Magnetization plateaus in weakly coupled dimer spin system

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I study a spin system consisting of strongly coupled dimers which are, in turn, weakly coupled in a plane by zigzag interactions. The model can be viewed as the strong-coupling limit of a two-dimensional zigzag chain structure typical, e.g., for the (ac) planes of $KCuCl_3$. It is shown that the magnetization curve in this model has plateaus at $\frac{1}{3}$ and $\frac{2}{3}$ of the saturation magnetization, and an additional plateau at $\frac{1}{2}$; the critical fields are calculated perturbatively. It is argued that for the three-dimensional lattice structure of the $KCuCl_3$ family the plateaus at $\frac{1}{4}$ and $\frac{3}{4}$ of the saturation can be favored in a similar way, which might be relevant to the recent experiments on NH_4CuCl_3 by Shiramura *et al.*, J. Phys. Soc. Jpn. **67**, 1548 (1998). [S0163-1829(99)07705-X]

I. INTRODUCTION

In the last couple of years there has been a growing interest in the phenomenon of magnetization plateaus in one-dimensional (1D) spin systems.[1–14] In a variety of models, the magnetization per site m_z as a function of the applied magnetic field H exhibits plateaus at certain values of m_z; at those plateaus the magnetization in units of the saturation $M=m_z/S$ (here S is the spin of a magnetic ion) is ''locked'' at some rational number (at least, up to now no indications of the possibility to have plateaus at irrational M were found). Oshikawa *et al.*[4] have shown that in purely one-dimensional systems the allowed values of M, at which the plateaus are possible, are given by the following condition:

$$QS(1-M)\in\mathbb{Z}, \tag{1}$$

where Q is the number of magnetic ions in the magnetic elementary cell of the ground state (which can be different from the elementary cell prescribed by the Hamiltonian[7,9]). This condition is necessary but not sufficient; in other words, if the plateau exists, it has to be at one of the values of M determined by Eq. (1), but in principle the plateaus do not have to exist at *all* values of M which are allowed by Eq. (1). Validity of the result (1) is supported by a considerable amount of numerical and analytical data, as well as by the recent experimental observation[8] of the $M=\frac{1}{2}$ plateau in the $S=1$ bond-alternating chains realized in $[Ni_2(Medpt)_2(\mu\text{-ox})(\mu\text{-}N_3)]ClO_4\cdot 0.5H_2O$.

The present paper is motivated by the recent high-field magnetization measurements[15] in the double-chain $S=\frac{1}{2}$ compound NH_4CuCl_3 that have revealed well-pronounced plateaus at $M=\frac{1}{4}$ and $M=\frac{3}{4}$ in the temperature range $T<1.5$ K. At room temperature, the crystal structure of this material coincides with that of $KCuCl_3$, and the main feature is the presence of the double chains of edge-sharing $CuCl_6$ octahedra, separated by NH_4^+ ions (see Figs. 2 and 3 of Ref. 16 or Fig. 1 of Ref. 17). Double chains composed of magnetic Cu^{2+} ions can be viewed as spin-$\frac{1}{2}$ bond-alternating antiferromagnetic (AF) zigzag chains with two different nearest-neighbor exchange constants J_2, J_3 and next-nearest-neighbor exchange J_1 (see Fig. 1 of Ref. 17, and Fig. 1 of the present paper); in what follows, the word ''chain'' is used in the meaning of a zigzag (double) chain. If one assumes that the essential physics in NH_4CuCl_3 is determined by single chains, then the appearance of plateaus at $\frac{1}{4}$ and $\frac{3}{4}$ of saturation means that due to a certain rather complicated spontaneous symmetry breaking the magnetic elementary cell contains $Q=8$ Cu^{2+} ions instead of $Q=2$ as suggested by the Hamiltonian symmetry; still, then the absence of plateau at $M=\frac{1}{2}$ that is characteristic for a single chain[9] is puzzling.

Values of the exchange constants in NH_4CuCl_3, as well as in the other materials of the so-called $KCuCl_3$ family,[16,17] are presently not known. There are certain arguments[18] based on fitting the susceptibility data for the isostructural compound $KCuCl_3$ that suggest that one of the nearest-neighbor couplings (J_3) is dominating.[19] On the other hand, neutron-scattering data for $KCuCl_3$ strongly suggest that interchain interactions are important.[20] From the susceptibility data,[17,15] one can guess that the intrachain couplings in NH_4CuCl_3 are much weaker than in $KCuCl_3$ and thus interchain couplings should play an even more important role; this assumption also fits well to the fact that the zero-field ground state of NH_4CuCl_3 is magnetic at low temperature.[15]

In the present paper I show that interchain couplings can considerably affect the number and positions of the plateaus. To demonstrate that, I consider a simple model describing a system of zigzag chains coupled in a plane; within the effective spinless fermion model valid in the limit of weakly coupled dimers it is shown that this model can exhibit magnetization plateaus at $\frac{1}{3}$, $\frac{1}{2}$, and $\frac{2}{3}$ of the saturation. The critical fields are calculated using the lowest-order perturbation theory around the ''atomic limit'' of the model. Physics of the plateau at $M=\frac{1}{2}$ is determined by single chains and is almost exactly the same as studied in Ref. 9, while the plateaus at $M=\frac{1}{3},\frac{2}{3}$ appear solely due to the two-dimensional interchain couplings. Generally speaking, the number and positions of the ''additional'' plateaus caused by interchain interactions strongly depend on the topology of the specific lattice. Possible relevance of this mechanism to the experiments on NH_4CuCl_3 is discussed: it is argued that the three-

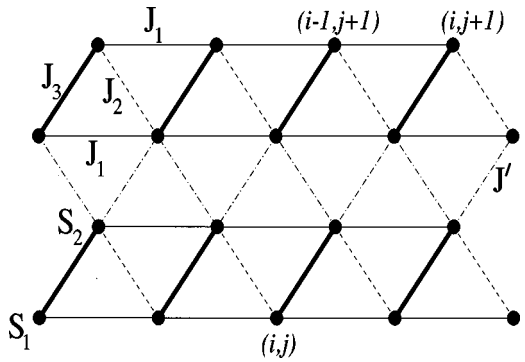

FIG. 1. Two-dimensional system of coupled zigzag chains described by the Hamiltonian (2), a structure that is realized in the (ac) planes of the $KCuCl_3$ family compounds (Refs. 16 and 17).

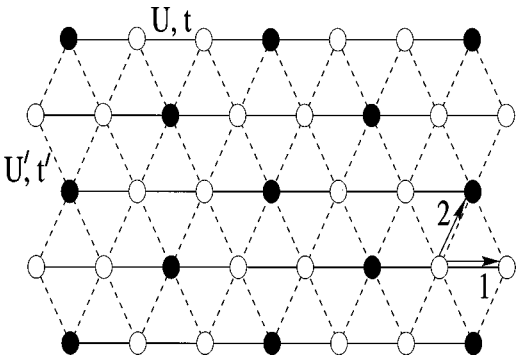

FIG. 2. Effective model of spinless fermions on a triangular lattice resulting from the system shown in Fig. 1, as described by Eq. (3). The structure with $M=\frac{1}{3}$ in the ''atomic limit'' is shown; open and filled circles denote, respectively, empty and filled sites (i.e., dimers in $|s\rangle$ and $|t_+\rangle$ states).

dimensional lattice structure of NH_4CuCl_3 naturally favors the appearance of plateaus at $\frac{1}{4}$ and $\frac{3}{4}$ of the saturation. We also discuss possible reasons of absence of the other plateaus in this material.

The paper is organized as follows: in Sec. II the effective model for a weakly coupled dimer system is introduced and the magnetization curve of the two-dimensional system of coupled chains is studied; Sec. III is devoted to the analysis of characteristic features of a three-dimensional coupled chain system typical for NH_4CuCl_3; Sec. IV contains discussion and concluding remarks.

II. TWO-DIMENSIONAL COUPLED CHAIN MODEL

Consider a system of zigzag chains coupled in a plane by frustrating zigzag interaction J', as shown in Fig. 1. Such a structure is typical, e.g., for (ac) planes of $KCuCl_3$. Let us further assume that the exchange constant J_3 is antiferromagnetic (AF) and much stronger than all the others; then the system can be viewed as a two-dimensional arrangement of dimers weakly coupled with each other. Assume further that the external magnetic field $\boldsymbol{H}$ is applied in the z direction. The model is described by the Hamiltonian

$$\begin{aligned}\hat{H}=\sum_{ij}\ \{&J_3\boldsymbol{S}_1^{(i,j)}\cdot\boldsymbol{S}_2^{(i,j)}+J_2\boldsymbol{S}_1^{(i,j)}\cdot\boldsymbol{S}_2^{(i-1,j)}\\&+J_1(\boldsymbol{S}_1^{(i,j)}\cdot\boldsymbol{S}_1^{(i+1,j)}+\boldsymbol{S}_2^{(i,j)}\cdot\boldsymbol{S}_2^{(i+1,j)})\\&+J'(\boldsymbol{S}_2^{(i,j)}\cdot\boldsymbol{S}_1^{(i,j+1)}+\boldsymbol{S}_2^{(i,j)}\cdot\boldsymbol{S}_1^{(i-1,j+1)})\\&+g\mu_B\boldsymbol{H}\cdot(\boldsymbol{S}_1^{(i,j)}+\boldsymbol{S}_2^{(i,j)})\},\end{aligned}\tag{2}$$

where g is the Landé factor and μ_B is the Bohr magneton. If one neglects weak interdimer interactions J_1, J_2, J' completely, than the lowest states at each dimer are the singlet $|s\rangle$ and the $S^z=+1$ triplet $|t_+\rangle$. We will be interested in the regime of strong fields $g\mu_BH\gtrsim J_3$; then the other two triplet states $|t_0\rangle$, $|t_-\rangle$ are high in energy and their contribution can be neglected, in the spirit of Refs. 9, 12, and 14. In this way one gets a reduced Hilbert space with only two degrees of freedom per dimer, and the problem can be formulated in terms of spinless fermions living on the asymmetric triangular lattice as shown in Fig. 2, with the following effective Hamiltonian $\hat{\mathcal{H}}$:

$$\begin{aligned}\hat{\mathcal{H}}=&\mu\sum_{ij}\ n_{ij}+U\sum_{ij}\ n_{ij}n_{i+1,j}+t\sum_{ij}\ (c_{ij}^{\dagger}c_{i+1,j}+\text{H.c.})\\&+U'\sum_{ij}\ n_{ij}(n_{i,j+1}+n_{i-1,j+1})\\&+t'\sum_{ij}\ (c_{ij}^{\dagger}c_{i,j+1}+c_{ij}^{\dagger}c_{i-1,j+1}+\text{H.c.}).\end{aligned}\tag{3}$$

Here $n_{ij}\equiv c_{ij}^{\dagger}c_{ij}$, and $n_{ij}=1$ corresponds to the $|t_+\rangle$ state on the (ij)th dimer, while absence of particle $(n_{ij}=0)$ corresponds to the singlet. The magnetization per site in the saturation units M coincides now with the concentration of fermions. The effective interaction constants U,U', hopping amplitudes t, t', and chemical potential μ are given by

$$\begin{aligned}&U=(2J_1+J_2)/4,\quad t=(2J_1-J_2)/4\\&U'=J'/4,\quad t'=-J'/4,\quad \mu=J_3-g\mu_BH.\end{aligned}\tag{4}$$

It is useful to note the particle-hole symmetry of the effective Hamiltonian: written in terms of hole operators, Eq. (3) preserves its form (up to a constant shift), with the change only in the value of the chemical potential

$$\mu\leftrightarrow\mu_{\text{hole}}=-\mu-2U-4U'.\tag{5}$$

In what follows we assume that the effective interaction constants U,U' are *positive*, i.e., the interaction is repulsive.

A. The ''atomic limit''

Consider first the ''atomic limit'' of the model (3), i.e., put hopping t,t' to zero. Then the problem becomes equivalent to the generalized Ising model in magnetic field, a class of models that is well studied in the theory of alloys (see, e.g., Ref. 21 and references therein). Generally, in this limit the interaction energy per site E_{int} is a piecewise linear function of the concentration M, and the curve $E_{\text{int}}(M)$ has ''kinks'' (jumps of the first derivative) at certain values of M. When the magnetic field increases, the chemical potential μ becomes negative, and particles start being ''pumped'' into the system until this process is stopped by the interaction. Kinks in $E_{\text{int}}(M)$ correspond to plateaus in the magnetization curve $M(H)$, and the width of a plateau is propor-

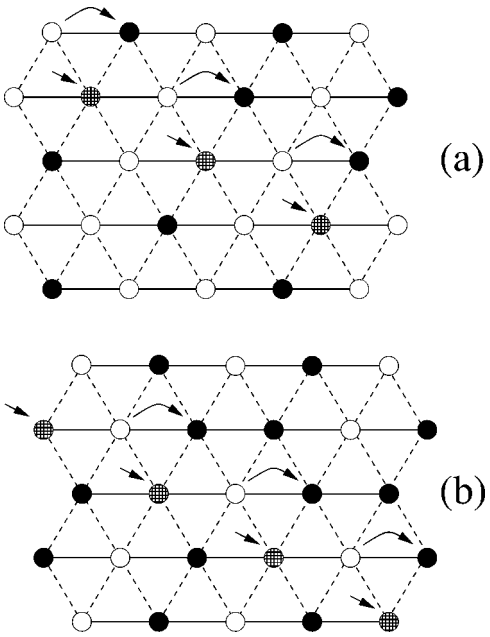

FIG. 3. Different processes of increasing the concentration in the atomic limit of the model (3) at $U'<U$: (a) the process leading from $M=\frac{1}{3}$ to the intermediate plateau at $M=\frac{1}{2}$; (b) the process increasing the magnetization above $M=\frac{1}{2}$. Half-filled and black circles represent, respectively, ''new'' particles inserted into the system and ''old'' ones that formed the $M=\frac{1}{3}$ or $M=\frac{1}{2}$ structure.

tional to the magnitude of jump in the first derivative. The atomic limit determines the possible number and positions of plateaus; hopping generally tends to smear out the plateaus, and below we will study this effect perturbatively.

In the atomic limit the model (3) is equivalent to the so-called anisotropic triangular nearest-neighbor Ising model in external field, and its ground state is known exactly.[22] Up to $M=\frac{1}{3}$ one can insert particles into the system without losing any interaction energy (see Fig. 2). Thus,

$$E_{\rm int}(M)=0, \quad M\leqslant\frac{1}{3}. \tag{6a}$$

Using the particle-hole symmetry, one immediately obtains from the above the behavior of $E_{\rm int}$ above $M=\frac{2}{3}$:

$$E_{\rm int}(M)=(2U+4U')\left(M-\frac{1}{2}\right), \quad M\geqslant\frac{2}{3}. \tag{6b}$$

In the ''intermediate'' interval of concentrations $\frac{1}{3}<M<\frac{2}{3}$ the situation depends on the ratio of interaction constants U and U'. If $U'<U$, then the lowest energy cost for inserting new particles above $M=\frac{1}{3}$ is $3U'$ per particle, which is determined by the *multiparticle* process of inserting at once a *row* of particles as shown in Fig. 3(a). This process leads from the state with $M=\frac{1}{3}$ to the state with $M=\frac{1}{2}$, and another multiparticle process shown in Fig. 3(b) leads from $M=\frac{1}{2}$ to $M=\frac{2}{3}$, taking the energy of $2U+U'$ per particle.

For $U'>U$, similar multiparticle processes exist that govern the increase of concentration above $M=\frac{1}{3}$ and $M=\frac{1}{2}$, with the energy cost of $3U$ and $4U'-U$ per particle, respectively [one can obtain the corresponding pictures by rotating the particle configurations of Figs. 3(a) and 3(b) by $\pi/3$ but keeping the solid and dotted lines fixed].

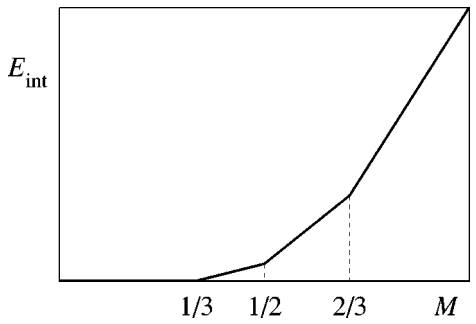

FIG. 4. A schematic look of the interaction energy per site $E_{\rm int}$ as a function of the concentration M, in the ''atomic limit'' of Eq. (3).

Taking all that into account, one obtains the following result for the interaction energy as a function of M in the $[\frac{1}{3},\frac{2}{3}]$ interval:

$$E_{\rm int}=\begin{cases} 3U'(M-\frac{1}{3}), & \frac{1}{3}<M\leqslant\frac{1}{2} \\ (2U+U')M-U, & \frac{1}{2}<M<\frac{2}{3} \end{cases} \tag{6c}$$

for $U'<U$, and

$$E_{\rm int}=\begin{cases} 3U(M-\frac{1}{3}), & \frac{1}{3}<M\leqslant\frac{1}{2} \\ (4U'-U)(M-\frac{1}{2})-\dfrac{U}{2}, & \frac{1}{2}<M<\frac{2}{3} \end{cases} \tag{6d}$$

for $U'>U$. The resulting behavior of $E_{\rm int}(M)$ is schematically shown in Fig. 4. Thus, generally three plateaus are possible at $M=\frac{1}{3}$, $\frac{1}{2}$, and $\frac{2}{3}$. In the limit of zero interchain interaction $U'=0$ only the plateau at $M=\frac{1}{2}$ survives, so that one may think of this plateau as coming from single chains, while plateaus at $\frac{1}{3}$ and $\frac{2}{3}$ appear due to 2D interchain coupling. The plateau at $M=\frac{1}{2}$ disappears when 2D interactions become isotropic (i.e., $U'=U$).

It should be remarked that the positions of those ''additional'' plateaus strongly depend on the topology of the lattice. For instance, if one considers a trellis lattice shown in Fig. 5 (this type of interchain coupling is realized, e.g., in $Sr_{n-1}Cu_{n+1}O_{2n}$ family[23]), then spinless fermions live not on a triangular, but on a square effective lattice, and the only possible plateau would be at $M=\frac{1}{2}$.

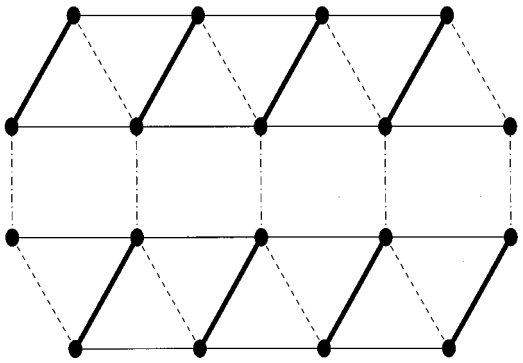

FIG. 5. Dimers coupled in a trellis lattice. This topology allows only one plateau at $M=\frac{1}{2}$.

B. The effect of hopping

Since in absence of hopping we know the solution exactly, it is natural to try to take hopping into account perturbatively. Then one can determine the critical fields marking the beginning and the end of each plateau (see Fig. 6).

To justify the use of perturbative approach with a hopping term as a perturbation, one has to demand that J_2 is close to $2J_1$, i.e., that a single zigzag chain is close to the Shastry-Sutherland line,[24] and that the interchain coupling is much smaller than J_1:

$$|2J_1 - J_2| \ll J_1, \quad J' \ll J_1. \tag{7}$$

Comparing Eq. (7) with Eq. (4), one can see that it does not make sense to consider the case $U' > U$, and in what follows we will assume that $U' \ll U$.

Let us start with the $M = \frac{1}{3}$ structure shown in Fig. 2, where one-third of the total number of sites N is occupied by particles. An increase of the magnetization from $M = \frac{1}{3}$ is determined by the multiparticle process of inserting a row of L new particles, $L \gg 1$ [see Fig. 3(a)]. Then there are no first-order corrections to $E_{N/3+L}$, and in the second order the gap $\Delta_{c3} = (E_{N/3+L} - E_{N/3})/L$ reads as

$$\Delta_{c3}^{U'<U} = \mu + 3U' + \frac{4t'^2}{2U - U'} + \frac{4t^2}{U} - \frac{t^2}{U'} - \frac{2t^2 - 12t'^2}{U + U'} + \frac{5t'^2}{U} + O(1/L). \tag{8}$$

Zero of the gap Δ_{c3} gives the critical field H_{c3}.

Similarly, for $U' < U$, increase of the magnetization from the $M = \frac{1}{2}$ state is determined by the multiparticle process shown in Fig. 3(b), which, up to the second order, yields the following expression for the gap $\Delta_{c5} = (E_{N/2+L} - E_{N/2})/L$:

$$\Delta_{c5} = \mu + 2U + U' + (4t'^2 - 3t^2)/U + 4t'^2/(U + U') + 3t^2/U' - 6t'^2/(2U - U') + O(1/L). \tag{9}$$

To obtain the critical field H_{c2}, one has to consider the set of degenerate states with one particle taken out of the structure shown in Fig. 2; the degeneracy is lifted only in the second order, giving the following dispersion $\varepsilon_{c2}(\boldsymbol{k}) = E_{N/3-1}(\boldsymbol{k}) - E_{N/3}$ of the $N/3-1$ excitation:

$$\varepsilon_{c2}(\boldsymbol{k}) = -\mu + \frac{8}{U'}[t^2 + t'^2 + tt'(\cos k_1 + \cos k_2)] + \frac{8t'^2}{U}[1 + \cos(k_1 - k_2)] - \frac{3t^2}{U'} - \frac{12t'^2}{U + U'}, \tag{10}$$

and the gap $\Delta_{c2} = \min \varepsilon_{c2}(\boldsymbol{k})$ is given by

$$\Delta_{c2} = -\mu - 3t^2/U' - 12t'^2/(U + U') + 8(|t| - |t'|)^2/U' + 16t'^2/U, \quad |t/t'| > 2U'/U, \tag{11}$$

$$\Delta_{c2} = -\mu + 5t^2/U' - 12t'^2/(U + U') - 4t^2U/U'^2 + 8t'^2/U', \quad |t/t'| < 2U'/U.$$

Finally, it is easy to calculate the value of the first critical field H_{c1} that indicates the point where the injection of triplets starts.[14] In the vicinity of H_{c1} the density of triplets is very low, so that one can just neglect the interaction terms in Eq. (3). The hopping part of the effective Hamiltonian yields the dispersion $E_1 - E_0 \equiv \varepsilon(\boldsymbol{k})$,

$$\varepsilon(\boldsymbol{k}) = \mu + 2t\cos k_1 + 2t'\{\cos k_2 + \cos(k_1 - k_2)\}.$$

The gap $\Delta_{c1} = \min \varepsilon(\boldsymbol{k})$ is given by

$$\mu_{c1} = \begin{cases} \mu - 2t - (t')^2/t & \text{at } t > 0 \\ \mu + 2t - 4|t'| & \text{at } t < 0. \end{cases} \tag{12}$$

All the other critical fields now can be obtained by exploiting the particle-hole symmetry (5), which connects the following pairs:

$$\mu_S \leftrightarrow \mu_{c1}, \quad \mu_{c5} \leftrightarrow \mu_{c4},$$

$$\mu_{c3} \leftrightarrow \mu_{c6}, \quad \mu_{c2} \leftrightarrow \mu_{c7}.$$

Zeros of the gaps given by formulas (8), (9), (11), and (12), together with the definition of μ as stated in Eq. (4), determine the critical fields. It should be remarked that in the derivation of Eq. (12) we did not use perturbation theory, so that the expression for H_{c1} is exact within the effective model (3). It can be improved, however, by including the processes involving the other two triplet states that were discarded in our consideration, in the spirit of the degenerate perturbation theory (see, e.g., Ref. 25).

III. THREE-DIMENSIONAL NH_4CuCl_3-TYPE LATTICE

Now let us proceed to a more complicated model, which could be useful for understanding the physics of magnetization process of NH_4CuCl_3. Consider a three-dimensional model that can be imagined as a set of two-dimensional systems (''layers'') from Sec. II stacked on top of each other, as shown schematically in Fig. 7. Besides exchange interactions $J_{1,2,3}, J'$ inside a single layer, we have now two additional interlayer interaction constants J'', J'''. This model corresponds to the lattice structure of $NH_4Cu\,Cl_3$,[16,17] with only nearest-neighbor exchange couplings taken into account. As before, we assume that all interactions are antiferromagnetic, and that J_3 is much stronger than the other exchange constants, so that the system can be viewed as a set of weakly coupled dimers. Along the same lines of reasoning as those used in the previous section, one obtains the effective spinless fermion model shown pictorially in Fig. 8; in addition to Eq. (4), there are now two more pairs of interaction and hopping constants,

$$U'' = \frac{1}{2}J'', \quad t'' = 0, \quad U''' = \frac{1}{4}J''' = -t'''. \tag{13}$$

There is again a particle-hole symmetry; relation (5) now has to be modified to

$$\mu \leftrightarrow \mu_{hole} = -\mu - 2U - 4U' - 4U'' - 4U'''. \tag{14}$$

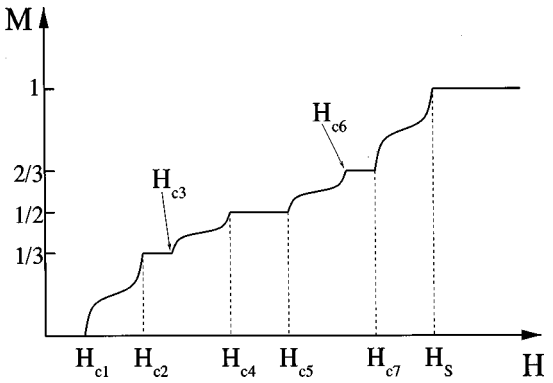

FIG. 6. Schematic plot of the magnetization M as a function of the applied field H for the effective model (3).

One can find that up to the concentration $M=\frac{1}{4}$ particles can be pumped into the system without interacting with each other. The corresponding structure with $M=\frac{1}{4}$ is shown in Fig. 8(a); it is easy to see that no new particles can be inserted without interaction. Thus, the interaction energy $E_{\rm int}$ as a function of M always has a kink at $M=\frac{1}{4}$, and a symmetrical kink should be present at $M=\frac{3}{4}$. Those are exactly the values of M at which the magnetization plateaus in NH_4CuCl_3 are observed.[15] However, it can be shown that in the atomic limit there are *always* other kinks at different values of M present in $E_{\rm int}(M)$. Simply comparing the energies of different structures and displaying them in the $E_{\rm int}(M)$ plot, one can see that there always exists certain $M=\frac{1}{2}$ structure whose energy is below $E_{\rm int}(\frac{1}{4})+E_{\rm int}(\frac{3}{4})$, i.e., the corresponding point in the $E_{\rm int}(M)$ plot lies below the line connecting the $M=\frac{1}{4}$ and $M=\frac{3}{4}$ structures; in the same way one can check that $M=\frac{1}{3}$ and $M=\frac{2}{3}$ structures can be also favored under certain conditions on the model parameters.

Since no other plateaus except $M=\frac{1}{4}$ and $M=\frac{3}{4}$ are observed in NH_4CuCl_3, this presents a problem. Let us consider a particular regime with the exchange constants satisfying the following inequalities:

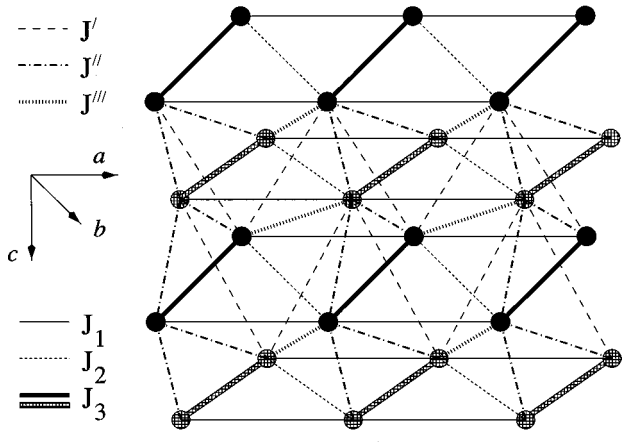

FIG. 7. Three-dimensionally coupled spin structure realized in $KCuCl_3$ family (Refs. 16 and 17). Filled circles denote $S=\frac{1}{2}$ spins of the lower layer in the (ac) plane, and half-filled circles represent spins of the upper layer.

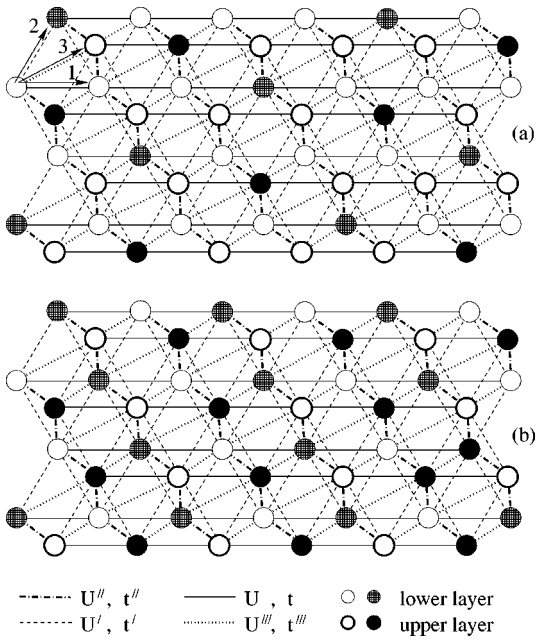

FIG. 8. Visual representation of the effective spinless fermion model resulting from the model shown in Fig. 6; the interaction constants U and hopping amplitudes t are indicated for each link and are given by formulas (4) and (13). Filled and empty circles represent occupied and empty sites; (a) the $M=\frac{1}{4}$ structure; (b) the $M=\frac{1}{2}$ structure that is realized under conditions (15).

$$J'''\leqslant J'', \quad J'\leqslant 2J''', \qquad J'\leqslant 2J_1+J_2+4(J''-J'''). \tag{15}$$

Then one can show that in the atomic limit the only additional plateau structure realized in between $M=\frac{1}{4}$ and $M=\frac{3}{4}$ is the $M=\frac{1}{2}$ structure shown in Fig. 8(b). Then, generally, there exist eight critical fields, as in the 2D problem considered in the previous section, which can be calculated perturbatively in the same way. The perturbation theory results in three dimensions are somewhat cumbersome even in the second order, so that they are listed in the Appendix.

The important point is that in the regime determined by Eq. (15) hopping reduces the width of the $M=\frac{1}{2}$ plateau already in the first order, while corrections to the width of $M=\frac{1}{4},\frac{3}{4}$ plateaus start from the second order. Thus, in the above regime there is only one additional plateau at $M=\frac{1}{2}$ that is smeared by hopping more effectively than the plateaus at $M=\frac{1}{4},\frac{3}{4}$. This could be a qualitative explanation of the real situation in NH_4CuCl_3, if one assumes that the additional plateau is completely wiped out.

IV. DISCUSSION AND SUMMARY

In this paper I show that in quasi-one-dimensional spin systems the location of plateaus in the dependence of magnetization on applied field can be considerably affected by the presence of two- or three-dimensional interchain couplings. As an example, a simple model of two-dimensionally coupled zigzag chains is considered in a dimer limit. It has been shown that, depending on the lattice structure and on the interplay between the model parameters, the system can exhibit plateaus at $\frac{1}{3}$ and $\frac{2}{3}$ of the saturation, in addition to the ''usual'' plateau at one half of the saturation determined by physics of a single chain.

We argue that this mechanism can be relevant for the recent intriguing high-field magnetization measurements[15] in NH_4CuCl_3, since the three-dimensional lattice structure of this material naturally favors the appearance of plateaus at $\frac{1}{4}$ and $\frac{3}{4}$ of the saturation, i.e., exactly at those positions where the plateaus were actually observed.

Unfortunately, the arguments presented here cannot be used for any quantitative predictions concerning the exchange constants in NH_4CuCl_3, because of the following reasons: (i) zero-field magnetic ground state of NH_4CuCl_3 implies that the first critical field $H_{c1} \leqslant 0$, and from Eq. (A8) one can clearly see that then J_3 cannot be much stronger than all the other couplings, as we assumed; (ii) to justify considering the hopping term as a perturbation, one would need to satisfy Eq. (7), and the condition $J_2 \to 2J_1$ does not seem plausible on the basis of the available crystal structure data.[17]

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APPENDIX: GAPS IN 3D MODEL

In the 3D model of Sec. III the magnetization curve looks similar to that presented in Fig. 6, except that now we have plateaus at $\frac{1}{4}$ and $\frac{3}{4}$ instead $\frac{1}{3}$ and $\frac{2}{3}$, respectively. The effect of hopping can be considered perturbatively, provided that Eq. (7) is satisfied together with the additional inequalities

$$J'' \ll J_1, \quad J''' \ll J_1. \tag{A1}$$

In the second order of perturbation theory, the dispersion $\varepsilon_{c3}(\boldsymbol{k})$ of the excitation with one extra particle inserted above the $M=\frac{1}{4}$ state, and the dispersion $\varepsilon_{c2}(\boldsymbol{k})$ of the excitation resulting from taking away one particle off the same $M=\frac{1}{4}$ state, can be represented in the following form:

$$\varepsilon_{c,i}(\boldsymbol{k}) = \varepsilon^0_{c,i} + A_i \cos(k_1+k_2) + B_i \cos 2(k_1-k_2) + 2C_i \cos(k_1-k_2)[\cos k_3 + \cos(k_1+k_2-k_3)], \tag{A2}$$

There are no first-order corrections to the energies of $N/4 \pm 1$ states. The corresponding constants for the $N/4+1$ excitation are given by the formulas

$$\varepsilon^0_{c3} = \mu + 2U' + 4U''' + \frac{t^2}{U''} + \frac{t'^2}{U'''} + \frac{2t^2}{U'+2U''} + \frac{6(t^2+t'^2)}{U-U'+2U''-4U'''} + \frac{16[t'^2+(t''')^2]}{U'+3U'''} - \frac{12t'^2}{U'+4U'''} + \frac{2t'^2}{2U-U'+2U''} + \frac{2t^2}{U+U'+2U''} - \frac{12t'^2}{U+2U''} - \frac{12t^2}{U'+2U''} + \frac{4t'^2}{U+3U''-U'''} + \frac{4t^2}{U'+3U''-U'''} + \frac{4t^2}{U'+2U''-U'''} + \frac{4(t''')^2}{U'+U'''} + \frac{4t'^2}{U+2U''-U'''} + \frac{2t'^2}{U-U'+2U''} - \frac{16(t''')^2}{2U'+3U'''}, \tag{A3}$$

$$A_3 = \frac{8tt'}{U-U'+2U''-4U'''} + \frac{4(t''')^2}{U'+U'''}, \tag{A4}$$

$$B_3 = \frac{t'^2}{U'''}, \quad C_3 = \frac{4t't'''}{U'+3U'''},$$

and for the $N/4-1$ excitation one obtains

$$\varepsilon^0_{c2} = -\mu + \frac{8(t''')^2}{U'} + \frac{2(t^2+t'^2)}{U''} + \frac{2t'^2}{U'''} + \frac{2t^2}{U'+U''} + \frac{2t'^2}{U+U''} + \frac{16[t'^2+(t''')^2]}{U'+3U'''} - \frac{6t'^2}{U+2U''} - \frac{12t'^2}{U'+4U'''} - \frac{14(t''')^2}{2U'+3U'''} - \frac{6t^2}{U'+2U''}, \tag{A5}$$

$$A_2 = \frac{4tt'}{U''} + \frac{4(t''')^2}{U'}, \quad B_2 = \frac{t'^2}{U'''}, \quad C_2 = \frac{8t't'''}{U'+3U'''}.$$

Closing of the gaps $\Delta_{c,i}=\min \varepsilon_{c,i}(\boldsymbol{k})$ $(i=2,3)$ determines the critical fields H_{c2},H_{c3} marking the beginning and the end of the $M=\frac{1}{4}$ plateau, respectively. Taking into account that $B_{2,3}$ and $C_{2,3}$ are always positive [by our assumption on antiferromagnetic character of all exchange interactions, see Eqs. (4) and (13)], one can obtain the following formulas for the gaps:

$$\Delta_c=\varepsilon_c^0+\begin{cases}A+B-4C, & B<C\\ A-B-2C^2/B, & B>C\end{cases}$$

at $A<0$, and for $A>0$ the result is a bit more complicated:

$$\Delta_c=\varepsilon_c^0+\begin{cases}-A-B, & C<(AB)^{1/2}\\ A+B-4C, & A<C,B<C\\ B-A-2C^2/A, & (AB)^{1/2}<C<A\\ A-B-2C^2/B, & (AB)^{1/2}<C<B.\end{cases}$$

(We have omitted the subscripts 2,3 above for the sake of clarity.) One can see that the expressions for Δ_{c2},Δ_{c3} ''explode'' when either of the interchain interaction constants U',U'',U''' vanishes; this corresponds to the obvious fact that the $M=\frac{1}{4}$ plateau ceases to exist if any of the interchain interactions is absent.

The energy $\varepsilon_{N/2+1}(\boldsymbol{k})=\varepsilon^{(1)}_{N/2+1}(\boldsymbol{k})+\varepsilon^{(2)}_{N/2+1}(\boldsymbol{k})$ of the excitation with one extra particle above the $M=\frac{1}{2}$ state contains both first- and second-order corrections. In the first-order approximation one obtains

$$\varepsilon^{(1)}_{N/2+1}=\mu+2(U+U'+2U'')+2t'\cos(k_1-k_2)+2t'''[\cos k_3+\cos(k_1+k_3)], \tag{A6}$$

and the second-order contribution is

$$\begin{aligned}\varepsilon^{(2)}_{N/2+1}=&\frac{t^2(1+\cos 2k_1)}{U''-U'''}+\frac{2t'^2(1+\cos 2k_2)}{U-U'+2(U''-U''')}+\frac{4t^2+4t'^2+8tt'\cos(k_1+k_2)}{U-U'+4(U''-U''')}+\frac{4t^2+4t'^2+8tt'\cos(k_1-k_2)}{U+4(U''-U''')}\\ &-\frac{22t^2}{U+4(U''-U''')}-\frac{22t'^2}{2U-U'+4(U''-U''')}+\frac{4t'^2}{2U-U'+3(U''-U''')}+\frac{4t^2}{U+3(U''-U''')}\\ &+\frac{4t'^2}{2U-U'+3U''-4U'''}+\frac{4t^2}{U+4U''-3U'''}+\frac{4t^2}{U+3U''-4U'''}+\frac{4t'^2}{2U-U'+4U''-3U'''}\\ &+\frac{2t^2}{U+U'+4(U''-U''')}+\frac{2t'^2}{2U+4(U''-U''')}.\end{aligned} \tag{A7}$$

Zero of the corresponding gap $\Delta_{c5}=\min \varepsilon_{N/2+1}(\boldsymbol{k})$, together with the formulas (4) and (13), determines the critical field H_{c5} marking the end of the $M=\frac{1}{2}$ plateau.

The energy $\varepsilon_{c1}(\boldsymbol{k})$ of the one-particle excitation above the nonmagnetic $(M=0)$ state is completely determined by the hopping part of the Hamiltonian and has the form

$$\varepsilon_{c1}(\boldsymbol{k})=\mu+2t\cos k_1+2t'[\cos k_2+\cos(k_1-k_2)]+2t'''[\cos k_3+\cos(k_1+k_2-k_3)]; \tag{A8}$$

the gap in the above dispersion closes at the critical field H_{c1} above which the magnetization starts to increase from zero.

Exploiting the particle-hole symmetry (14) allows one to connect the following pairs of critical fields:

$$H_S\leftrightarrow H_{c1},\quad H_{c5}\leftrightarrow H_{c4},\qquad H_{c3}\leftrightarrow H_{c6},\quad H_{c2}\leftrightarrow H_{c7}. \tag{A9}$$

Setting H_{c4} equal to H_{c5}, we can obtain a condition for ''closing'' the $M=\frac{1}{2}$ plateau; restricting ourselves to the first-order expressions for the sake of simplicity, we get

$$J_1+\frac{1}{2}J_2+J''\leqslant J'+3J'''. \tag{A10}$$

However, it is easy to see that Eq. (A10) is already outside the range of validity of the perturbation theory determined by Eqs. (7) and (A1), so that this condition should be understood only as a rough estimate indicating that J',J''' have to be sufficiently strong to wipe out the plateau at $M=\frac{1}{2}$.

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[1] K. Hida, J. Phys. Soc. Jpn. **63**, 2359 (1994).
[2] K. Okamoto, Solid State Commun. **98**, 245 (1996).
[3] T. Tonegawa, T. Nakao, and M. Kaburagi, J. Phys. Soc. Jpn. **65**, 3317 (1996).
[4] M. Oshikawa, M. Yamanaka, and I. Affleck, Phys. Rev. Lett. **78**, 1984 (1997).
[5] K. Totsuka, Phys. Lett. A **228**, 103 (1997).
[6] D. C. Cabra, A. Honecker, and P. Pujol, Phys. Rev. Lett. **79**, 5126 (1997); Phys. Rev. B **58**, 6241 (1998).
[7] T. Tonegawa, T. Nishida, and M. Kaburagi, in *Proceedings of the 5th International Conference on Research in High Magnetic*

Fields, Sydney, 1997 edited by R. G. Clark [Physica B **246-247** (1998)].
[8] Y. Narumi, M. Hagiwara, R. Sato, K. Kindo, H. Nakano, and M. Takahashi, in *Proceedings of the 5th International Conference on Research in High Magnetic Fields, Sydney, 1997* (Ref. 7).
[9] K. Totsuka, Phys. Rev. B **57**, 3454 (1998).
[10] H. Nakano and M. Takahashi, J. Phys. Soc. Jpn. **67**, 1126 (1998).
[11] T. Sakai and M. Takahashi, Phys. Rev. B **57**, R3201 (1998).
[12] K. Totsuka, Eur. Phys. J. B **5**, 705 (1998).
[13] D. C. Cabra and M. D. Grynberg, cond-mat/9803368 (unpublished).
[14] F. Mila, cond-mat/9805029 (unpublished).
[15] W. Shiramura, K. Takatsu, B. Kurniawan, H. Tanaka, H. Uekusa, Y. Ohashi, K. Takizawa, H. Mitamura, and T. Goto, J. Phys. Soc. Jpn. **67**, 1548 (1998).
[16] H. Tanaka, K. Takatsu, W. Shiramura, and T. Ono, J. Phys. Soc. Jpn. **65**, 1945 (1996).
[17] K. Takatsu, W. Shiramura, and H. Tanaka, J. Phys. Soc. Jpn. **66**, 1611 (1997).
[18] T. Nakamura and K. Okamoto, Phys. Rev. B **58**, 2411 (1998).
[19] Those arguments, however, have to be taken with caution since they are obtained by fitting in a purely one-dimensional model.
[20] T. Kato, K. Takatsu, H. Tanaka, W. Shiramura, M. Mori, K. Nakajima, and K. Kakurai, J. Phys. Soc. Jpn. **67**, 752 (1998).
[21] F. Ducastelle, *Order amd Phase Stability in Alloys* (North-Holland, Amsterdam, 1991), Chap. 3.
[22] K. Y. Lin and F. Y. Wu, Z. Phys. B **33**, 181 (1979).
[23] M. Takano, Z. Hiroi, M. Azuma, and Y. Takeda, Jpn. J. Appl. Phys., Part 1 **7**, 3 (1992); Z. Hiroi, M. Azuma, M. Takano, and Y. Bando, J. Solid State Chem. **95**, 230 (1991).
[24] B. S. Shastry and B. Sutherland, Phys. Rev. Lett. **47**, 964 (1981).
[25] Kunj Tandon, Siddhartha Lal, Swapan K. Pati, S. Ramasesha, and Diptiman Sen, cond-mat/9806111 (unpublished).