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Anisotropic S=1 Magnetic Chain in an External Field: Low Lying Energy Levels and ESR Transition Rates

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We investigate the spectrum of elementary excitations for $S = 1$ Heisenberg chains with single ion anisotropy in an external magnetic field and their signatures in ESR transition rates. We find that the order of levels as found previously for wavevector $q = \pi$ (Haldane gap) changes drastically at $q \neq \pi$, leading to an accidental degeneracy at $q \approx 0.75\pi$. We calculate the dependence on magnetic field of ESR transition rates for transitions inside the group of lowest excited states (one magnon states) as well as for ground state transitions to some low-energy two magnon states. Characteristic variations of intensity allow to identify the participating levels. Numerical calculations for $S = 1$ chains up to 14 spins confirm the approximations used in obtaining the analytical results.

§1. Introduction

In the last decade a lot of interest has been devoted to one-dimensional (1D) spin systems with isotropic exchange interactions and a correlation induced gap in the excitation spectrum. The prototype of these systems is the $S = 1$ antiferromagnetic Heisenberg- (Haldane) chain.[1] The basic structure of the ground state of this system was identified as valence bond state (VBS state) with the help of a simple model including also biquadratic exchange interactions, for which the ground state is exactly known.[2] The excitation spectrum consists of magnons with an energy gap Δ at wavevector $q = \pi$. A more realistic hamiltonian for systems of experimental interest which includes single-ion anisotropies and an external magnetic field is (in the following we take the value of the antiferromagnetic exchange coupling as energy unit and measure magnetic fields in units of $g\mu_B$)

$$H = \sum_{n=1}^{L} \Big(\vec{S}_n \cdot \vec{S}_{n+1} + D(S_n^z)^2 + E\left((S_n^x)^2 - (S_n^y)^2\right) - \vec{B}\cdot\vec{S}_n \Big). \quad (1)$$

This $S = 1$ chain has been the subject of a number of investigations in recent years: For $D = E = 0$ the gap closes for a critical magnetic field $B_c = \Delta_0$, where Δ_0 is the zero field gap, and the system undergoes a transition to a new phase which has the characteristics of a Luttinger liquid: gapless spectrum, power law correlations. Without external field and for $E = 0$ the system is in the Haldane phase for $D < D_c$ and in the large$-D$ phase for $D > D_c$, where $D_c \approx 1$.[4] The combined effect of anisotropies and external magnetic fields on the energy gap has been investigated by Golinelli et al.[5] Systems described by the hamiltonian of eq.(1) such as NENP and NINO were investigated already some years ago[6–9] and understood after invoking the existence of an internal staggered magnetic field. Recently, new anisotropic $S = 1$ systems, which do not show this internal staggered filed were investigated by ESR experiments: Honda et al[10] have performed measurements on the material NDMAP, an $S = 1$ Heisenberg chain in the Haldane phase (subcritical single ion anisotropies), which by a sufficiently strong magnetic field may be driven into the high field phase, and Orendac et al have investigated the material NENC,[11] an $S = 1$ Heisenberg chain in the large $D-$phase. We have reconsidered the dynamics of $S = 1$ chains governed by the hamiltonian of eq.(1) (with $D > 0, E > 0$) with the aim to contribute to the interpretation of the ESR spectra in these materials. In the following we report the first part of this investigation which refers to the dynamics in the Haldane phase. In §2 we extend the work of ref. 5 to cover the complete spectrum of one- and two-magnon excitations and in §3 we calculate and discuss ESR transition rates.

We conclude the introduction with a discussion of the symmetries of the 1D system defined by the hamiltonian of eq.(1). We always assume translational symmetry, thus the wavevector q is always one good quantum number. The complete rotational symmetry in spin space present for the ideal Haldane chain (with the total spin S_{tot} and its projection S_{tot}^z as good quantum numbers, implying threefold degenerate triplet ($S = 1$) excitations for each wavevector) is reduced to rotational symmetry with respect to the $z-$axis for $D \neq 0$ and/or $B_z \neq 0$. Then states are classified by the total spin projection $S_{tot}^z = \sum_n S_n^z$ and the basic excited triplet is split into one doublet (which is further Zeeman splitted for finite B_z) and one singlet (at $q = \pi$ for $D > 0$ in the Haldane phase the doublet is lowest and in the large$-D$ phase the singlet is lowest). For finite external field and/or $E \neq 0$ the symmetry is reduced further and only reflection symmetry is left. Reflection in spin space with respect to axis α is defined by the operator

$$R_\pi^\alpha = \prod_n e^{i\pi S_n^\alpha}. \quad (2)$$

Without external field, the hamiltonian of eq.(1) commutes with R_α for arbitrary α, and for finite external field in direction α, the hamiltonian commutes with R_α for arbitrary D, E. The corresponding quantum numbers are $r_\alpha = \pm 1$, which we will refer to as spin parity quantum number.

§2. Excitations in the Haldane Phase

Whereas an approximate quantitative description for ground state and elementary excitations of Haldane-like chains is difficult to obtain, Golinelli et al[5] have shown that the effect of anisotropies and external magnetic field on the low energy properties can be investigated rather successfully to lowest order when the Haldane state in the isotropic limit is appropriately parametrized. We shortly review their results in the following: The ground state energy is obtained to lowest order in D as $E_g^{(1)} = E_0 + \frac{2}{3}DL$ and is independent of B, the first $B-$dependent correction appears in order DB^2. All spin parity quantum numbers r_α are $+1$. The change in excitation energies resulting from the anisotropies and magnetic field is conveniently calculated using a basis of $S = 1$ states with well defined spin parities. For the lowest excited states, which develop from the degenerate Haldane triplet under the influence of anisotropy and magnetic field, these eigenstates of R_α are expressed in the standard S_{tot}^z basis with states $|S_{tot} = 1, S_{tot}^z\rangle$ as follows:

$$\begin{aligned} |x\rangle &= \frac{1}{\sqrt{2}}\left(|S_{tot}^z = +1\rangle - |S_{tot}^z = -1\rangle\right), \\ &r_x = +1,\ r_y = r_z = -1, \\ |y\rangle &= \frac{1}{\sqrt{2}}\left(|S_{tot}^z = +1\rangle + |S_{tot}^z = -1\rangle\right), \\ &r_x = -1,\ r_y = +1,\ r_z = -1, \\ |z\rangle &= |S_{tot}^z = 0\rangle, \\ &r_x = r_y = -1,\ r_z = +1. \end{aligned} \tag{3}$$

When the effects of anisotropies and field are calculated using these states, the properties of the Haldane state enter only in the form of two parameters, (i) energy gap Δ_0 at zero anisotropy and field and (ii)

$$\kappa = \sum_n \left(\frac{2}{3} - \langle x|(S_n^z)^2|x\rangle\right), \tag{4}$$

which is finite in the thermodynamic limit $L \to \infty$. At zero magnetic field the triplet splits under the influence of the anisotropies D and E producing three gap energies

$$\begin{aligned} \Delta_x &= \Delta_0 - \kappa D + 3\kappa E, \\ \Delta_y &= \Delta_0 - \kappa D - 3\kappa E, \\ \Delta_z &= \Delta_0 + 2\kappa D. \end{aligned} \tag{5}$$

The effect of the anisotropies and the external field on these lowest excited states is now obtained from a diagonalization in the subspace of the states of eq.(2). This amounts to assuming $D, E \ll 1$ and the magnetic field B well below the critical field B_c. The resulting energies have been discussed in ref. 5: when the external field is in one of the coordinate directions (α), the corresponding quantum number r_α survives as good quantum number, i.e. two of the basis states mix wheras the energy of the third one remains unaffected by the magnetic field.

In the following we extend these results of ref. 5 in several ways: (i) the wavefunctions of the excited states are written down, demonstrating the mixing of the different components of S_{tot}^z (which is no more a good number) and preparing the discussion of ESR transition rates in §3, (ii) the full, wavevector dependent spectrum is discussed and (iii) the combined effect of anisotropy and magnetic field on two magnon states is discussed.

2.1 One-magnon states: wave functions and full spectrum

In the variational approach of ref. 5 energies as well as wave functions of the one-magnon states are expressed by the gap energies Δ_α at $H = 0, D = E = 0$ and by the parameter κ. For the external field in $x-$direction the result is (we assume for definiteness $B_x = B \geq 0$, $\Delta_z - \Delta_y \geq 0$)

$$\epsilon_x = \Delta_x; \qquad |\psi_x\rangle = |x\rangle, \tag{6}$$

$$\begin{aligned} \epsilon_{yz(1,2)} &= \frac{1}{2}\left(\Delta_z + \Delta_y \mp \sqrt{(\Delta_z - \Delta_y)^2 + 4B^2}\right); \\ |\psi_{yz(1)}\rangle &= a|y\rangle + b|z\rangle, \\ |\psi_{yz(2)}\rangle &= b|y\rangle - a|z\rangle, \end{aligned} \tag{7}$$

where

$$\begin{aligned} a &= \frac{1}{\sqrt{2}}\left(1 + \frac{u}{\sqrt{u^2+1}}\right)^{\frac{1}{2}}, \\ b &= \frac{1}{\sqrt{2}}\left(1 - \frac{u}{\sqrt{u^2+1}}\right)^{\frac{1}{2}}, \\ u &= \frac{\Delta_z - \Delta_y}{2B}. \end{aligned} \tag{8}$$

The lowest excitation energy is $\epsilon_{yz(1)}$. $|\psi_x\rangle$ is eigenstate of S_{tot}^x with eigenvalue 0, $|\psi_{yz(1),yz(2)}\rangle$ approach eigenstates of S_{tot}^x with eigenvalues ± 1 for $u \to 0$, i.e. large field (Zeeman states). Spin parity r_x is the only remaining good quantum number with values $r_x = +1$ for $|\psi_x\rangle$ and $r_x = -1$ for $|\psi_{yz(1)}\rangle$ and $|\psi_{yz(2)}\rangle$. Magnon energies and wave functions for the external field in $z-$direction are obtained by interchanging the coordinates x and z in eqs.(6-8) above.

The results for energies and wave functions remain formally valid for the first excited state with arbitrary wavevector q when the gap energies Δ_α are replaced by the zero field excitation energies $\omega_\alpha(q)$ at the wavevector considered. This means that the parameter κ, which contains the necessary information about the system in the isotropic, no field limit has to be taken as $q-$dependent. The result of a numerical calculation of $\kappa(q)$ is given in Fig. 1 for $L = 10, 12, 14$. Whereas the energies $\omega_\alpha(q)$ are found to exhibit strong $L-$dependence, a comparison of the results for different L shows that $\kappa(q)$ is practically $L-$independent. The most interesting feature is that $\kappa(q)$ changes sign at $q \approx 0.75\pi$. This implies that the order of levels at wavevector π, where the doublet is lower than the singlet, is reversed for $q < q_r, q_r \approx 0.75\pi$. At $q = q_r$ the spectrum of excitations shows an accidental degeneracy.

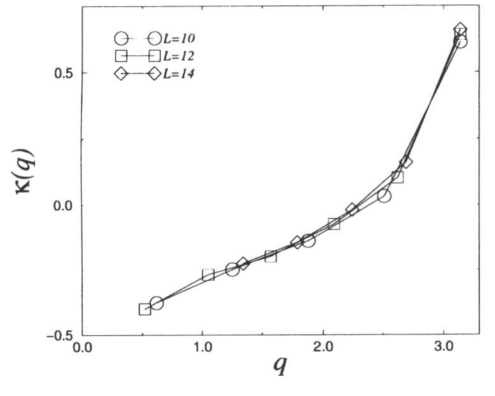

Fig. 1. The parameter $\kappa(q)$ determining the splitting of one-magnon energies due to anisotropy

An analytical estimate for $\kappa(q)$ is obtained using the simplest model for the Haldane triplet, the crackion model[12,13] which in the AKLT limit gives the following expression:

$$\kappa(q) = \frac{1}{12}\frac{9\cos 2q + 6\cos q + 1}{3\cos q + 5}.$$

This result is valid at the AKLT point (with biquadratic exchange of strength $\beta = -\frac{1}{3}$) in the crackion approximation and its quantitative value is very limited away from this point as is seen by the result $\kappa(\pi) = \frac{1}{6}$, substantially different from the numerical value $\kappa(\pi) \approx \frac{2}{3}$ for $\beta = 0$. However, it is interesting that $\kappa(q)$ changes sign at $q \approx 0.83\pi$ indicating the same type of level crossing which is found in the numerical results. These results for one magnon states at finite wavevector are meaningful only if the wavevector is sufficiently large to guarantee that the lowest excitation is not a two-magnon state, i.e. $q_0 < q < \pi$ with $q_0 \approx \frac{1}{2}\pi$.

2.2 Two-magnon states

Two-magnon states can be discussed in a way analogous to the one-magnon states starting from the nine states obtained as combinations of the basis states of eqs.(3). As explicit example we give in the following the lowest energy two-magnon states with wavevector $q = 0$ which are obtained in the unperturbed limit by adding two one-magnon quanta with $q = \pi$. With $B = 0, E = 0, D \neq 0$ we obtain one level with energy $2\Delta_0 + 4\kappa$ with $S^z_{tot} = 0$, a fourfold degenerate level with energy $2\Delta_0 + \kappa$ and $S^z_{tot} = \pm 1$ (each of these twofold) and a fourfold degenerate level with energy $2\Delta_0 - 2\kappa$ and and $S^z_{tot} = 0$ (twofold) and $S^z_{tot} = \pm 2$. The energies of these states in the external field are obtained trivially by adding the energies of the two constituent quanta as given in eqs.(6) and (7) and an excitation continuum results as usual. However, total spin S_{tot} is no more a good quantum number, the eigenstates are linear combinations of states $|S_{tot}, M_{tot}\rangle$ with total spin $S_{tot} = 0, 1$ or 2 and the mixing coefficients depend on the magnetic field. The two magnon state lowest in energy, $|\psi_{yz(1),yz(1)}\rangle$ is obtained by combining two quanta with energy $\epsilon_{yz(1)}$, and the second lowest, $|\psi_{yz(1),x}\rangle$ by combining two quanta with energy $\epsilon_{yz(1)}$ and ϵ_x. For the unnormalized wavefunction of these two states we obtain (again for external field in x−direction)

$$\begin{aligned}
|yz(1), yz(1)\rangle &= (\Delta_z - \Delta_y)\left(\frac{1}{2}|2;+2\rangle + \frac{1}{2}|2;-2\rangle\right.\\
&\quad \left.-\frac{1}{\sqrt{6}}|2;0\rangle + \frac{2}{\sqrt{3}}|0;0\rangle\right)\\
&\quad +\sqrt{(\Delta_z - \Delta_y)^2 + 4B^2}\left(\frac{1}{2}|2;+2\rangle\right.\\
&\quad \left.+\frac{1}{2}|2;-2\rangle - \frac{3}{\sqrt{6}}|2;0\rangle + \frac{2}{\sqrt{3}}|0;0\rangle\right)\\
&\quad +B\left(|2;+1\rangle + |2;-1\rangle\right),\\
|yz(1), x\rangle &= \left((\Delta_y - \Delta_z) + \sqrt{(\Delta_z - \Delta_y)^2 + 4B^2}\right)\\
&\quad \left(|2;+1\rangle - |2;-1\rangle + |1;+1\rangle + |1;-1\rangle\right)\\
&\quad +B\left(|2;+2\rangle - |2;-2\rangle + \sqrt{2}|1;0\rangle\right). \qquad (9)
\end{aligned}$$

Here $|S_{tot}, M_{tot}\rangle$ is the state obtained by combining two triplet excitations (states in the isotropic limit) with wavevector π each to a state with total spin S_{tot} and spin projection M_{tot}. The first of these states has spin parity $r_x = +1$, the second one $r_x = -1$. It is seen that the angular momentum of the two magnon states is a mixture of all possible values $S_{tot} = 0, 1, 2$ owing to the effect of anisotropy and magnetic field; this opens the possibility to have ESR absorption lines from the ground state to two-magnon excited states as will be discussed in the next section.

§3. ESR Transition Rates

In this section we present results for magnetic dipole transition rates as observable in ESR experiments. Magnetic dipole transitions between states i and f are determined by the matrix elements of the components of total spin $S^\beta_{tot} = \sum_n S^\beta_n$, $T^\beta_{f,i} \propto \langle f|\, S^\beta_{tot}\, |i\rangle$ and the transition rate is

$$R_{f,i} = |\sum_\beta h_\beta T^\beta_{f,i}|^2, \qquad (10)$$

where we have denoted the components of the microwave radiation field as h_β. For the external magnetic field $\vec{B}$ in direction α we have the following selection rules

$\Delta q = 0$, i.e. conservation of total momentum,
$\Delta S_{tot} = 0$, i.e. conservation of total angular momentum,
$r^{(f)}_\alpha = r^{(i)}_\alpha$ for $\vec{h} \propto \vec{e}_\alpha$, resp. $r^{(f)}_\alpha = -r^{(i)}_\alpha$ for $\vec{h} \perp \vec{e}_\alpha$.

When transitions from the ground state are considered, $|i\rangle = |g\rangle$, the angular momentum selection rule allows low energy transitions only when the ground state has admixtures of higher angular momenta since S^α_{tot} always annihilates $|g\rangle$ when $|g\rangle$ is a singlet. Moreover, the momentum selection rule allows only transitions to two-magnon states at wavevecor $q = 0$. Transitions between states with identical wave vector in the excited triplet are allowed when the spin parity selection rule is satisfied. In the remainder of this section, the transitions rates for the various cases of experimental interest are calculated using the results of §2.

Transitions between members of the first excited triplet:

For the external magnetic field in $x-$ and $z-$directions the states are given in §2 and the relevant transition elements are obtained as given in Table I (only nonzero elements are listed). a, b are given in eq.(8). In order to test the validity of the approximation, we present in Fig. 2 for $\vec{B} \propto \vec{e}_x, \vec{h} \propto \vec{e}_z$ the results of a numerical calculation of the transition rates between the states $|yz(1)\rangle \rightarrow |x\rangle$ (lowest to middle state, upper curve) and of the transition $|x\rangle \rightarrow |yz(2)\rangle$ (middle to upper state, lower curve) for a chain with $L = 14$, i.e. the quantities a^2 and b^2. It is seen that the dependence on magnetic field is reproduced perfectly by the analytic results and it becomes clear that, by using different wavevector directions and by varying the external magnetic field in direction and magnitude, levels can be identified and the interpretation of the spectra can be checked even for unpolarized radiation.

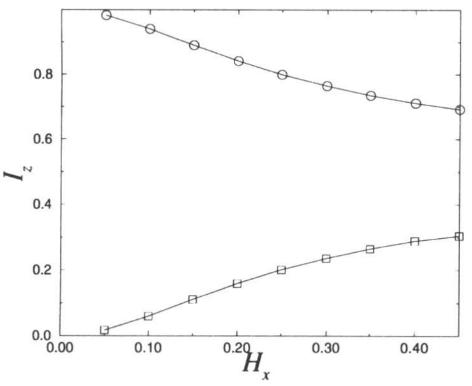

Fig. 2. ESR-transition rates for transitions between members of the first excited triplet, see text

Ground state transitions:

A finite transition rate for transitions from the ground state requires the presence of higher spin admixtures to the ground state as induced by the combined effect of anisotropy and magnetic field in the following way: We start by considering finite (but subcritical) magnetic fields at vanishing anisotropy, such that the ground state does not depend on the magnetic field and treat an anisotropy $D \neq 0$ in perturbation theory, assuming $D \ll 1$ as in section II. The first order correction in D to the Haldane ground state wave function $|g_0\rangle$ for a subcritical magnetic field in $x-$direction is then obtained

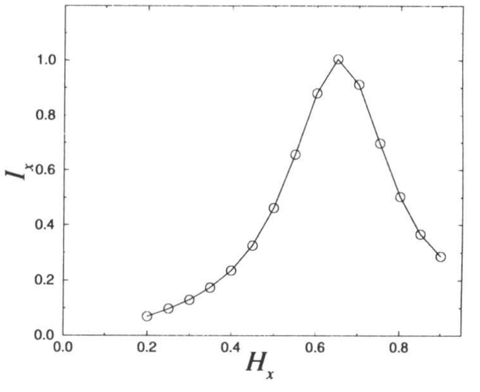

Fig. 3. ESR-transition rates for transitions from the ground state to the lowest two-magnon state, see text

as

$$\begin{aligned}|g\rangle &= |g_0\rangle + D((|exc(1); S_{tot} = 2, S^z_{tot} = +2\rangle \\ &\quad +|exc(1); S_{tot} = 2, S^z_{tot} = -2\rangle) \\ &\quad +|exc(2); S_{tot} = 2, S^z_{tot} = +0\rangle),\end{aligned}$$

where $|exc(1,2); S_{tot}, S^z_{tot}\rangle$ are states with definite angular momentum properties, but otherwise unknown. A complete calculation of the matrix elements relevant for ESR transition rates requires knowledge of these states in detail, the dependence on external magnetic field, however, is determined completely by the angular momentum coefficents and is obtained as follows:

Transition from the ground state to the lowest two-magnon state at $q = 0$ with spin parity $r_x = +1$: A nonzero matrix element is obtained only when the microwave field $\vec{h}$ is in $x-$direction and the field dependent part of the transition amplitude is

$$\begin{aligned}T^x_{yz(1),yz(1);g} &\propto \langle yz(1), yz(1)|S^x_{tot}|g\rangle \\ &= \text{constant } D\,B. \qquad (11)\end{aligned}$$

The energy of this transition is twice the gap energy and approaches zero at the critical field. The transition rate vanishes when the magnetic field approaches zero which illustrates that it is the combined effect of anisotropy and magnetic field which makes the transition rate finite: For vanishing magnetic field S^z_{tot} is a good quantum number and matrix elements become zero.

Transition from the ground state to the second lowest two magnon state at $q = 0$ with spin parity $r_x = -1$: A nonzero matrix element is obtained only when the microwave field $\vec{h}$ is perpendicular to the $x-$direction and the field dependent part of the transition amplitude is

$$\begin{aligned}\vec{h} \propto \vec{e}_y: \quad T^y_{yz(1),x;g} &\propto \langle yz(1), x|S^y_{tot}|g\rangle \\ &= \text{constant } D\,B\left(\sqrt{u^2+1} - u\right), \\ \vec{h} \propto \vec{e}_z: \quad T^z_{yz(1),x;g} &\propto \langle yz(1), x|S^z_{tot}|g\rangle \\ &= \text{constant } D\,B, \qquad (12)\end{aligned}$$

where the notation of eq.(8) has been used. The energy

direction of $\vec{B}$	transition	direction of $\vec{h}$	matrix element	intensity for $\vec{k} \propto \vec{e}_y$	intensity for $\vec{k} \propto \vec{e}_z$
$\vec{B} \propto \vec{e}_x$	$yz(1) \to (x)$	$\propto \vec{e}_y$	$-ib$	a^2	b^2
		$\propto \vec{e}_z$	a		
	$(x) \to yz(2)$	$\propto \vec{e}_y$	ia	b^2	a^2
		$\propto \vec{e}_z$	b		
	$yz(1) \to yz(2)$	$\propto \vec{e}_x$	$\frac{1}{\sqrt{2}}(b^2 - a^2)$	$\frac{1}{2}(b^2 - a^2)^2$	$\frac{1}{2}(b^2 - a^2)^2$
$\vec{B} \propto \vec{e}_z$	$yx(1) \to yx(2)$	$\propto \vec{e}_z$	$\frac{1}{2}(a^2 - b^2)$	$\frac{1}{4}(b^2 - a^2)^2$	0
	$yx(1) \to (z)$	$\propto \vec{e}_x$	$\frac{1}{\sqrt{2}}b$	$\frac{1}{2}b^2$	$\frac{1}{2}(a^2 + b^2)$
		$\propto \vec{e}_y$	$\frac{i}{\sqrt{2}}a$		
	$yx(2) \to (z)$	$\propto \vec{e}_x$	$-\frac{1}{\sqrt{2}}b$	$\frac{1}{2}b^2$	$\frac{1}{2}(a^2 + b^2)$
		$\propto \vec{e}_y$	$\frac{i}{\sqrt{2}}a$		

Table 1. ESR transition matrix elements T_{if} and intensities (for unpolarized microwave radiation) R_{if} with external magnetic field in $x-$ and $z-$directions.

of this transition is above the gap energy by Δ_x, thus it approaches Δ_x at the critical field. It should be noted that this transition coincides with the transition $yz(1) \to (x)$ inside the excited triplet in spin parity change and, when the critical field is approached, also in energy.

In Fig. 3 we present the result of a numerical calculation of the transition rate from the ground state to the lowest two magnon state for $L = 14$. The transition rate increases quadratically with magnetic field up to $B \approx 0.4$, for higher fields some deviations occur which probably result from the low excitation energies and small system size. It is also seen that at the critical field (which appears shifted to larger magnetic fields due to finite size effects) a level crossing occurs and the transition rate to the lowest $q = 0$ level above the critical field decreases with B. These effects will be dealt with in a forthcoming publication devoted to the high field phase.

§4. Conclusion

We have applied the simple analytical approach of ref. 5 to calculate the magnetic field dependence of ESR transition rates in the Haldane phase of anisotropic $S = 1$ chains in an external magnetic field. Transitions are possible both within states belonging to the anisotropy split first excited triplet, as well as between the ground state and low-lying two magnon states. Characteristic variations of intensity allow to identify the participating levels. Numerical calculations for $S = 1$ chains up to 14 spins confirm the analytical results.

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