

**Laser-induced polarization of a quantum spin system in the steady-state regime**

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The effect of the circularly polarized laser field on quantum spin systems in the steady-state regime, in which relaxation plays the central role, has been studied. The dynamical mean-field-like theory predicts several general results for the behavior of the time-average magnetization caused by the laser field. The induced magnetization oscillates with the frequency of the laser field (while Rabi-like oscillations, which modulate the latter in the dynamical regime, are damped by the relaxation in the steady-state regime). At high frequencies, that magnetization is determined by the value to which the relaxation process is directed. At low frequencies the slope of that magnetization as a function of the frequency is determined by the strength of the laser field. The anisotropy determines the resonance behavior of the time-averaged magnetization in both the ferromagnetic and antiferromagnetic cases with nonzero magnetic anisotropy. Nonlinear effects (in the magnitude of the laser field) have been considered. The effect of the laser field on quantum spin systems is maximal in resonance, where the time-averaged magnetization, caused by the laser field, is changed essentially. Out of resonance the changes in the magnetization are relatively small. The resonance effect is caused by the nonzero magnetic anisotropy. The resonance frequency is small (proportional to the anisotropy value) for spin systems with ferromagnetic interactions and enhanced by exchange interactions in the spin systems with antiferromagnetic couplings. We show that it is worthwhile to study the laser-field-induced magnetization of quantum spin systems caused by the high-frequency laser field in the steady-state regime in “easy-axis” antiferromagnetic spin systems (e.g., in Ising-like antiferromagnetic spin-chain materials). The effects of the Dzyaloshinskii-Moriya interaction and the spin-frustration couplings (in the case of the zigzag spin chain) have been analyzed.

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Dynamics and relaxation under the effect of laser fields have attracted much attention during recent years. Ultrafast manipulation with laser fields can lead to states of matter which do not exist in equilibrium. Recently, several applications of intense laser fields to systems of interacting particles have been predicted theoretically and realized in experiments, including Mott insulators [1], superconducting systems [2], systems of ultracold atoms [3], and topological transitions [4]. An application of the intense laser field can change, e.g., the repulsion between electrons to attraction because it induces inversion of the population, corresponding to “negative” effective temperatures, which implies changing the sign of the interaction between particles [5]. The laser field (especially in the terahertz regime) can provide a direct way of manipulating the motion of charges and spins at the femtosecond time scale [6]. Terahertz-induced magnetic dynamics can address spins via the Zeeman interaction [7].

To study magnetic properties of quantum spin systems one usually applies the static magnetic field and measures the induced magnetization as a function of the applied field. However, for interacting spin systems such a study requires very high values of magnetic fields, up to 100 T. Recently, studying magnetic systems under the action of high-frequency laser fields was proposed [8]. The idea explores the unitary transformation of the circularly polarized laser field to an effective magnetic field applied perpendicular to the polarization plane, with the effective magnitude of the magnetic field equal to the frequency of the laser. It was shown [8] that such a circular polarization is the key ingredient

of the laser-induced magnetization of spin systems. It turns out that a laser with the frequency of the terahertz range can typically produce such an effective magnetic field (in the rotating frame) of the order of 40 T, while the magnitude of that laser field is usually less than 0.5 T. This way, stronger effective fields can be obtained by increasing the frequency of the laser. Similar results were predicted for the linear polarized laser field [9]. Recently, a similar approach was used in Ref. [10] for the theoretical investigation of gapped spin-1 systems. Notice that in Refs. [8,10] the effect of laser fields on spin systems was considered in the dynamical regime, not taking into account possible relaxation processes. However, it is clear that damping can essentially change the behavior of the laser-induced magnetization (see Ref. [9] for the linear laser polarization).

In this work the effect of the circularly polarized laser field on several quantum spin systems in the steady-state regime has been studied. That regime takes place at large time scales. The induced magnetization oscillates with the frequency of the laser field in the steady-state regime. It is different from the dynamical regime (small time scales), in which oscillations with the laser frequency are modulated by Rabi-like oscillations. The latter are smeared out by the relaxation at large time scales. The magnetic anisotropy causes the resonance behavior of the time-averaged magnetization. Nonlinear effects (in the magnitude of the laser field) can cause jumps and cusps in the frequency dependence of the steady-state value of the time-averaged magnetization. Such nonlinear effects are stronger for ferromagnetic interactions and weaker for the antiferromagnetic case. The resonance frequency is relatively small (proportional to the anisotropy

value) for spin systems with ferromagnetic interactions, and it is enhanced by the exchange interaction in spin systems with antiferromagnetic couplings. That is why we suggest studying the laser-field-induced magnetization of quantum spin systems caused by the high-frequency laser field in the steady-state regime, namely, in “easy-axis” antiferromagnetic spin systems. The other main statement of our work is the following. The effective Hamiltonian of the spin system under the action of the circularly polarized laser field in a rotating frame determines only the *dynamics* of spins, not the thermodynamics. The action of the circular polarized laser field drastically differs from the one of the static magnetic field. The latter changes the thermodynamic properties of the system *together* with dynamics. As usual, for the Hermitian Hamiltonian the laser field induces oscillations of spins of the considered system and thus the oscillation of the projection of the magnetization. That is why the circularly polarized laser field (i.e., the periodic driving force acting on the spins of a magnet) can induce significant enhancement of the projection of the magnetization only for some period of time (smaller than the inverse frequency of the laser field). Then, the projection of the magnetization reaches its maximal value and then becomes smaller and so on. The circular polarized laser field itself does not induce stationary changes of the magnetization of the spin system. It can cause only the mean steady-state value of the magnetization, about which the latter oscillates under the action of the driving force. Such a mean value (as well as the magnitude of oscillations) depends on many parameters of the system, including the presence (absence) of the magnetic anisotropy, the sign of the isotropic spin-spin interactions, and the value and form of relaxation in the system. On the other hand, the static magnetic field induces *stationary* changes of the projection of the magnetization together with the standard Zeeman oscillations of the magnetization.

## II. LASER-INDUCED MAGNETIZATION: THE STEADY STATE

Consider the quantum spin system with interactions between spins. Suppose the system is situated in the circularly polarized electromagnetic field of a laser. The Hamiltonian of the system can be written as

$$\mathcal{H} = \mathcal{H}_{\text{is}} + \mathcal{H}_{\text{an}} - h \sum_n [\cos(\omega t) S_n^x - \sin(\omega t) S_n^y], \quad (1)$$

where  $S_n^{x,y,z}$  are the operators of the projections of the spin situated at the site  $n$ ,  $t$  is time,  $h = g\mu_B h_0$  and  $\omega$  are the magnitude ( $h_0$ ) and the frequency of the circular polarized laser field ( $g$  is the effective  $g$  factor, and  $\mu_B$  is Bohr’s magneton),  $\mathcal{H}_{\text{is}} = J \sum_{n,\delta} \mathbf{S}_n \cdot \mathbf{S}_{n+\delta}$  is the part of the Hamiltonian which describes isotropic (exchange) interactions with the coupling constant  $J$  between spins of the system, and, finally,  $\mathcal{H}_{\text{an}}$  describes the magnetic anisotropy. Here we limit ourselves to the uniaxial anisotropy, for example,  $\mathcal{H}_{\text{an}} = \sum_{n,\delta} A_\delta S_n^z S_{n+\delta}^z$ , where  $A_\delta$  are anisotropy constants. (For  $\delta = 0$  the model describes the single-ion anisotropy, while for  $\delta \neq 0$  it describes the interion anisotropy).

It is well known that the dynamics of any measurable values in quantum mechanics can be described in two ways. In the first approach one can write the Heisenberg equations of motion for

operators of those quantities and average the obtained results either with the wave function (for the pure state) or with the density matrix (for the mixed state) of the system taken at a fixed time. In the second approach one can find the solution of the nonstationary Schrödinger equation for the wave function of the system (or the Liouville equation for the density matrix) and then average the considered operator with the obtained time-dependent wave function or density matrix. Naturally, if one realizes these procedures exactly, the result must be the same. Let us use the second approach. The Liouville equation for density matrix  $\rho$  has the form

$$i\hbar\dot{\rho} = [\mathcal{H}, \rho]. \quad (2)$$

To consider the effect of the circular polarized laser field it is useful [8] to perform the unitary transformation  $\rho = U\rho_1 U^{-1}$ , where  $U = \exp(i\omega t \sum_n S_n^z)$ . For the spin system it is equivalent to the transition to the rotating frame. Then the equation of motion for  $\rho_1$  can be written as

$$i\hbar\dot{\rho}_1 = [\mathcal{H}_1, \rho_1], \quad (3)$$

where  $\mathcal{H}_1 = U^{-1}\mathcal{H}U + \hbar\omega \sum_n S_n^z$ . For the system described by the Hamiltonian (1) the Hamiltonian  $\mathcal{H}_1$  can be written exactly,  $\mathcal{H}_1 = \mathcal{H}_{\text{is}} + \mathcal{H}_{\text{an}} - h \sum_n S_n^x + \hbar\omega \sum_n S_n^z$ .

Usually, however, the spin system is not isolated. For example, there are processes which export the energy from the system, i.e., relaxation processes. Relaxation can be considered in a number of ways. For simplicity it is possible to use the relaxation first introduced in Ref. [11]. Namely, one can write the Liouville equation of motion for the density matrix as the “quantum Boltzmann kinetic equation” for the density matrix in the approximation of relaxation time in the form

$$i\hbar\dot{\rho} = [\mathcal{H}, \rho] - i\hbar\gamma(\rho - \rho_0). \quad (4)$$

Here the relaxation of the density matrix with the rate  $\gamma$  to the state with the density matrix  $\rho_0$  was introduced. The reason for the relaxation of the density matrix is the interaction of the considered system with some environment; such an interaction withdraws energy from the considered system. For example, for the studied quantum spin system the crystal lattice (i.e., the elastic subsystem) can serve as such an environment. This approximation implies equal relaxation times for all eigenmodes of the system. It is equivalent to the Bloch form of the relaxation in the theory of the nuclear magnetic resonance [12]. (Two relaxation times as in Bloch’s approach can be easily introduced in the above scheme by using different relaxation rates for diagonal and nondiagonal components of the density matrix.) In general, one can use the Lindblad master equation [13] for the description of relaxation processes, which describes several methods of dissipation. In particular, one can speak about Bloch-like terms (with the relaxation rate) in the Lindblad equations and other terms which are similar to the Torrey phenomenological theory [14], which adds diffusion processes to the Bloch equations. It is possible to show that the effect of relaxation in the Bloch form is similar to the effect of relaxation in the Landau-Lifshitz form for spin systems [15]. Here we are interested mostly in the homogeneous response and can neglect the spatial dependence of relaxation. So in the limit of small relaxation rates the quantum Boltzmann equation and the Lindblad equation yield similar results. In our approach

the temperature dependence in the thermal equilibrium appears via  $\rho_0 = \exp(-\mathcal{H}_0/T)[\text{Tr} \exp(-\mathcal{H}_0/T)]^{-1}$ , where  $T$  is the temperature (the Boltzmann constant is taken to be equal to 1) and  $\mathcal{H}_0$  is the Hamiltonian that describes the state to which the system relaxes. Obviously, after the unitary transformation we can write

$$i\hbar\dot{\rho}_1 = [\mathcal{H}_1, \rho_1] - i\hbar\gamma(\rho_1 - U\rho_0U^{-1}). \quad (5)$$

### III. STEADY-STATE MAGNETIZATION IN MAGNETICALLY ISOTROPIC SYSTEMS

First, let us consider the spin system without magnetic anisotropy,  $A_\delta = 0$ . All projections of the total spin  $S^{x,y,z} = (1/N)\sum_n S_n^{x,y,z}$  ( $N$  is the number of spins) commute with the Hamiltonian  $\mathcal{H}_{\text{is}}$ . In that case it is possible to write equations of motion for the average values of operators of spin projectors with the density matrix  $\langle S^{x,y,z} \rangle = \text{Tr}(\rho S^{x,y,z})$  (they can be called the quantum Boltzmann equations too) as

$$\begin{aligned} i\hbar \left[ \frac{\partial \langle \tilde{S}^\pm \rangle}{\partial t} + \gamma(\langle \tilde{S}^\pm \rangle - \tilde{S}_0^\pm) \right] &= \mp \hbar\omega \langle \tilde{S}^\pm \rangle \mp \hbar \langle S^z \rangle, \\ i\hbar \left[ \frac{\partial \langle S^z \rangle}{\partial t} + \gamma(\langle S^z \rangle - S_0^z) \right] &= -\frac{\hbar}{2}(\langle \tilde{S}^+ \rangle - \langle \tilde{S}^- \rangle). \end{aligned} \quad (6)$$

The approach is similar to the Bloch equations. Here the standard notations  $S^\pm = S^x \pm iS^y$ ,  $\tilde{S}^\pm = US^\pm U^{-1}$ , and  $\tilde{S}_0^\pm = US_0^\pm U^{-1}$  are used. Notice that, obviously,  $S^z = \tilde{S}^z$  and  $S_0^z = \tilde{S}_0^z$ . Here we supposed that the system relaxes to the state with the values of spin projections  $S_0^{x,y,z}$ . The latter can be determined, e.g., by the Hamiltonian of the system in the thermal equilibrium (i.e., not taking into account the effect of the laser field) [9] or in some other way [11]. One can see that  $S_0^{x,y,z}$  determine the temperature dependence of the system in thermal equilibrium in this approach. The solution of these equations describes the dynamics of the isotropic quantum spin system under the action of the circular polarized laser field if the relaxation is taken into account in the simplest form. Notice that the set of equations (6) is closed.

It is instructive to look for the solution of these equations in two important limits. First, one can be interested in the dynamical regime of the spin system under the action of the laser field. It happens for  $t \ll \gamma^{-1}$ . In this limit it is possible to neglect terms proportional to  $\gamma$  on the left-hand sides of Eqs. (6). Such a situation was studied in Refs. [8,10]. The solution (we suppose an initially nonzero value of  $\langle S^z \rangle$ ) describes oscillations of  $S^{x,y,z}$  with the frequency of the laser field  $\omega$  modulated by the Rabi-like frequency  $\Omega = \sqrt{\omega^2 + (\hbar/\hbar)^2}$ . For example, the  $z$  component of the laser-induced spin moment (the magnetization is proportional to that component multiplied by  $g\mu_B$ ) is

$$\langle S^z \rangle \sim \frac{\hbar}{\hbar\Omega} \left\{ \cos(\omega t) - \cos[(\omega - \Omega)t] - \frac{\hbar}{\hbar\Omega} [1 - \cos(\Omega t)] \right\}. \quad (7)$$

Notice that here the effective field  $\sqrt{\omega^2 + (\hbar/\hbar)^2}$  is much larger than in the case when the static magnetic field  $H$  is present and the Rabi-like frequency is  $\sqrt{(\omega - H/\hbar)^2 + (\hbar/\hbar)^2}$ , which in resonance is small,  $\Omega \sim \hbar/\hbar$ . For the standard for laser field situations,  $\hbar\omega > \hbar$ ;

hence, the values of spin projections oscillate almost with the frequency  $\omega + \Omega \approx 2\omega$ , modulated by the low frequency  $|\omega - \Omega| \approx \hbar^2/2\hbar^2\omega$ . By time averaging Eq. (7) we get  $\langle \tilde{S}^z \rangle \equiv \lim_{A \rightarrow \infty} (1/A) \int_0^A \langle S^z \rangle dt \sim -(\hbar/\hbar\Omega)^2$ . At small  $t \ll |\omega|^{-1}$  Eq. (7) can be expanded, yielding  $\langle S^z \rangle \sim (\hbar/2)(t/\hbar)^2[\hbar\omega - \hbar]$ . The latter essentially implies the results of Refs. [8,10].

However, in this work we are interested in the steady-state regime,  $t \gg \gamma^{-1}$ . For that case it is possible to neglect the terms with time derivatives in Eqs. (6). Equations (6) are the system of inhomogeneous linear differential equations (LDEs). It is known that the solution of an inhomogeneous LDE can be constructed as the general solution of the respective homogeneous LDE (the complementary function) plus a particular integral of the inhomogeneous LDE. The complementary functions of Eqs. (6) are oscillating-in-time functions, which decay in time as  $\exp(-\gamma t)$ . Hence, we can neglect this part for  $t \gg \gamma^{-1}$ . On the other hand, the solution of Eqs. (6) presented below with neglected time derivatives has particular integrals of Eqs. (6) considered as the set of inhomogeneous LDEs. Namely, those particular integrals describe the steady state that is of interest for our purposes. The total time evolution of the system, described by Eqs. (6), at large times is also determined by the (oscillating and decaying in time) complementary functions, i.e., by Eq. (7) multiplied by  $\exp(-\gamma t)$ . A similar time evolution can be seen, for example, in Ref. [9], where the dynamics of the spin system under the action of the linear polarized laser was studied. One can see below that in the steady-state regime ( $t \gg \gamma^{-1}$ ) there are oscillations of the magnetization with the frequency of the laser field  $\omega$  for nonzero  $S_0^\pm$ . The solutions for the average spin projectors in the steady-state regime can be written as

$$\begin{aligned} \langle S^z \rangle &= S_0^z - \hbar X, \\ \langle \tilde{S}^x \rangle &= \tilde{S}_0^x - \hbar\omega X, \\ \langle \tilde{S}^y \rangle &= \hbar\gamma X, \end{aligned} \quad (8)$$

where

$$X = \frac{\hbar S_0^z + \hbar\omega \tilde{S}_0^x + \hbar\gamma \tilde{S}_0^y}{\hbar^2 + (\hbar\omega)^2 + (\hbar\gamma)^2}. \quad (9)$$

Hence, in the steady-state regime average values of the projectors of spin operators oscillate with the frequency of the circular polarized laser field  $\omega$  [notice that  $\tilde{S}^x \rightarrow S^x \cos(\omega t) + S^y \sin(\omega t)$  and  $\tilde{S}^y \rightarrow S^y \cos(\omega t) - S^x \sin(\omega t)$ , with similar relations for  $\tilde{S}_0^{x,y}$ ]. We see that in the steady-state regime the Rabi oscillations with the frequency  $\Omega$  are damped by the relaxation (see Ref. [9] for the case of the linear polarized laser field), and the average values of spin projections oscillate only with the laser frequency.

Consider the ensemble-averaged value of the projection of the spin, along which the effective magnetic field of the laser  $\hbar\omega$  in the rotating frame is directed,  $\langle S^z \rangle$  (the one studied in Refs. [8,10]). It oscillates with the frequency  $\omega$  about its mean value (the latter can be calculated by averaging  $\langle S^z \rangle$  with respect to time; see above)

$$\langle \tilde{S}^z \rangle = S_0^z \left( 1 - \frac{\hbar^2}{\hbar^2 + (\hbar\omega)^2 + (\hbar\gamma)^2} \right). \quad (10)$$

Figure 1 shows the dependence of such a mean value of the total spin moment of the isotropic system (normalized by  $S_0^z$ )

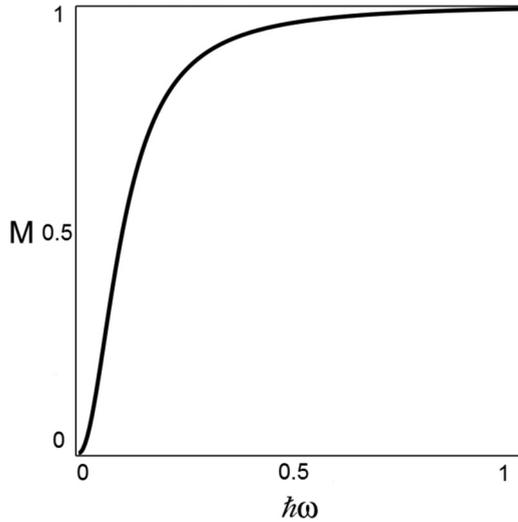


FIG. 1. The normalized average value  $M = \langle \bar{S}^z \rangle / S_0^z$  of the time-averaged projection of the total spin of the system with isotropic spin-spin (ferromagnetic) interactions as a function of the frequency of the applied circular polarized laser field  $\omega$  in the steady-state regime. We used the parameters  $\hbar\gamma = 0.01|J|$  and  $h = 0.1|J|$ . (We normalize all energy values to the value of the isotropic exchange interaction  $|J| = 1$ .)

as a function of the laser frequency in the steady-state regime. The slope of the curve at low frequency is proportional to the power of the laser field (determined by  $h^2$ ), and it depends on the ratio  $(h/\hbar\gamma)^2$ . For very small  $h$  the average value of the total spin moment can be approximated by  $S_0^z$  for most frequencies. Notice that this answer [Eq. (10)] is universal. It does not depend on which kind of isotropic exchange interaction is considered (ferromagnetic, antiferromagnetic, inhomogeneous, spin frustrated), and it is independent of which kinds of low-energy excitations are present in the system (whether they are gapless or gapped). The only necessary condition for the existence of that answer is the absence of the magnetic anisotropy. It is important to notice also that, on the other hand, for the isotropic antiferromagnetic system without external static magnetic field we have  $S_0^{x,y,z} \equiv 0$ , and hence,  $\langle S^z \rangle = 0$  for any values of the frequency and magnitude of the circularly polarized laser field,  $\omega$  and  $h$ . In this sense Fig. 1 is rather related to the spin system with the ferromagnetic isotropic interaction between spins.

#### IV. EFFECT OF THE MAGNETIC ANISOTROPY: FERROMAGNETIC SYSTEMS

Now, let us consider the effect of the nonzero magnetic anisotropy.

Let us first study the steady-state regime of the effect of the circular polarized laser field on the quantum spin system with the ferromagnetic spin-spin interaction. Here it is more convenient to write Heisenberg equations of motion for spin operators and then average them with the density matrix taken at some fixed time. When writing the equations of motion for spin projections, one can see that the situation drastically differs from the isotropic case. Other operators, bilinear in spin-projection operators, e.g.,  $\pm A_\delta(S_{n-\delta}^z S_n^\pm + S_n^\pm S_{n+\delta}^z)$ ,

appear on the right-hand sides of Heisenberg equations of motion for spin projections. Then, to study the dynamics, one needs to write Heisenberg equations of motion for such pair-correlation functions. The equations for the latter will have cubic terms in spin-projection operators on their right-hand sides. The simplest way to take into account such terms is to use the dynamical mean-field-like approximation [9,16]. In that method we truncate the above-mentioned procedure by approximating averages of quadratic forms of spin-projection operators by the products of two averages of spin-projection operators. Then, naturally, one needs to determine self-consistently the values of those averages of spin projections. From now on, for simplicity we consider the case  $S = 1/2$  with  $A_\delta = A$  for fixed  $\delta$  (for example, for the nearest-neighbor coupling in the ferromagnetic chain we have  $\delta = \pm 1$ ; we consider  $A < 0$ , i.e., the easy-axis anisotropy for the ferromagnetic exchange coupling  $J < 0$ ). The generalization for the case of any value of spin  $S$  is straightforward.

After averaging with the density matrix and after the introduction of relaxation into equations of motion, analogous to the isotropic case (6), we obtain the quantum Boltzmann equations,

$$i\hbar \left[ \frac{\partial \langle \tilde{S}^\pm \rangle}{\partial t} + \gamma (\langle \tilde{S}^\pm \rangle - \tilde{S}_0^\pm) \right] = \mp (\hbar\omega - ZA \langle S^z \rangle) \langle \tilde{S}^\pm \rangle \mp h \langle S^z \rangle, \\ i\hbar \left[ \frac{\partial \langle S^z \rangle}{\partial t} + \gamma (\langle S^z \rangle - S_0^z) \right] = -\frac{h}{2} (\langle \tilde{S}^+ \rangle - \langle \tilde{S}^- \rangle), \quad (11)$$

where  $Z$  is the coordination number (for example, for the ferromagnetic spin chain we have  $Z = 2$ ). Now our goal is to look for the steady-state solution of this set of equations for  $t \gg \gamma^{-1}$ . The solution shows that all components of the average values of spin projections oscillate with the frequency of the circularly polarized laser field  $\omega$  (the oscillations with the Rabi frequency  $\Omega$  are damped in the steady-state regime). The averaged in time (mean) value of the projection of the spin along the anisotropy axis is determined from the solution of the self-consistency equation

$$\langle \bar{S}^z \rangle = S_0^z \left( 1 - \frac{h^2}{h^2 + (\hbar\gamma)^2 + (\hbar\omega - ZA \langle \bar{S}^z \rangle)^2} \right). \quad (12)$$

It is the cubic equation with respect to  $\langle \bar{S}^z \rangle$ . It is possible to obtain solutions of that equation in the closed form (e.g., via the modified Cardano's formula). However, the exact analytic form of those solutions is very complicated, and we will not present it here. For simplicity, we present the results of calculations in graphical form for several values of parameters in Fig. 2.

Again, as for the isotropic case, for large values of the laser frequency the mean value of the spin projection tends to  $S_0^z$ . Also at low frequencies the slope of the dependence  $M(\omega)$  is determined by the value of the laser power  $h^2$ . However, the magnetic anisotropy can cause specific features in the frequency dependence of the averaged value of the spin projection in the steady-state regime. Namely, the resonance behavior appears: The frequency dependence of the mean value of the magnetization, caused by the laser, shows the depression related to the resonance  $\hbar\omega = ZA \langle S^z \rangle$  at low values of the laser strength. The resonance is, obviously, caused by the nonzero magnetic anisotropy. It is important

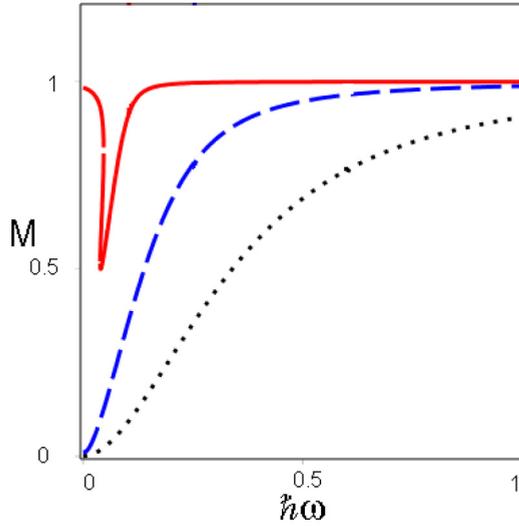


FIG. 2. The solutions of the self-consistency equation for normalized average value  $M = \langle \bar{S}^z \rangle / S_0^z$  of the time-averaged projection of the total spin of the ferromagnetic spin-1/2 chain ( $Z = 2$ ) with anisotropic spin-spin interactions  $A = 0.1|J|$  as a function of the frequency of the applied circular polarized laser field  $\omega$  in the steady-state regime. We used  $\hbar\gamma = 0.01|J|$  and various values of the power of the laser field. The solution for  $h = 0.01|J|$  is shown as the red solid line, the solution for  $h = 0.1|J|$  is shown as the blue dashed line, and the solution for  $h = 0.3|J|$  is shown as the black dotted line.

to point out that in the dynamical regime,  $t < \gamma^{-1}$ , such a resonance behavior yields a small value of the Rabi frequency  $\Omega = \sqrt{(\omega - ZA\langle S^z \rangle / \hbar)^2 + (h/\hbar)^2}$  which in resonance is determined by  $h/\hbar$ . Then, one can see that, depending on the strength of the laser field, several different situations can exist. For small values of  $h$  there can be a transition (jump) between solutions. For higher values of the laser power the jump is transformed into cusps. Figure 3 shows the behavior of  $M(\omega)$  more precisely for small values of  $\omega$  for small  $h$ . Consider the behavior of  $M(\omega)$  according to Fig. 3. At small values of  $\omega$  there are three solutions related to one value of  $\omega$  in the considered range of parameters. To avoid this degeneracy, let us consider how the value of  $M(\omega)$  can be changed when we increase  $\omega$  from 0 to  $0.5|J|$  (as in Fig. 3, exact solutions). First,  $M(\omega)$  decreases smoothly [according to the first root of Eq. (12), shown as the blue long-dashed line in Fig. 3] until some value of  $\omega$ , at which there has to be a jump to the second root (shown as the red solid line in Fig. 3). On the other hand, with  $\omega$  decreasing from  $0.5|J|$  to 0,  $M(\omega)$  decreases smoothly [according to the first and second solutions of Eq. (12), shown by the blue long-dashed and red solid lines in Fig. 3], and then at some other value of  $\omega$  it jumps to larger values, i.e., to the first solution, shown by the blue long-dashed line. Hence, according to this scenario, the value of  $M(\omega)$  is mostly determined by the first and the second roots of Eq. (12) (shown by the blue long-dashed and red solid lines in Fig. 3), and the third root (the green dotted line) presents a nonphysical solution. The real values of  $\omega$ , at which such jumps exist, cannot be determined exactly: They may be determined by additional processes not considered here. The situation is reminiscent of the theory of

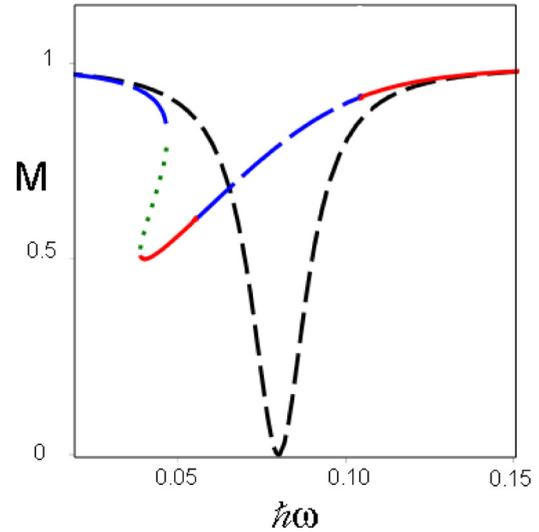


FIG. 3. The blue long-dashed line shows the first solution of the nonlinear equation [Eq. (12)] for  $h = 0.01|J|$  (see Fig. 2). The red solid line shows the second solution, and the green dotted line is related to the third solution. The black dashed line shows the approximate solution calculated according to Eq. (13). Other parameters are the same as in Fig. 2.

the first-order phase transitions, where we need to choose the physical solution between possible roots of the cubic equation.

For comparison in Fig. 3 we also present the behavior of  $M(\omega)$  for the ferromagnetic case for small  $h^2$ , in which region one can write

$$\langle \bar{S}^z \rangle \approx S_0^z \left( 1 - \frac{h^2}{(\hbar\gamma)^2 + (\hbar\omega - ZAS_0^z)^2} \right). \quad (13)$$

In the approximate solution smooth behavior at resonance instead of the jump is manifested. Out of resonance the difference between the exact and approximate solutions (see Fig. 3) is small, while in resonance the difference is large. Notice that in the dynamical regime  $t < \gamma^{-1}$  the mean  $z$  component of the laser-field-induced spin moment formally coincides with Eq. (13) for  $\gamma \rightarrow 0$ .

How can the features related to nonlinear effects be interpreted? To understand their nature, it is instructive to present the self-consistency equation (12) in a more convenient form for analysis (the depressed cubic equation),

$$x^3 + px + q = 0. \quad (14)$$

Here the change in variable (known as the Tschirnhausen transformation)

$$\langle \bar{S}^z \rangle = \frac{2\hbar\omega + ZAS_0^z - 3x}{3ZA} \quad (15)$$

is used. The governing parameters are

$$3q = -2(\hbar\gamma)^2(\hbar\omega - ZAS_0^z) - h^2(2\hbar\omega + ZAS_0^z) + \frac{2(\hbar\omega - ZAS_0^z)^3}{9}, \quad (16)$$

$$p = (\hbar\gamma)^2 + h^2 - \frac{(\hbar\omega - ZAS_0^z)^2}{3}.$$

The discriminant  $\Delta^2 = (p/3)^3 + (q/2)^2$  is nonpositive, so according to the modified Cardano's formula three real solutions exist for the cubic equation (12). For the realistic set of parameters  $0 < S_0^z \leq S$ ,  $|A| \sim 0.1|J|$  and for  $\hbar\gamma \ll |J|, |A|$ , two of those solutions describe mostly physically reasonable situations (see Figs. 2 and 3). The third one shows physically unreasonable behavior (as a function of the frequency and the magnitude of the laser field). That is why we have chosen only those parts of formal solutions for the mean magnetization which describe physically reasonable behavior.

It is known that, depending on the values of governing parameters  $p$  and  $q$ , the solution of the cubic equation can have several features. For the values of parameters  $(p/3)^3 + (q/2)^2 = 0$  the jump takes place between two values of solutions. On the other hand, the jump transforms into the cusp for  $p_c = q_c = 0$ . At  $q = 0$  the behavior of  $\langle \tilde{S}^z \rangle$  is equal to  $(2\hbar\omega/3ZA) + (S_0^z/3) - (p^{1/2}/ZA)$ , and for  $q = 0$  it is  $\langle \tilde{S}^z \rangle = (2\hbar\omega/3ZA) + (S_0^z/3) - (q^{1/3}/ZA)$ . Naturally, the values of exponents belong to the mean-field universality class due to the approach we used. At  $p = q = 0$  we have the triple real root. The nonlinearity is clearly caused by nonzero magnetic anisotropy.

## V. UNIAXIAL ANTIFERROMAGNETIC SYSTEMS

Now, let us consider the antiferromagnetic spin-1/2 system under the action of the circular polarized laser field in the steady-state regime.

Formally, we can first consider the answer presented above for the ferromagnetic system with the magnetic anisotropy. Notice that for the antiferromagnetic case  $J > 0$  we have to consider  $A > 0$  for the case of the easy-axis magnetic anisotropy. However, in the thermal equilibrium, for the antiferromagnetic system without static magnetic field  $H = 0$  one has  $S_0^{x,y,z} = 0$  for  $A \neq 0$ , and hence, the answer for magnetization caused by the circular polarized laser field in the antiferromagnetic spin system in our dynamical mean-field approximation is *zero*. It does not depend on the parameters of the laser field ( $\omega$  and  $h$ ) or on  $\gamma$ .

To obtain a nonzero answer for the laser-induced magnetization for the antiferromagnetic system with the nonzero magnetic anisotropy, we have to use the two-sublattice approximation (see Ref. [16]). Such an approximation takes into account that while we have  $S_0^{x,y,z} = 0$  for the antiferromagnetic system, for some values of parameters the average values of the nearest-neighboring *site* spins are nonzero.

To be concrete, let us consider the spin chain with the nearest- and next-nearest-neighbor interactions (which is equivalent to the zigzag ladder). The Hamiltonian of such a system can be written as

$$\begin{aligned} \mathcal{H}_{\text{af}} = & \sum_n \left[ J(\mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n} + \mathbf{S}_{2,n} \cdot \mathbf{S}_{1,n+1}) \right. \\ & - A(S_{1,n}^z S_{2,n}^z + S_{2,n}^z S_{1,n+1}^z) \\ & \left. + \sum_{j=1,2} (J_N \mathbf{S}_{j,n} \cdot \mathbf{S}_{j,n+1} - A_N S_{j,n}^z S_{j,n+1}^z) \right], \end{aligned} \quad (17)$$

where  $j = 1, 2$  numerate two legs of the spin ladder,  $n$  numerates the position in the leg,  $J > 0$  is the isotropic antiferromagnetic exchange between legs (or nearest-neighbor exchange in the chain), and  $J_N$  is the exchange along legs (or the next-nearest-neighbor exchange along legs).  $A < 0$  and  $A_N < 0$  (the easy-axis case) determine the magnetic anisotropy constants related to  $J$  and  $J_N$ , respectively.

For this case the quantum Boltzmann equations can be written as

$$\begin{aligned} i\hbar \left[ \frac{\partial \langle \tilde{S}_{1,2}^\pm \rangle}{\partial t} + \gamma (\langle \tilde{S}_{1,2}^\pm \rangle - \tilde{S}_{0,1,2}^\pm) \right] \\ = \mp B \langle \tilde{S}_{1,2}^\pm \rangle \pm 2J \langle S_{1,2}^z \rangle \langle \tilde{S}_{2,1}^\pm \rangle \mp h \langle S_{1,2}^z \rangle, \\ i\hbar \left[ \frac{\partial \langle S_{1,2}^z \rangle}{\partial t} + \gamma (\langle S_{1,2}^z \rangle - S_{0,1,2}^z) \right] = -\frac{h}{2} (\langle \tilde{S}_{1,2}^+ \rangle - \langle \tilde{S}_{1,2}^- \rangle), \end{aligned} \quad (18)$$

where  $B = \hbar\omega + 2A_N \langle S_{1,2}^z \rangle + 2(J + A) \langle S_{2,1}^z \rangle$ . Here  $S_{1,2}^{\pm,z}$  are the projections of the spin in each elementary cell of the antiferromagnetic spin-1/2 chain (rung of the ladder). Equations (18) do not contain the contribution from  $J_N$ . First, it is caused by the application of the two-sublattice approximation (in fact, the terms proportional to  $J$  appear because we artificially divide our system into two sublattices). Second, the contribution from the terms proportional to  $J_N$  produces higher-order corrections in  $h$  to the laser-induced magnetization (see Ref. [17]).

Again, the analysis of the solution of Eqs. (18) in the steady-state regime  $t \gg \gamma^{-1}$  reveals that the average values of the spin projections of the antiferromagnetic chain oscillate with the frequency of the circular polarized laser field  $\omega$  (Rabi oscillations are smeared out by the relaxation). The average-in-time value of the average spin projection  $\langle S^z \rangle \equiv \langle (S_1^z + S_2^z) \rangle / 2$  of the antiferromagnetic chain can be found as the solution of the self-consistency equations

$$\begin{aligned} \langle S^z \rangle = & \frac{(S_{0,1}^z + S_{0,2}^z)}{2} - \frac{h^2}{2} \\ & \times \left[ \frac{F_1}{(\hbar\gamma)^2 + (\hbar\omega - x_1)^2} + \frac{F_2}{(\hbar\gamma)^2 + (\hbar\omega - x_2)^2} \right], \\ S_a = & \frac{(S_{0,1}^z - S_{0,2}^z)}{2} - \{ [\hbar\omega + 4(J + A) \langle S^z \rangle]^2 \\ & - 2(J + |A - A_N|)^2 (\langle S^z \rangle^2 - S_a^2) \} \\ & \times \frac{h^2}{[(\hbar\gamma)^2 + (\hbar\omega - x_1)^2][(\hbar\gamma)^2 + (\hbar\omega - x_2)^2]}, \end{aligned} \quad (19)$$

where

$$x_{1,2} = 2(J + A) \langle S^z \rangle \mp F, \quad (20)$$

with

$$F = 2\sqrt{(J - A_N)^2 \langle S^z \rangle^2 + |A - A_N| (2J + |A - A_N|) S_a^2} \quad (21)$$

and with  $S_a = \langle S_1^z - S_2^z \rangle / 2$  and, finally,

$$F_{1,2} = \frac{\langle S^z \rangle}{2} \pm \frac{(J - A_N) \langle S^z \rangle^2 - |A - A_N| S_a^2}{F}. \quad (22)$$

Unfortunately, it is impossible to solve Eqs. (19) in the closed form for any magnitude of the laser field  $h$ . However, we

can obtain the approximate solution. For the antiferromagnetic system with the easy-axis magnetic anisotropy it is the most natural to study the situation in which  $S_{0,1}^z \approx -S_{0,2}^z \equiv S_0$  [18]. The largest value for  $S_0$  is expected for Ising-like antiferromagnetic chains. Consider the small- $h$  regime, which is natural for the laser excitation. In this regime we get

$$S_a \approx S_0 - \frac{2h^2 S_0^2 (J + |A - A_N|)}{\Delta}, \quad (23)$$

$$\langle S^z \rangle = -\frac{h^2 \hbar \omega |A - A_N| S_0^2}{\Delta},$$

where

$$\Delta = [(\hbar\gamma)^2 + (\hbar\omega - F)^2][(\hbar\gamma)^2 + (\hbar\omega + F)^2]. \quad (24)$$

Neglecting the  $h$  dependence of  $S_a$ , i.e.,  $S_a \approx S_0$ , we finally obtain for the averaged-in-time  $z$  component of the spin moment (related to the magnetization) of the antiferromagnetic chain under the action of the circular polarized laser field in the steady-state regime  $\langle S^z \rangle \approx -[h^2 S_0^2 \hbar \omega |A - A_N| / \Delta]$ , where in the expression for  $\Delta$  we use  $F = 2S_0 \sqrt{|A - A_N|(2J + |A - A_N|)}$ . The temperature dependence in our approach comes from  $S_0(T)$  (see above). Obviously, the value of  $S_0$  is maximal at  $T = 0$  and decreases with the growth of temperature; hence, we expect the maximum of the effect at low temperatures.

Figure 4 presents the mean value of the normalized  $z$  component of the spin moment of the antiferromagnetic chain (ladder) as a function of the laser frequency. One clearly sees the main features characteristic of the steady-state regime of the response of any quantum spin system to the circularly polarized laser field. Namely, the value  $M = \langle S^z \rangle / S_0$  is approximately zero away from the resonance frequency. Also, the response to the laser field clearly has a resonance character, and the resonance is determined by the nonzero magnetic

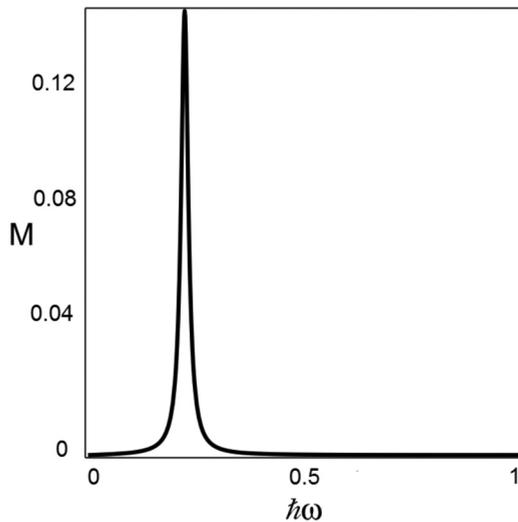


FIG. 4. The normalized time-averaged value  $M = \langle S^z \rangle / S_0$  of the projection of the total spin of the antiferromagnetic spin-1/2 chain (spin ladder) with anisotropic spin-spin interactions as a function of the frequency of the applied circular polarized laser field  $\omega$  in the steady-state regime. We used  $\hbar\gamma = 0.01|J|$ ,  $h = 0.01|J|$ ,  $S_0 = 0.4$ ,  $J = 1$ , and  $(A - A_N) = -0.1|J|$ .

anisotropy. Again, we can point out that in the dynamical regime  $t < \gamma^{-1}$  a nonzero anisotropy causes Rabi oscillations (which are damped in the steady-state regime) with the small Rabi frequency  $\Omega_1 = \sqrt{(\omega - F/2\hbar)^2 + (h/\hbar)^2}$ ; in resonance the latter is approximately determined by the magnitude of the laser field  $h/\hbar$ . Notice that there is another Rabi frequency for the antiferromagnetic case,  $\Omega_2 = \sqrt{(\omega + F/2\hbar)^2 + (h/\hbar)^2}$ , but that frequency is large compared to  $\Omega_1$ . However, differences exist when making a comparison to the ferromagnetic case. The resonance frequency for the antiferromagnetic case is determined not only by the magnetic anisotropy itself, like in the ferromagnetic case, but by the square root of the product of the anisotropy and the exchange plus anisotropy. Such a dependence is characteristic of the easy-axis magnetic systems [17]. Obviously, for zero magnetic anisotropy the obtained results coincide with the previously considered isotropic case for small  $h$  values. The strongest effects will be manifested in Ising-like spin-1/2 antiferromagnetic chains. The Ising case can be trivially obtained by taking the limit  $J = J_N \rightarrow 0$ . It turns out that the absolute value of the laser-induced steady-state magnetization even in resonance is smaller for the antiferromagnetic case than for the ferromagnetic one. This fact is caused by the difference in the nature of the interactions between spins: The ferromagnetic interaction obviously enhances the total magnetization (even for the laser-induced situation), while the antiferromagnetic interaction tends to the minimization of the total magnetization. Formally, the difference between resonance responses can be seen from the comparison of Eqs. (13) and (19). Namely, for the antiferromagnetic case two resonance terms partly cancel each other's effect on the total magnetization of the system. The case of the simple (nonzigzag) antiferromagnetic spin chain in the two-sublattice approximation can be formally considered by taking the limit  $J_N = A_N \rightarrow 0$ .

The effect of the spin frustration caused by next-nearest-neighbor interactions is in the renormalization of the value of the effective anisotropy. In particular, the resonance frequency tends to zero for  $A_N = A$ . Also, the time-averaged (mean) value of the magnetization of the antiferromagnetic chain as a response to the circularly polarized laser field can be zero for  $A_N = A$  in the steady-state regime. As for nonlinear effects, we expect ones similar to those for the ferromagnetic situation. However, these effects will be weaker because in the antiferromagnetic case resonance and antiresonance terms (with  $x_{1,2}$ ) partly cancel each other out (see above).

We can also consider the situation with the easy-plane magnetic anisotropy, which implies opposite signs for  $A$  and  $A_N$  in Eqs. (17) and (18). Here it is clear that  $S_{0,1,2}^z = 0$ . Hence, for the easy-plane antiferromagnetic systems the laser-induced magnetization can be related only with nonzero  $S_{0,1,2}^\pm(T)$  (which are expected to be maximal in the ground state). However, our analysis of the solutions of the self-consistency equations shows that in this case for the circularly polarized laser field we get  $\langle S^z \rangle = 0$  in the steady-state regime.

Our results can be generalized for the case of the Dzyaloshinskii-Moriya (DM) interactions present in the system. Here we consider the case in which the Dzyaloshinskii vector is directed along the  $z$  axis; that is, the DM coupling distinguishes the same axis as the magnetic anisotropy. In this

case we have to add to the Hamiltonian of the antiferromagnetic chain the term

$$\mathcal{H}_{\text{DM}} = \sum_n \left[ D(S_{1,n}^x S_{2,n}^y - S_{1,n}^y S_{2,n}^x + S_{2,n}^x S_{1,n+1}^y - S_{2,n}^y S_{1,n+1}^x) + D_N \sum_{j=1,2} (S_{j,n}^x S_{j,n+1}^y - S_{j,n}^y S_{j,n+1}^x) \right], \quad (25)$$

with obvious notations. The effect of the DM terms is in the introduction of the replacements  $A \rightarrow A - \sqrt{D^2 + J^2} + J$  and  $A_N \rightarrow A_N - \sqrt{D_N^2 + J_N^2} + J_N$  in the equations for the response of the antiferromagnetic spin-1/2 chain to the circularly polarized laser field in the steady-state regime. Notice that we do not consider large DM constants here, so that the above combinations remain positive.

Finally, let us consider the situation in which the direction of the anisotropy (or the Dzyaloshinskii vector) does not coincide with the direction of the laser field. Here, for example, the shift of the resonance has to be multiplied by  $1 - 3 \cos^2 \theta$ , where  $\theta$  is the angle between the direction of the laser field and the axis, distinguished by the anisotropy. Other results seem to be generic because the contribution of the nonresonance terms is small in the steady-state regime. However, there is a possibility of parametriclike resonances, which will manifest properties of the response to the circular polarized laser field similar to the ones for the linear polarized laser (see Ref. [9]).

## VI. SUMMARY

In summary, the effect of the circular polarized laser field on several quantum spin systems in the steady-state regime, in which relaxation plays the central role, has been studied. The steady-state regime occurs at large time scales for  $t \gg \gamma^{-1}$ . Our dynamical mean-field-like theory predicts several general results for the behavior of the time-averaged (mean) magnetization (the spin moment multiplied by  $g\mu_B$ ) caused by the circular polarized laser field. First, the induced magnetization oscillates with the frequency of the laser field (while Rabi-like oscillations, which modulate the latter in the dynamical regime for  $t \ll \gamma^{-1}$ , are damped). Second, at high frequencies, that magnetization is determined by the value to which the relaxation process is directed. Third, at low frequencies the slope of that magnetization as a function of the frequency is determined by the strength of the laser field. The anisotropy determines the resonance behavior of the time-averaged magnetization for both the ferromagnetic

and antiferromagnetic spin-spin interactions with the nonzero magnetic anisotropy. The strongest effects have to exist in Ising-like spin-1/2 antiferromagnetic chains [19]. Nonlinear effects (in the magnitude of the laser field  $h$ ) can cause jumps and cusps in the behavior of the steady-state value of the time-averaged magnetization due to the circular polarized laser field. Such nonlinear effects are stronger for ferromagnetic interactions and weaker for the antiferromagnetic case. The resonance frequencies are determined by the magnetic anisotropy in the ferromagnetic case and by the square root of the product of the magnetic anisotropy and the exchange coupling plus anisotropy for the antiferromagnetic case. The effects of the Dzyaloshinskii-Moriya interaction, the spin-frustration couplings (in the case of the zigzag spin chain), and the influence of the tilting of the direction of the laser field with respect to the axis of the magnetic anisotropy (or Dzyaloshinskii vector) have been analyzed.

In conclusion, the effect of the circularly polarized laser field in the steady-state regime on quantum spin systems is maximal in resonance, where the time-averaged magnetization, caused by the laser field, is changed essentially. Out of resonance the changes in the magnetization are relatively small. The resonance effect is caused by the nonzero magnetic anisotropy. The resonance frequency is relatively small (proportional to the anisotropy value) for spin systems with ferromagnetic interactions, and it is enhanced in spin systems with antiferromagnetic couplings. The effective Hamiltonian of the spin system under the action of the circularly polarized laser field in a rotating frame determines only the dynamics of spins, not the thermodynamics. The action of the circular polarized laser field drastically differs from that of the static magnetic field. The latter changes thermodynamic properties of the system together with the dynamics. Hence, to observe the maximal laser-induced magnetization we suggest studying quantum spin systems in the steady-state regime in easy-axis antiferromagnetic or ferromagnetic spin systems, like Ising antiferromagnetic spin-chain materials [19] in resonance. The resonance frequency is high (which is important for experiments with lasers) for the antiferromagnetic spin systems. The obtained results are generic and can be compared with the data of possible experiments on the laser field effect on quantum spin systems in the steady-state regime.

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