Uniform and staggered magnetizations induced by Dzyaloshinskii-Moriya interactions in isolated and coupled spin-1/2 dimers in a magnetic field

S. Miyahara,[1] J.-B. Fouet,[2] S. R. Manmana,[3,4] R. M. Noack,[4] H. Mayaffre,[5] I. Sheikin,[6] C. Berthier,[5,6] and F. Mila[7]
[1]*Department of Physics and Mathematics, Aoyama Gakuin University, Sagamihara, Kanagawa 229-8558, Japan*
[2]*Institut Romand de Recherche Numérique en Physique des Matériaux (IRRMA), PPH-Ecublens, CH-1015 Lausanne, Switzerland*
[3]*Institut für Theoretische Physik III, Universität Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Germany*
[4]*Fachbereich Physik, Philipps Universität Marburg, D-35032 Marburg, Germany*
[5]*Laboratoire de Spectrométrie Physique, Université J. Fourier & UMR5588 CNRS, BP 87, 38402 Saint Martin d'Hères, France*
[6]*Grenoble High Magnetic Field Laboratory, CNRS, BP 166, F-38042 Grenoble Cedex 09, France*
[7]*Institute of Theoretical Physics, École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*
(Received 19 October 2006; revised manuscript received 1 February 2007; published 1 May 2007)

We investigate the interplay of Dzyaloshinskii-Moriya interactions and an external field in spin-1/2 dimers. For isolated dimers and at low field, we derive simple expressions for the staggered and uniform magnetizations which show that the orientation of the uniform magnetization can deviate significantly from that of the external field. In fact, in the limit where the **D** vector of the Dzyaloshinskii-Moriya interaction is parallel to the external field, the uniform magnetization actually becomes *perpendicular* to the field. For larger fields, we show that the staggered magnetization of an isolated dimer has a maximum close to one-half the polarization, with a large maximal value of $0.35g\mu_B$ in the limit of very small Dzyaloshinskii-Moriya interaction. We investigate the effect of interdimer coupling in the context of ladders with density-matrix renormalization-group (DMRG) calculations and show that, as long as the values of the Dzyaloshinskii-Moriya interaction and of the exchange interaction are compatible with respect to the development of a staggered magnetization, the simple picture that emerges for isolated dimers is also valid for weakly coupled dimers with minor modifications. The results are compared with torque measurements on $Cu_2(C_5H_{12}N_2)_2Cl_4$.

DOI: 10.1103/PhysRevB.75.184402 PACS number(s): 75.10.Jm, 75.10.Pq, 75.40.Mg, 75.30.Kz

I. INTRODUCTION

In Mott insulators, the Heisenberg interaction $J\mathbf{S}_i\cdot\mathbf{S}_j$ is in most cases the dominant source of coupling between local moments, and most theoretical investigations are based on modeling in which only this type of interaction is included. It has been known for a very long time, however, that other, less symmetric, interactions are present. For instance, unless there is an inversion center on a bond, spin-orbit coupling induces an antisymmetric interaction of the form $\mathbf{D}\cdot(\mathbf{S}_i\times\mathbf{S}_j)$, which is known as the Dzyaloshinskii-Moriya (DM) interaction.[1,2] Since it breaks the fundamental SU(2) symmetry of the Heisenberg interactions, the DM interaction is at the origin of many deviations from pure Heisenberg behavior, such as canting[3] or small gaps.[4–10] It is also known to have a dramatic impact on the properties of antiferromagnets in a magnetic field. Numerous experimental investigations of quantum antiferromagnets currently in progress in large field facilities call for a detailed understanding of this problem.[11–15] Several issues have recently been the subject of rather intensive research. For instance, the impact on triplon Bose-Einstein condensation[16–18] of DM interactions has been analyzed.[19] The interplay of frustration and DM interactions has also received significant attention.[20–23] The consequence of the breaking of SU(2) symmetry on the excitation spectrum is also well understood thanks to the work of several people including some of the present authors.[4–10] It is by now well established that a DM interaction can open a gap in otherwise gapless regions. The scaling of this gap with the magnitude of the DM interaction has been worked out for several cases.[6,8]

Surprisingly, however, the other important consequence of the breaking of the SU(2) symmetry on the ground-state properties of weakly coupled dimers, namely the development of a local magnetization, has not received much attention so far, although it is of immediate relevance to several compounds. It was shown in the case of $SrCu_2(BO_3)_2$ that a DM interaction can lead to the development of a measurable (and in fact quite large) staggered magnetization,[20] but a simple picture of how the magnitude and the orientation of the DM interaction with respect to the magnetic field influences these properties has not yet emerged. Besides, the fact that a DM interaction can lead to the development of a transverse uniform magnetization and its impact on torque measurements of the magnetization have not been investigated in detail. All these questions are central to the understanding of several systems of current interest. In particular, recent NMR results by Clémancey *et al.*[24] have revealed the presence of a staggered magnetization in the dimer compound $Cu_2(C_5H_{12}N_2)_2Cl_4$ [abbreviation: Cu(Hp)Cl], and the interpretation of these results requires a precise investigation of the effect of DM interactions on weakly coupled dimer systems.

In this paper, our goal is threefold. First of all, we want to put the theoretical results of Ref. 24 in a broader perspective, investigating all aspects of the local magnetization, and not simply the staggered magnetization that has been detected in the NMR experiment reported in Ref. 24. We also want to strengthen the case for the model proposed in Ref. 24 by reporting torque measurements which can be very naturally explained in the context of that model. Finally, a more general goal is to come up with a simple picture of the develop-

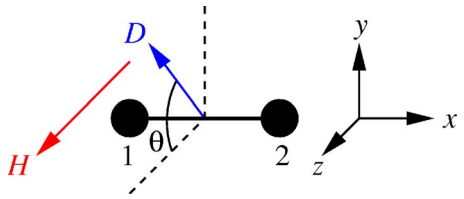

FIG. 1. (Color online) Pictorial representation of the model of Eq. (1) of a dimer with DM interaction in a magnetic field.

ment of local magnetization in the presence of DM interactions.

To this end, we first look at the case of an isolated dimer, and derive simple expressions in the limits of weak and strong magnetic field which we believe are very useful to get a simple picture of subtle issues such as the effect of the relative orientation of the magnetic field and the **D** vector of the DM interaction on the uniform magnetization. We then turn to the case of coupled dimers and concentrate on a simple ladder geometry. This choice is motivated partly by the potential relevance of this geometry to actual compounds such as Cu(Hp)Cl, and by the possibility to obtain very accurate results using the density-matrix renormalization-group method (DMRG) (Refs. 25–27) in this quasi-one dimensional geometry. We report in great detail on several quantities of direct experimental relevance such as the excitation gap or the uniform magnetization. Finally, we report on different torque measurements on Cu(Hp)Cl and discuss them in the light of these results.

II. ISOLATED DIMER

The problem of an isolated dimer in a magnetic field in the presence of a DM interaction is defined by the Hamiltonian

$$\mathcal{H} = J\mathbf{S}_1 \cdot \mathbf{S}_2 + \mathbf{D} \cdot (\mathbf{S}_1 \times \mathbf{S}_2) - g\mu_B H(S_1^z + S_2^z). \qquad (1)$$

The z axis has been chosen to be that of the magnetic field, and the yz plane as the plane defined by the magnetic field and the **D** vector. In actual systems, the direction of the **D** vector relative to the bond connecting the two sites is fixed by the microscopic arrangement of atoms and orbitals, and it is the orientation of the magnetic field that can be varied with respect to the crystal, but the convention of having the magnetic field along the z axis makes the discussion somewhat simpler. The **D** vector is written as $\mathbf{D}=(0, D\sin\theta, D\cos\theta)$. The model is illustrated in Fig. 1 in the case where both the field and the **D** are perpendicular to the dimer, but all results are valid whatever the actual orientation of the dimer (see the discussion of symmetry below).

The isolated dimer problem is, of course, very simple. The Hilbert space is of dimension 4, and it will prove convenient to work in the basis,

$$|s\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle),$$

$$|t_1\rangle = |\uparrow\uparrow\rangle,$$

$$|t_0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle),$$

$$|t_{-1}\rangle = |\downarrow\downarrow\rangle. \qquad (2)$$

In order to come up with an intuitive picture of how local magnetizations develop, we will first discuss the problem from the point of view of symmetry. We will then derive useful expressions for small D in weak field and close to saturation, and we will present plots of some representative results obtained by exact diagonalizations in the last paragraph of that section.

A. Symmetry analysis

Without DM interaction, the Hamiltonian is invariant under the real-space permutation of sites 1 and 2 denoted P_{12} in the following. Without a magnetic field, the Hamiltonian is invariant under all rotations in spin space, and under time reversal $\mathcal{T}$, which changes the sign of all components of spin operators. All these operations commute with each other. In a magnetic field, the SU(2) symmetry of the Heisenberg model is reduced to a U(1) symmetry corresponding to a rotation in spin space around the field direction $R_z(\alpha)$, and time reversal is not a symmetry any more.

As soon as a DM interaction is introduced, the Hamiltonian is no longer invariant with respect to the permutation P_{12}. If the magnetic field is parallel to the **D** vector, a rotation in spin space around their common direction is still a symmetry, but in the general case, all elementary symmetries are lost. However, the operation that exchanges sites 1 and 2 and simultaneously changes the sign of the x component of the spin operators (the component perpendicular to both the field and the **D** vector) is a symmetry operation, as can be easily checked directly in Eq. (1). This operation can be described as the composition of a rotation by π around the direction perpendicular to both the field and the **D** vector $R_x(\pi)$, the time-reversal operation $\mathcal{T}$, and the permutation P_{12}. As a consequence, the expectation values of local spin operators in any eigenstate of the Hamiltonian satisfy the relations

$$\langle S_1^x\rangle = -\langle S_2^x\rangle,$$

$$\langle S_1^y\rangle = \langle S_2^y\rangle,$$

$$\langle S_1^z\rangle = \langle S_2^z\rangle. \qquad (3)$$

These relations imply that the staggered magnetization per site, defined as $\mathbf{m}_s=(\langle\mathbf{S}_1-\mathbf{S}_2\rangle)/2$, is perpendicular to the plane defined by the magnetic field and the **D** vector, while the uniform magnetization per site defined by $\mathbf{m}_u=(\langle\mathbf{S}_1+\mathbf{S}_2\rangle)/2$ must lie in that plane.

If the **D** vector is parallel to the field, the U(1) rotational symmetry (in spin space) is still present, which can be easily checked since $S_z^{tot}=S_1^z+S_2^z$ commutes with $S_1^xS_2^y-S_1^yS_2^x$. The states $|t_{-1}\rangle$ and $|t_1\rangle$ are still eigenstates with energies $J/4\pm g\mu_B H$, but $|s\rangle$ and $|t_0\rangle$ get coupled. The staggered magnetization is identically zero, while the uniform magnetiza-

tion jumps abruptly from 0 to $2g\mu_B\hat{z}$ ($\hat{z}$ is the direction of the applied magnetic field) at a critical field H_c larger than its $D=0$ value $J/g\mu_B$.

B. Low-field limit

In the limit $D/J \ll 1$ and below the saturation field $H_c = J/g\mu_B$, the ground-state wave function, up to second order in D/J, reads

$$|\phi_0\rangle = \left(1 - \frac{D^2}{4J^2}\right)|s\rangle - \frac{D\sin\theta}{2\sqrt{2}(J - g\mu_B H)}|t_1\rangle + i\frac{D\cos\theta}{2J}|t_0\rangle - \frac{D\sin\theta}{2\sqrt{2}(J + g\mu_B H)}|t_{-1}\rangle. \quad (4)$$

In the low-field limit, first-order perturbation theory in H can be used to derive simple expressions for the expectation value of the various spin operators:

$$\langle S_1^x\rangle = -\langle S_2^x\rangle = \frac{g\mu_B HD\sin\theta}{2J^2},$$

$$\langle S_1^y\rangle = \langle S_2^y\rangle = -\frac{g\mu_B HD^2\cos\theta\sin\theta}{4J^3},$$

$$\langle S_1^z\rangle = \langle S_2^z\rangle = \frac{g\mu_B HD^2\sin^2\theta}{4J^3}. \quad (5)$$

These expressions lead to compact and suggestive expressions for the uniform and staggered magnetizations:

$$\mathbf{m}_u = \frac{g\mu_B}{4J^3}(\mathbf{D}\times\mathbf{H})\times\mathbf{D},$$

$$\mathbf{m}_s = \frac{g\mu_B}{2J^2}(\mathbf{D}\times\mathbf{H}). \quad (6)$$

As required by symmetry, the staggered magnetization is perpendicular to both the field and the **D** vector. As far as the uniform magnetization is concerned, symmetry only requires that it lies in the plane of the magnetic field and of the **D** vector, but in the low-field limit, Eq. (6) shows that it is perpendicular to the **D** vector. So the uniform magnetization is in general *not* parallel to the magnetic field, as it would be in a system with SU(2) symmetry, and it can in fact deviate strongly: In the limit where the **D** vector becomes parallel to the field, the uniform magnetization becomes perpendicular to the magnetic field, a rather anomalous behavior that should have important consequences for torque measurements of the magnetization.

Another remarkable feature of these results is that the staggered magnetization is first order in D, while the uniform magnetization is second order. Thus at low field the response is dominated by the staggered magnetization, as already observed in $SrCu(BO_3)_2$.

Finally, let us emphasize that, as implied by Eq. (6) as well as by the symmetry arguments developed in Sec. II A, the uniform and staggered magnetizations have universal expressions in terms of the magnetic field and of the **D** vector, which are valid regardless of the orientation of the dimer with respect to them.

C. Critical field

At the critical field $H_c = J/g\mu_B$, one has to turn to degenerate perturbation theory since, for $D=0$, $|s\rangle$ and $|t_1\rangle$ are degenerate. When the **D** vector is not parallel to the field, these states get coupled by an off-diagonal term $D\sin\theta$. In the limit $D\to 0$, the ground-state wave function is then simply given by $\phi_0 = (|s\rangle - |t_1\rangle)/\sqrt{2}$, and the staggered magnetization per site is equal to $(\sqrt{2}/4)g\mu_B \simeq 0.35 g\mu_B$, independently of the angle θ. Such a large value in the limit $D\to 0$ might come as a surprise since, when $D=0$, the staggered magnetization should be identically zero. But in fact, when D is small, the staggered magnetization becomes peaked around H_c, in an interval of width of the order of $D\sin\theta$, which shrinks to zero in the limit where $D\to 0$. So, for $H \neq H_c$, the staggered magnetization indeed goes to zero when $D\to 0$. In addition at the critical field $H=H_c$, the staggered magnetization is ill defined in the case $D=0$ since the ground state becomes degenerate.

When $D \ll J$, the uniform magnetization at this field is equal to $g\mu_B$, which corresponds to half the polarization value. When the angle between D and H is not $\pi/2$, a small uniform component develops along y due to the coupling of $|s\rangle$ with $|t_0\rangle$. This transverse (with respect to the field) uniform magnetization is given by $m_u^y = -(\sqrt{2}/4)\cos\theta(D/J)g\mu_B \simeq -0.35\cos\theta(D/J)g\mu_B$. In contrast to the small-field result, it is now linear in D, but remains much smaller than the staggered magnetization, which is of order 1.

D. Exact results

To get an idea of the accuracy of the expressions obtained at low field and close to the saturation field, we have plotted in Fig. 2 the exact value of m_s^x, m_u^y, and m_u^z for a representative case ($D/J=0.04$ and $\theta=\pi/4$). The small-field expression is quantitatively accurate up to $H \simeq 0.25J/g\mu_B$, and the width of the peak of the staggered magnetization and the maximal value of m_u^y are indeed of order D.

III. COUPLED DIMERS (LADDER)

A. Model

In this section, our goal is to check to which extent the properties of a system of weakly coupled dimers resemble those of isolated dimers. In particular, the transition between zero magnetization and polarization takes place through an extended region of magnetic field of the order of the interdimer coupling, and we would like to know how the system behaves within and outside this region. We will attack this problem numerically, and in order to perform simulations on large systems, we have chosen to work in a ladder geometry and to use the DMRG. The model is defined by the Hamiltonian

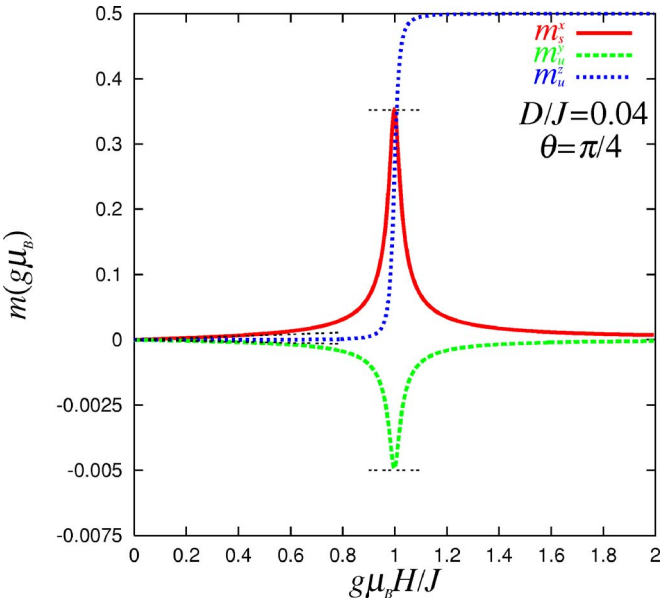

FIG. 2. (Color online) Field dependence of the uniform and staggered magnetizations per site m_u^y, m_u^z, and m_s^x of the isolated dimer model for $D/J=0.04$ and $\theta=\pi/4$. The dashed lines are the analytical results derived in the limit $D/J \ll 1$. Note the difference of scale for positive and negative magnetizations.

$$\mathcal{H} = J\sum_i \mathbf{S}_{i,1}\cdot\mathbf{S}_{i,2} + \sum_i (-1)^i \mathbf{D}\cdot(\mathbf{S}_{i,1}\times\mathbf{S}_{i,2}) + J_\parallel \sum_i (\mathbf{S}_{i,1}\cdot\mathbf{S}_{i+1,1} + \mathbf{S}_{i,2}\cdot\mathbf{S}_{i+1,2}) - g\mu_B H\sum_i (S_{i,1}^z + S_{i,2}^z). \tag{7}$$

As for the isolated dimer, the **D** vector is assumed to lie in the yz plane, i.e., $\mathbf{D}=(0, D\sin\theta, D\cos\theta)$. Our choice of an alternating **D** vector from one rung to the other (see Fig. 3) is motivated by symmetry considerations. Indeed, in a canonical ladder, the middle of each rung is an inversion center, and the DM interaction vanishes by symmetry. A simple way to allow for the DM interaction to become finite without modifying the symmetry of the exchange couplings is to assume that some buckling is present along the ladder, as sketched in Fig. 4. In that case, the only mirror plane that contains a bond is the xz plane, and a DM interaction with a **D** vector parallel to y is allowed by symmetry. But, in this geometry, the presence of a C_2 axis (see Fig. 4) implies that the **D** vector alternates from one rung to the other. The buckling realized in Cu(Hp)Cl is slightly more subtle (successive rungs are connected by an inversion symmetry in the middle of a plaquette), but this symmetry also implies alternating **D** vectors. Note, however, that other ways of breaking the inversion symmetry of the rungs can lead to other arrangements of **D** vectors.

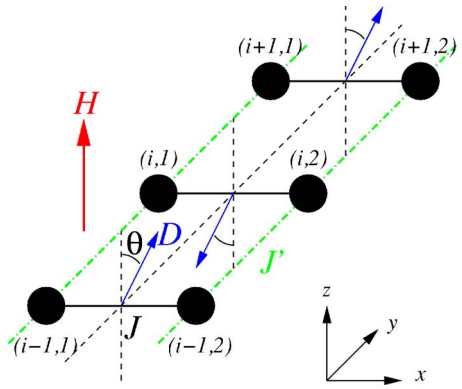

FIG. 3. (Color online) Ladder with staggered DM interaction.

Another motivation to work with alternating **D** vectors is to keep the perturbation caused by the interdimer coupling as small as possible. In that respect, this choice is natural. Indeed, as we have seen in the previous section, the presence of a **D** vector on a rung induces a staggered magnetization. If the **D** vectors of neighboring rungs i and $i+1$ are equal, the moments $\langle\mathbf{S}_{i,1}\rangle$ and $\langle\mathbf{S}_{i+1,1}\rangle$ will also be equal, which is in conflict with antiferromagnetic interrung exchange interactions. If, on the contrary, the **D** vectors are opposite on neighboring rungs, the local moments will adopt configurations that are compatible with the exchange.

Note that a ladder structure is always consistent with DM interactions along the legs of the ladder. In the context of weakly coupled dimers, these interactions, which should be a small fraction of the exchange, are expected to be extremely small, and we have checked that the effect of a small interrung DM interaction is indeed negligible on the results discussed below.

B. Symmetry analysis

While symmetry considerations related to the microscopic origin of the DM interaction will usually force it to lie in a

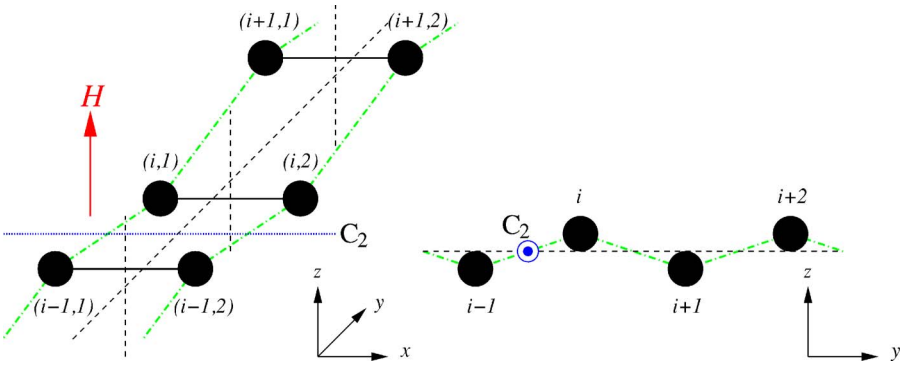

FIG. 4. (Color online) Structure of a buckled ladder. In such a ladder, a staggered DM interaction in the y direction is allowed by symmetry.

specific direction with respect to the lattice, as for a single dimer, the discussion of symmetry is simpler if real space and spin space are separated. For the general case where the magnetic field and the **D** vectors are not parallel, the Hamiltonian has two types of symmetries acting only in real space: (i) the inversion centers around the middles of the plaquettes formed by two consecutive rungs; (ii) all even translations along the ladder direction. It also has a symmetry that acts in both real space and spin space (the global version of the symmetry identified for a dimer), namely (iii) the operation that exchanges sites $(i,1)$ and $(i,2)$ or all dimers and simultaneously changes the sign of the x component of all spin operators. As long as these symmetries are not broken, the following relations between the expectation values of local spin operators on two neighboring rungs are expected to be satisfied:

$$\langle S^x_{i,1}\rangle = -\langle S^x_{i,2}\rangle = -\langle S^x_{i+1,1}\rangle = \langle S^x_{i+1,2}\rangle,$$

$$\langle S^y_{i,1}\rangle = \langle S^y_{i,2}\rangle = \langle S^y_{i+1,1}\rangle = \langle S^y_{i+1,2}\rangle,$$

$$\langle S^z_{i,1}\rangle = \langle S^z_{i,2}\rangle = \langle S^z_{i+1,1}\rangle = \langle S^z_{i+1,2}\rangle, \quad (8)$$

where x is the direction perpendicular to both the field and the **D** vector. As for the single dimer case, these relations are valid regardless of the actual orientation of the lattice with respect to the magnetic field and **D** vector. They only require the DM interaction to alternate from one rung to the next. We thus define the staggered and uniform magnetizations per site as

$$\mathbf{m}_s = (1/N)\sum_i (-1)^i(\langle \mathbf{S}_{i,1}\rangle - \langle \mathbf{S}_{i,2}\rangle),$$

$$\mathbf{m}_u = (1/N)\sum_i (\langle \mathbf{S}_{i,1}\rangle + \langle \mathbf{S}_{i,2}\rangle), \quad (9)$$

where N is the total number of sites, and with the convention that the angle θ is positive for i even. As in the isolated dimer case, the staggered magnetization $\mathbf{m}_s$ is along the x axis, while the uniform magnetization $\mathbf{m}_u$ lies in the yz plane.

C. Uniform and staggered magnetizations

Let us now turn to the discussion of the numerical results we have obtained for the model of Eq. (7). We are interested in the regime $D<J_\parallel<J$. For $D=0$, the model is a simple ladder in a field, and the properties are well understood. There is, of course, no staggered magnetization because of the U(1) symmetry, and the uniform magnetization is parallel to the field for the same reason. It vanishes below a critical field H_{c1}, takes off with a square-root singularity, and reaches saturation with another square-root singularity at a second critical field H_{c2}. The difference $H_{c2}-H_{c1}$ scales with $J_\parallel$. Since, apart from this scaling, the properties depend very little on $J_\parallel$, we quote results for a single value of $J_\parallel$, and having in mind the compound Cu(Hp)Cl,[11] we have chosen $J_\parallel/J=0.2$. For that ratio, the critical fields in the absence of DM interactions are given by $g\mu_B H_{c1}=0.82J$ and $g\mu_B H_{c2}=1.40J$.

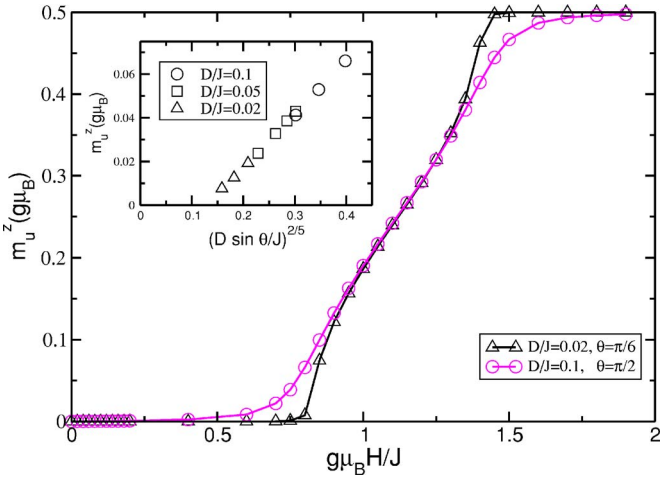

FIG. 5. (Color online) Examples of the variation of the uniform magnetization along the z axis with the field. Inset: Plot of m_u^z as a function of $(D \sin \theta/J)^{2/5}$ slightly below H_{c1}, which confirms the scaling predicted in Ref. 8.

For the model with DM interaction, we have performed exact diagonalization (ED),[27] up to 20 sites (10 rungs), and DMRG calculations on ladders with up to 80 rungs. The results evolve smoothly with the size, and we only quote DMRG results obtained for 80-rung clusters (finite-size effects for the gap are discussed in the next section). Note that in those calculations, S_z is not a good quantum number. This is well known to reduce greatly the maximal size available to exact diagonalizations, but this also has an impact on the number of states we were able to keep during the DMRG runs. Here, we diagonalize (by the Davidson method) a matrix of size $4m^2$ at each DMRG step. In a standard DMRG run where S_z is a good quantum number, the matrix of the effective Hamiltonian in the variational basis is block-diagonal, which can speed up the diagonalization by a factor of 10 or more. The memory needed is also larger at fixed m than for the standard DMRG. For those reasons, most of the calculations were done with up to $m=600$ states kept during five sweeps, and only up to $N=80$ sites. The discarded weight was of the order of 10^{-10} when we targeted two states to extract the gap, and of the order of 10^{-12} or less when we targeted a single state to extract correlations. We also performed a few runs with m up to 800 in order to confirm that the numerical data were well converged.

The z component of the magnetization is displayed in Fig. 5 for several values of D and θ. It is reminiscent of that for $D=0$; however, when $D\neq 0$, the magnetization develops as soon as the magnetic field is switched on, only reaching saturation asymptotically in the limit of infinite field. The square-root singularities are removed. It was shown in Ref. 10 that, at H_{c1}, the magnetization should depend on the magnitude of the D vector as $m_u^z \propto (D \sin \theta)^{2/5}$, in agreement with the present results (see the inset of Fig. 5).

When $\theta \neq \pi/2$ (i.e., $D_z \neq 0$), a uniform magnetization along the y axis also develops, as in the isolated dimer case. Figure 6 shows the magnetization along the y axis and the angle ϕ_u between the uniform magnetization and the z axis as a function of the magnetic field for $\theta=\pi/6$. At low field,

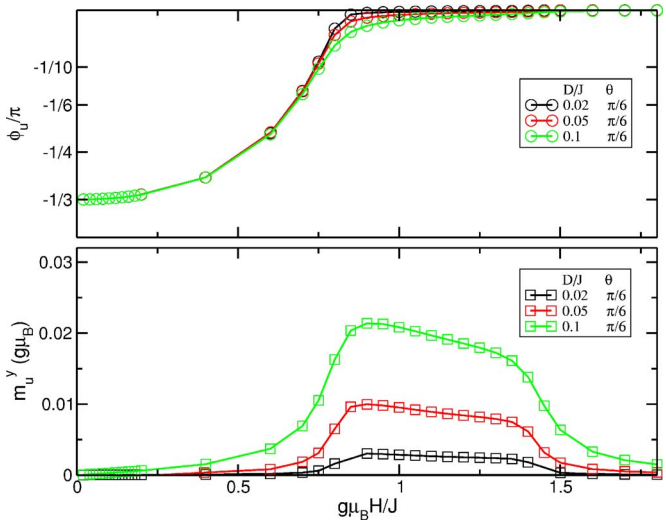

FIG. 6. (Color online) Lower panel: y component of the uniform magnetization as a function of the field for $\theta=\pi/6$ and various values of D/J. Significant values appear between H_{c1} and H_{c2}, and far outside this interval as soon as D/J is not too small. Upper panel: Angle ϕ_u between the magnetic field and the uniform magnetization $\mathbf{m}_u$ as a function of the field for $\theta=\pi/6$ and several values of D/J. Note that $\theta-\phi_u$ goes to $\pi/2$ in the low-field limit, in agreement with the prediction for an isolated dimer [Eq. (6)].

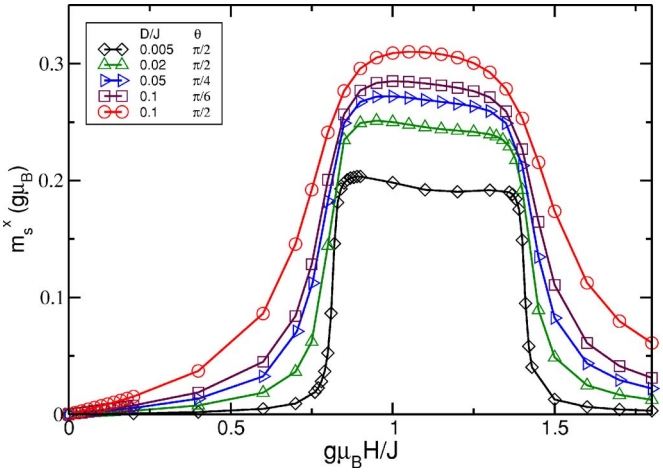

FIG. 7. (Color online) Staggered magnetization as a function of the magnetic field for several values of D/J and θ. Large values are achieved between H_{c1} and H_{c2}, and far outside this interval as soon as D/J is not too small. The value between H_{c1} and H_{c2} depends relatively weakly on D/J and θ, and is of the same order as the maximal value in the case of an isolated dimer ($0.35g\mu_B$). In contrast, the value outside this interval depends very strongly on the magnitude of $D \sin\theta$.

the uniform magnetization is orthogonal to the DM vector, again as for an isolated dimer. The magnetization along y is maximal between the two critical fields. Its value in that range is clearly much smaller than the component along the field (ϕ_u becomes very small near H_{c1}), but this extra contribution to the uniform magnetization will produce a torque that should be detectable experimentally given the very high sensitivity of torque measurements.

The staggered magnetization along x exhibits a kind of plateau in the intermediate phase between H_{c_1} and H_{c_2} (Fig. 7). Its magnitude inside the plateau is of the order of the maximal value of the isolated dimer ($0.35g\mu_B$), and it depends relatively weakly on D. In contrast, the extent of the tails outside this plateau region increases rapidly with D. Remarkably, the magnetization per spin along x is larger than along z up to H_{c1} and even slightly above. Note that the staggered magnetization depends essentially on the value of D_y and is very weakly affected by the value of D_z.

D. Gap

The effect of a SU (2) breaking interaction on a ladder has been studied in Ref. 10. It strongly depends on the nature of the plateau phase. For the transition from the zero or full polarization to the gapless phase, the effective-field theory is expected to be the same as for the spin chain close to saturation, and the gaps at H_{c1} and H_{c2} should open as $(D \sin\theta)^{4/5}$, as shown in Ref. 8. This prediction clearly agrees with the results for $\theta=\pi/2$ shown in Fig. 8 (lower right panel). Size effects are already very small for $N=80$ sites, as can be seen in Fig. 8 (lower left panel). Between the two critical fields, the gap is expected to remain finite. (The closing of the gap in Ref. 10 was caused by a breaking of the Z_2 symmetry which does not occur here as there is no $m=1/2$ plateau when $D/J=0$). The effect of the z component of D is expected to be very small. This is also confirmed by our DMRG results (not shown).

Note that, because of the presence of a gap, the finite-size effects are expected to be negligible if the cluster size is larger than the correlation length. Since the correlation is inversely proportional to the gap, the finite-size effects are expected to be maximal at the two critical fields where the gap is minimum. As shown in the lower left panel of Fig. 8, these effects are already completely negligible for 40 sites, and all the results presented in this paper can be considered to accurately represent the thermodynamic limit.

E. Application to experiments

The existence of a field induced staggered magnetization in dimer systems has been established by NMR in $SrCu_2(BO)_3$ (Ref. 20) and in Cu(Hp)Cl.[24] In this latter compound, in which $H_{c1}=7.5$ T and $H_{c2}=13$ T, the field dependence of $\mathbf{m}_s$ at 50 mK between 5 and 15 T has been very well reproduced by our calculation on a ladder for reasonable parameter values: $D/J=0.05$, $J_\parallel/J=0.2$, $J=13$ K. The field-induced transverse uniform magnetization $\mathbf{m}_u$, however, is an order of magnitude smaller than $\mathbf{m}_s$, and very difficult to detect in NMR measurements. On the contrary, magnetic torque measurements are not sensitive to the transverse staggered magnetization, but are ideally suited to detect the presence of $\mathbf{m}_u$, as explained in the following. Let us recall that in the presence of an external magnetic field H applied along the $\mathbf{z}$ axis, a crystal which is able to rotate around an axis $\mathbf{i}$ is submitted to a torque which depends on the orientation of $\mathbf{z}$ and $\mathbf{i}$ with respect to the principal axis of the susceptibility

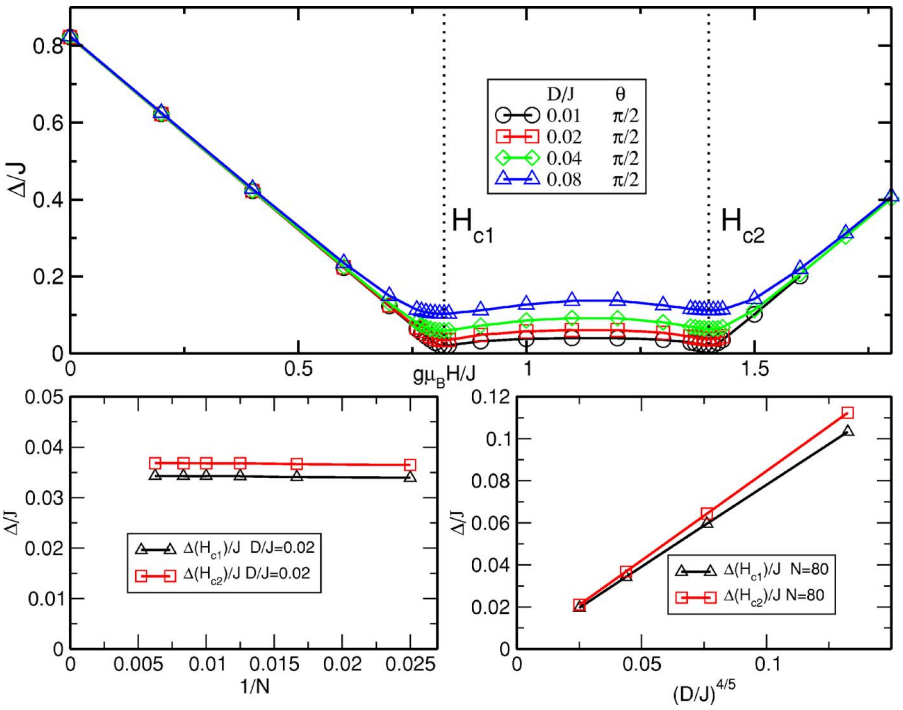

FIG. 8. (Color online) Upper panel: Field dependence of the excitation gap $\triangle$ for $J_{\parallel}/J=0.2$ and several values of D/J: $D/J=0.01$ and $\theta=\pi/2$ (black circle), $D/J=0.02$ and $\theta=\pi/2$ (red square), $D/J=0.04$ and $\theta=\pi/2$ (green diamond), and $D/J=0.08$ and $\theta=\pi/2$ (blue triangle) for $N=80$ (DMRG). Lower left panel: Scaling of the excitation gap as a function of $1/N$ for $D/J=0.02$ and $\theta=\pi/2$. Lower right panel: Scaling of the gap as a function of D/J for $N=80$ (see text).

tensor χ. The torque is equal to zero each time the magnetic energy $E_M=-H^2\mathbf{z}\chi\mathbf{z}$ passes through an extremum when rotating the crystal around $\mathbf{i}$. Let us consider the simple geometry where $\mathbf{i}$ is perpendicular both to the applied field and to the easy axis of the susceptibility tensor, which we shall assume to have an axial symmetry. In that case, τ can be simply expressed as

$$\tau=(\chi_{\perp}-\chi_{\parallel})\sin 2\theta H^2, \tag{10}$$

where θ is the angle between $\mathbf{z}$ and the easy axis of χ. In the absence of Dzyaloshinskii-Moriya interaction, the anisotropy of the magnetic susceptibility of a system of coupled dimers of spins 1/2 is expected to come only from the macroscopic resultant of the individual g tensors. Such an anisotropy is both temperature and field independent. However, as shown above, a staggered DM interaction along the rungs of a ladder can also induce a component of the magnetization with a component m_u^y perpendicular to the applied magnetic field, which strongly varies with H and disappears only well above H_{c2} (Fig. 6). Except for very peculiar orientations of $\mathbf{D}$, this component will exist even if the field is applied along the easy axis, so that only m_u^y will contribute to the torque, which should thus vanish for fields much larger than H_{c2}.

In order to test this idea, magnetic torque measurements have been performed at low temperature (410 mK) on the same compound Cu(Hp)Cl in which the presence of field induced staggered magnetization $\mathbf{m}_s$ was observed by NMR.[24] Experiments were carried out in a resistive magnet and τ was measured from zero up to 23 T. A small crystal of Cu(Hp)Cl was glued on a beryllium bronze cantilever, the displacements of which were measured capacitively. The cantilever could be rotated *in situ* with respect to the applied field H. The orientation of the crystal was adjusted so that $\tau=0$ at the highest values of H, as shown in the inset of Fig. 9, which fully cancels the contribution due to the anisotropy of the g tensor. This orientation indeed corresponds to $H\|[100]$. In spite of this, a large additional contribution shows up between the two critical fields, and extends well outside the intermediate region. For comparison, the calculated component of the uniform magnetization perpendicular to the field of a ladder with $J_{\parallel}/J=0.2$, $D_y/J=0.05$, and $D_z/J=0.086$ ($\theta=\pi/6$) is depicted on the same plot, with scales adjusted to get the same value at H_{c1}. The values of D_y and $J_{\parallel}$ are those used in Ref. 24 to fit the staggered magnetization, while the results depend very little on D_z up to an overall scale factor. The two curves are in good qualitative

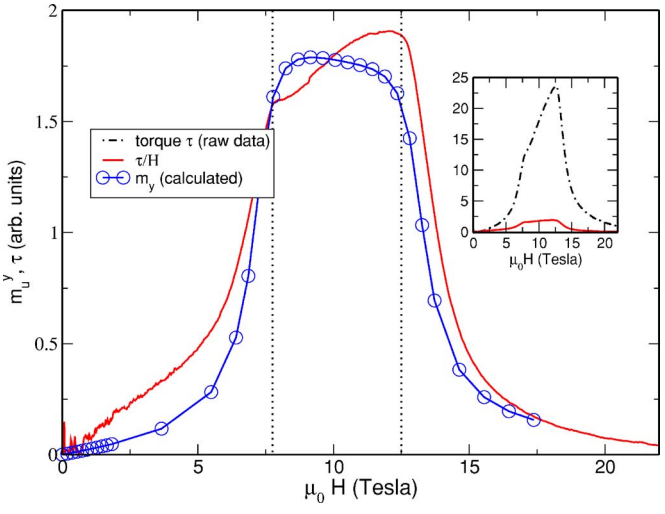

FIG. 9. (Color online) Transverse uniform magnetization m_u^y for $D_y/J=0.05$, $D_z/J=0.086$, and $J_{\parallel}/J=0.2$ (blue circles) and torque divided by field [experimental curve obtained on Cu(Hp)Cl] as a function of the field (red line). Inset: torque measurement raw data (dashed-dotted black line) and torque divided by field (red line).

agreement, especially considering the fact that the only adjustable parameter is the overall scale factor. In order to go beyond this qualitative agreement, it would be necessary to consider several additional effects. First of all, inelastic neutron-scattering data have challenged the description of this system as a simple ladder,[28] and further couplings (still not definitely identified) should presumably be included. In addition, there is a transition into a three-dimensional (3D) ordered phase[11] between H_{c1} and H_{c2}, and although the precise nature of the ordering is still unknown, it is very likely it will affect the uniform magnetization. The onset of the 3D ordering at H_{c1} is clearly visible on the experimental measurements shown in Fig. 9. Obviously, at the present stage, too little is known about these additional effects to be able to take them into account, and this is left for future investigation.

IV. CONCLUSIONS

If spin-1/2 dimers are coupled in such a way that there is no inversion center at the middle of the bond, very significant modifications of the physics in a magnetic field have to be expected. Indeed, unless it is forbidden by symmetry, a DM interaction will always be present, and the analysis reported in this paper shows that even a tiny DM interaction can modify some aspects of the physics rather dramatically. This is especially true for the staggered magnetization, which immediately acquires large values in the intermediate phase where the system gets polarized, and which can take on significant values outside this phase for physically relevant values of the DM interaction. This is also true for the uniform magnetization as soon as the **D** vector of the DM interaction and the field are neither parallel nor perpendicular. In that case, a component of the uniform magnetization perpendicular to the magnetic field appears, which can induce a measurable torque on the sample. This has been proven for an isolated dimer and for a ladder with staggered DM interactions, but these conclusions are expected to hold true for all coupled-dimer systems as long as the **D** vectors are arranged in such a way that there is no competition with Heisenberg exchange as far as the development of a staggered magnetization is concerned. It is our hope that these results will help understand some of the strange properties observed in coupled-dimer systems.

ACKNOWLEDGMENTS

We acknowledge useful discussions with Karlo Penc and Oleg Tchernyshyov. This work was supported by the Grant-in-Aids for Scientific Research on Priority Areas "Invention of Anomalous Quantum Materials" and for Aoyama Gakuin University 21st COE Program from the Ministry of Education, Culture, Sports, Science and Technology of Japan, by the Swiss National Fund, by MaNEP, and by the SFB 382.

[1] I. E. Dzyaloshinskii, J. Phys. Chem. Solids **4**, 241 (1958).
[2] T. Moriya, Phys. Rev. Lett. **4**, 228 (1960).
[3] D. Coffey, K. S. Bedell, and S. A. Trugman, Phys. Rev. B **42**, 6509 (1990).
[4] T. Sakai and H. Shiba, J. Phys. Soc. Jpn. **63**, 867 (1994).
[5] D. C. Dender, P. R. Hammar, D. H. Reich, C. Broholm, and G. Aeppli, Phys. Rev. Lett. **79**, 1750 (1997).
[6] M. Oshikawa and I. Affleck, Phys. Rev. Lett. **79**, 2883 (1997).
[7] J. Z. Zhao, X. Q. Wang, T. Xiang, Z. B. Su, and L. Yu, Phys. Rev. Lett. **90**, 207204 (2003).
[8] J.-B. Fouet, O. Tchernyshyov, and F. Mila, Phys. Rev. B **70**, 174427 (2004).
[9] A. L. Chernyshev, Phys. Rev. B **72**, 174414 (2005).
[10] J.-B. Fouet, F. Mila, D. Clarke, H. Youk, O. Tchernyshyov, P. Fendley, and R. M. Noack, Phys. Rev. B **73**, 214405 (2006).
[11] G. Chaboussant, M.-H. Julien, Y. Fagot-Revurat, M. Hanson, L. P. Lévy, C. Berthier, M. Horvatić, and O. Piovesana, Eur. Phys. J. B **6**, 167 (1998).
[12] H. Kageyama, K. Yoshimura, R. Stern, N. V. Mushnikov, K. Onizuda, M. Kato, K. Kosuge, C.-P. Slichter, T. Goto, and Y. Ueda, Phys. Rev. Lett. **82**, 3168 (1999).
[13] O. Cépas, K. Kakurai, L. P. Regnault, T. Ziman, J. P. Boucher, N. Aso, M. Nishi, H. Kageyama, and Y. Ueda, Phys. Rev. Lett. **87**, 167205 (2001).
[14] R. Coldea, D. A. Tennant, K. Habicht, P. Smeibidl, C. Wolters, and Z. Tylczynski, Phys. Rev. Lett. **88**, 137203 (2002).
[15] M. Jaime, V. F. Correa, N. Harrison, C. D. Batista, N. Kawashima, Y. Kazuma, G. A. Jorge, R. Stein, I. Heinmaa, S. A. Zvyagin, Y. Sasago, and K. Uchinokura, Phys. Rev. Lett. **93**, 087203 (2004).
[16] I. Affleck, Phys. Rev. B **41**, 6697 (1990); **43**, 3215 (1991).
[17] T. Giamarchi and A. M. Tsvelik, Phys. Rev. B **59**, 11398 (1999).
[18] M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. **89**, 077203 (2002).
[19] J. Sirker, A. Weisse, and O. P. Sushkov, Europhys. Lett. **68**, 275 (2004).
[20] K. Kodama, S. Miyahara, M. Takigawa, M. Horvatic, C. Berthier, F. Mila, H. Kageyama, and Y. Ueda, J. Phys.: Condens. Matter **17**, L61 (2005).
[21] M. Elhajal, B. Canals, and C. Lacroix, Phys. Rev. B **66**, 014422 (2002).
[22] M. Elhajal, B. Canals, R. Sunyer, and C. Lacroix, Phys. Rev. B **71**, 094420 (2005).
[23] V. N. Kotov, M. Elhajal, M. E. Zhitomirsky, F. Mila, Phys. Rev. B **72**, 014421 (2005).
[24] M. Clémancey, H. Mayaffre, C. Berthier, M. Horvatić, J.-B. Fouet, S. Miyahara, F. Mila, B. Chiari, O. Piovesana, Phys. Rev. Lett. **97**, 167204 (2006).
[25] S. R. White, Phys. Rev. Lett. **69**, 2863 (1992); Phys. Rev. B **48**, 10345 (1993).
[26] U. Schollwöck, Rev. Mod. Phys. **77**, 259 (2005).
[27] R. M. Noack and S. R. Manmana, in *Lectures on the Physics of Highly Correlated Electron Systems IX: Ninth Training Course in the Physics of Correlated Electron Systems and High-Tc Superconductors*, edited by A. Avella and F. Mancini, AIP Conf. Proc. No. 789 (AIP, Melville, NY, 2005), p. 93.
[28] M. B. Stone, Y. Chen, J. Rittner, H. Yardimci, D. H. Reich, C. Broholm, D. V. Ferraris, and T. Lectka, Phys. Rev. B **65**, 064423 (2002).