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Magnetic properties of J–J–J' quantum Heisenberg chains with spin $S = 1/2$, 1, 3/2 and 2 in a magnetic field

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Abstract
By means of the density matrix renormalization group (DMRG) method, the magnetic properties of the J–J–J' quantum Heisenberg chains with spin $S = 1/2$, 1, 3/2 and 2 in the ground states are investigated in the presence of a magnetic field. Two different cases are considered. (a) When J is antiferromagnetic and J' is ferromagnetic (i.e. the AF–AF–F chain), the system is a ferrimagnet. The plateaus of the magnetization are observed. It is found that the width of the plateaus decreases with increasing ferromagnetic coupling, and disappears when J'/J passes a critical value. The saturated field is observed to be independent of the ferromagnetic coupling. (b) When J is ferromagnetic and J' is antiferromagnetic (i.e. the F–F–AF chain), the system becomes an antiferromagnet. The plateaus of the magnetization are also seen. The width of the plateaus decreases with decreasing antiferromagnetic coupling, and disappears when J'/J passes a critical value. Though the ground state properties are quite different, the magnetization plateaus in both cases tend to disappear when the ferromagnetic coupling becomes more dominant. Besides, no fundamental difference between the systems with spin half-integer and integer has been found.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Low-dimensional quantum spin systems have been attracting both experimental and theoretical interest in the last decades due to an interplay of strong quantum fluctuations and topology.

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Several theoretical predictions for the low-dimensional quantum spin chains have been verified by experimental studies. For the Heisenberg antiferromagnetic (HAF) chains with spin $S = \textit{half-integer}$, the celebrated Lieb, Schultz and Mattis theorem showed that the excitation of the system is gapless [1]. For the HAF chains with spin $S = \textit{integer}$, the excitation from the singlet ground state to the triplet excited state was conjectured to be gapful, now known as the Haldane conjecture [2]. Another interesting phenomenon in HAF spin chains is the occurrence of the magnetization plateaus, that can be viewed as an essentially macroscopic quantum phenomenon, and has gained much attention recently. A decade ago, Hida considered an $S = 1/2$ HAF chain with exchange coupling of three-site translational invariance in the presence of an applied magnetic field, and uncovered a plateau in the magnetization curve at one-third of the saturation magnetization [3]. Slightly after Hida's numerical calculation on the plateau of the $S = 1/2$ F–F–AF chain, an analytical approach was done by Okamoto [4]. These results lead to a more general necessary condition for the appearance of the magnetization plateaus which was proved by Oshikawa *et al* [5]. It tells us that for the HAF spin chains with $S = \textit{integer}$ or half-integer, the magnetization curve can have plateaus at which the magnetization per site m is topologically quantized by

$$n(S - m) = \textit{integer}, \tag{1}$$

where S is the magnitude of the spin, and n is the period of the ground state determined by the explicit spatial structure of the Hamiltonian. At the plateaus, the spin gaps open, that can be in some sense regarded as a kind of generalization of the Haldane conjecture. Similar to the quantum Hall effect, the magnetization plateau is another striking example of the macroscopic quantum phenomenon, in which magnetization is quantized to fractional values of the saturated magnetization value and is a function of the magnetic field.

The magnetization plateaus are predicted and observed in many low-dimensional spin systems. Among others, a magnetization plateau at half the saturation magnetization was observed in $S = 1$ HAF bond-alternating chain compounds $[\mathrm{Ni_2(dpt)_2}(\mu - \mathrm{ox})(\mu - \mathrm{N_3})](\mathrm{PF_6})$ (abbreviated as NDOAP) and $[\mathrm{Ni}(333 - \mathrm{tet})(\mu - \mathrm{NO_2})]\mathrm{ClO_4}$ (abbreviated as NTENP), where the experimental result is in agreement with the numerical calculations [6–8]; the ferrimagnetic mixed spin chains such as the bimetallic chain $\mathrm{MM'(pbaOH)(H_2O)}n\mathrm{H_2O}$ and the organic compound $\mathrm{Mn(hfas)_{23}(3R)_2}$ show quantum magnetization plateaus [9–13]; the magnetization plateaus are also found in p-merized chains and ladders [14–17]. The effect of randomness on magnetization process also attracts much theoretical interest [18, 19]. The one-dimensional (1D) helical spin system $\mathrm{Co(hfac)_2NITPhOMe}$ shows some interesting magnetic behaviours, where one of the unusual properties is that the magnetization shows plateaus at zero and one-third of the saturation if a magnetic field is applied along the helical axis, but no plateaus if the field is applied in the plane perpendicular to that axis [20]. Another intriguing topic in 1D spin systems is concerned with the spin-orbital model, such as $\mathrm{Na_2Ti_2Sb_2O}$ and $\mathrm{NaV_2O_5}$, where the orbital degree of freedom plays an important role in the appearance of plateaus [21–24].

Despite the existence of considerable theoretical insight, a satisfactory understanding of the experimental situation is still sparse. Experimentally, polynuclear azido-bridged derivatives are a rich source of new magnetic systems. Because of the extreme versatility of the coordination modes of the azido ligand, these magnetic systems may coordinate as $\mu_{1,3}$–$\mathrm{N_3}$ (end-to-end, EE), $\mu_{1,1}$–$\mathrm{N_3}$ (end-on, EO), or even in more exotic modes as $\mu_{1,1,1}$–$\mathrm{N_3}$ or $\mu_{1,1,3}$–$\mathrm{N_3}$ [25–27]. The similar degree of stability of the EE or EO coordination modes often favours a variety of topologies or dimensionalities. The antiferromagnetic (AF) interaction and the ferromagnetic (F) coupling are generally found in the EE and EO mode, respectively. The exotic topologies with alternating patterns EE/EE/EO have been reported [28], which offers exciting research prospects.

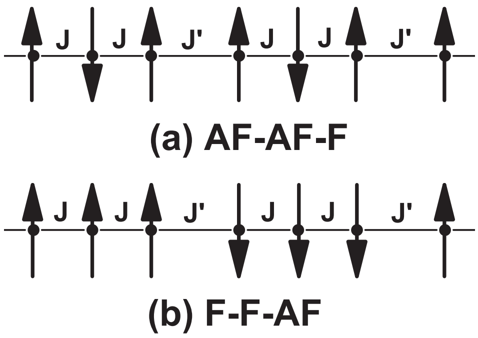

Figure 1. The spin arrangements of the J–J–J' chain, where ↑ denotes spin up, ↓ denotes spin down, J and J' are exchange couplings. (a) AF–AF–F chain, $J = J_{\mathrm{AF}}$, $J' = J_{\mathrm{F}}$; (b) F–F–AF chain, $J = J_{\mathrm{F}}$, $J' = J_{\mathrm{AF}}$.

In this paper, motivated by the exotic magnetic properties of a period $n = 3$ quantum spin system such as EE/EE/EO, we shall investigate the ground state of a 1D J–J–J' quantum spin chain with spin $S = 1/2$, 1, 3/2 and 2, respectively. It is found that in the magnetic process magnetization plateaus are observed, in agreement with equation (1). The width of the magnetization plateaus is found to depend on the ratio of J'/J. The fundamental difference of the properties between the systems with spin half-integer and integer is not observed.

The rest of this paper is outlined as follows. In section 2, we introduce the model Hamiltonian for the 1D J–J–J' Heisenberg spin chain. In section 3, we present our numerical results of the ground state of the system. A brief summary is given in section 4.

2. Model

Motivated by the experimental study of the magnetic coordinated compounds [25–27], let us investigate the 1D J–J–J' Heisenberg quantum spin system. The Hamiltonian of the system reads

$$H = \sum_j (J\mathbf{S}_{3j-2}\cdot\mathbf{S}_{3j-1} + J\mathbf{S}_{3j-1}\cdot\mathbf{S}_{3j} + J'\mathbf{S}_{3j}\cdot\mathbf{S}_{3j+1}) - h\sum_j S_j^z, \tag{2}$$

where J and J' are exchange integrals with $J, J' > 0$ denoting the AF coupling and $J, J' < 0$ the F coupling, h is the external magnetic field and we take $g\mu_{\mathrm{B}} = 1$. The schematic spin arrangement of the system is shown in figure 1.

When $J = J' = J_{\mathrm{AF}}$, the system becomes a uniform HAF spin chain. In this case, it is well known that for $S =$ *half-integer* there is no magnetization plateau in the magnetic process before saturation; for $S =$ *integer*, owing to the existence of the Haldane gap, there appears a plateau at zero magnetization below the lower critical field where the spin gap closes. When $J = J' = J_{\mathrm{F}}$, the system becomes a uniform Heisenberg ferromagnetic chain. It has no magnetization plateaus before saturation in this case.

When $J \neq J'$, the system shows complex behaviours. As indicated in figure 1, there are two interesting Cases. (a) $J = J_{\mathrm{AF}}$, $J' = J_{\mathrm{F}}$. It is a ferrimagnetic spin chain (i.e. the configuration is AF–AF–F). (b) $J = J_{\mathrm{F}}$, $J' = J_{\mathrm{AF}}$. It is an antiferromagnetic system (i.e. the configuration is F–F–AF). In order to probe the fundamental difference of properties of the systems with spin half-odd integer and integer during the magnetizing process, we shall consider $S = 1/2$, 1, 3/2 and 2 for each case below.

3. DMRG results

The density matrix renormalization group (DMRG) [30] was proposed more than ten years ago; it is nowadays a powerful numerical method invoked to study the ground state and low-lying states of low-dimensional lattice systems. In the following, the magnetic properties of the ground state of the spin chain with configurations (a) and (b) in figure 1 with open boundary conditions are investigated by the DMRG method. In our calculations, we took the chain length $L = 60$, the number of states kept per block $N = 60$ for spin $S = 1/2$ and 1, and $N = 80$ for spin $S = 3/2$ and 2. The truncation error is less than 10^{-3} in all cases.

3.1. AF–AF–F chain

Let us first consider the AF–AF–F chain, as shown in figure 1(a). In this case, $J = J_{\mathrm{AF}}$ and $J' = J_{\mathrm{F}}$. Obviously, it is a ferrimagnetic chain. When $J_{\mathrm{F}}/J_{\mathrm{AF}}$ is much less than unity, the antiferromagnetic coupling is dominant; while $J_{\mathrm{F}}/J_{\mathrm{AF}}$ is much greater than unity, the ferromagnetic coupling becomes dominant. Without loss of generality, we shall take the ratio $J_{\mathrm{F}}/J_{\mathrm{AF}}$ in the range of [0.1, 10] in this subsection.

Figure 2 shows the magnetization process of the AF–AF–F chain with spin $S = 1/2$, 1, 3/2 and 2, respectively. For $S = 1/2$, the two plateaus at $m = 1/6$ and $m = 1/2$ (saturation plateau) are obtained, consistent with the necessary condition given by equation (1) with $n = 3$. For $S = 1$, the three plateaus at $m = 1/3, 2/3$ and 1 (saturation plateau) are seen. For $S = 3/2$ the four plateaus at $m = 1/2$, 5/6, 7/6 and 3/2 (saturation plateau) are observed, and for $S = 2$ the five plateaus at $m = 2/3$, 1, 4/3, 5/3 and 2 are obtained. The number of the plateaus is $(2S + 1)$. It is clear that the smaller the ratio $J_{\mathrm{F}}/J_{\mathrm{AF}}$ is, the more obvious the plateaus are, that appears to be independent of the magnitude of spin. In other words, when the AF interaction in the system plays a dominant role, the magnetization plateau becomes wider; when the ferromagnetic interaction is dominant, the width of the plateau gets narrower, as indicated in figure 2. In between the plateaus, the magnetization per site increases with increasing field for all cases with different spins. In this system, the saturated field does not change with the ratio $J_{\mathrm{F}}/J_{\mathrm{AF}}$. A further discussion on this point can be found in section 4. For the present system with different spins $S = 1/2$, 1, 3/2 and 2, qualitatively similar behaviours of the magnetization process are observed.

The appearance of the magnetization plateaus can be understood from the spatial dependence of the averaged local magnetic moment, as discussed in [29]. Figure 3 presents the spatial variation of $\langle S_j^z\rangle$ in the ground state for the AF–AF–F chain with spin $S = 3/2$ and 2, respectively. For both cases, the ratio $J_{\mathrm{F}}/J_{\mathrm{AF}} = 0.1$ is taken. Figure 3(a) shows the case with $S = 3/2$. At the external field $h/J_{\mathrm{AF}} = 2$, the expectation values $\langle S_j^z\rangle$ versus site j follow the sequence such that $\{\ldots, (1.2268, 1.2268, -0.9535), \ldots\}$, resulting in the magnetization per site $m = \sum_{j=1}^{N}(\langle S_{3j-2}^z\rangle + \langle S_{3j-1}^z\rangle + \langle S_{3j}^z\rangle)/3N = 1/2$, corresponding to the plateau $m = 1/2$. When the field increases to $h/J_{\mathrm{AF}} = 3$, the behaviour of $\langle S_j^z\rangle$ becomes $\{\ldots, (1.2280, 1.2280, 0.0440), \ldots\}$, giving rise to $m = 5/6$; at $h/J_{\mathrm{AF}} = 4$, $\langle S_j^z\rangle$ versus j behaves as $\{\ldots, (1.3390, 1.3390, 0.8219), \ldots\}$, leading to $m = 7/6$. Figure 3(b) shows the case with $S = 2$. Similarly, at $h/J_{\mathrm{AF}} = 2.5$, the expectation value $\langle S_j^z\rangle$ versus site j behaves as $\{\ldots, (1.7178, 1.7241, -1.4448), \ldots\}$, resulting in $m = 2/3$; at $h/J_{\mathrm{AF}} = 3.5$, $\langle S_j^z\rangle$ varies according to $\{\ldots, (1.6478, 1.6478, -0.2956), \ldots\}$, giving $m = 1$; at $h/J_{\mathrm{AF}} = 4.5$, $\langle S_j^z\rangle$ behaves as $\{\ldots, (1.7137, 1.7127, 0.5746), \ldots\}$, leading to $m = 4/3$; and at $h/J_{\mathrm{AF}} = 5.5$, it becomes $\{\ldots, (1.8390, 1.8390, 1.3220), \ldots\}$, giving rise to $m = 5/3$. These m values just correspond to the magnetization plateaus. In addition, the spin configurations for the parameters above are stable against the small local field $h_{\mathrm{loc}}/J_{\mathrm{AF}} = 0.1$, which is set at $j = 1$ site and parallel to the external magnetic field h.

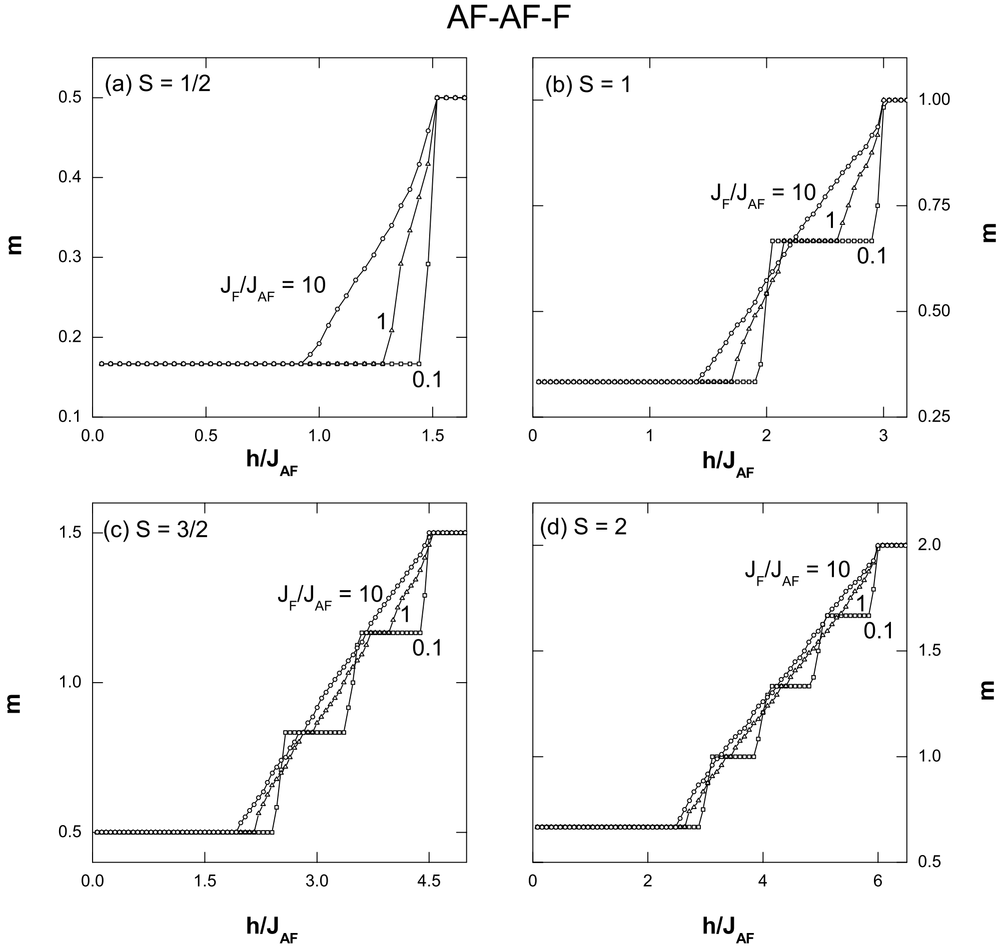

Figure 2. Magnetic curves of the AF–AF–F chain of $S = 1/2$, 1, 3/2 and 2, respectively, where h is the applied magnetic field and m is the magnetization per site. $J = J_{\mathrm{AF}} = 1$, $J' = J_{\mathrm{F}}$.

Figure 4 illustrates the phase diagram in the $h_c - J_F/J_{AF}$ plane for the AF–AF–F chain with spin $S = 1/2$, 1, 3/2 and 2, respectively. In the shaded regions, the magnetization plateaus appear, implying that the excitations from the ground state are gapful. When J_F/J_{AF} increases beyond the critical value $(J_F/J_{AF})_c$ at which the plateau vanishes, i.e. the ferromagnetic coupling becomes more dominant, the magnetization plateaus tend to disappear. The plateau–non-plateau transition could be of the Berezinskii–Kosterlitz–Thouless (BKT)-type, and the width of the plateau $m = \Delta h_c$ near the transition point behaves as

$$\Delta h_c = D \exp\left(-C\Big/\sqrt{(J_F/J_{AF})_c - J_F/J_{AF}}\right), \tag{3}$$

where D and C are constants [31].

As the width of the plateau might be extremely small near the transition point, a direct estimation of the critical point from the numerical data may be somewhat inaccurate. However, equation (3) can be used to estimate the plateau–non-plateau transition point $(J_F/J_{AF})_c$ from the raw numerical data, which yields $(J_F/J_{AF})_c = 4.5$ for $S = 1$ and $m = 2/3$, 2.2 for $S = 3/2$ and $m = 7/6$ and 1.2 for $S = 2$ and $m = 5/3$, respectively, as shown in figure 5. The larger the spin S is, the smaller the critical value of $(J_F/J_{AF})_c$ is. It is also noticeable that for a given

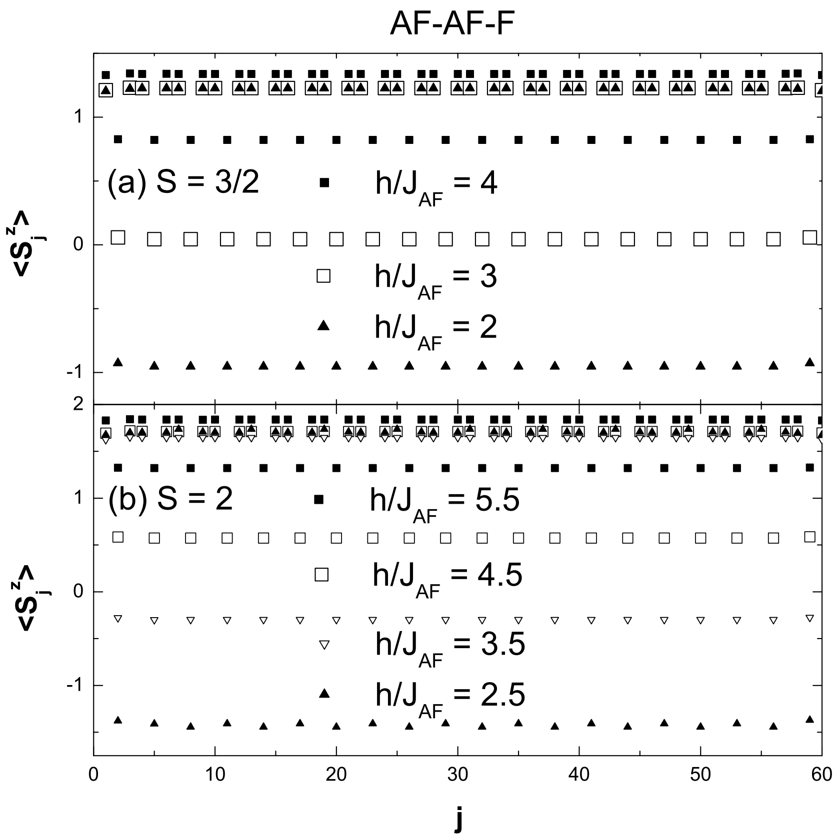

Figure 3. The spatial variation of $\langle S_j^z \rangle$ in the ground state for the finite AF–AF–F chain with length $L = 60$ at $J_F/J_{AF} = 0.1$. (a) For $S = 3/2$, the external field is taken as $h/J_{AF} = 2$, 3 and 4, respectively. (b) For $S = 2$, the external field is taken as $h/J_{AF} = 2.5$, 3.5, 4.5 and 5.5, respectively.

S, the critical values for different plateaus are slightly different. No fundamental difference between the systems with spin integer and half-integer is observed.

3.2. F–F–AF chain

Now we consider the case with $J = J_F$, $J' = J_{AF}$, namely, the system is an F–F–AF chain, as shown in figure 1(b). It is an antiferromagnet. In contrast to the AF–AF–F chain, when J_{AF}/J_F is much less than unity, the ferromagnetic coupling is dominant; while J_{AF}/J_F is greater than unity, the antiferromagnetic interaction predominates. We shall take the ratio J_{AF}/J_F in the range of [0.1, 8] in this subsection.

Figure 6 shows the magnetization process of the F–F–AF chain with different spins. For $S = 1/2$, we have observed the two plateaus at $m = 1/6$ and $1/2$, consistent with the condition of equation (1) with $n = 3$. For $S = 1$, the three plateaus at $m = 1/3$, $2/3$ and 1 (saturation plateau) are observed. For $S = 3/2$, the four plateaus at $m = 1/2$, $5/6$, $7/6$ and $3/2$ (saturation plateau) are seen, and for $S = 2$, the five plateaus at $m = 2/3$, 1, $4/3$, $5/3$ and 2 are obtained. It is seen that the smaller the ratio J_{AF}/J_F is, the less obvious the plateaus (except for the saturation plateaus) are. In other words, when the AF coupling is predominant in this system,

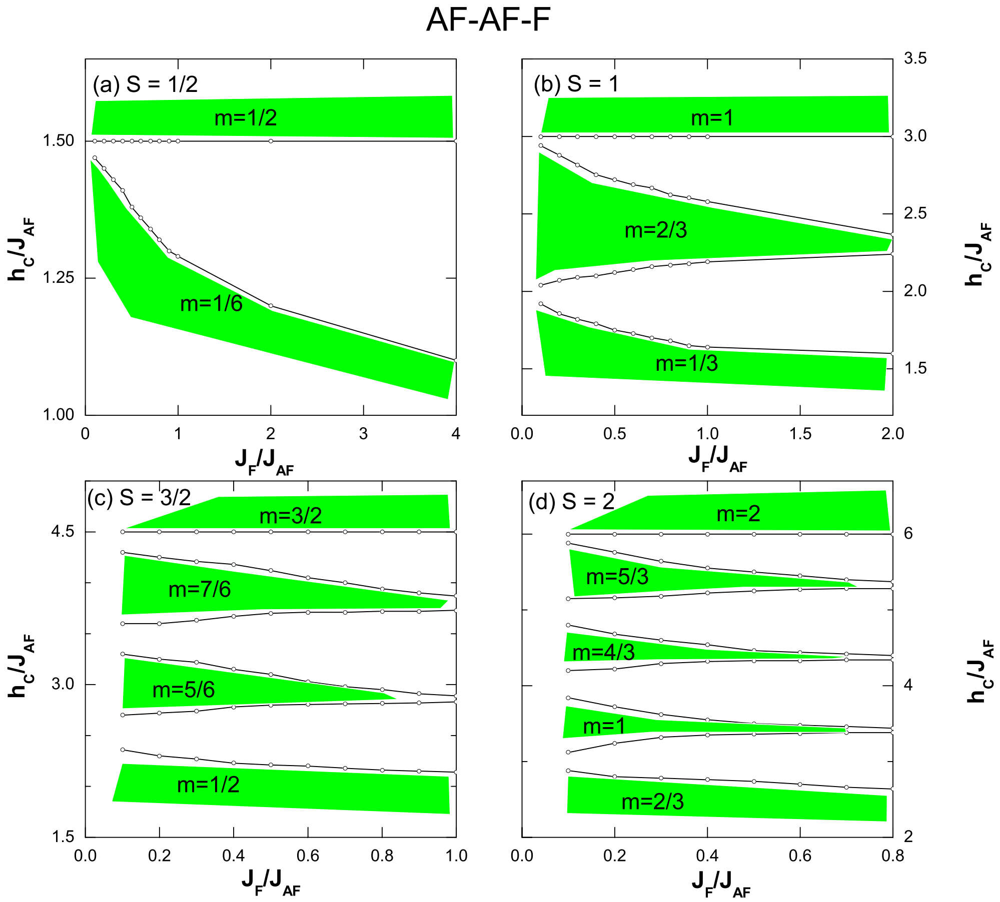

Figure 4. Phase diagram in the h_c–J_F/J_{AF} plane for the AF–AF–F chain with $S = 1/2$, 1, 3/2 and 2, respectively. In the shaded regions, the magnetization plateaus labelled with the quantized values of m appear.

the plateau is more obvious, and the width of the plateau is much greater. In between the plateaus, the magnetization increases rapidly with the magnetic field. From figure 6, one may find that the plateaus at $m = 0$ for $S = 1$, $m = 1/6$ for $S = 3/2$ and $m = 0$ and 1/3 for $S = 2$, which are also allowed by equation (1) with $n = 3$, are not observed. This is because equation (1) is a necessary condition for the occurrence of the magnetization plateaus. Therefore, the number of plateaus is still $(2S+1)$. As the present system is an antiferromagnet, the magnetic curves look different to those of the AF–AF–F chain. The saturation fields depend closely on the magnitude of the ratio J_{AF}/J_F, in contrast to the AF–AF–F chain. In addition, the similar behaviours for the system with different spins are obtained, and no fundamental difference of the properties of the system with spin integer and half-integer is observed.

Figure 7 shows the behaviour of $\langle S_j^z \rangle$ against j for the F–F–AF chain with spin $S = 3/2$ and 2, respectively. For both cases, the ratio $J_{AF}/J_F = 8$ is set. Figure 7(a) shows the case with $S = 3/2$. For the external field $h/J_F = 5$, the spatial variation of $\langle S_j^z \rangle$ is $\{\ldots, (0.0366, 0.0365, 1.4269), \ldots\}$, giving rise to $m = 1/2$. As the field is $h/J_F = 10$, the behaviour of $\langle S_j^z \rangle$ against j becomes $\{\ldots, (0.5071, 0.5071, 1.4857), \ldots\}$, resulting in $m = 5/6$. For $h/J_F = 20$, the spin configuration is $\{\ldots, (1.0018, 1.0018, 1.4964), \ldots\}$,

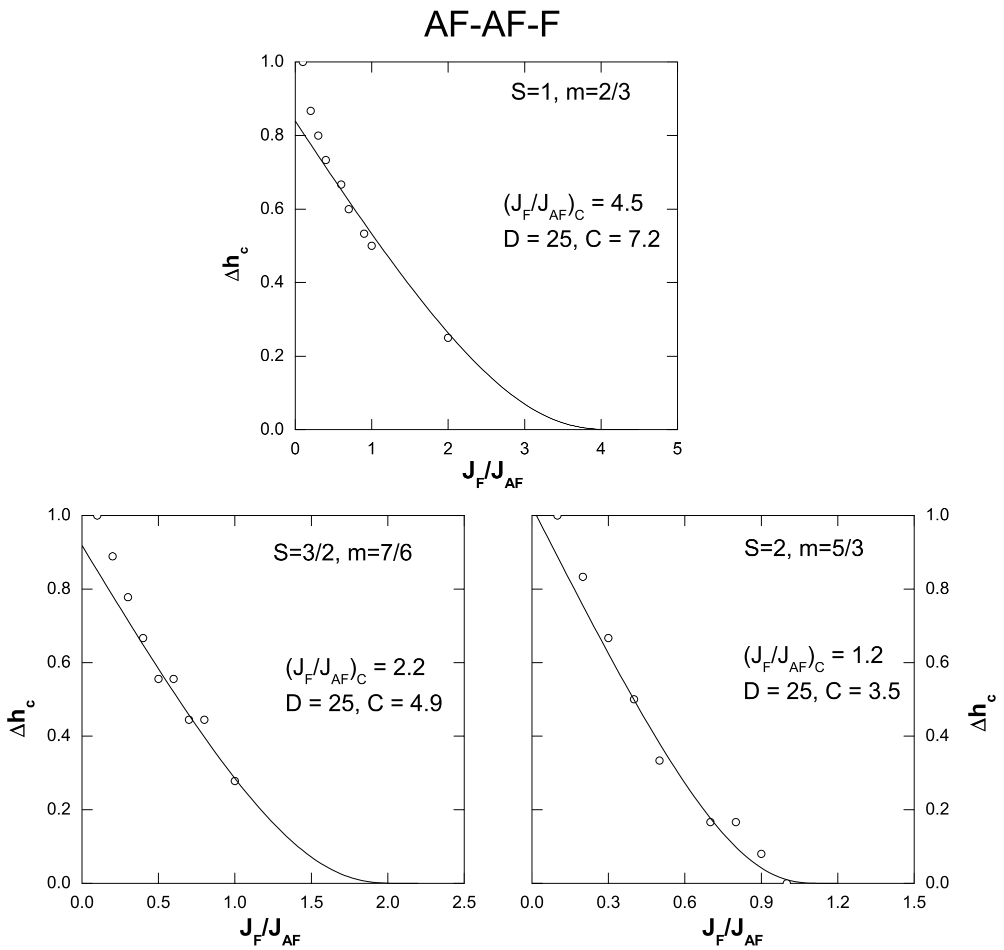

Figure 5. The plateau–non-plateau transition point $(J_F/J_{AF})_c$ for the AF–AF–F chain with $S = 1$, 3/2 and 2, respectively, where D and C are constants in equation (3), and $\Delta h_c(J_F/J_{AF})$ are scaled by the value $\Delta h_c(0.1)$.

giving $m = 7/6$. Figure 7(b) shows the case with $S = 2$. For $h/J_F = 5$, the expectation values $\langle S_j^z\rangle$ are $\{\ldots, (0.0871, 0.0863, 1.8265), \ldots\}$, leading to $m = 2/3$. When $h/J_F = 10$, the behaviour of $\langle S_j^z\rangle$ versus j becomes $\{\ldots, (0.5181, 0.5183, 1.9636), \ldots\}$, giving $m = 1$; at $h/J_F = 20$, it becomes $\{\ldots, (1.0060, 1.0060, 1.9882), \ldots\}$, yielding $m = 4/3$; at $h/J_F = 25$, the spin configuration is $\{\ldots, (1.5018, 1.5018, 1.9964), \ldots\}$, giving $m = 5/3$. The m values just correspond to the magnetization plateaus. In addition, the spin configurations for the parameters above are stable against the small local field $h_{loc}/J_F = 0.1$ at the $j = 1$ site, which is parallel to the external magnetic field h.

Figure 8 presents the phase diagram in the $h_c - J_{AF}/J_F$ plane for the F–F–AF chain with spin $S = 1/2$, 1, 3/2 and 2, respectively. In the shaded regions, the magnetization plateaus occur. At the plateaus, the spin excitations from the ground state are gapful. When J_{AF}/J_F increases beyond the critical value $(J_{AF}/J_F)_c$ at which the plateau occurs, i.e. the AF coupling becomes more dominant, the magnetization plateaus tend to appear. The plateau–non-plateau transition is also expected to be of the BKT type, and the width of the plateau $m = \Delta h_c$ near

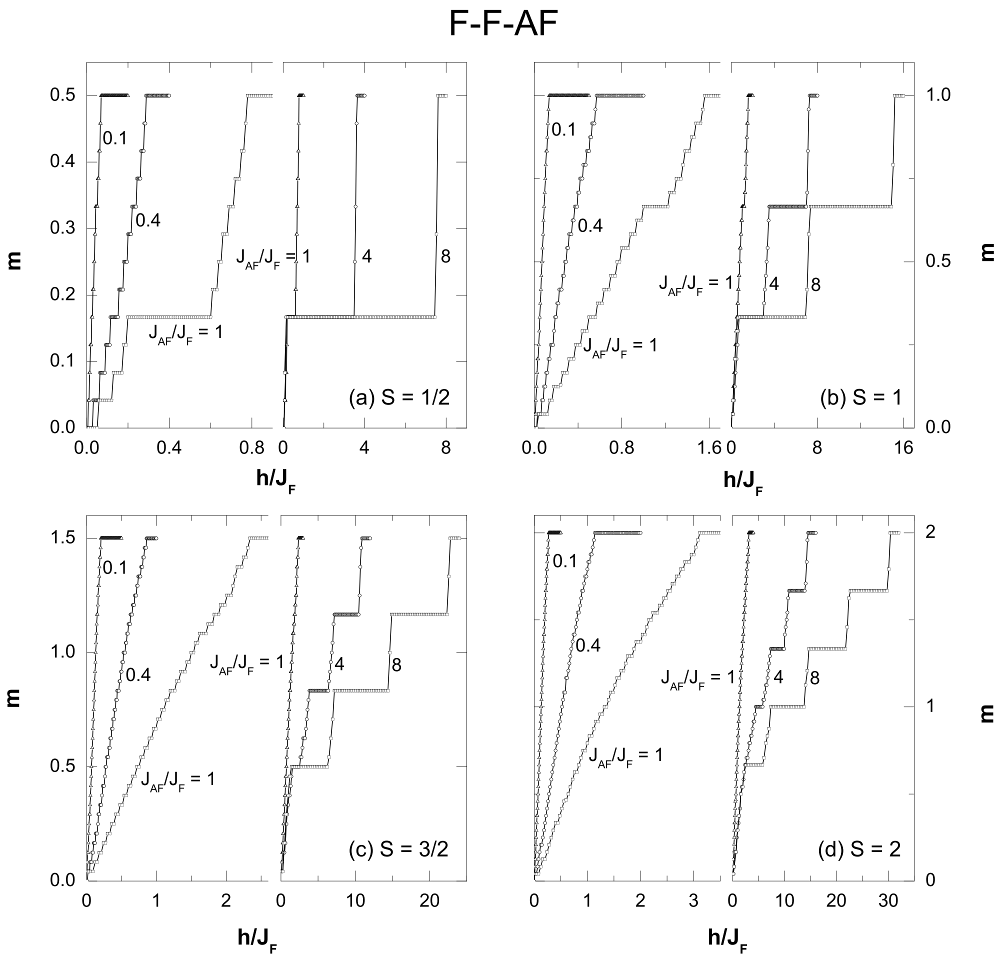

Figure 6. Magnetization process of the F–F–AF chain with $S = 1/2$, 1, 3/2 and 2, respectively, where h is the applied magnetic field, and m is the magnetization per site. $J = J_F = 1$, $J' = J_{AF}$.

the transition point behaves as

$$\Delta h_c = D \exp\left(-C \Big/ \sqrt{J_{AF}/J_F - (J_{AF}/J_F)_c}\right), \tag{4}$$

where D and C are two constants [31].

As the width of the plateau might be extremely small near the transition point, equation (4) is used to estimate the plateau–non-plateau transition point $(J_{AF}/J_F)_c$ from the raw numerical data. It gives $(J_{AF}/J_F)_c = 0.09$ for $S = 1/2$ and $m = 1/6$, 0.13 for $S = 1$ and $m = 2/3$, 0.18 for $S = 3/2$ and $m = 7/6$ and 0.24 for $S = 2$ and $m = 5/3$, respectively, as shown in figure 9. It should be pointed out that the breakdown of the magnetization plateau of the $S = 1/2$ F–F–AF chain has been studied, with the estimation of the critical value $(J_{AF}/J_F)_c = 0.065$ [31]. It can be seen that the larger the spin S is, the greater the value of $(J_{AF}/J_F)_c$ is. For a given S, the value of $(J_{AF}/J_F)_c$ for different plateaus is slightly different. In addition, no fundamental difference between the F–F–AF systems with spin integer and half-integer is also observed.

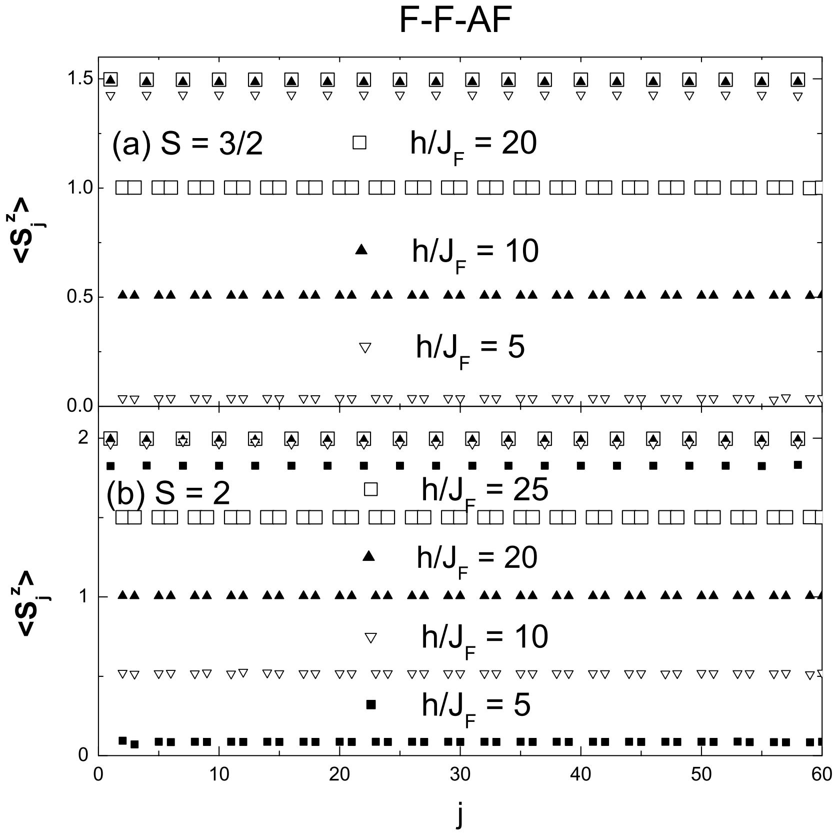

Figure 7. The spatial variation of $\langle S_j^z \rangle$ in the ground state for the finite F–F–AF chain with length $L = 60$ at $J_{\mathrm{AF}}/J_{\mathrm{F}} = 8$. (a) For $S = 3/2$, the external field is $h/J_{\mathrm{F}} = 5$, 10, 20, respectively. (b) For $S = 2$, the external field is $h/J_{\mathrm{F}} = 5$, 10, 20 and 25, respectively.

4. Summary and discussion

In this paper, by using the DMRG method we have numerically studied the magnetic properties in the ground states of the J–J–J' trimerized quantum Heisenberg chains with spin $S = 1/2$, 1, 3/2 and 2, respectively. The two different cases are considered: (a) $J = J_{\mathrm{AF}}$ and $J' = J_{\mathrm{F}}$, i.e., the AF–AF–F trimerized ferrimagnetic chain. The magnetization plateaus are observed when the AF coupling is dominated. The positions of these plateaus are allowed by equation (1) with $n = 3$, and the number of the plateaus is to be $(2S+1)$. For a certain spin S and a plateau ($m \neq S$), the width of magnetization plateaus decreases with increasing the F coupling, and becomes zero when the ratio $J_{\mathrm{F}}/J_{\mathrm{AF}}$ passes a critical value, e.g. $(J_{\mathrm{F}}/J_{\mathrm{AF}})_{\mathrm{c}} = 4.5$ for $S = 1$ and $m = 2/3$, 2.2 for $S = 3/2$ and $m = 7/6$ and 1.2 for $S = 2$ and $m = 5/3$, respectively. The larger the spin S is, the smaller the critical value of $(J_{\mathrm{F}}/J_{\mathrm{AF}})_{\mathrm{c}}$ is. For a given S, the value of $(J_{\mathrm{F}}/J_{\mathrm{AF}})_{\mathrm{c}}$ for different plateaus is slightly different. Furthermore, the saturation field does not change with the F coupling. (b) $J = J_{\mathrm{F}}$ and $J' = J_{\mathrm{AF}}$, i.e., the F–F–AF trimerized antiferromagnetic chain. The critical value in the plateau–non-plateau transition is about $(J_{\mathrm{AF}}/J_{\mathrm{F}})_{\mathrm{c}} = 0.09$ for $S = 1/2$ and $m = 1/6$, 0.13 for $S = 1$ and $m = 2/3$, 0.18 for $S = 3/2$ and $m = 7/6$ and 0.24 for $S = 2$ and $m = 5/3$, respectively. For a given S, the value of $(J_{\mathrm{AF}}/J_{\mathrm{F}})_{\mathrm{c}}$ for different plateaus is slightly different. We would like to point out

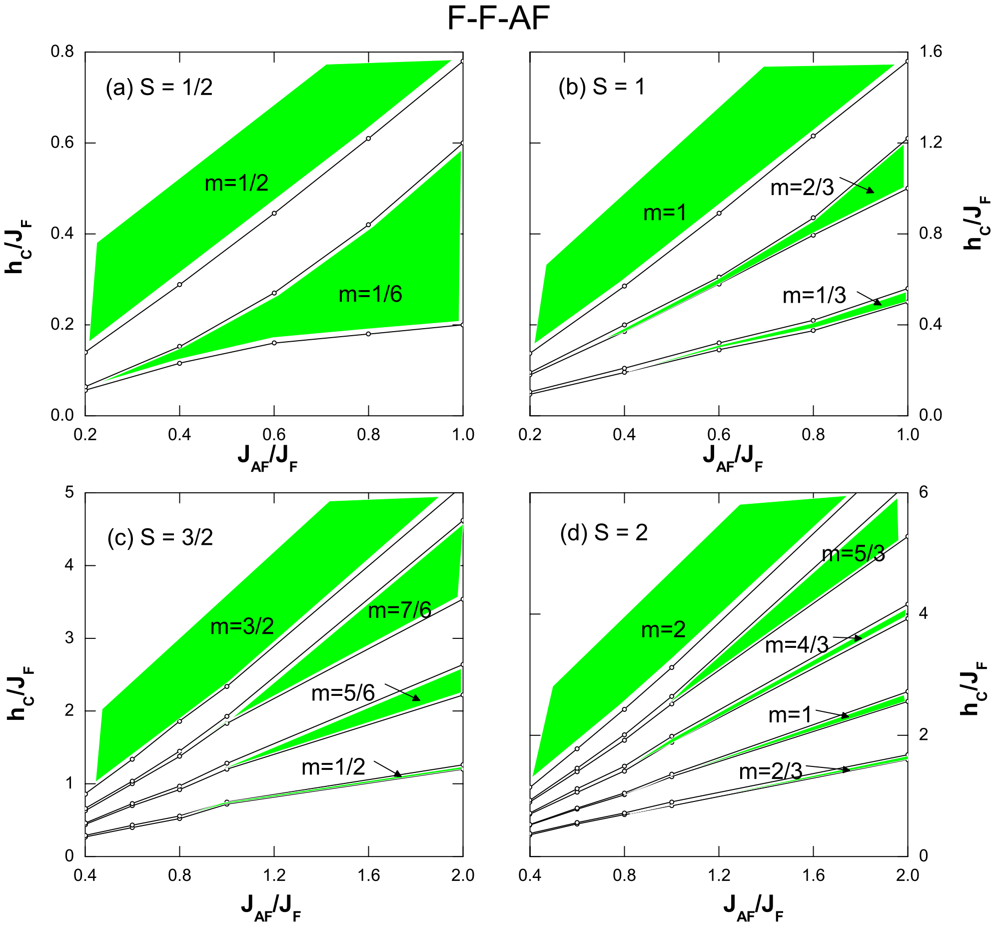

Figure 8. Phase diagram in the h_c–J_{AF}/J_F plane for the F–F–AF chain with $S = 1/2$, 1, 3/2 and 2, respectively. In the shaded regions, the magnetization plateaus labelled with the quantized values of m appear.

that though the ground state properties of the F–F–AF chain are quite different from those of the AF–AF–F chain, as the former is a antiferromagnet, while the latter is a ferrimagnet, the magnetization plateaus in both cases tend to disappear when the ferromagnetic coupling becomes more dominant.

As the width of the plateau might be extremely small near the transition point, a direct estimation of the critical point from the numerical data may be somewhat inaccurate, as mentioned above. However, near the transition point the width of the plateau is expected to behave as $D\exp(-C/\sqrt{|J'/J - (J'/J)_c|})$, where D and C are two constants. According to this property, $(J'/J)_c$ can be estimated from the raw numerical data. Such an estimation is consistent with the previous study, showing that our present calculations are reliable.

The appearance of the magnetization plateaus can be understood from the spatial dependence of the averaged local magnetic moment. For the AF–AF–F chain with spin $S = 3/2$, as the ratio $J_F/J_{AF} = 0.1$ and the external field $h/J_{AF} = 2$ are taken, the spatial variation of $\langle S_j^z\rangle$ versus site j in the ground state follow the sequence such that $\{\ldots, (1.2268, 1.2268, -0.9535), \ldots\}$, resulting in the magnetization per site $m =$

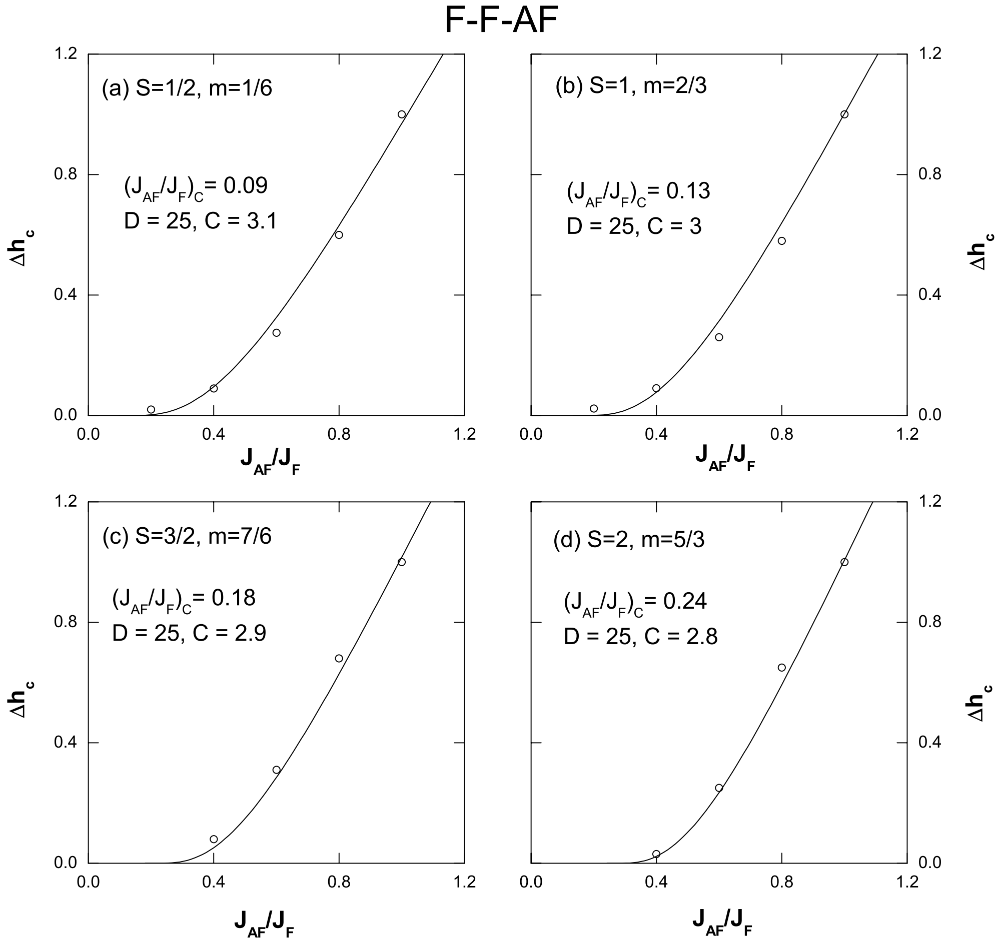

Figure 9. The plateau–non-plateau transition point $(J_{\mathrm{AF}}/J_{\mathrm{F}})_{\mathrm{c}}$ for the F–F–AF chain with $S = 1/2$, 1, $3/2$ and 2, respectively, where D and C are constants in equation (4), and $\Delta h_{\mathrm{c}}(J_{\mathrm{AF}}/J_{\mathrm{F}})$ are scaled by the value $\Delta h_{\mathrm{c}}(1.0)$.

$\sum_{j=1}^{N}(\langle S_{3j-2}^{z}\rangle + \langle S_{3j-1}^{z}\rangle + \langle S_{3j}^{z}\rangle)/3N = 1/2$, corresponding to the plateau $m = 1/2$. Similar cases are found when the external fields $h/J_{\mathrm{AF}} = 3$ and 4 are set. For the AF–AF–F chain with spin $S = 2$, F–F–AF chain with spin $S = 3/2$ and 2, similar spatial dependences of the averaged local magnetic moment are observed. It should be noted that the spin configuration for the parameters above are stable against the small local field $h_{\mathrm{loc}}/J_{\mathrm{AF}} = 0.1$ for AF–AF–F chain and $h_{\mathrm{loc}}/J_{\mathrm{F}} = 0.1$ for F–F–AF chain, which is set at $j = 1$ site and parallel to the external magnetic field h.

By the spin-wave method starting from the fully polarized state, the saturation magnetic field of the dimerized and quadrumerized AF chains can be obtained analytically [16]. Inspired by this method, we have deduced an expression for the trimerized chains. The energy difference ΔE between the lowest energy with $M^z = (3N-1)S$ and that with $M^z = 3NS$ is given by the lowest eigenvalue of the following matrix:

$$\begin{pmatrix} -(J'+1) & J'\mathrm{e}^{\mathrm{i}k} & \mathrm{e}^{-\mathrm{i}k} \\ J'\mathrm{e}^{-\mathrm{i}k} & -(J'+1) & \mathrm{e}^{\mathrm{i}k} \\ \mathrm{e}^{\mathrm{i}k} & \mathrm{e}^{-\mathrm{i}k} & -2 \end{pmatrix}$$

where k is the wavenumber, $J = 1$, J' and S are the same notation as mentioned above. We have found that the energy difference ΔE is the lowest when $k = \pi$. This gives rise to the saturation field $H_s = -\Delta E = (3 + 2J' + \sqrt{9 - 4J' + 4(J')^2})S/2$. When $J' = 1$, it becomes a simple AF chain, $H_s = 4S$. When $J' < 0$, it becomes an AF–AF–F chain, and H_s is nearly independent of J'. These results are also in agreement with the result of the quadrumerized chain [16]. Most importantly, all these analytic results are in agreement with our numerical results.

In trimerized Heisenberg chains, the total number of the plateaus is $2S + 1$, which implies that the OYA condition is only a necessary condition in trimerized chains. This observation can be understood straightforward in the case of ferrimagnet, i.e. the AF–AF–F chain. As shown in figure 1(a), the possible minimum magnetization value is $m = S/3$, so the plateaus whose magnetization value is smaller than $S/3$ cannot naturally appear. For example, for $S = 3/2$, the plateaus permitted by the OYA condition should be $m = 1/6$, $1/2$, $5/6$, $6/7$ and $3/2$, respectively. As the plateau $m = 1/6$ is smaller than the minimum magnetization of $S/3 = 1/2$, it does not appear. Then, the number of the emerging plateaus is $4 = 2S + 1$. However, this argumentation seems not to apply to the case of the antiferromagnet, i.e. the F–F–AF chain. In figure 1(b), the possible minimum magnetization value is $m = 0$. The plateaus $m = 1/6$ of $S = 3/2$ would be expected to emerge during the magnetization, but they do not appear. The reason why the plateau $m = 1/6$ of $S = 3/2$ vanishes is under exploration. It is worthy of stressing that for a certain spin S the number of disappearances of plateaus is smaller than that of the emergence of plateaus.

The obtained results of the two cases reveal that the degree of the inhomogeneity of the couplings measured by the ratio J'/J, not just the period of the quantum spin chain indicated in equation (1), determines its ground-state properties. In addition, no fundamental difference of the properties of the system with spin integer and half-integer is observed. It should be noted that the magnetization plateaus are only related to the AF coupling. In cases (a) and (b), when the F coupling is much stronger than the AF one, there are no magnetization plateaus at all, though the ratio of the couplings can still be large. Clearly, these magnetization plateaus come from the quantum origin, and are closely related to the competition between the exchange couplings. The thermodynamic properties of this system are being studied at present.

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