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Stabilization of magnetic polarons in antiferromagnetic semiconductors by extended spin distortions

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Abstract. We study the problem of a magnetic polaron in an one-dimensional antiferromagnetic semiconductor (ferron). We obtain an analytical solution for the distortion produced in the antiferromagnetic structure due to the presence of a charge carrier bound to an impurity. The region in which the charge carrier is trapped is of the order of the lattice constant (small ferron) but the distortion of the magnetic structure extends over a much larger distance. It is shown that the presence of this distortion makes the ferron more stable, and introduces a new length scale in the problem.

PACS. 75.10.Pq Spin chain models – 75.50.Pp Magnetic semiconductors – 75.30.Hx Magnetic impurity

1 Introduction

Quite a while ago Nagaev [1] has shown that the minimal energy for a charge carrier moving in an antiferromagnetic background is obtained when the electron modifies the magnetic background and is self-trapped in a region with canted antiferromagnetic or ferromagnetic order. The term ferron (a magnetic polaron in an antiferromagnetic background) was coined there to name this new quasiparticle. Depending on the radius of the self trapping region, it is possible to differentiate between small ferron, localized in a region of the order of the lattice constant, and large ferron localized at larger scales. Small bound ferrons are expected to be typical of the low doping region of the phase diagram of manganites, when the material is antiferromagnetic and insulating. Reviews can be found in references [2–4]. Treatment of magnetic polarons in Hubbard and $t-J$ models can be found in references [5,6], respectively.

There are also experimental data that confirm the existence of bound ferrons in antiferromagnetic semiconductors such as underdoped manganites. A review of them can be found in references [7,8]. In reference [9], a liquid-like spatial distribution of magnetic droplets in $La_{1-x}Ca_xMn_3$ with $x = 0.05$, 0.08 is reported. Ferromagnetic rich-hole droplets with a diameter of $4-5$ lattice units isotropically distributed with a mean distance of 9 lattice units among them are observed in an antiferromagnetic poor-hole background. Also it is reported that these magnetic droplets are coupled together through the antiferromagnetic background. In reference [10], a ^{55}Mn NMR study on the same compound within the doping range $x < 0.23$, confirms the electronic phase separation and report the existence of thin boundaries between antiferromagnetic and ferromagnetic domains. These magnetic droplets contain a number of conduction electrons larger than one (about 30), but this is not essential in describing the spin distortion that they create in the antiferromagnetic background.

Almost in all the calculations of the ferrons it was assumed that the region of the distortion of the magnetic structure coincides with the range of localization of the electron. Often these regions were simply taken as spheres of radius R, to be determined self-consistently. However, as first pointed out by De Gennes [11], the distortion of the magnetic order around a magnetic defect (i.e. ferron) may decay slowly with distance. In this paper we study what would be the "back effect" of this slowly decaying magnetic distortion on the conditions of the electron localization and on the properties of the resulting self-trapped state. First numerical calculations addressing this problem have been carried out recently by Nagaev [12]. In this paper the magnetic anisotropy, always present in real materials, was neglected, and he did not manage to obtain the results in a closed form and did not get proper estimates for the radius and the energy of the ferron taking this effect into account.

In this article, we obtain the analytical solution for the distortion created by a localized conduction electron trapped in a region of the order of the lattice constant in an one-dimensional antiferromagnetic semiconductor. The main virtue of our calculation is than reveals the existence of a region surrounding the trapping region which acts as an antiferromagnetic domain wall, as it was anticipated in reference [12]. We calculated the properties of the ferron in this situation and show that the account of an extended magnetic distortion around it actually leads to an increase of the ferron stability and its binding energy. Our result could offer a key to understand the features of

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the phase diagram of antiferromagnetic semiconductors, such as manganites at the small doping range $x < 0.1$.

2 Results

To study the problem of the magnetic polaron formation, we start from the Hamiltonian proposed by Nagaev [12]. We analyze the one-dimensional case because it can be treated analytically and its solution provides a full physical insight into the problem. Estimates to the three-dimensional case are given below. We consider an one-dimensional chain of magnetic ions along the y-axis, described by the double exchange (or Vonsovsky s-d) model. An uniaxial magnetic anisotropy term is added with x being the easy axis. The magnetic structure of the magnetic ions without the conduction electron is represented by two sub-lattices with the spin up along the easy axis (that means $S^x = S$) for ions at even sites ($g = 2n$), and spin down (that means $S^x = -S$) for ions at odd sites ($g = 2n + 1$). All distances are measured in terms of the lattice constant. A non-magnetic donor impurity is added to the chain at the point $y = -1/2$. Its conduction electron is bound to it and it can jump between their two magnetic neighbors (bound ferron). This disturbs the pure antiferromagnetic order along the chain, even though the conduction electron is trapped only on these two magnetic ions. The Hamiltonian of the system can be represented by:

$$\begin{aligned} H_{sd} = &-t\left(a^+_{-1,\sigma}a_{0,\sigma} + a^+_{0,\sigma}a_{-1,\sigma}\right) \\ &- A\sum_{g=-1,0}\left(\mathbf{s}\mathbf{S}_g\right)_{\sigma,\sigma'} a^+_{g,\sigma}a_{g,\sigma'} \\ &- I\sum_g \mathbf{S}_g\mathbf{S}_{g+1} - K'\sum_g \left(S^x_g\right)^2 \end{aligned} \quad (1)$$

where $a^+_{g,\sigma}, a_{g,\sigma}$ are the conduction electron operator corresponding to the site g and spin projection σ, $\mathbf{s}$ the conduction electron spin operator, $\mathbf{S}_g$ is the spin operator of the magnetic ion at site g (d-spins). The d-d exchange integral I is assumed negative in order to get the antiferromagnetic ordering. The anisotropy constant K' is considered positive.

Hamiltonian (1) reflects the fact that the conduction electron is restricted to move only over the site $g = -1, 0$. This correspond to take the Coulomb attraction between conduction electron and the impurity as a deep rectangular well of a size of two lattice constants. This approximation is valid when the Coulomb attraction between conduction electron and impurity is of the order of the largest parameter in the Hamiltonian (1). This situation is typical of charged impurities in magnetic semiconductors. The energy of Coulomb interaction between the conduction electron and the impurity is an additive constant which does not depend on the d-spin configuration and, for this reason, omitted in the calculation.

Depending on the relative value of the parameters $W = 2zt$ and AS, z being the number of first neighbors and S the magnitude of the d-spin, we have two different situations. In the case $W \gg AS$, we talk about a wide-band semiconductor. In the case $W \ll AS$, we talk about a double exchange semiconductor.

Our goal is to obtain an expression for the magnetic energy of the system of d-spins, both in the case of wide-band and double exchange semiconductors. As $S \geq 2$ for the compounds of interest (typically, manganites), the d-spins are considered classically. Their orientations are described in a coordinate system centered in the position of each magnetic ion. As it was stated above, it is assumed that the conduction electron is in the lowest bound state in the space spanned by the operators of the sites $g = -1, 0$. Also we assume that the magnetic moment of the ferron is directed along z-axis, that is, the conduction electron spin acts as an effective magnetic field along the z-axis for the d-spin system. In this case, the following symmetries hold for the d-spin system: $S^y_g = 0$, and $S^z_g = S^z_{-(g+1)}$. Then $\mathbf{S}_g = S\left((-1)^g \sin\theta_g, 0, \cos\theta_g\right)$. It is important to notice that here, although the d-spin lies in the $z - x$ plane, the angle θ is not the polar angle, but the spherical coordinate.

To obtain the magnetic energy of the d-spin system the electronic part of the Hamiltonian (1) must be averaged out. To do this, we assume that the electronic wave function is the ground state wave function for the dominant term in the electronic Hamiltonian, i.e. the hopping term in the wide-band case, and the s-d exchange term in the double exchange case. The other term is treated as a perturbation [17]. In the case of wide-band semiconductor, the hopping term is diagonalised and its ground state is $|\Phi\rangle = \frac{1}{\sqrt{2}}\left(a^+_{-1,1/2} + a^+_{0,1/2}\right)|0\rangle$. In the case of double exchange semiconductor, the ground state is the same but the operators $a^+_{g,1/2}$ being the operators with the spin projection along the direction of the vector $\mathbf{S}_g$, instead of the laboratory z-axis. We treat the case of a wide-band semiconductor first. Then the magnetic energy of the d-spin system is:

$$\begin{aligned} E = &J\sum_g \cos\left(\theta_g + \theta_{g+1}\right) \\ &- L[\cos\theta_{-1} + \cos\theta_0] - K\sum_g \sin^2\theta_g - t \end{aligned} \quad (2)$$

where: $J = -IS^2$, $L = AS/4$, and $K = K'S^2$.

Minimizing the equation (2) with respect to the angles θ_g, a set of non-linear equations is obtained:

$$\begin{aligned} &J\sin\left(\theta_g + \theta_{g+1}\right) + J\sin\left(\theta_{g-1} + \theta_g\right) \\ &\quad - L\sin\theta_g[\delta_{g,-1} + \delta_{g,0}] + K\sin\left(2\theta_g\right) = 0. \end{aligned} \quad (3)$$

There is a boundary condition $\theta_{g\to\pm\infty} = \pi/2$ if the chain is long enough, that means if $KN \gg L$, with N being the number of magnetic ions of the chain. Further, the above symmetry conditions imply that only the sites with $g \geq 0$ must be considered.

For the double exchange case, the same set of equations is obtained but the term in L in equation (2) must be changed by the standard effective hopping of the double exchange model, $-t\cos\left(\frac{\theta_{-1}+\theta_0}{2}\right)$ [18], and the constant

term $-t$ must be changed by $-2L$. Using the symmetry condition $\theta_{-1} = \theta_0$, this is equivalent to change L by $t/2$ in equation (3). Therefore both cases can be treated on the same footing.

Following the paper by Néel [13], we look for a differential equation for the d-spin distortion. If we were dealing with a ferromagnetic d-d exchange, the set of equations (3) would describe a domain wall. To obtain a differential equation for our problem an additional step is needed. Instead of working with the angles θ_g, we perform a rotation of an angle π around each y-axis if the site is an odd site, and no rotation if the site is an even site. This corresponds to make the following changes in the angles: $\theta_{2n} \rightarrow \theta_{2n}, \quad \theta_{2n+1} \rightarrow \pi - \theta_{2n+1}$. We assume that the length of variation of the angle θ is larger than the lattice constant. We treat θ as a continuous function over the y-axis and perform a power expansion in the lattice constant. Then a differential equation is obtained. Taking into account the above symmetries, we have to solve only for the positive semi-axis. Further, we divide the problem in two parts. For $y > 0$, we have:

$$J\frac{d^2\theta}{dy^2} + K\sin 2\theta = 0. \tag{4}$$

For $y = 0$ and using again the symmetries, we obtain:

$$J\left.\frac{d\theta}{dy}\right|_{y=0} + (J+K)\sin 2\theta_0 - L\sin\theta_0 = 0 \tag{5}$$

where $\theta_0 = \theta(y=0)$. This is the sine-Gordon equation with a boundary condition at the origin. The simple coupling between d-spin system and conduction electron, with a linear term in L in equation (5), can be traced back to the simplified choice of the wave function of the conduction electron. For a more realistic wave function (that means a more realistic Coulomb attraction between conduction electron and impurity), a more complicated coupling would be obtained. The linear coupling appears because the localization region in which the conduction electron moves has an homogeneous canting order. This is the only possible ordering in the region of two lattice sites. In a more general situation the different backgrounds contribute to the coupling between both subsystems. However, when $W, AS \gg J$, as in real materials, the contribution mainly comes from these canted ferromagnetic backgrounds and therefore our approximation is justified.

We solve equation (4) with the boundary condition that $\theta_{y\rightarrow+\infty} \rightarrow \pi/2$. Multiplying by $2\frac{d\theta}{dy}$ and integrating once, we obtain:

$$J\left(\frac{d\theta}{dy}\right)^2 - K\cos 2\theta = C \tag{6}$$

where C is the integration constant. Following reference [14], we made:

$$\begin{aligned} C &= J\left(\frac{d\theta}{dy}\right)^2\Bigg|_{y=0} - K\cos 2\theta_0 \\ &= \frac{1}{J}\left[L\sin\theta_0 - (J+K)\sin 2\theta_0\right]^2 - K\cos 2\theta_0. \end{aligned} \tag{7}$$

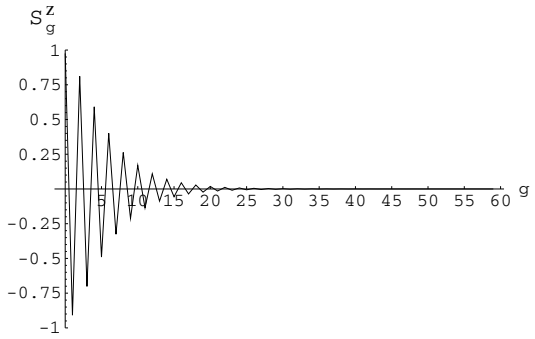

Fig. 1. Magnetization along z-axis, S_g^z, (in units of S) with $L = 3$ and anisotropy $K = 2.5 \times 10^{-2}$ (both in J units). Only one half, $g \geq 0$, of the chain is shown. To minimize d-d exchange energy neighbouring spin are nearly antiparallel, $S_g^z = -S_{g+1}^z$. Despite of this two different conditions are satisfied: at the center of the chain $g = 0$, $|S_g^z| \approx 1$, but at the end of the chain, $S_g^z \approx 0$. This structure gives a full energy gain in L with a small lost in exchange and anisotropy energies.

The last step is to take into account equation (5). Now we made the following change:

$$\sin\theta = \frac{f(y)-1}{f(y)+1}. \tag{8}$$

The differential equation (6) is cast into the form:

$$\begin{aligned} \frac{1}{(f+1)^2}\left[\frac{1}{f}\left(\frac{df}{dy}\right)^2 + \frac{2K}{J}(f-1)^2\right] = \\ \left[\frac{L}{J}\frac{f_0-1}{f_0+1} - 4\frac{J+K}{J}\frac{f_0-1}{(f_0+1)^2}\sqrt{f_0}\right]^2 \\ + \frac{2K}{J}\left(\frac{f_0-1}{f_0+1}\right)^2 \end{aligned} \tag{9}$$

where we define $f_0 = f(y=0)$. The solution of equation (9) is:

$$f(y) = \exp(a+by) \tag{10}$$

with a, b being two real constants to be determined. Note that a, $b > 0$ guarantees that the angle θ lies in the range $(0, \pi/2)$ for y belonging to the domain $[0, +\infty)$. Also note that at the infinite θ goes to $\pi/2$, as required. The differential equation is identically satisfied if:

$$b^2 = \frac{8K}{J} \tag{11}$$

and:

$$\sqrt{\frac{8K}{J}}\frac{\sqrt{f_0}}{f_0-1} = \frac{L}{J} - 4\frac{J+K}{J}\frac{\sqrt{f_0}}{f_0+1} \tag{12}$$

which is numerically solved to obtain f_0, or alternatively a. In reference [14], the solution of the equation (4) with the boundary condition (5) is treated in detail.

In Figure 1 we plot the magnetization along the z-axis, S_g^z, for the values of $L = 3, K = 2.5 \times 10^{-2}$ (both in J units). This is obtained by inverting the above

Table 1. Ground state energies (in J units), canting angle in the core, θ_0, and total magnetization along z-axis (in S units), M_z, for a 1D magnetic polaron with, and without compensating region, using $L = 3$, $K = 2.5 \times 10^{-2}$ (both in J units).

	Ground state energy	θ_0	M_z
With comp. reg.	-3.60101	0.22011	$1.00019S$
Without comp. reg.	-2.68811	0.63688	$1.60791S$

changes to the original angles. Note that only one half of the chain is shown. It can be seen how while neighboring d-spins are almost antiparallel to minimize d-d exchange energy, that is $S_g^z \approx -S_{g+1}^z$, the magnetic moment of each sublattice rotates to satisfy two different conditions: at the center of the chain, $g = 0$, d-spins are along z-axis and $|S_g^z| \approx 1$, and at the end of the chain d-spin are along x-axis and $S_g^z \approx 0$.

We calculate the total magnetization that appears in the d-spins system along the z-axis. The result is shown in Table 1. As can be seen, the existence of the magnetic distortion outside the localization region of the charge carrier leads to a partial compensation in the total magnetization.

With the analytical solution one can easily calculate the radius of the magnetic distortion. As can be seen from the solution, equations (8, 9), the radius is not properly defined, because the distortion created by the conduction electron in the magnetic system of d-spins reaches the complete chain. We choose to define the radius as the distance at which the straight line with slope equal to $\frac{d\theta}{dy}|_{y=0}$ and passing throughout the origin at θ_0, reaches the value $\pi/2$. This underestimates the radius, but has the obvious advantage that in this way, the radius depends only on the value of the constant a, or alternatively on the angle θ_0:

$$R = \left(\frac{\pi}{2} - \theta_0\right) \sqrt{\frac{J}{2K}} \sec\theta_0. \tag{13}$$

Also note that the value of $\theta_0 \approx 0$ and therefore $R \approx \pi\sqrt{J/8K}$. We obtain $R = 6.18981$ (in lattice constant units) for $K = 2.5 \times 10^{-2}$ (in J units).

With the analytical solution it is also easy to calculate the energy of the ferron. The equation for the energy, equation (2), contains also the energy for the system of d-spins even in the case with no conduction electron present. This energy has a value of $E_0 = -(J+K)N$. We define the energy of the magnetic polaron as $E - E_0$. As the transformation that we perform to obtain the differential equation (6) is a canonical transformation, we can use it again to calculate the energy. Taking this into account we transform the equation (2) and make again a power expansion in the lattice constant:

$$E = E_{\text{core}} - 2\int_0^{\frac{N}{2}} dy \left\{ J\left[1 - \frac{1}{2}\left(\frac{d\theta}{dy}\right)^2\right] + K\sin^2\theta \right\} \tag{14}$$

where $E_{\text{core}} = J(1+\cos 2\theta_0) - 2L\cos\theta_0 - t$ [19]. Therefore the polaron energy is:

$$E_{\text{pol}} = E - E_0 = E_{\text{core}} + \sqrt{8JK}\,(1 - \sin\theta_0) \tag{15}$$

as expected from the result for the ferromagnetic domain wall. Also note again that the value of θ_0 is small and therefore $E_{\text{pol}} \approx -2(L-J) - t + \sqrt{8JK}$, not only in the case of wide-band, but also in the case of double exchange semiconductor.

The motivation for the energy calculation is to demonstrate that the ground state energy of a bound ferron with a magnetization compensating region is much lower than the ground state energy of a bound ferron without such a long-range distortion. In Table 1, we present the result for the ground state energy of the magnetic polaron calculated from the equation (15). We also show for comparison the result in the case without compensating region. This latter was calculated solving the set of equations (3) numerically, and imposing $\theta_1 = \pi/2$. As expected the energy coming from equation (15) is lower, meaning that a true bound ferron is much more stable than bound ferrons considered previously in the literature, as it was once again anticipated by Nagaev in [12].

As can be seen from the equation (15), the main part of the energy of the magnetic polaron is concentrated in the core, that means at the sites $g = -1, 0$, where the charge carrier is trapped. The energy in the compensating region, that means outside the sites $g = -1, 0$, is very small and positive. This seem to be in contradiction with the previous discussion, in which the presence of a compensating region was presented as energetically favored. The physical explanation of this behavior is the following. The main energy scale in the problem that is coupled to the magnetic ordering is L, the s-d interaction, in a wide-band semiconductor. It tends to put the d-spins in the core as parallel to the z-axis as possible. But the d-spins in the core are connected to the rest of the chain through the d-d exchange term. The role of the compensating region is to isolate the d-spins in the core from the rest of the chain. This allows the d-spins in the core to be parallel to the z-axis with a high gain in s-d exchange energy (E_{core} is high and negative) and a little loss due to the perturbation of the antiferromagnetic ordering on the remaining part of the chain (the energy of the compensating region is positive but small). To better explain this point, we also show in Table 1, the canting angle of the d-spins of the core, θ_0, for a bound ferron with compensation region, and for a bound ferron without compensating region. As can be seen, the presence of the magnetization compensation region with the structure described above strongly reduces the value of canting angle. It is important to note that the existence of extended spin distortions does not rest on the presence in the model of the anisotropy term. A similar solution can be found by taking the limit $K = 0$ in equations (4, 5), but the radius of the distorted spin region goes to infinity. Also, because of the rotational invariance of the model with $K = 0$, the ferron energy is independent of the relative orientation of the magnetic moment of ferron to the long-range magnetic ordering of the sample. The presence of the anisotropy term breaks this degeneracy and makes the radius of the distorted spin region take a physical finite value.

Apart for the solution treated here, there is another possible solution for the d-spin structure [16]. This second solution is characterized by the absence of extended spin distortions. It corresponds to rotate by an angle π around y-axis the d-spins of one half, say the negative one, $g < 0$, of the chain. This makes that the magnetization in the trapping region will be directed along x-axis, instead of z-axis. Mathematically, it corresponds to choose a different symmetry condition for the d-spin system, namely $S^y_g = 0$, and $S^x_g = S^x_{-(g+1)}$. This solution saves more anisotropy energy and allows for a lower canting angle in the core, being at first instance the ground state in the range of parameters chosen here. However, as the Heisenberg model has to be solved over a compactified ring, that is, the d-spin at $g \longrightarrow +\infty$ is linked to the d-spin at $g \longrightarrow -\infty$, one has to add an extra energy $2J$ to the polaron energy, equation (14). Therefore as $J \gg K$ the ground state always corresponds to the solution treated here. In 2D (3D), this cost in energy is at least $8J$ ($24J$) if one assumes localization regions of the conduction electron of 4 (8) sites. This correspond to the situation in which one creates the ferron simply by rotating the d-spins of one half of the sites in the localization region by π. If one tries to move this abrupt barrier to the infinity the cost in energy goes to infinity because the boundary in 2D (3D) has dimension 1 (2), not being simply a point. The only possibility to avoid this large energy lost would be to place a second ferron near the first one, to compensate the rotation by π in part of the chain. While this is easy in 1D, it is not possible in 2D, 3D for a general ferron distribution, causing the frustration of the antiferromagnetic ordering, and a lost in energy of the order of J. This second kind of solutions also implies the breakdown of the long-range antiferromagnetic order at very low densities of ferrons, which is not observed in real compounds [15]. Note also that with the solution we focus on in the manuscript, the energy lost is of the order of $\sqrt{JK}$, and the long-range antiferromagnetic survives up to densities of the order of $\sqrt{K/J}$. Therefore, the solution treated in this article seems the relevant one to the physics of real compounds.

The three-dimensional case cannot be solved analytically. We only give a simple estimate for the radius of the distortion for an antiferromagnetic coupling. We use the three-dimensional analogue of the Hamiltonian (1). The case $K' = 0$ is treated in reference [11]. To introduce the anisotropy we use a simple variational method. We assume that the distortion angle is given by the spherically-symmetrical asymptotic solution of De Gennes, $\theta(r) \sim 1/r^2$ for $r < R$, and that there is no distortion for $r > R$. The optimal value of R is obtained by minimization. For R large, we obtain $R \sim (J/K)^{\frac{1}{6}}$. This provides an order of magnitude for the radius of the distortion. With this estimate for the radius of the distortion is now clear that, in real three-dimensional materials and over experimental doping ranges, the (one-electron) ferrons treated here must overlap, forming magnetic droplets with a number of conduction electrons larger than one, as reported in the experiment. Therefore it is expected that the main conclusions of this article hold for the three-dimensional case and for real materials, although to discuss the formation of these magnetic droplets would be necessary to include in our model the repulsive Coulomb interaction between conduction electrons.

To summarize, we have found the detailed structure of the one-dimensional d-spin system in the region surrounding of a bound ferron, completing the previous results of [1,11,12]. The main result is the appearance of a new length scale, namely the extent of the magnetic distortion created by the charge carrier. The existence of this distortion makes the ferron more stable. This may determine, together with the Coulomb interaction, the spatial distribution of magnetic droplets and their coupling in antiferromagnetic semiconductors, such as underdoped manganites. Also it could be related to the onset of the electronic phase separation at the very low doping range observed in these compounds.

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17. An exact solution for the full electronic Hamiltonian is avaliable, but we prefer to use this approximation, by proposes of clarity. The exact solution can not be used in the analytical approach we made here
18. Note that this is a two site problem, and therefore no additional phases are needed
19. As t does not enter in the equation to determine the magnetic structure (3), its numerical value is set to zero. Here it is shown only for completeness. Also for the double exchange case, one has to change L by $t/2$, and conversely