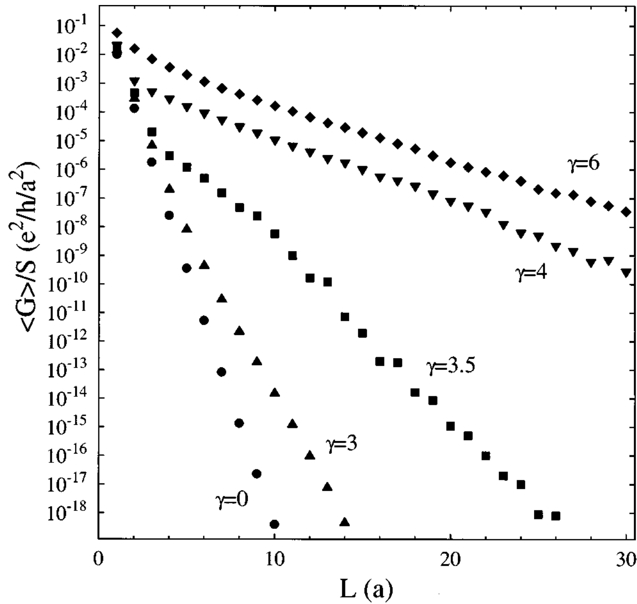

FIG. 4. Normalized conductance $\langle G\rangle/S$ versus insulator thickness L for $N=5\times10^4$.

tribution histogram, where only a few random configurations contribute. Making an analogy with the conductance in strongly disordered systems,[13] we can conclude that these configurations provide highly conducting ''channels,'' i.e., chains or ''necklaces'' of resonant electronic states connecting the two metals through the insulator.

Figure 4 shows the conductance averaged over $N=5\times10^4$ random configurations of disorder versus the thickness of the insulating layer L for various values of γ. When $\gamma=0$ the conductance decreases very rapidly with L. Adding a monolayer of the insulator reduces the conductance by two orders of magnitude. The slope of the curves gradually decreases with increasing disorder. This is due to the increasing

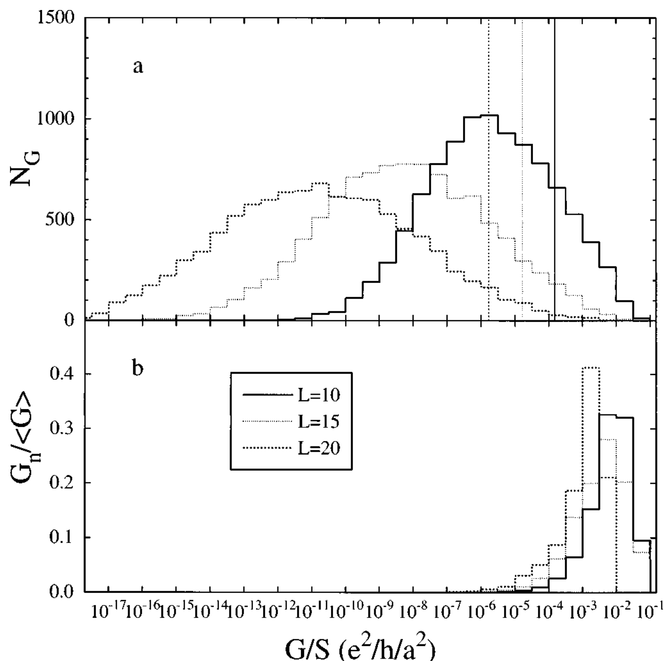

FIG. 5. Same as in Fig. 3 for $\gamma=6$ and various L.

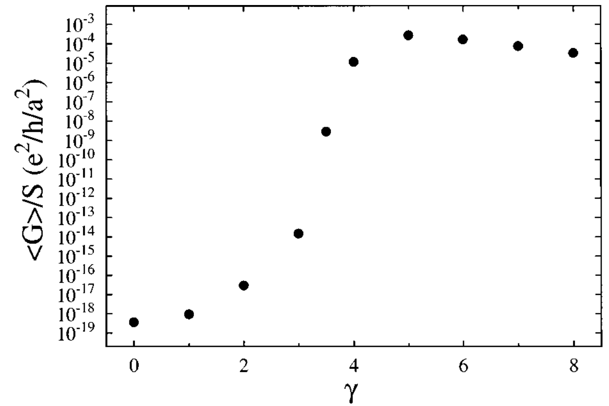

FIG. 6. Normalized conductance $\langle G\rangle/S$ as a function of γ for $N=5\times10^4$.

number of electronic states within the band gap of the insulator. These electronic states assist the tunneling reducing effectively the height of the tunneling barrier.

According to the original experimental results of Ref. 1 resistances of tunnel junctions with an alumina spacer layer of about 2 nm thickness and a junction area S of 6×10^{-4} cm^2 range from 10^2 to 10^5 Ω. Taking a lattice parameter a of 0.2 nm, we find that the experimental values of G/S range from 10^{-10} to 10^{-12} in units of $e^2/(ha^2)$. These values are consistent with our computations for $L=2$ nm $(10a)$, where $\langle G\rangle/S$ changes from 10^{-14} to 10^{-8} when γ varies from 3 to 3.5. We conclude, therefore, that the experiments on spin-polarized tunneling are performed under conditions of strong disorder within the insulator, with $\gamma=3$ to 3.5 being representative values characterizing the experiments in Ref. 1.

At relatively large values of L all the curves presented in Fig. 4 can reasonably well be fitted by the exponential-decay function $C\exp(-2\kappa L)$ which describes tunneling through a mean potential barrier $U=\hbar^2\kappa^2/(2m)$. For $\gamma=3.5$, Fig. 4 gives $U\approx0.3$ eV. This value is much smaller than those obtained by fitting the current-voltage data using the Simmons theory of tunneling.[14] We note, however, that a typical value of $U=2$ eV (Refs. 1 and 10) would give rise to a decrease in the conductance by a factor of 20, if the insulator thickness is increased by a monolayer. This is not the case for alumina-based tunnel junctions.[15]

Although the dependence of the conductance on L can be fitted by assuming tunneling through an effective barrier of fixed height, the actual mechanism of tunneling through a disordered insulator is much more complex. As seen from Fig. 5(a), the conductance distribution histogram broadens with increasing thickness of the insulating layer. The mean conductances $\langle G\rangle$, which are shown by the vertical lines in Fig. 5(a), are shifting further away from the maximum of the distributions. This implies that with increasing L fewer random disorder configurations contribute to the tunneling. The effective barrier height which characterizes these random configurations is lower than the mean potential barrier U extracted from the fitting of the $G(L)$ curves and it decreases with L. This can be seen by comparing the conductance distributions shown in Fig. 5(a) to the relative contribution of conductances to $\langle G\rangle$ shown in Fig. 5(b). As seen, with increasing L from 10 to 20 the mean conductance $\langle G\rangle$ de-

creases by two orders of magnitude whereas the maximum in the distribution of $G_n/\langle G\rangle$ changes by less than an order of magnitude. We conclude, therefore, that the effective potential barrier of the conducting channels which determine the tunneling current in the regime of high disorder decreases with increasing insulator thickness. As we see below, this has an important effect on spin polarization of the conductance.

Figure 6 shows the dependence of the conductance on the amount of disorder within the insulator γ. As seen, first, the conductance grows with increasing γ. The growth becomes dramatic when γ exceeds 3. This reflects the fact that at this amount of disorder the number of electronic states near the Fermi energy increases significantly, which assists the process of tunneling. At $\gamma=5$ the conductance reaches a saturation and starts to decrease at higher disorder, which is an indication of the localization regime of conductance.[16]

For calculations of the spin polarization P of the conductance the on-site atomic energies for the down-spin electrons of the left ferromagnetic metal were shifted by $\delta=3$ and $\delta=1.5$ below those for the up-spin electrons, as shown in Fig. 1, and the right metal was assumed to be nonmagnetic. The spin polarization P was defined as $P=(\langle G_\uparrow\rangle-\langle G_\downarrow\rangle)/(\langle G_\uparrow\rangle+\langle G_\downarrow\rangle)$, where $\langle G_\uparrow\rangle$ and $\langle G_\downarrow\rangle$ are the mean conductances for the up- and down-spin electrons, respectively. In order to verify how the spin polarization depends on the choice of the nonmagnetic metal, the on-site energy E_m of the metal was taken to be equal to 0 and $-\delta$. First, we discuss the results of our computations for $E_m=0$ which are shown in Fig. 7(a) by circles.

We see that for a perfect insulator, when $\gamma=0$, the values of P are very high, namely, $P=0.97$ for $\delta=3$ and $P=0.78$ for $\delta=1.5$. These high values can be explained by two factors. Each of these factors is connected with the dependence of the conductance on the parallel momentum $k_\parallel$, which is conserved in the process of tunneling through a perfect periodic system. First, because of the negative shift of the down-spin band, the electronic states of the insulator with $k_\parallel$, which corresponds to nonzero density of states of the metal at the Fermi energy, lie at higher energies for the down-spin electrons than those for the up-spin electrons, providing a higher potential barrier for the former. The difference in the height of the barrier between the up- and down-spin electrons increases with increasing δ. Second, the electronic states of the left and right metals are localized in different regions of the Brillouin zone when electrons tunnel between bands with significantly different on-site energies. In this case the electrons have to tunnel from regions of the Brillouin zone with a high density of states (DOS) to regions with a low DOS and vice versa. This leads to a smaller conductance for the down-spin electrons.

As seen from Fig. 7(a), when γ increases the spin-polarization of the conductance for $E_m=0$ decreases. This behavior can be explained by the fact that electrons propagating through the insulator lose their memory of $k_\parallel$ due to the scattering by the disorder. Because of this, the two factors which were decisive for the high values of P in the case of a perfect insulator are becoming less important. First, the spin dependence of the effective barrier becomes less pronounced, due to intermixing of electronic states within the insulator. Second, electrons can now tunnel to different regions of the Brillouin zone, because the transverse momentum is no longer conserved. This second factor results in a stronger increase of the conductance when tunneling occurs between bands with different on-site energies. The degree of both effects depends on the amount of disorder (and the thickness of the insulator). It seen from Fig. 7(a), that the spin polarization decreases rapidly when γ exceeds 3. As we discussed above, at this amount of disorder the conductance grows very rapidly (see Fig. 6) due to the increasing number of tunnel-assisted electronic states near the Fermi energy. At higher disorder the spin polarization displays a tendency to saturate.

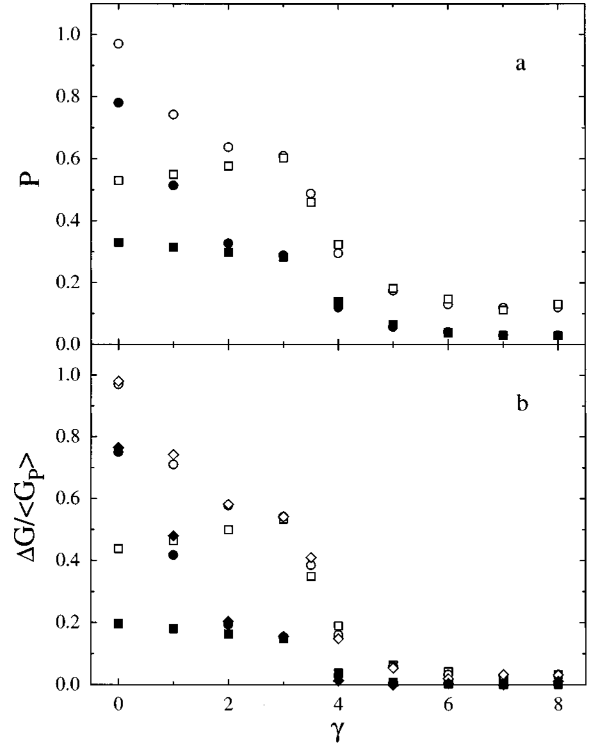

FIG. 7. Spin-polarization P (a) and tunneling magnetoresistance $\Delta G/\langle G_P\rangle$ (b) as a function of γ for $\delta=1.5$ (full symbols) and $\delta=3$ (open symbols), $E_m=0$ (circles), and $E_m=-\delta$ (squares) for $N=5\times10^4$. In the bottom panel a direct calculation of TMR (diamonds) is compared with the calculation by formula $2P^2/(1+P^2)$ (circles and squares).

As is obvious from Fig. 7(a), at low disorder the values of P are very different for the different on-site energies of the nonmagnetic metal, i.e., for $E_m=0$ and $E_m=-\delta$. In general, therefore, the spin polarization of the tunneling current depends on the particular nonmagnetic metal which is used in tunnel junctions. However, with increasing disorder the difference in spin polarization becomes smaller and for $\gamma\geqslant3$ the values of P for the different on-site energies of the nonmagnetic metal become equal within the computational error. We note that the increase of P with γ at low disorder for the case $\delta=3$ and $E_m=-3$ is connected with the second factor, described above for $E_m=0$, which now contributes to the spin polarization constructively.

Figure 7(b) shows the results of calculations of the tunneling magnetoresistance as a function of γ. The TMR was defined as the difference between the mean conductances for the parallel and antiparallel alignments of the ferromagnets

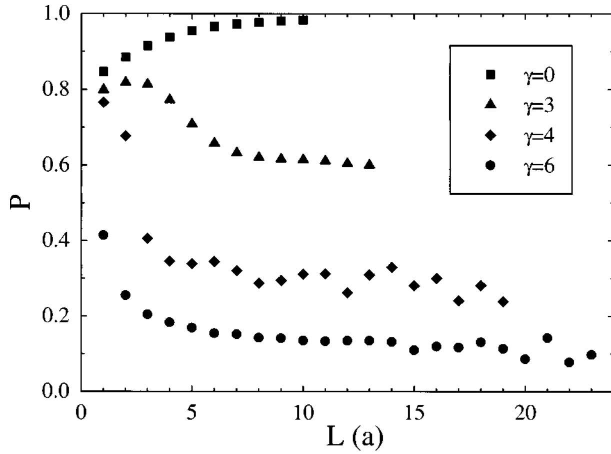

FIG. 8. Spin polarization of tunneling P as a function of the insulator thickness L for various values γ and $N=5\times10^4$.

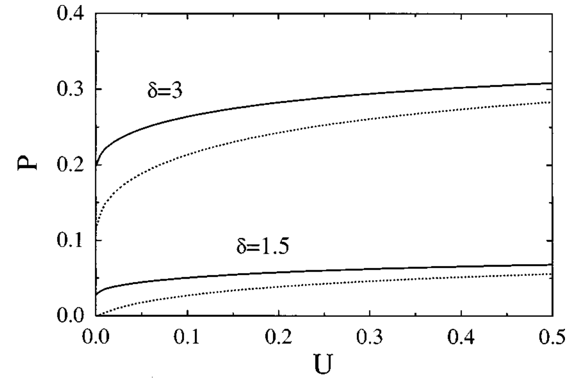

FIG. 9. Spin polarization P of tunneling through the one-dimensional chain of atoms as a function of the effective potential barrier U for $\delta=3$ and $\delta=1.5$ calculated for $E_m=0$ (solid lines) and $E_m=-\delta$ (dashed lines).

ΔG normalized to the mean conductance for the antiparallel alignment $\langle G_P\rangle$. The calculations were performed by modeling the band structure for the parallel and antiparallel magnetizations of the ferromagnets, as shown in Fig. 1, and compared to the results obtained using the Julliere's formula,[3] which has the form of $2P^2/(1+P^2)$ for the case when the same ferromagnets are adjacent to both surfaces of the insulator. We see that the values of TMR computed directly are in very good agreement with those calculated from Julliere's formula both for $E_m=0$ and $E_m=-\delta$ when $\gamma\geq3$ and disagree for $E_m=-\delta$ at lower disorder. As we saw above by comparing experimental and calculated values of the conductance, the experiments on TMR are performed in the regime of relatively strong disorder with $\gamma\sim3-3.5$. At this amount of disorder, our results are consistent with the finding that the majority of experimental results on TMR can be interpreted in terms of the values of the spin polarizations obtained by Tedrow and Meservey.[4] It follows from our modeling, however, that the spin polarization is determined not only by the ferromagnet but depends on the amount of disorder within the insulator and, as we will see below, on the insulator thickness.

Finally, we have calculated the dependence of the spin-polarization of the tunneling current on the insulator thickness for different values of γ. These results are displayed in Fig. 8. We see that at $\gamma=0$ the spin polarization increases approaching unity at high thicknesses. This is opposite to what we found for the case of the disordered insulator, where at sufficiently large L the polarization decreases. The enhancement of P at small thicknesses of the insulator is the result of correlations in $k_{\parallel}$ space. At high thicknesses the spin polarization displays a tendency to saturation, although the calculation errors do not allow us to make this conclusion definite. Below we discuss the calculated values of spin polarization of tunneling in the regime of high disorder and the mechanism of the decrease in the spin polarization with increasing insulator thickness.

It is known from the analysis of the conductance in strongly disordered systems that the electric current flows through quasi-one-dimensional chains of electronic states which span the distance between two metal electrodes more or less directly.[13] The flow of the current is concentrated at a few sites (i.e., chain ends) across the insulator-metal interface. The breakdown of the $k_{\parallel}$ conservation and localization of the tunneling current at certain atomic sites make the local interfacial densities of states important parameters which influence the process of tunneling. If the potential barrier characterizing the conductance through the channels was small, the electronic states of the metals would be decoupled and the tunneling current would be proportional to the product of the local DOS of the metal electrodes. This immediately would lead to the result $P=(D_{\uparrow}-D_{\downarrow})/(D_{\uparrow}+D_{\downarrow})$, where $D_{\uparrow}$ and $D_{\downarrow}$ are the local DOS at the interface layer of the ferromagnet for the up-spin and down-spin electrons respectively (note that this definition of P was used by Julliere[4]). However, this is not the case. For $\delta=3$ the local DOS for the up- and down-spins electrons are $D_{\uparrow}=0.166$ and $D_{\downarrow}=0.079$, that gives $P=0.355$ which is higher than the value of 0.13 obtained for $\gamma=6$ ($L=10$). For $\delta=1.5$ the local DOS are $D_{\uparrow}=0.166$ and $D_{\downarrow}=0.139$, giving $P=0.088$ which is again higher than the value of 0.04 obtained for $\gamma=6$. We see, therefore, that at high disorder within the insulator the spin polarization is lower than that predicted by the assumption that the tunneling current is proportional to the product of the DOS of the ferromagnets for a given spin.

The explanation of this fact is that the tunneling current is determined by a few conducting channels which are characterized by a *very low* effective barrier. Due to this, the electronic states of the metals are coupled through the channels and the conductance is no longer proportional to the product of the DOS. In order to simulate this behavior we have performed a calculation of the tunneling through a one-dimensional chain of atoms. The on-site energies E_0 of the atoms in the chain were taken to provide a tunneling barrier. Since the band width of an infinite chain of atoms within a singe-band nearest-neighbor tight-binding model is 4 (in units of the hopping integral), we choose $|E_0|\geq2$. In this case the effective potential barrier experienced by electrons propagating through the atomic chain is equal to $U=|E_0|-2$. The quoted conductances are the average for the cases $+E_0$ and $-E_0$. Figure 9 shows the results for the spin polarization P as a function of U for $\delta=3$ and $\delta=1.5$ and for two on-site atomic energies of the nonmagnetic metal, i.e., $E_m=0$ and $E_m=-\delta$. As P rapidly tends to a constant with increasing chain length L, the calculations were performed for a fixed length $L=20$. We see that the values of P are

lower than 0.355 for $\delta=3$ and 0.088 for $\delta=1.5$ predicted by the densities of states. These latter numbers are asymptotic values for the spin polarization at large U. Therefore, our qualitative model explains the lower values of P at high disorder.

Another important conclusion which follows from Fig. 9, is that the spin polarization of tunneling through the one-dimensional chain of atoms decreases with decreasing height of the potential barrier U. As seen, the decrease is especially strong at very small U. This explains the fact that spin polarization decreases with increasing barrier thickness when tunneling occurs through a disordered insulator, because the effective barrier height characterizing the conducting channels in the regime of high disorder decreases with the thickness.

In conclusion we have shown that disorder within the insulator has a dramatic effect on spin-polarized electron tunneling. In comparison with perfect periodic systems where the tunneling current is homogeneously distributed across the tunnel junction, in disordered systems the current flows through a few regions of the insulator where local disorder configuration provides highly conducting channels for electron transport. These conducting channels can be characterized by a low height of the effective potential barrier, which decreases with increasing disorder and with increasing thickness of the insulator. Due to this, the spin polarization of the tunneling current decreases with increasing disorder and insulator thickness. Although the spin polarization of tunneling is no longer determined only by the intrinsic properties of the ferromagnet, i.e., by the density of states for a given spin, the tunneling magnetoresistance is in agreement with the Julliere's formula $2P^2/(1+P^2)$, where P is defined as the spin polarization of the tunneling current from the ferromagnet to a nonmagnetic metal.

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