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Lifetime of magnetic excitations in supported ferromagnetic and antiferromagnetic spin-$\frac{1}{2}$ Heisenberg chains

J. P. Gauyacq[1,*] and N. Lorente[2]

[1]*Institut des Sciences Moléculaires d'Orsay, ISMO, Unité mixte de recherches CNRS-Université Paris-Sud, UMR 8214, Bâtiment 351, Université Paris-Sud, 91405 Orsay Cedex, France*

[2]*Centre d'Investigació en Nanociéncia i Nanotecnologia (CSIC-ICN), Campus de la UAB, E-08193 Bellaterra, Spain*

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The lifetime of magnetic excitations in finite 1D-supported Heisenberg chains of magnetic atoms is studied theoretically for a model system formed of $S = \frac{1}{2}$ spins. Both ferromagnetic and antiferromagnetic cases are considered as well as open chains and rings of atoms. Different chain lengths are considered allowing extrapolation to infinite chains. All the excited magnetic states in the finite chains and rings are studied, not only the spin-wave mode. The magnetic excitations decay by electron-hole pair creation in the substrate. As the main result, for all the systems considered, the decay rate appears to vary approximately proportionally to the excitation energy of the state, with a proportionality constant independent of the strength of the Heisenberg exchange term. In certain finite systems, a stable state is evidenced at low energy, associated with a special spin coupling structure.

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I. INTRODUCTION

The development of high-resolution very low-temperature scanning tunneling microscopy (STM) opened a new way towards the detailed study of magnetic degrees of freedom in adsorbates at surfaces. In particular, inelastic electron tunneling spectroscopy (IETS) is a very valuable tool to determine the various energy levels of an adsorbate system, the conductance above an adsorbate as a function of applied bias presents steps at the energy position of the excited magnetic states of the system. This led to detailed studies of various adsorbate/surface magnetic properties.[1–10] In the case of chains of adsorbed atoms,[3,5] the spectroscopic information on the system could be interpreted using a Heisenberg Hamiltonian, so that it is tempting to assign the various excited states in a finite size structure to spin waves quantized in the system. The experimental achievements prompted a series of theoretical developments of the magnetic excitation process by tunneling electrons using perturbative,[11–14] nonperturbative,[15] multielectron,[16–18] and strong coupling approaches.[19,20] The strong coupling approach has been applied in particular to a model theoretical study of the excitation of chains of spin-$\frac{1}{2}$ adsorbates[21,22] (antiferromagnetic, ferromagnetic, and frustrated ferromagnetic cases). It showed that tunneling electrons were very efficient in inducing magnetic excitation in chains, similar to the individual adsorbate case and that detailed information on the spin wave excitation in an infinite chain (energy and momentum spectra) could be inferred from studies on finite size chains.[21] Spin waves have been the subject of many studies in particular via their excitation by neutron scattering.[23–26] In neutron scattering, the excitation concerns the entire chain, and the corresponding dynamic spin structure factor has been described in a series of papers (Refs. 27–30 and references therein). In contrast to the neutron scattering case where the exchanged wavenumber can be defined, for tunneling electrons, the primary excitation is local; the electron tunnels through a given atom in the chain, which thus receives the excitation; however, this atom is magnetically coupled to the rest of the chain so that the local excitation results in magnetic excitations delocalized along the chain with all possible wavenumbers.

All these studies on individual adsorbates and on nanostructures showed the existence of many excited magnetic states and yielded information on their spectroscopy and on the dynamics of their excitation by tunneling electrons. The excited states correspond to different orientations of the adsorbate magnetic moments with respect to the substrate and thus open fascinating possibilities for the miniaturization of electronics devices. However, in this context, a very important feature of these excited states is their lifetime, i.e. the spin relaxation time, which determines the extent to which such excited states could participate in a surface process. The inverse of the state lifetime is also an important contribution to the level width and also determines the extent to which the excited state can be observed experimentally. Much less is known on the lifetime of the excited magnetic states than on their energy. On the experimental side, spin relaxation times for magnetic states excited by tunneling electrons have been studied via the variation of the conductance in the case of very large electron current;[31] then multiple excitations induced by successive tunneling electrons compete with the excited state decay rate and thus yield a way of determining the state lifetimes. These turned out to be rather short in the sub-ns range for the studied system, Mn on CuN/Cu(100).[31] Another experimental study succeeded in a direct pump-probe measure of the time dependence of the magnetic excitation in the case of Fe and Cu atoms coadsorbed on CuN/Cu(100) surface.[32] The measured lifetime was substantially longer, in the few hundred ns range; this was attributed both to a favorable spin configuration (large easy axis anisotropy) of the Fe adsorbate and to the effect of the coadsorbed Cu adsorbate. These two examples concerned systems in which the magnetic adsorbate was separated from the metal substrate by a CuN monolayer which can be thought to provide an efficient insulation from the metal and thus to stabilize magnetic excitations. Indeed, in the same way as tunneling electrons are very efficient in inducing transitions among magnetic states in an adsorbate, electrons from the substrate that continuously hit the adsorbate

can also induce efficient magnetic de-excitations via electron-hole pair creation. As a consequence, an insulating layer separating the adsorbate from the metallic substrate can be thought to increase the spin relaxation time in adsorbates, in a way similar to the case of excited electronic states at surfaces.[33,34] Experimental studies of individual magnetic atoms or dimers directly adsorbed on a metal[8,9,35] revealed very broad structures in the conductance that were assigned to very short-lived excited magnetic states, with a lifetime in the tens of fs range. On the theoretical side, several studies of the decay of the magnetization excitation via electron-hole pair creation were performed on the systems mentioned above confirming the large difference between systems adsorbed on an insulating layer or directly on the metal substrate.[9,36,37]

The aim of this paper is to study the lifetime of magnetic excitations in finite Heisenberg chains of magnetic atoms adsorbed on a surface. It is a model study for finite-sized chains of spin-$\frac{1}{2}$ atoms coupled via ferromagnetic or antiferromagnetic Heisenberg couplings. The lifetime of the spin waves but also of all the other excited states in the system are computed. It is found that in both the ferro- and antiferromagnetic cases, the lifetime of the excited magnetic states follow a rather simple law, thus allowing the extrapolation of the results obtained for finite chains to the case of an infinite chain. The method used is directly inspired from our earlier work on individual magnetization excitation lifetimes and is briefly presented in Sec. II. Sec. III presents the results for ferro- and antiferromagnetic spin waves. Higher-lying states are discussed in Sec. IV for rings of atoms and in Sec. V for open chains of atoms. Sec. VI discusses the case of a very long-lived state appearing in the spin-$\frac{1}{2}$ chains, and the paper ends with a concluding summary.

II. METHOD

This paper uses the strong coupling approach that has been developed to treat magnetic excitation/de-excitation in adsorbates at surfaces.[19–21,36] Actually, it further elaborates on two earlier works: excitation of spin waves in Heisenberg chains[21] and de-excitation in individual adsorbates[36] to treat the de-excitation of spin waves by tunneling electrons in Heisenberg chains. Only a brief description of the method is given here.

We study a finite size ensemble of local spins (a chain of N atoms with $S = \frac{1}{2}$) coupled by a Heisenberg Hamiltonian:

$$H = \sum_{i=1}^{N-1} J\, \vec{S}_i\, \vec{S}_{i+1}. \tag{1}$$

In the present model study, we chose a simple system: the exchange coupling J is constant and only concerns first neighbors. Two different kinds of systems are examined: open linear chains of atoms [with Hamiltonian Eq. (1)] and rings of atoms with Hamiltonian Eq. (2):

$$H = \sum_{i=1}^{N} J\, \vec{S}_i \cdot \vec{S}_{i+1}, \quad \text{with} \quad \vec{S}_{N+1} \equiv \vec{S}_1. \tag{2}$$

The latter, with the loop condition, is an attempt to describe an infinite periodic chain of atoms[38] and so to reach a description of spin waves. Here, J is positive or negative and thus describes ferro- or antiferromagnetic cases. Below, J is used as the energy unit of the problem. We only considered chains and rings made of spin $\frac{1}{2}$. Although larger spins could, in principle, be studied with the present approach, the number of possible states increases much more rapidly with N for spins larger than $\frac{1}{2}$, making it very difficult to handle long chains. This paper aims at studying the decay rates of spin waves in infinite chains (modeled by rings) and tries to find common trends in the decay rates of the entire spectrum of excited states in a system, and for this reason we limited our study to the spin $\frac{1}{2}$ case.

Hamiltonian Eqs. (1) and (2) are diagonalized in a basis of products of local spins of the form $|M_1, M_2, ..., M_{N-1}, M_N\rangle$, where M_i is the projection of the spin of the atom at site i on the quantization axis ($M_i = \pm\frac{1}{2}$), to yield the eigen-states of the chain $|\Phi_j\rangle$, associated to the eigen-energies E_j. As discussed in earlier works,[26,38–40] the rings [Hamiltonian Eq. (2)] provide an excellent description of the ferromagnetic case and in particular of the spin waves, whereas the convergence with N, the total number of atoms, is rather slow in the antiferromagnetic case. However, the characteristics of the excitation by tunneling electrons in both cases can be inferred from calculations on finite-sized rings.[21]

The lifetime τ_j of the excited state Φ_j is the inverse of the total decay rate, $\Gamma_{\text{Tot},j}$, sum of the partial decay rates, $\Gamma_{j,f}$ (decay of the excited state j toward all the lower lying f states). The decay corresponds to the superelastic (final electron energy larger than its initial energy) scattering of substrate electrons by the adsorbate. The decay rate for a vanishing temperature can be expressed by the T transition matrix for electron scattering by one of the atoms in the chain as (see discussion in Refs. 36 and 41):

$$\begin{aligned}\frac{1}{\tau_j} &= \Gamma_{\text{Tot},j} = \sum_f \Gamma_{j,f} \\ &= \sum_f \frac{2\pi}{\hbar}\,\delta\Omega_f \sum_{k_j,k_f,m_j,m_f} |\langle k_f, m_f, \Phi_f | T | k_j, m_j, \Phi_j\rangle|^2 \\ &\quad \times \delta(\varepsilon_j - \varepsilon_f)\delta(\varepsilon_j - E_F),\end{aligned} \tag{3}$$

where Φ_f are the final states of the decay, associated to the energy transfer $\delta\Omega_f = E_j - E_f$. The total energy is $E_T = E_j + \varepsilon_j = E_f + \varepsilon_f$. The initial state and final states of the substrate electrons are labeled by their energy ε_j and ε_f, their wavenumbers k_j and k_f, and by their spin projections on the quantization axis m_j and m_f. Equation (3) is derived under two assumptions (see details in Refs. 36 and 41): (i) the system temperature is assumed to vanish, and (ii) the T transition matrix elements are assumed to be constant in the small energy interval involved in the decay process. In the present strong-coupling approach, the T transition matrix elements reduce (see below) to products of a flux factor by a spin-transition probability; the latter only involves spin coupling coefficients that are independent of the electron energy. Assumption (ii) then only concerns the flux of substrate electrons hitting the adsorbate, which can be assumed to be independent of energy in the small energy domain involved in the decay.

Similar to Refs. 36, it is assumed that the inelastic scattering of the substrate electrons with the chain (ring)

of magnetic atoms only occurs with one atom of the chain at a time and that this scattering is fast so that a sudden approximation, neglecting the Heisenberg Hamiltonian during the scattering process, can be used. The T transition matrix is then diagonal in the basis set formed by the eigen-states of $\vec{S}_T^2$ and $S_{T,z}$ (quantum numbers S_T and M_T), where $\vec{S}_T$ is the total spin of the scattering atom + electron system; in the present model system with spin-$\frac{1}{2}$ atoms, S_T is equal to zero or 1. The decay rate can then be re-expressed as:

$$\begin{aligned}\frac{1}{\tau_j} &= \sum_f \frac{2\pi\delta\Omega_f}{\hbar} \sum_{k_j,k_f,m_j,m_f} \delta(\varepsilon_j-\varepsilon_f)\delta(\varepsilon_j-E_F) \\ &\quad\times \left| \sum_{S_T} \langle k_f|T^{S_T}|k_j\rangle \sum_{M_T} \langle m_f,\Phi_f|S_T,M_T\rangle\langle S_T,M_T|m_j,\Phi_j\rangle \right|^2 \\ &= \sum_f \frac{2\pi\delta\Omega_f}{\hbar} \sum_{k_j,k_f,m_j,m_f} \delta(\varepsilon_j-\varepsilon_f)\delta(\varepsilon_j-E_F) \left| \sum_{S_T} \langle k_f|T^{S_T}|k_j\rangle A^{S_T}_{j,m_j\to f,m_f} \right|^2 . \end{aligned} \tag{4}$$

The term $A^{S_T}_{j,m_j\to f,m_f}$ is the amplitude for the $j\to f$ transition in the S_T symmetry; it is given by:

$$A^{S_T}_{j,m_j\to f,m_f} = \sum_{M_T} \langle m_f,\Phi_f|S_T,M_T\rangle\langle S_T,M_T|m_j,\Phi_j\rangle. \tag{5}$$

It only contains spin coupling coefficients, i.e. it expresses how one can go from the initial state to the final state via the (S_T, M_T) intermediate symmetry. If only one intermediate S_T symmetry is contributing to the decay, Eq. (4) can be further simplified into:

$$\begin{aligned}\frac{1}{\tau_j} &= \sum_f \frac{2\pi\delta\Omega_f}{\hbar} \sum_{k_j,k_f} \delta(\varepsilon_j-\varepsilon_f)\delta(\varepsilon_j-E_F) |\langle k_f|T^{S_T}|k_j\rangle|^2 \left(\sum_{m_j,m_f} \left| A^{S_T}_{j,m_j\to f,m_f} \right|^2 \right) \\ &= \sum_f \frac{2\pi\delta\Omega_f}{\hbar} T^{S_T}(E_F) \left(\sum_{m_j,m_f} \left| A^{S_T}_{j,m_j\to f,m_f} \right|^2 \right) = \sum_f \frac{2\pi\delta\Omega_f}{\hbar} T^{S_T}(E_F)\, P_{\rm Spin}(S_T, j\to f). \end{aligned} \tag{6}$$

Under this form, the decay rate for the $j\to f$ transition reduces to the product of the number of electrons hitting the adsorbate per second that can accommodate the $\delta\Omega_f$ inelasticity, by a spin-transition probability $P_{\rm Spin}$. For the present model study, we assumed that the two S_T symmetries contribute to the decay, and similar to Ref. 36, we used a statistical expression neglecting the interference terms between transitions within the two S_T symmetries that can be seen in Eq. (4). Finally, the decay rate of state j is obtained as:

$$\frac{1}{\tau_j} = \sum_f \Gamma_{j,f} = T_{\rm total}(E_F) \sum_f \frac{\delta\Omega_f}{h} P_{\rm Spin}(j\to f), \tag{7}$$

where $T_{\rm Total}(E_F)/h$ is the total electron flux hitting the adsorbate per unit of energy and per unit of time, it appears as a general factor for the decay rate of all magnetic states in the system.

It is given by:

$$\begin{aligned} T_{\rm total}(E_F) &= \sum_{S_T} (2\pi)^2 \sum_{k_j,k_f} \delta(\varepsilon_j-\varepsilon_f)\delta(\varepsilon_j-E_F) \\ &\quad\times |\langle k_f|T^{S_T}|k_j\rangle|^2. \end{aligned} \tag{8}$$

Here, $P_{\rm Spin}(j\to f)$ is a mean decay efficiency over the two S_T symmetries; it only depends on spin-coupling coefficients. The above expression corresponds to decay by scattering of a substrate electron on one given atom in the chain, and contributions for the different atoms have to be added. In the case of an open chain of atoms, these contributions are *a priori* different for the different sites in the chain, whereas in a ring, all atoms are equivalent, and one simply has to multiply the above rate by N the number of atoms in the ring.

Equation (7) is very similar to those we obtained for excitation by tunneling electrons.[20] Its physical interpretation is simple: there is a continuous flow of electrons coming from the substrate and bouncing from the adsorbate back into the substrate; their flux is governed by $T_{\rm Total}(E_F)$, and their efficiency in inducing the $(j\to f)$ de-excitation is given by the probability $P_{\rm Spin}(j\to f)$, leading to Eq. (7). In our study on Mn adsorbates on CuN/Cu(100),[36] $T_{\rm Total}(E_F)$ was found equal 1.1 (in a.u.) at the Fermi energy [as mentioned above, Ref. 36 gives the general theory and presents an application of how to compute $T_{\rm Total}(E_F)$ in a given system]. For the present model study, we took this global factor equal to unity, so that the present model should be typical for a chain of magnetic atoms adsorbed on an insulating monolayer on a metal. Thus, this paper cannot aim at quantitative results for a precise system because of the absence of a quantitative value of $T_{\rm Total}(E_F)$ for a given system. However, the $T_{\rm Total}(E_F)$ quantity is a common

factor for all the decay rates in a system, and as shown below, we can obtain general information on the characteristics of magnetic excitation lifetime and in particular on the lifetime dependence on the excitation energy.

One can also stress that the above decay process is very efficient. Indeed the excitation or de-excitation probability of a scattering electron [the $P_{\mathrm{Spin}}(j \rightarrow f)$ quantity in the present case] can be very large;[19,20,36] this is linked to the physical nature of the spin-transition process, which reduces to angular momentum conservation in the recoil of the scattering electron. It is linked with other scattering processes involving efficient angular momentum transfer, both in gas phase and surface problems.[42–45] This makes the magnetic excitation/de-excitation process very different from other processes, such as vibrational excitation.[46–48] We can also stress that, in the strong-coupling approach we use, an exchange interaction does not have to be explicitly introduced; simply assuming the exchange interaction to be very large allows obtaining the magnetic transition probabilities from spin-coupling coefficients.

We can note that the present approach does not include a tip contribution to the de-excitation process. Indeed, in most cases, the current flowing through the junction is weaker than the substrate electron flow bathing the adsorbate. Actually, contributions from the tip can be added as excitation and de-excitation processes induced by the tunneling electrons. This is exactly the way these were considered in the analysis of experiment,[31] as well as in our earlier work on Mn adsorbates lifetimes.[36] The system is driven out of equilibrium, but a rate equation description of the system is possible so that the de-excitation processes induced by the tip electron have the same character as the excitation processes induced by tunneling electrons; they thus look different from the spontaneous decay induced by substrate electrons discussed in this paper.

III. LIFETIME OF SPIN WAVES IN SPIN-1/2 HEISENBERG CHAINS

A. Ferromagnetic rings

We consider the case of a ring of N atoms with a ferromagnetic coupling. The ground state is degenerated and associated with a total spin equal to $N/2$, i.e. all the spins are aligned; the first excited state corresponds to the spin-wave mode; it is associated with a total spin $(N/2 - 1)$. In this case, a finite ring of atoms can be considered as a piece of an infinite chain of atoms, and a calculation with a ring of N atoms yields $N/2$ points in the k spectrum of the spin wave. Figure 1 shows the present results for the decay rate of the spin wave (units of J) as a function of the wavenumber k; the different k values were obtained for different values of N. Here, k is shown in units of π/a, where a is the interatomic distance. It appears that the discrete points for the decay rate of a spin wave of wavenumber k, $\Gamma(k)$, almost follow a continuous curve, the small deviations being attributed to differences in the number of open channels for different N, so that Fig. 1 can be considered as displaying the k dependence of the spin-wave decay rate. It appears that the decay rate depends very strongly on k, from vanishingly small at low k to a large value at the edge of the Brillouin zone. This behavior of $\Gamma(k)$ is reminiscent

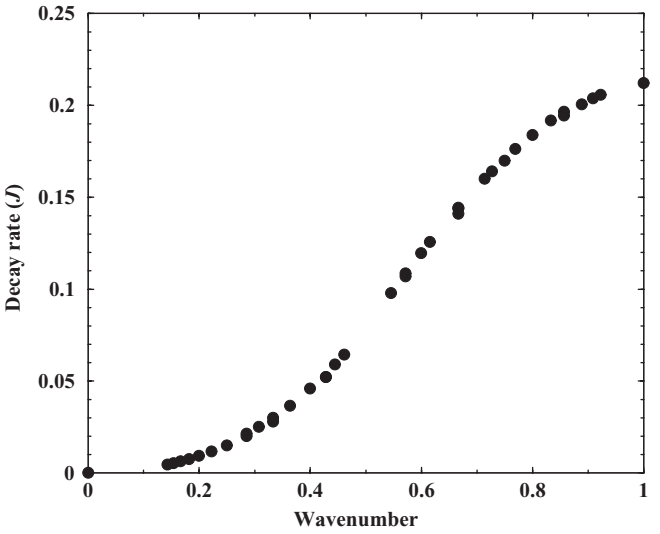

FIG. 1. Decay rate of the spin wave in a ferromagnetic ring of atoms as a function of the wavenumber. The decay rate is expressed in units of J, the ferromagnetic Heisenberg coupling, and the wavenumber in units of π/a (a is the distance between atoms in the ring). The different k values were obtained with calculations with different numbers of atoms in the ring.

of the energy dispersion $E(k)$ of the ferromagnetic spin wave (Refs. 38 and 26), and Fig. 2 shows the ratio $\Gamma(k)/E(k)$. It appears that the ratio is constant over the entire k range within a factor two. This quasiscaling of the decay rate with the energy can be understood by noting that the decay rate contains the excitation energy ($\delta\Omega_f$) as a factor; indeed several decay channels can exist so that it cannot be perfectly proportional, but still this scaling catches most of the variation seen in Fig. 1.

There are several ways of discussing the decay rate. Looking at the absolute value of the lifetime allows discussing

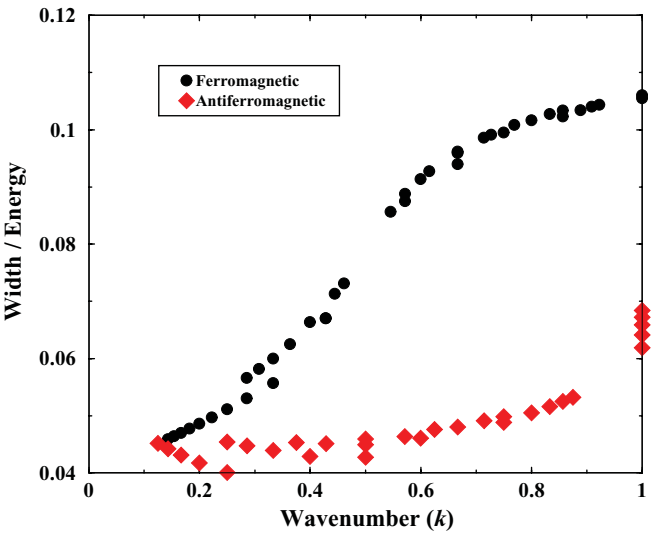

FIG. 2. (Color online) Ratio of the decay rate to the excitation energy of the spin-wave states in a Heisenberg ring of atoms, as function of k, the wavenumber (in units of π/a). Black circles: ferromagnetic case; red diamonds: antiferromagnetic case. The different results were obtained with calculations with different numbers of atoms in the ring.

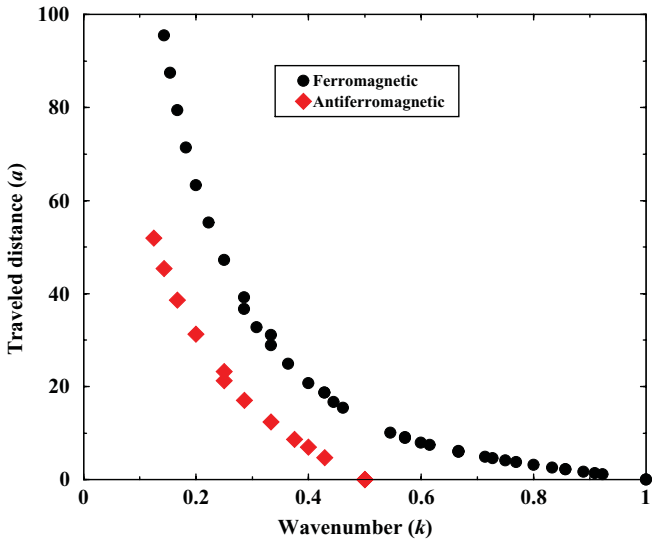

FIG. 3. (Color online) Mean distance traveled by a spin wave in an infinite Heisenberg chain of atoms before it decays, as a function of the wavenumber k. The distance is expressed in units of a, the interatomic distance in the chain, and the wavenumber in units of π/a. Black circles: ferromagnetic case. Red diamonds: antiferromagnetic case (the mean traveled distance can only be defined for $k < 0.5\,\pi/a$ in this case).

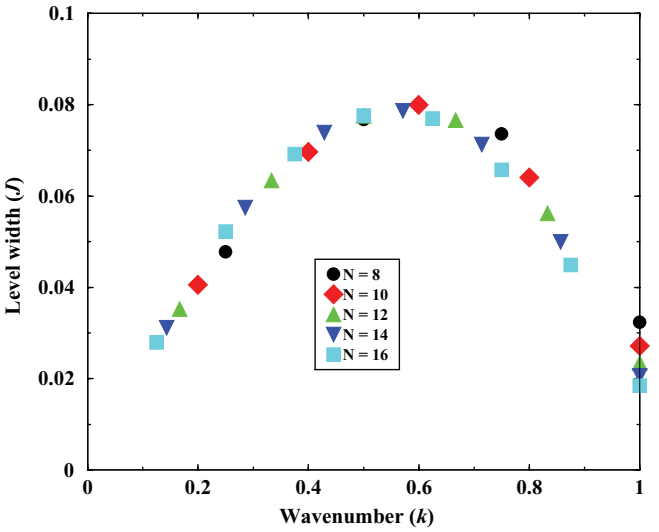

FIG. 4. (Color online) Decay rate of the spin-wave states of an antiferromagnetic ring as a function of the wavenumber (in units of π/a). The decay rate is expressed in units of J, the antiferromagnetic Heisenberg coupling. The different k values are obtained in calculations with different numbers N of atoms in the ring (see insert).

the visibility of the wave in the time domain. Looking at the ratio $\Gamma(k)/E(k)$ yields the visibility of the wave on an energy spectrum, e.g. in a magnetic IETS context; it is almost constant in k. Another way, displayed in Fig. 3, consists in defining the mean distance traveled by a spin wave before decaying. This a very practical notion[49] that gives direct information about the traveling wave character of the spin waves: how far can a spin wave go along the magnetic atom chains? It is defined as the product of the group velocity of the wave by the wave lifetime:

$$d(k) = \frac{dE}{dk}\tau(k). \tag{9}$$

Here, $d(k)$ is shown in Fig. 3 in units of a. With the above definition, it is independent of the ferromagnetic coupling J, but is inversely proportional to the flux factor $T_{\text{Total}}(E_F)$. It is then typical for the general ferromagnetic chain of atoms adsorbed on an insulating monolayer on a metal. As a first result, the distance is large, the wave traveling over tens of atoms before decaying. The distance diverges like k^{-1} as k goes to zero; however, one must remember that a $k = 0$ wave is not really propagating. In contrast at the edge of the Brillouin zone, this distance vanishes (this is due to the vanishing of the group velocity of the wave at the edge of the Brillouin zone), and one cannot consider the excited magnetic states at large k as waves traveling along the chain.

B. Antiferromagnetic rings

In the case of antiferromagnetic couplings in a ring with an even number of magnetic atoms, due to the large correlations in this system,[26] the characteristics of the ground state and of spin waves only converge slowly toward those in the infinite system, in contrast to the situation of a ferromagnetic ring, though, it is possible to extract general rules for the excitation of spin waves and higher states by tunneling electrons from calculations on finite rings.[21] In this system, the ground state of the system is associated to a total spin of the system equal to zero and the spin waves to a total spin equal to one. The spin waves thus decay towards lower-lying states with a total spin equal to zero, one, or two. Figure 4 presents $\Gamma(k)$, the decay rate of the spin waves (in units of J) as a function of the wavenumber k. The different discrete k values are obtained with different numbers of atoms in the ring (only even values of N are considered). The different points almost fall on a common smooth curve, but the convergence worsens at large k, as was already found in our earlier study on excitation processes.[21] This is particularly visible at the edge of the Brillouin zone; actually, in this point, the excitation energy of the spin wave vanishes for an infinite Heisenberg chain, and so the decay rate should also vanish. As in the ferromagnetic case, the k dependence of Γ resembles the dispersion law of the spin wave. This is confirmed in Fig. 2, which presents the ratio $\Gamma(k)/E(k)$; it is found to be almost constant with a variation of the order of $\pm 10\%$, if we exclude the point at the edge of the Brillouin zone that is known to be very slowly converging with N. This quasiproportionality between the decay rate and the excitation energy of the spin wave is a very remarkable result, which could be very useful for extrapolating widths/lifetimes for spin waves.

Figure 3 also presents the mean distance traveled by an antiferromagnetic spin wave along the chain of atoms. It can only be defined in the range $k < \pi/2a$, for which the group velocity is positive, and it vanishes at $k = \pi/2a$. As in the ferromagnetic case, this distance $d(k)$ is independent of the strength of the magnetic coupling J and is inversely proportional to the electron flux term $T_{\text{Total}}(E_F)$. We can then conclude from Fig. 3 that the distance traveled in the antiferromagnetic case is large but shorter than that found in the ferromagnetic case for the same coupling to the substrate. The distance diverges like k^{-1} as k goes to zero. This last

feature might be linked with the present model using a simple Heisenberg chain; it would disappear in the presence of an energy gap for the spin wave.

IV. LIFETIME OF ALL THE EXCITED STATES IN RINGS OF ATOMS

A. Ferromagnetic rings

We now consider all the excited states in a ferromagnetic ring, not only the spin waves. Each excited state can *a priori* decay toward lower-lying states with a selection rule on the total spin of the system $\Delta S_{\mathrm{Tot}} = 0, \pm 1$. In particular, only the spin waves can directly decay to the ground state since they are the only ones that can be excited from the ground state by a scattering electron; all other states can only decay to excited states, possibly leading to complex decay patterns from the original excited state to the ground state. Here, we compute the decay rate of each excited state; it corresponds to the time evolution of the population in this excited state and not to the time evolution of the return of the population into the ground state.

Figure 5(a) presents the decay rate of the excited states in a ferromagnetic ring of atoms as a function of the state excitation energy. Both the decay rate and the excitation energy are expressed in units of J, the Heisenberg ferromagnetic coupling. The results for three different values of N are shown, $N = 10$, 11, and 12. As the two main striking features, (i) the results are very close for the different N, and (ii) all the rates almost fall on a single smooth curve. These two features are indicative of a general scaling law for the decay rate as a function of the excitation energy. The relation is linear for the largest energy and bends a little at very low excitation energy as can be seen in Fig. 5(b), which presents an enlarged view of the results for $N = 14$ at low excitation energy; the scatter of points is also larger at low excitation energy. The results for the spin waves follow the general law, typically lying in the upper part of the cloud of points in Fig. 5(a) (not shown explicitly) and in Fig. 5(b). The quasiproportionality between the decay rate and the excitation energy is quite remarkable. Indeed, the different excited states correspond to very different spin structures (actually all the possible couplings are present); though, their decay via electron-hole pair creation obeys the same quantitative scaling law. One can stress that since both the decay rate and the excitation energy are proportional to J, the result in Fig. 5(a) is independent of the strength of the Heisenberg coupling. Changing the substrate would lead to a change in $T_{\mathrm{Total}}(E_F)$, i.e. to a global scaling of the width of the states, without altering the quasiproportionality law.

One can also notice in Fig. 5(b) that one of the excited states has a vanishing width. One such stable state can be found for all even values of N (for odd N, only a long-lived state is found); they lie in the low-energy part of the spectrum in Fig. 5(a) and are further discussed in Sec. VI.

B. Antiferromagnetic rings

Figure 6 presents the decay rate of the excited states in an antiferromagnetic ring of atoms as a function of the state excitation energy. Both the decay rate and the excitation energy are expressed in units of J, the Heisenberg antiferromagnetic

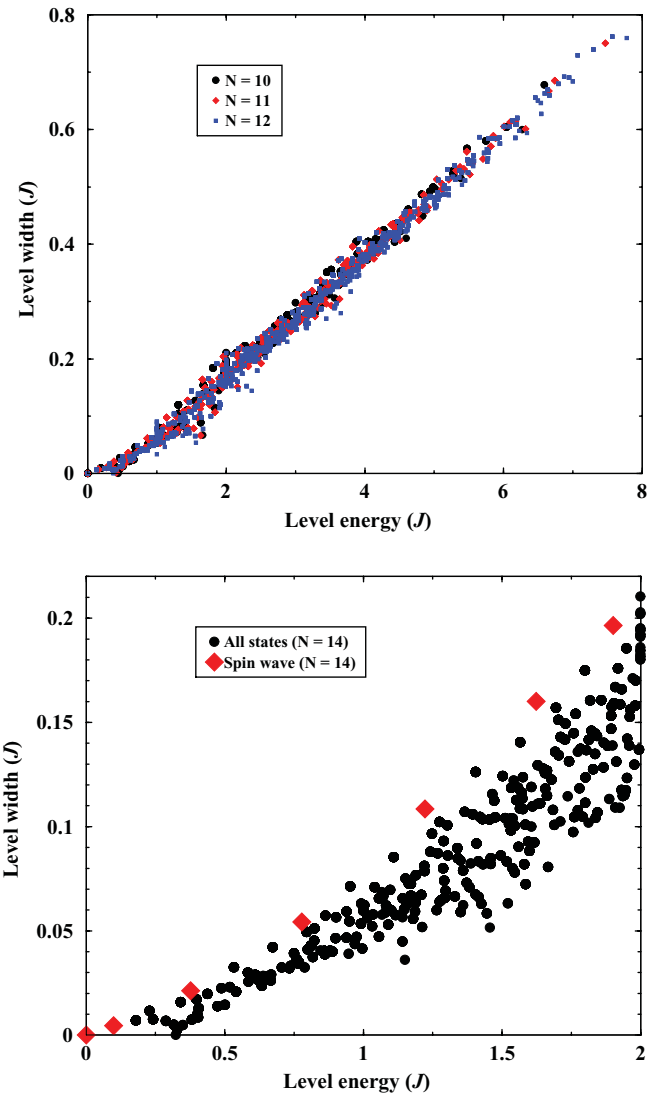

FIG. 5. (Color online) (a) Decay rate of the excited states in a ferromagnetic ring of atoms as a function of the excitation energy of the state. Both the decay rate and the energy are expressed in units of J, the Heisenberg ferromagnetic coupling. Results for three different numbers of atoms in the ring ($N = 10$, 11, and 12, see insert) are shown. (b) Decay rate of the excited states in a ferromagnetic ring of atoms as a function of the excitation energy of the state. Both the decay rate and the energy are expressed in units of J, the Heisenberg ferromagnetic coupling. The number of atoms in the ring is $N = 14$. Results for the spin wave are shown as red diamonds, and the results for all other excited states are shown as full black circles.

coupling. The results for three different values of N are shown, $N = 8$, 10, and 12. The same striking features as seen in the ferromagnetic case are again present: (i) the results are very close for the different N, and (ii) all the rates almost fall on a single smooth curve. The scaling even appears to be slightly clearer than in Fig. 5(a). In particular, the results for the spin waves (dark blue/gray line for $N = 12$) fall perfectly inside the cloud of points. Figure 6 also shows the results for the second spin-wave mode (light blue/light gray line); this mode has been defined in Ref. 21 as the second set of excited states when the excited states are ranked according to

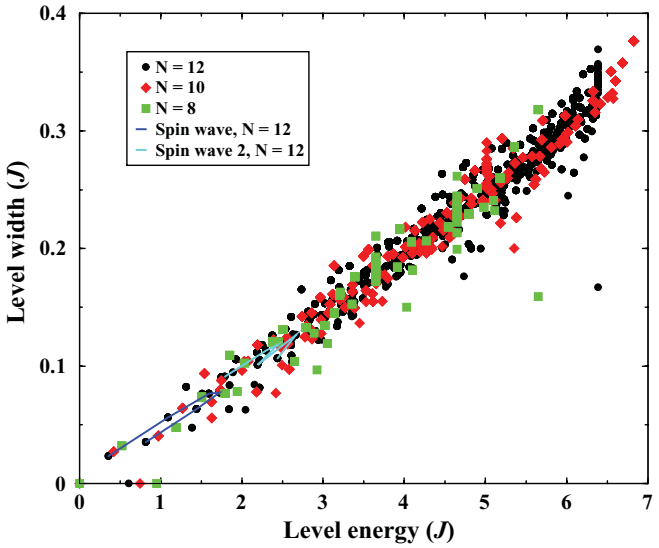

FIG. 6. (Color online) Decay rate of the excited states in an antiferromagnetic ring of atoms as a function of the excitation energy of the state. Both the decay rate and the energy are expressed in units of J, the Heisenberg antiferromagnetic coupling. Results for three different numbers of atoms in the ring ($N = 8$, 10, and 12, see insert) are shown. Results for the spin wave are also shown as well as for the second spin-wave mode (see text and insert).

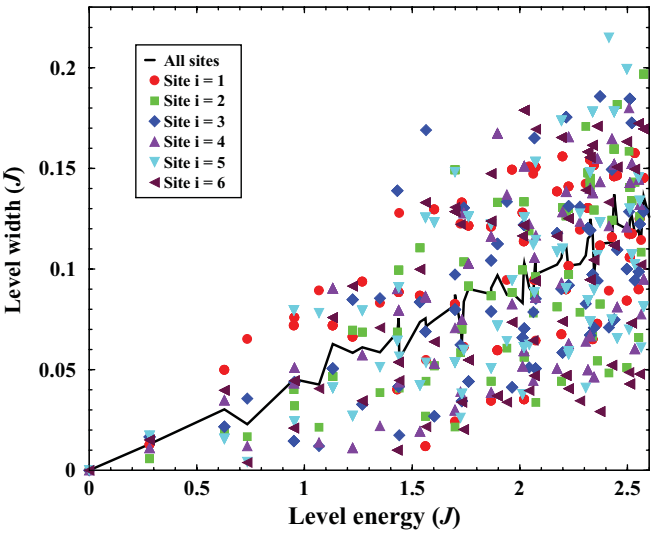

FIG. 7. (Color online) Decay rate of all the excited states of an open antiferromagnetic chain of atoms as a function of the state excitation energy. Both the decay rate and the excitation energy are expressed in units of J, the antiferromagnetic Heisenberg coupling. The number of atoms in the chain is $N = 12$. Decay of the excited states occurs via the superelastic collision of a substrate electron on one of the atoms in the chain. The contribution of the various atoms in the chain to the decay are presented by the various symbols (see insert), they have been multiplied by N to compare with the total decay rate of the excited states shown by the full black line.

their excitation probability by tunneling electrons. This second mode was interpreted as resulting from the quantization of the two-spinon continuum[28,29] in the finite-sized ring. The decay rate of this second mode also falls in the middle of the cloud of points of all states. The lifetime of all the excited states in an antiferromagnetic chain thus appears to be proportional to the excitation energy, although the spin structures of the various states are very different; in particular, the spin-wave modes which are the only states that can be excited from the ground state by a colliding electron exhibit decay rates very similar to all the other states that are not coupled directly to the ground state.

Similar to the ferromagnetic case, we can notice a single exception to the above rule: there exists a stable state in the excited state spectrum at low energy in Fig. 6; only one such state appears for each N value. They are further discussed in Sec. VI.

V. LIFETIME OF EXCITED STATES IN OPEN CHAINS

We also studied the decay of the excited states in an open chain of magnetic atoms described by Hamiltonian Eq. (1). In contrast to the case of rings, which were studied mainly as an attempt to discuss infinite chains, in particular spin waves, open chains of atoms should correspond to actual systems on surfaces. The situation is different from that of rings of atoms discussed above, the excited states cannot be attributed a wavenumber and cannot be considered as traveling waves. Though, as seen in Ref. 21, the excitation spectrum of an open chain by electrons tunneling through an atom close to the chain center bears strong resemblances with the excitation spectrum for a ring of atoms, so that we can expect strong similarities for the decay rates in the ring and open-chain cases.

Figure 7 presents the decay rate for all the excited states in an open antiferromagnetic chain of $N = 12$ atoms as a function of the excitation energy of the states. Both the state decay rate and excitation energy are expressed in units of J, the antiferromagnetic coupling term. In the case of a chain of atoms, the decay of the excited state is induced by the superelastic scattering of a substrate electron on one of the atoms in the chain. All the atoms in the chain are different (besides the symmetry with respect to the chain center), and thus the contributions to the decay rate from scattering through the different atoms in the chain are different. This is displayed on Fig. 7, where the contributions from atoms 1–6 in an $N = 12$ chain are shown (site 1 is at the end of the chain, and site 6 is the closest to the center). The individual contributions from the different atoms have been multiplied by a factor 12 to allow the comparison on the same figure of the total decay rate with the individual contributions. The various contributions are very different, covering a large area on the figure; however, summing the contributions to get the total decay rate has a strong averaging effect, and the total decay rate in Fig. 7 exhibits the same decay rate/energy proportionality rule as found for the ring. So the same remarkable feature as seen in Figs. 5(a) and 6 is also present in the case of an open chain of atoms.

The scaling law in the open-chain case is further illustrated in Fig. 8(a). It appears that identical scaling laws are found for even and odd numbers of atoms, although the structures of the two chains and in particular of their ground state are very different. There is a very large difference between the excitation processes in odd and even antiferromagnetic chains,[3,5,12,17,21] though it is possible to show strong links

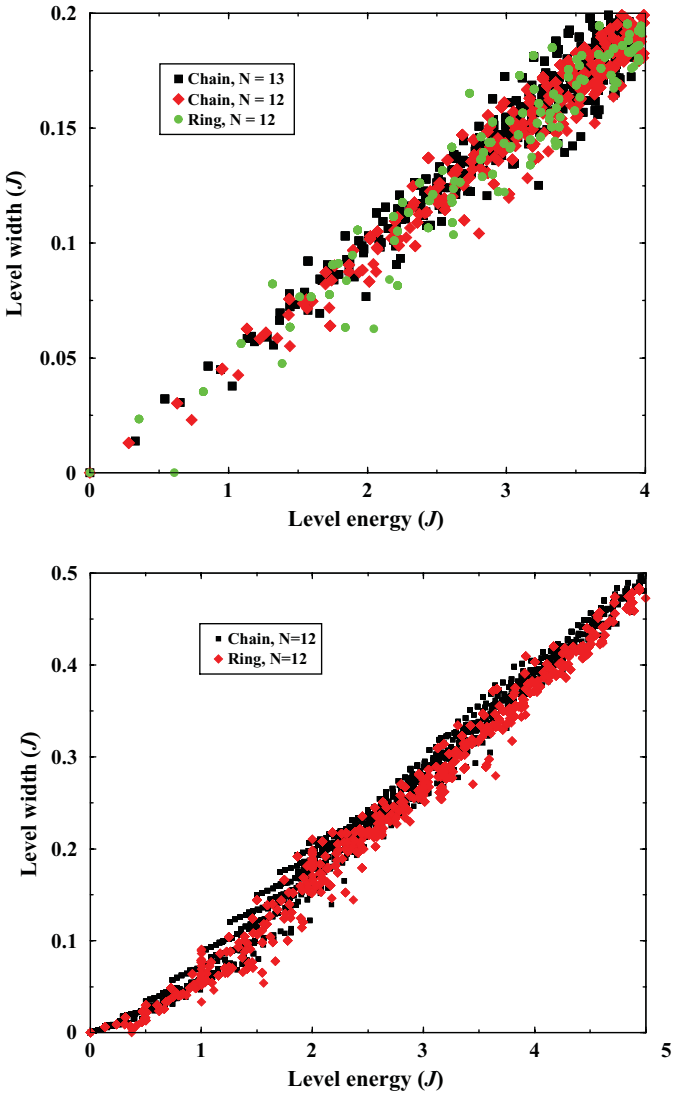

FIG. 8. (Color online) (a) Decay rate of all the excited states of an antiferromagnetic chain (ring) of atoms as a function of the state excitation energy. Both the decay rate and the excitation energy are expressed in units of J, the antiferromagnetic Heisenberg coupling. Various systems are shown on the figure: an open chain with 13 atoms (black squares), an open chain of 12 atoms (red diamonds), and a ring of 12 atoms (green circles). (b) Decay rate of all the excited states of a ferromagnetic chain (ring) of atoms as a function of the state excitation energy. Both the decay rate and the excitation energy are expressed in units of J, the ferromagnetic Heisenberg coupling. Various systems are shown on the figure: an open chain with 12 atoms (black squares), and a ring of 12 atoms (red diamonds).

between the two.[21] However, the decay rates of the excited states appear to be quantitatively equivalent in the two cases. Figure 8(a) also shows a comparison between an open chain and a ring, both with $N = 12$ atoms. It appears that the two results are very similar, the two sets of decay rates scale with the same proportionality factor between rates and energies (one can notice a difference between the open chain and ring sets of results: only the ring exhibits a stable state). So again, we find that open chains and rings are very similar even for not-too-large systems ($N = 12$ in the present case). The boundary condition is different at the chain end in open chains and rings; although it significantly affects the magnetic structure of the chain, it does not influence its dynamic properties for electron collisions much.

Figure 8(b) shows the decay rate of all the excited magnetic states in the case of ferromagnetic coupling. The results for both an open chain and a ring are shown for $N = 12$ ferromagnetic atoms. Qualitatively, the ferromagnetic open chains exhibit the same features as the antiferromagnetic chains discussed above: the contribution to the decay rate from one given site along the chain varies greatly from one site to another in the open chain case, but the total decay rate of the states is roughly proportional to the excitation energy of the state. The proportionality constant between total decay rate and excitation energy is almost the same in the open chain and ring cases. In addition, similar to the case of antiferromagnetic chains [see Fig. 8(a)], this proportionality constant is roughly independent of the chain length for the long chains (results not shown here for ferromagnetic chains).

Two main results appear in the above results [Figs. 8(a) and 8(b)] when looking at the decay rates of a large number of states on a large energy scale: (i) the decay rate of the excited states appear to be approximately proportional to the excitation energy of the states, and (ii) the proportionality constant does not depend on the chain length, even in the short chain range we considered. We can stress that the scaling is not perfect; actual results spread around the proportionality law, with a few special states such as those discussed in the next section. These properties appear in all the cases we studied: chains and rings with Heisenberg ferromagnetic or antiferromagnetic couplings of spin $S = \frac{1}{2}$ atoms. These properties survive if a magnetic field is applied to the system [see below in Fig. 9(b)]; this feature is not surprising since a magnetic field is not modifying the spin structure of the states but only the relative energy of the states (this, however, can change the number of possible decay channels of a given state). We performed some test calculations on other systems to probe the general character of this proportionality. Test calculations were performed on rings of spin $\frac{1}{2}$ with a frustrated ferromagnetic structure (chains of atoms with ferromagnetic first neighbor couplings and antiferromagnetic second neighbor couplings, see model in Ref. 22 and references therein); despite the very different magnetic structures and energy spectra met in these systems, the same kind of decay rate/energy scaling was found, except maybe at very low excitation energy.

Property (ii) is not so surprising in view of the convergence of the chain properties with N, the length of the chain (see the convergence of the excitation spectra in Ref. 21). However, property (i) can appear surprising. Indeed, the spin structures of the various states of a chain (or ring) are very different, and we would have expected different decay schemes and different behaviors for the different classes of states. In Eq. (7), the rate is given by the sum of products of a de-excitation energy by a spin-transition factor. Decay to the ground state is not the dominant decay route for most of the excited states (e.g. for finite ferromagnetic rings, only the very few spin-wave states can decay directly to the ground state); so a simple phase space argument cannot account for the observed proportionality. We rather tentatively attribute this proportionality to an averaging effect. A high-lying state can

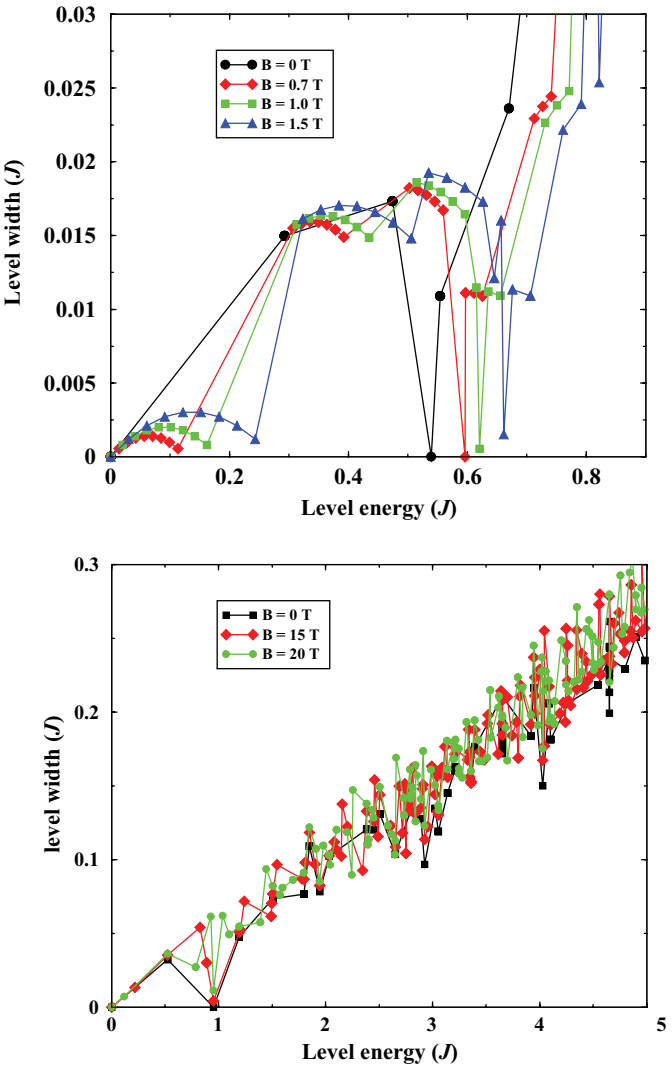

FIG. 9. (Color online) (a) Decay rate of the low-lying states of a ferromagnetic ring of eight atoms as a function of the level excitation energy. Both the level width and the level energy are expressed in units of J, the ferromagnetic Heisenberg coupling. Black circles: results in the absence of an applied magnetic field B. Red diamonds, green squares, and blue triangles: effect of an increasing applied B field (see insert). (b) Decay rate of the excited states of an antiferromagnetic ring of eight atoms as a function of the level excitation energy. Both the level width and the level energy are expressed in units of J, the antiferromagnetic Heisenberg coupling. Black squares: results in the absence of an applied magnetic field B. Red diamonds and green circles: effect of an increasing applied B field (see insert).

decay to a certain number of lower-lying states, each of these partial decays corresponding to a certain energy relaxation and spin-coupling factor. When the excitation energy increases, the number of possible de-excitation routes increases, leading to an averaging in the total decay rate, washing out the peculiarities of the spin coupling in each excited state. One can then expect to have a decay rate increasing monotonically with the excitation energy; this property should be rather robust and survive in many systems, in particular for chains with spins larger than $\frac{1}{2}$. A very qualitative and tentative view of the scaling consists in considering individual spin-flips in a chain of $S = \frac{1}{2}$ atoms. A high-lying excited state can be seen to be a certain number of spin-flips n_{sf} away from the ground state. Each substrate electron colliding on an atom of the chain is able to relax one of these spin-flips. The excitation energy can be assumed to be on the average proportional to n_{sf}; the decay rate can also be assumed to be proportional to n_{sf}, the number of possible relaxation routes induced by a substrate electron, and this justifies the proportionality observed in our results.

VI. LONG-LIVED STATES IN SPIN-1/2 HEISENBERG CHAINS

In the previous sections, we have noticed the existence of stable or very long-lived states in certain systems at low excitation energy. In all cases, at most one such special state appears in a given system. In the ferromagnetic case, a stable state is found for even numbers of atoms in a ring, and a very long-lived state is found for odd numbers [see e.g. Fig. 5(b)]. In the antiferromagnetic case, a stable state appears for even N rings (see Fig. 6), none appears in the antiferromagnetic open chains (see Fig. 8).

This oddity is due to a special spin coupling structure of the state. Figure 9(a) presents the decay rate of the low-lying states of a ring of $N = 8$ ferromagnetic atoms as a function of the state excitation energy. Results with an applied B magnetic field are also shown in order to illustrate the effect at play. In this system, the ground state is a $S_{\mathrm{Tot}} = 4$ state, ninefold degenerate in the absence of a B field. When a field is applied, the degeneracy of the ground state is lifted resulting in a manifold of nine states, closely spaced at low field. Each excited state in this manifold acquires a finite decay rate at finite B, due to the possibility of decaying to a lower state with the selection rule $\Delta M_{\mathrm{Tot}} = \pm 1$. This decay rate is small due to the small energy involved in the decay. The first group of excited states close to 0.3 J at $B = 0$ is a spin wave, associated with $S_{\mathrm{Tot}} = 3$ and sevenfold degenerated; it also splits with an applied B field, without much change in the decay rate, as expected. The second group of excited states is associated with $S_{\mathrm{Tot}} = 2$. The third excited state close to 0.54 J at $B = 0$ is the stable state. It does not split with a finite B field and is associated with $S_{\mathrm{Tot}} = 0$. Its excitation energy [abscissa in Fig. 9(a)] varies with B due to the variation of the energy of the ground state. For $B = 0$, the only states lower in energy are associated with $S_{\mathrm{Tot}} = 4, 3$ and 2 so that this $S_{\mathrm{Tot}} = 0$ state cannot decay via electron collisions to these lower states, and it is stable. The situation changes when the applied B field is large enough: then due to the Zeeman term, one sublevel from a higher lying $S_{\mathrm{Tot}} = 1$ manifold crosses the $S_{\mathrm{Tot}} = 0$ state, which thus acquires a finite width. This width is small due to the small energy involved in the decay, and it increases with B beyond this threshold. We can stress that, in the present case, the applied B field does not modify the magnetic structure of the states; it only modifies their energies; it lifts the degeneracies and changes the relative order of the states making possible the decay of the $S_{\mathrm{Tot}} = 0$ state. Analysis of the wave function of the stable state in the $N = 8$ case shows that it corresponds to the correlation mixing of several structures with two ferromagnetic contiguous zones: one zone with four spins up and one zone with four spins down. This stable state is found for all values of N, the number of

atoms in the ferromagnetic ring; its energy goes down as N increases. In the case of a ferromagnetic open chain, the energy ordering of the states with different S_{Tot} is different from that for a ring; there exists a $S_{\mathrm{Tot}} = 1$ state below the $S_{\mathrm{Tot}} = 0$ state, which thus has a finite decay rate.

Figure 9(b) illustrates the case of the stable state in the antiferromagnetic case. It presents the decay rate of the excited states in the case of an $N = 8$ ring. Similar to Fig. 9(a), a B field has been applied to allow a relative energy change among the levels and thus to allow the quenching of the special state. In the three cases in Fig. 9(b), the ground state is a $S_{\mathrm{Tot}} = 0$ state. At $B = 0$, the first excited state close to 0.5 J is a spin wave associated to $S_{\mathrm{Tot}} = 1$ and a momentum k equal to π/a, i.e. at the edge of the Brillouin zone. The second excited state with an excitation energy close to J is the stable state; it is associated with $S_{\mathrm{Tot}} = 0$ and with a momentum k equal to π/a. At $B = 0$, the second excited state cannot decay toward the ground state ($S_{\mathrm{Tot}} = 0$ to $S_{\mathrm{Tot}} = 0$ is forbidden) nor to the first excited state ($\Delta k = 0$ transitions do not exist in the antiferromagnetic case, see Ref. 21; see also a discussion in Ref. 22), and so it is stable. When the applied B field is increased, the third excited state (a spin wave associated with $S_{\mathrm{Tot}} = 1$ and $k = 0.8\,\pi/a$) splits in energy, and one of its components crosses the stable state, which thus acquires a finite lifetime, which decreases as B further increases [see Fig. 9(b)]. This stable state at $B = 0$ is found for all even values of N, the number of atoms in the antiferromagnetic ring; its energy goes down as N increases. In the case of an antiferromagnetic open chain, the energy ordering of the states with different S_{Tot} is different from that for a ring, and k is not a good quantum number anymore; and consequently, the special $S_{\mathrm{Tot}} = 0$ state has a finite decay rate.

VII. CONCLUDING SUMMARY

We have reported on a theoretical study of the lifetime of the excited states in finite chains and rings of spin-$\frac{1}{2}$ magnetic atoms (ferromagnetic and antiferromagnetic Heisenberg coupling). The decay of the excited magnetic states proceeds via the superelastic scattering of a substrate electron on one of the atoms in the chain, i.e. by an electron-hole pair creation mechanism. The study was not limited to the usual spin-wave states, and all the excited states have been considered, irrespective of their spin structure. The calculations were performed with different chain and ring lengths, allowing drawing some general conclusions for infinite systems.

As the main results:

– The decay rate of the excited magnetic states appears to be approximately proportional to the excitation energy of the states in all the cases we studied: ferromagnetic and antiferromagnetic, chains and rings of atoms. This result can seem to be *a priori* surprising; we tentatively attribute it to an averaging effect present when we consider a large number of high-lying excited states. The proportionality constant is found to be roughly the same for closed rings and open chains.

– The results for different chain lengths were found to be very similar. This confirms the rather fast convergence of this magnetic property with the chain length, similar to what was observed for the chain excitation by tunneling electrons.[21] It also allows drawing conclusions on the behavior of the lifetime of spin waves in an infinite system.

– The lifetime of the spin waves appear very similar to those of all the excited states in the chains, exhibiting the same approximate decay rate/energy ratio.

– We expect the proportionality result to have a rather general character. We found it in all the systems we studied: chains and rings of spin $\frac{1}{2}$ with ferromagnetic, antiferromagnetic, and frustrated ferromagnetic couplings, although these display very different magnetic structures. The proportionality constant is independent of the J Heisenberg coupling in the chain (J appears as a global energy scale in the problem). It depends on $T_{\mathrm{Total}}(E_F)$ factor, the flux of substrate electrons at the adsorbate, that enters as a global factor in the decay rate. Changing from one adsorbate system to another with the same spin structure would simply result in a change of this flux and thus to a global multiplicative factor on the decay rate of all states.

– It is possible to define the mean distance traveled by a spin wave along an infinite chain before decaying. This distance is independent of J, the Heisenberg coupling. For the present numerical application, which should be representative of adsorbed chains partly decoupled from the substrate, the distance traveled is rather long, allowing one to define a large k range where the spin waves are actually propagating along the chain.

– The decay appears to be faster in the ferromagnetic case than in the antiferromagnetic case. However, due to the different dispersion laws, ferromagnetic spin waves travel farther than antiferromagnetic spin waves.

– In finite systems, a stable state can appear in the low energy range of the excitation spectrum. They correspond to special coupling schemes in the spin chains for which the selection rule for the decay by electron-hole pair creation cannot be fulfilled. Applying a magnetic field allows quenching of these special states above a certain threshold field.

*jean-pierre.gauyacq@u-psud.fr

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