Formation of phases with inverted spin orientation in magnetic crystals at rapid cooling

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Phase transitions of a specific type in a magnetic crystal that take the form of the appearance of regions where the direction of the magnetization is opposite to that of the rest of the bulk are studied. The phase transition arises because of the strong exchange interaction between spins in the magnetic crystal upon meeting the conditions of (1) the presence of external pumping, which generates an excess of magnons as compared with their equilibrium number, and (2) the contribution of diffusion processes into the relaxation processes of the magnetization (nonlocal relaxation). The processes of formation of the phases with the inverse direction of magnetization intensify upon rapid cooling of the sample. It is shown that, for a certain set of the parameters, such phases can arise at rapid cooling even in the absence of the magnon pumping.

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I. INTRODUCTION

Nonequilibrium nonlinear systems are characterized by a multitude of processes occurring within them. Among them, a special place is occupied by processes in which the properties of the system change abruptly at a certain value of a changing external parameter [1,2]. Such transformations of systems are called *kinetic phase transitions*. Kinetic phase transitions are extremely diverse. Often, they are implemented due to new interactions that arise in nonequilibrium conditions and disappear when the system comes to equilibrium. However, there are specific nonequilibrium phase transitions like classical phase transitions but occurring in a system of particles with a finite lifetime. If the particles interact with each other, they may form different phases. With a finite lifetime of particles, the existence of these phases is possible if there is an external source for the appearance of new particles instead of disappearing ones. Therefore, such a system is in nonequilibrium. In condensed media, such particles are excited states that can move in the medium and consist of many elementary particles. They are called *quasiparticles*. Examples of such particles (quasiparticles) are excitons, electron-hole pairs in semiconductors, radiation defects (for example, vacancies and interstitial atoms), and magnons in magnets. Systems with such particles exist in the presence of an external source that creates them and disappear due to the finite lifetime of the particles when the source is turned off. In the presence of attraction between particles, they form phases whose properties can differ greatly from the properties of phase transitions in a system of particles with an infinite lifetime, studied in the problem of phase transitions in a system of stable particles. The study of nonequilibrium phase transitions is relevant since it provides information about the excited states of the system and gives new nontrivial effects that can be of applied importance and that can be controlled by external influences.

This paper is devoted to the study of the appearance and effects of manifestation of structures in a nonequilibrium system of magnons. Many nonlinear phenomena have been discovered in a system of magnon quasiparticles in magnetic materials using pumping [3–5]. In Ref. [6], experimental data of the spectra study in yttrium iron garnet by the Brillouin light scattering (BLS) method, when magnons were pumped by an alternating longitudinal magnetic field, were interpreted as the observation of Bose-Einstein condensations (BECs) of magnons at room temperature. The main argument for this was the observation of a singularity in the population of the region of states by magnons corresponding to the energy minimum in the magnon spectrum. Subsequently, other interesting facts were discovered: observation of spontaneous coherence and saturation in BEC [7], the observation of a periodic distribution of magnon structures in space [8], and the appearance of a sharp jump in the BLS intensity when pumping was turned off [9].

There were some works with another interpretation of these results about accumulation of magnons in the lowest state [10,11]. Authors Ref. [10] analyzed the kinetics of quasiparticles introduced into the system along with the presence of quasiparticles excited under equilibrium conditions and obtained the conditions under which BEC can occur at any sufficiently high temperature. From calculation of authors of Ref. [11] follows that condensation of a magnon may be described using the Rayleigh-Jeans statistic. In Ref. [12], the experimental results obtained in Refs. [6–9] were explained as manifestations of the effects of self-organization and the appearance of spatial structures in a system of high-density magnons, not as a phenomenon of BEC. During this interpretation, the author relied the explanation of various effects in spectra in double quantum wells based on an AlGaAs crystal with high exciton density. Experimental results, obtained by Timofeev [13] and Butov *et al.* [14] were interpretated in Refs. [15–18] not by the BEC of excitons, but by the formation of specific structures of exciton condensed phases in quantum wells. In Ref. [12], when explaining the experiments [6–9], it was assumed that there is an attraction

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interaction between magnons. As a result, in a magnetic crystal supersaturated with magnons, they tend to accumulate in certain spatial regions, forming phases like phase formation in the solutions supersaturated by some component [19,20]. The direction of the magnetic moment in phases is opposite to the direction of the magnetic field. The question of the interaction between magnons is complicated due to the collective nature of the excitations. The magnon energy is formed by various interactions: exchange, magnetic dipole-dipole, spin-orbital interactions, and interaction with an external magnetic field. At small concentration, the interaction energy of two magnons, depending on the system, may have a different sign. However, at high concentrations, according to Ref. [21], an attraction interaction between magnons dominates, but magnons are poorly determined quantum states when their concentration is high. At room and higher temperature, the number of flipped spines consists of more than several tens of percent, and the concept of magnons as elementary excitations cannot describe the system. The purpose of this paper is to study the spatial distribution of magnetization in a wide range of temperatures. During the investigated rapid cooling processes, temperature is changed from temperature close to phase transition value until room temperature. All spin excitations give a contribution to a formation of the magnetization. According to Ref. [22], the concept of magnons can be used up to the phase transition temperature but only for magnons with small wave vectors. However, few such magnons in comparison with all inverted spins do not contribute to the static magnetization of magnetic crystal. Therefore, in Ref. [12] and here, the system is described in terms of the magnetization as an order parameter obeying the Landau-Lifshitz (LL) equation, and the interaction between spins is already included in this equation in an effective magnetic field and is not considered explicitly. An important point in this paper is to consider the diffusion motion of spins at a high concentration of spin excitations. The presence of the spin diffusion leads to an appearance of the additional relaxation term in the basic equation. It is this type of term that is important in the phenomenological theory of phase transitions [19,20] and leads to the appearance of inhomogeneous solutions in the equations for the magnetization and to the appearance of new phases in magnetic solid when the magnetization deviates from the equilibrium value due to the presence of pumping. The presence of inhomogeneous structures should influence different processes in magnetic solids, particularly BLS. The solutions of the equation for the spatial distribution of magnetization, obtained considering the influence of pumping and spin diffusion, can afterwards be used to determine the spectrum of low-lying spin excitations, and magnons are between them.

Authors of experimental papers [23–26] that appeared recently have reported studies of yttrium iron garnet in which the effects observed in Refs. [6–9] and explained as a manifestation of magnon BEC were more pronounced upon rapid cooling of the sample. In this paper, the studies of Ref. [12] are extended to include the processes occurring during rapid cooling of the sample. It is shown that the appearance of inhomogeneous magnetic structures with flipped spin orientation in the magnetic crystal is enhanced by rapid cooling, and the effects observed in Refs. [23–26] can be explained without involving the BEC of magnons. The possibility of a creation of phases with inverse spins upon rapid cooling in the case of absence of magnon pumping is investigated.

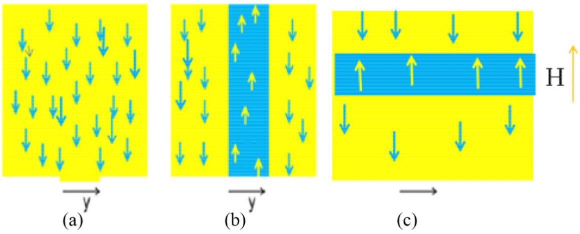

FIG. 1. The distribution of magnon magnetic moments in a crystal: (a) Distributions of moments of excited spins in the case without structure formation. (b) and (c) Distribution of moments in the case of formation of two types of structures. The continuous regions (light and dark) present the areas with most lattice cell moments oriented along the field (light) and opposite to the field (dark). The individual lattice cell moments are not shown because there are too many of them. (a) presents the uniform distribution of magnon moments. (b) and (c) show an alternative distribution of the magnetization, in which the states with a domain (the dark strips) have moments of the lattice cells oriented opposite to the magnetic field arises. The light arrows in the dark region show the magnetic moments of the magnons in the domain.

II. MODEL OF INVESTIGATED SYSTEM

In this paper, a ferromagnetic plate is studied in a constant magnetic field with a strength **H** parallel to the surface of the plate with a direction along the axis z in the presence of an external source of magnons in the sample. One of the sources of an additional amount of magnons, in comparison with its equilibrium value, is a variable longitudinal magnetic field (parametric pumping). The presence of additional magnons leads to a decrease in magnetization of the sample. Various types of instabilities may arise in a nonequilibrium system. In this paper, we are interested in the possibility of spontaneous appearance of spatial inhomogeneities in the distribution of magnetization and the appearance of regions (phases) in the sample with different orientations of the magnetic moments of atoms, caused by the redistribution of magnons in space due to their interaction and motion. In a nonequilibrium system of a ferromagnet under pumping conditions, magnons can form spatially inhomogeneous structures of various shapes. In view of the complexity of the equation for magnetization, we will consider the simplest inhomogeneous structures. The domain is not only the simplest object to consider but also often realized in experiments and satisfies the symmetry of the form of the slab of the magnet. In this case, the problem can be reduced to a one-dimensional problem. In this paper, a structure in the form of a domain was studied, and magnetization of the domain is antiparallel to the magnetic field (Fig. 1).

We proceed from the equation for the change in time of the magnetic moment **M** per unit volume:

$$\left(\frac{\partial \mathbf{M}}{\partial t}\right) = -\gamma[\mathbf{M}, \mathbf{H}_{\rm eff}] + \mathbf{R} + \mathbf{P}, \quad (1)$$

where γ is the gyromagnetic ratio, the effective magnetic field $\mathbf{H}_{\rm eff}$ is determined by the relation:

$$\mathbf{H}_{\rm eff} = -\frac{\delta F}{\delta \mathbf{M}}, \quad (2)$$

and F is the density of free energy of the ferromagnet, which is

$$F = F_{\text{exch}} + \frac{K}{2}(\nabla \mathbf{M})^2 - \mathbf{M} \cdot \mathbf{H} - \frac{1}{2}\mathbf{M} \cdot \mathbf{H}^{(m)}. \tag{3}$$

The first two terms in the right side of this formula describe the uniform and nonuniform parts of the exchange interaction contribution in the free energy. In this paper, we consider states with a strong deviation of the magnetization from the equilibrium value and in a wide temperature range. In such states, the system has many inverted spins and cannot be described by the introduction of many magnons due to their interaction and the formation of various multimagnon complexes. Therefore, to obtain the free energy of the magnetic system, we use the formula obtained from the Heisenberg Hamiltonian for the interaction between spins in the molecular field approximation, which makes it possible to describe the temperature dependence of the magnetization in the entire temperature range, except for a narrow region near the phase transition temperature, which is insignificant for this paper. In the Heisenberg expression and in mean field approximation, we have

$$F_{\text{exch}} = N\left\{\frac{T_c M^2}{2M_s^2} - T\ln\left[2\cosh\left(\frac{MT_c}{M_s T}\right)\right]\right\}, \tag{4}$$

where T is the temperature of the system, expressed in energy units, T_c is the ferromagnet-paramagnet phase transition temperature, N is the number of the crystal cells in a unit volume, $M_s = N\mu_o$ is the saturation magnetic moment per unit volume, and μ_o is the magnetic moment of spin. Note that Eq. (4) holds for a simple model of magnetic crystals with a fixed value of the magnetic moment in the cell, i.e., for a ferromagnet. However, for low-energy modes and at not-too-high magnetic fields, the magnetic moments of atoms of different sublattices rotate around the magnetization vector, which is conserved, and therefore, the physical manifestations of a ferromagnet and a ferrimagnet coincide [27].

The second term in Eq. (3) describes the energy of the inhomogeneity of the exchange interaction. The last term in Eq. (3) is the energy of the interaction of the magnetic moment with the magnetic field $\mathbf{H}^{(m)}$ created by the magnetic crystal. For a large-sized slab, the magnetic field created by the inclusion is zero $\mathbf{H}^{(m)} = 0$. Also, M_z depends on y [$M_z = M_z(y)$] for the domain orientation in Fig. 1(b) and depends on z [$M_z = M_z(z)$] for domain orientation in Fig. 1(c).

The vector $\mathbf{R}$ in Eq. (1) is the relaxation term defined by the formula:

$$\mathbf{R} = -\gamma_{R1}[\mathbf{n_M}, (\mathbf{n_M}, \mathbf{H}_{\text{eff}})] + \gamma_R \mathbf{H}_{\text{eff}} + \left(\frac{\partial \mathbf{M}}{\partial t}\right)_D, \tag{5}$$

where $n_{\mathbf{M}} = \mathbf{M}/M$. The first term in Eq. (5) describes the relaxation in the LL equation. This term is equivalent to the relaxation term of the Landau-Lifshitz-Gilbert equation. The conservation of the absolute value of the magnetization holds with both types of relaxation terms, and neither term can describe the longitudinal dynamics of the magnetization.

The second and the third terms in Eq. (5) are important for the description of the evolution of the nonuniform changes of the longitudinal magnetization at pumping, which is studied in this paper. The second term is given in the main equations of Ref. [28]. The expression for the third term was obtained in Ref. [12] for the diffusion motions of magnons at a low concentration of these quasiparticles, when they are good quantum numbers. It can also be derived for a high concentration of flipped spins in the case of the hopping mechanism of spin motion. For the domain orientation presented in Fig. 1(b), when normal to the domain surface is oriented along axis y, the time derivative of magnetization takes the following form:

$$\left(\frac{\partial M_z}{\partial t}\right)_D = -\frac{2DM_s\mu_o}{T}\left(1 - M^2/M_s^2\right)\frac{\partial^2 H_{\text{eff}}}{\partial y^2}, \tag{6}$$

where D is the spin-diffusion coefficient. In the case of an isotropic crystal, equation (6) differs only by the immaterial coefficient from the formula for nonlocal relaxation obtained by Baryakhtar [29] based on the crystal symmetry and the Onsager principle (see formula 1.5 in [30]). According to the formula for Baryakhtar's relaxation term, the change in magnetization with time is proportional to the second derivative of effective magnetic field with respect to the coordinate. This term was used for consideration of different processes in magnetic crystals [29–35]. In Ref. [12], it was applied to the consideration of a phase transition in nonequilibrium magnets. Such a term, proportional to the second derivative with respect to the coordinate, is important in the phenomenological description of phase transitions [19]. It is the inclusion of this type of dependence in the equation of motion for the magnetization in Ref. [12] that led to the buildup of a phase with the inverse spin orientation. The magnetism equation with an inhomogeneous relaxation term proportional to the second coordinate derivative of the effective magnetic field is called the Landau-Lifshitz-Baryakhtar equation.

The third term in Eq. (1) determines the pumping-induced change of the magnetization of the unit volume of the sample in unit time. This term is responsible for the spatial redistribution of magnons. We shall not consider the processes of the magnon creation by the external microwave field and shall make only some approximation for $\mathbf{P}$. In our consideration, the pumping is some mechanism that reverses the spin orientation. The magnons created by the pumping come very quickly to the quasiequilibrium state due to the magnon-magnon and magnon-phonon interactions. The pumping, creating magnons, causes the decrease of the magnetization. An additional magnetization appears in the system, directed opposite to the magnetic field. Thus, the value of $\mathbf{P}$ should be negative with respect to the crystal magnetization and positive in the regions where the magnetization changes sign. Also, it should be equal to zero at the point where $M_z = 0$. We shall approximate the pumping in a simplified form presented by the formula:

$$P_z = -G_o M_z, \tag{7}$$

where G_o is a positive phenomenological parameter. Its value may be obtained from the experimental decrease of the magnetization due to the pumping. We performed calculations for other approximations of the pump value P (for example, for a value of P independent of M). The pumping was chosen such that it led to a decrease in the magnetization of the medium. In this case, the calculation results give the qualitative behavior of the system, the same as that obtained when Eq. (7) is used for pumping.

As in the Ref. [12], we neglect the anisotropy energy of the magnetic interaction.

Considering the orientation of the magnetic field and effective magnetic field along the z axis and introducing dimensionless variables, we obtain from Eqs. (1)–(7) the following equation for the magnetization:

$$\frac{\partial \tilde{M}_z}{\partial \tilde{t}} = (1-\tilde{M}^2)\frac{\partial^2}{\partial \tilde{y}^2}\left(\tilde{M}_z - th(\tilde{M}_z T_c/T) - \tilde{H} - \frac{\partial^2 \tilde{M}_z}{\partial \tilde{y}^2}\right) - q\left(\tilde{M}_z - th(\tilde{M}_z T_c/T) - \tilde{H} - \frac{\partial^2 \tilde{M}_z}{\partial \tilde{y}^2}\right) - \tilde{G}_o \tilde{M}_z, \quad (8)$$

where

$$\tilde{M}_z = \frac{M_z}{M_s}, \quad \tilde{T} = \frac{T}{T_c}, \quad \tilde{H} = \frac{\mu_o H}{T_c}, \quad \Gamma_D = \frac{2T_c D}{d^2 T},$$
$$t = \frac{\tilde{t}}{\Gamma_D}, \quad q = \gamma_R \frac{\gamma_R T d^2}{2\mu_o M_s D}, \quad \tilde{G}_0 = \frac{G_o}{\Gamma_D}. \quad (9)$$

Let us give estimates of the coefficients of Eq. (8). For the following values of the crystal parameters:

$$\gamma_R = 10^6\ s^{-1}, \quad T_c = 560\,\mathrm{K}, \quad d = 3\times 10^{-8}\,\mathrm{cm},$$
$$M_s = 140\,\mathrm{Gs}, \quad H = 1000\,\mathrm{Oe}, \quad T = 300\,\mathrm{K}, \quad (10)$$
$$D = 10\,\mathrm{cm}^2/\mathrm{s},$$

quantities Γ_D, q, and H have the following values $\Gamma_D \sim 4.14\times 10^{16}\,\mathrm{s}^{-1}$, $q = 1.44\times 10^{-6}$, $\tilde{H} = 0.00012$.

III. STABILITY OF UNIFORM SOLUTION

The stationary homogeneous value of $\tilde{M}_z^o$ stems from the solution of Eq. (8):

$$q\left[\tilde{M}_z^o - th\left(\frac{\tilde{M}_z^o T_c}{T}\right) - \tilde{H}\right] - G_o \tilde{M}_z^o = 0. \quad (11)$$

Substituting this stationary value of the magnetic moment into Eq. (8) and adding a fluctuation in the form $\tilde{M}_z = \tilde{M}_z^o + m\exp[i\tilde{k}\tilde{y} + \lambda(\tilde{k})t]$, we obtain the equation for the damping increment $\lambda(\tilde{k})$:

$$\lambda(\tilde{k}) = (\tilde{k}^2 + \tilde{\gamma}_R)\left[\frac{1}{\tilde{T}}\mathrm{sech}^2\left(\frac{\tilde{M}_z^o}{\tilde{T}}\right) - \tilde{k}^2 - 1\right] - \tilde{G}_o. \quad (12)$$

The calculations of Fig. 2 were carried out for the values of the parameters of the system given by Eq. (10). As the figure illustrates that the value of $\lambda(\tilde{k})$ becomes positive at some $\tilde{k}$, and this means that the solution with a uniform distribution of magnetization becomes unstable, a periodic distribution of magnetization with a wave vector $\tilde{k}$ appears in the system. The dimensionless value of magnetization at this pumping $G_o = 7.843\times 10^{-7}$ is equal to $\tilde{M}_{zc}^o = 0.44485$ (Fig. 3). This value is much less than the saturation magnetization at room temperature ($\tilde{M}_z^o = \tilde{M}_s^o \sim 1$), and it is hardly achievable with any of the various methods of magnon pumping. However, instability can manifest itself, as we will see, during rapid cooling of the sample.

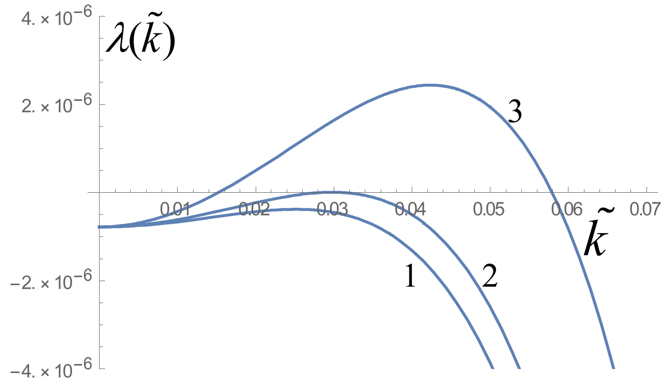

FIG. 2. The dependences $\lambda(\tilde{k})$ on $\tilde{k}$ for the three values of the pump parameters G_o: (1) 7.839×10^{-7}, (2) 7.843×10^{-7}, and (3) 7.855×10^{-7}.

The found boundary of stability of the homogeneous state lies very close to the spinodal curve of the magnetic state. A very small shift from the spinodal is caused by the nonequilibrium pumping term in the Eq. (8), and the shift is not seen in Fig. 3. The spinodal curve is determined by the relation $\frac{\partial^2 F}{\partial M^2} = 0$, where the magnetization M is the order parameter of the system. At such a condition, the uniform solution loses stability [19]. Figure 3 shows the dependences of the spinodal curve on temperature (solid curve 3) and magnetization in the equilibrium state (solid curve 1) obtained using Eqs. (3), (4), and (11) at $\tilde{G}_o = 0$ and taken, for example, in the nonequilibrium state at $\tilde{G}_o = 3\times 10^{-8}$ (dashed curve 2 in Fig. 3); choosing $\tilde{G}_o$ as such, the decrease in magnetization is 2.5% of the magnetization of the sample at $\tilde{T} = 0.5357$.

Nonequilibrium states with magnetization located between curves 1 and 3 (between spinodal and binodal) curves created by magnon pumping can be both spatially homogeneous and inhomogeneous. The latter can be formed in the presence

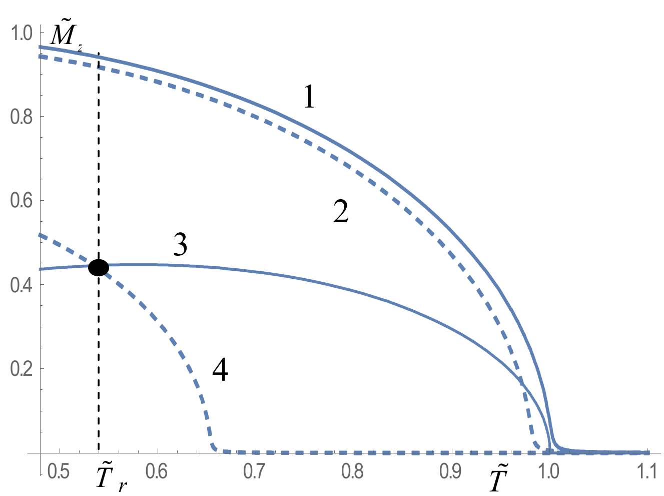

FIG. 3. Temperature dependence of the magnetization (1) in the equilibrium state, (3) in the spinodal curve, and (2) and (4) in the presence of magnon pumping. The black dot shows the point where the homogeneous state loses stability at room temperature at $G_o = 7.843\times 10^{-7}$ according Fig. 2.

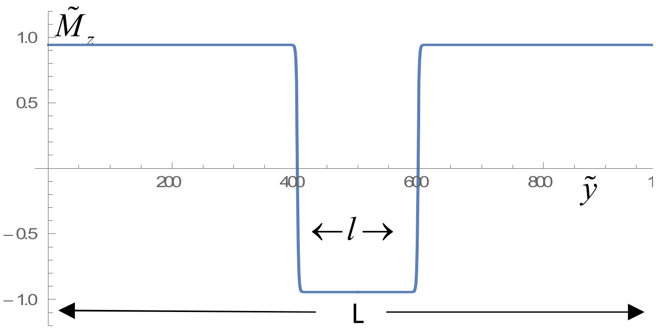

FIG. 4. Spatial distribution of magnetization in a domain-shaped phase. The OZ axis is directed perpendicular to the figure. The parameters are represented by Eq. (10).

of fluctuations. The homogeneous states with magnetization below the spinodal are unstable.

IV. STATIONARY STRUCTURES IN MAGNETIC CRYSTAL UNDER MAGNON PUMPING

Domain-shaped inhomogeneous structures (phases) in a ferromagnet at the magnon pumping were considered in Ref. [12] for the temperatures close to the phase transition temperature.

Calculations for the arbitrary range of temperatures but in the approximation of a self-consistent field for a ferromagnet [with the exchange energy presented in Eq. (4)] give qualitatively similar results. Therefore, we only provide a few examples.

According to the calculations presented in the previous section, in a magnetic crystal, within a wide range of the magnon pumping value, two types of magnetization states can occur, either homogeneous or inhomogeneous. Inhomogeneous structures develop in the presence of fluctuations exceeding a certain magnitude. Fluctuations can be of thermodynamic or imperfect origin. In this paper, spatially inhomogeneous stationary solutions were obtained by the method of initial fluctuations which is described in Ref. [12]. The stable solutions were found from the numerical solution of the evolution equation [here, Eq. (8)] using initial conditions in the shape of the probing profiles having forms close to the required structure form. If the solution converges—that is, ceases to change with time at $t \to \infty$ — and does not depend on the choice of the initial profile when it is varied within certain limits, then this is one of the stable solutions to the investigated equation. In this way, one can find the magnetization profile in the shape of a single peak of the new phase, for example, using the Gaussian form of the initial fluctuation or a series of peaks in a periodic array. The stationary solution obtained in such a way can also later be used to study the dynamics of changes in the magnetization induced by the nonstationary external fields. Figure 4 shows an example of the spatial dependence of the magnetization for a certain value of the pumping.

The domain is a phase with the magnetization orientation opposite that of the matrix magnetization. Domains appear at certain threshold values of pumping $\tilde{G}_{o\,\mathrm{min}}$ with a certain minimum thickness. For parameters given by Eq. (10), the minimum value of the domain thickness is 6×10^{-7} cm. As seen from Fig. 5, if the pumping rate increases, the size of the domain grows; starting from some value, the growth reaches saturation and slows down. The number of flipped spins in a domain per unit time decreases due to relaxation processes. The domain can be stable if these losses are compensated by the magnons captured by the domain from the surrounding space. The inverted spin number in the domain is proportional to its size.

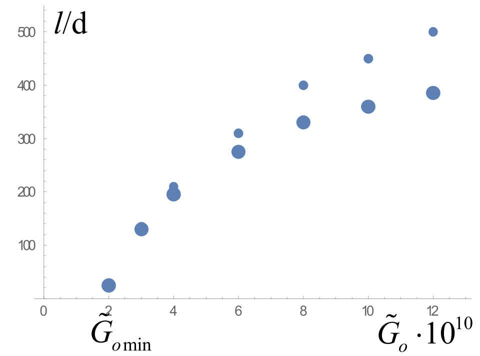

FIG. 5. Dependence of the thickness l of the phase in the form of domain on pumping for different values of the width of a sample L: $L = 1000\,d$ for big points, $L = 1500\,d$ for small points. Parameters are chosen according to Eq. (10).

Magnons can be captured from a region remote from the domain at a distance less than the diffusion displacement length of the magnon. Therefore, for a small number of domains, when the distances between them are greater than the diffusion displacement length, the domain sizes are determined only by the pumping value and do not depend on the sample size. At a high domain density, pump-excited magnons are already distributed among many domains, and this affects the domain sizes.

The sizes of regions with flipped spins are small. This is due to the short-range nature of the exchange forces. Also, for this reason, the domain boundary is sharp. To demonstrate it, Fig. 6 shows the spatial dependence of the magnetization on an increased scale for the coordinates. The domain wall has a thickness of the order of 2×10^{-7} cm.

Since the domain is thin, the light with the wavelength greater than the domain width is scattered by individual spins

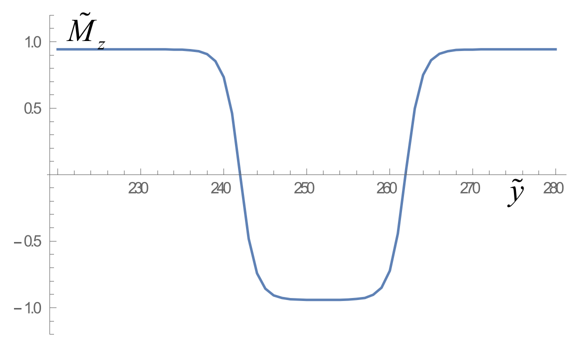

FIG. 6. Spatial dependence of the magnetization in the vicinity of the domain on an enlarged scale of coordinates.

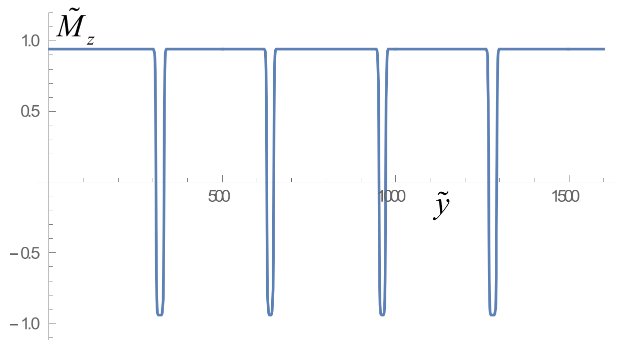

FIG. 7. Periodic structure of phases with inversion magnetization calculated with the pumping parameter equal to $\tilde{G}_o = 2 \times 10^{-10}$. Other parameters are defined by Eq. (10).

located throughout the entire thickness of the domain or, along it at the distances of the order of the wavelength, will add up coherently. At the same time, the fields scattered by individual spins randomly dispersed in the crystal are compensated. Therefore, the formation of structures should lead to the enhancement of the scattering at certain frequencies with an increase of the size of the inclusion.

Figure 7 shows an example of a periodic array of phases with an inverted magnetization. Such periodic magnetization distribution was observed in Ref. [8] and has been explained by a presence of two BEC systems in magnetic crystal, and an interference between their wave function leads to arising periodical distribution of magnetization. In our model, the periodical structure appearance follows from the fact that unstable particles with attractive interaction form periodical structures if their density exceeds some critical value. It is shown for radiation defects (vacancies) in Ref. [36] and for excitons in Ref. [37]. The attractive interaction between spins in magnets is included in the effective magnetic field of the LL equation.

V. PECULIARITIES OF THE BEHAVIOR OF THE CONDENSED PHASE WHEN PUMPING IS TURNED OFF

In Ref. [9], an interesting phenomenon was observed. It is an enhancement of signal of BLS in a magnetic crystal when pumping of magnons is turned off. The phenomenon is explained by the authors [9] as an enhancement of the effects of BEC. Using the model proposed here of spatial self-organization processes in nonequilibrium conditions, we show that, after the pump is quickly turned off, the processes of spatial condensation of flipped spins intensify for some time, the growth of the condensed phase sizes occurs, and that manifests itself in increasing of BLS which has been observed in Ref. [9].

Let us consider a qualitative explanation of the phenomenon. After pumping is turned off, the parameters of the system (temperature, magnon relaxation times, etc.) change. The steady-state temperature of the sample T_2 due to irradiation is higher than the temperature T_r of the cooled sample after pumping is turned off. Therefore, the magnetization at the moment when the pump is turned off is less than the equilibrium magnetization at the temperature T_r because, firstly, there is no negative contribution of magnon pumping to the magnetization, and secondly, due to the heating of the sample during irradiation, the magnetization at the temperature T_2 is lower than at the temperature T_r. After switching off the pumping, the temperature T and the magnetization M relax to their equilibrium values. The time for establishing equilibrium of the lattice subsystem occurs mainly due to the phonon-phonon interaction. The time for establishing equilibrium in the magnetic subsystem occurs due to interactions in the magnetic subsystem and the interaction of the magnetic subsystem with the lattice. At rapid cooling, the time to establish equilibrium in the lattice system is less than the time to establish equilibrium in the spin system. Therefore, for some time, an excess of flipped spins will be realized in the sample in comparison with the equilibrium value for a given temperature. In the presence of an excess of magnons greater than a certain value, inhomogeneous structures can form in the system, the presence of which can lead to increasing BLS which was observed in Ref. [9]. This excess of flipped spins disappears when the equilibrium magnetization is established. Thus, the appearance of the structures may explain the experiments of the enhancement of the signal of BLS [9] when magnon pumping is turned off.

Using the equation for the magnetization, Eq. (8), we consider the formation of structures after pumping was turned off in the form of domains with the orientation of the magnetic moments of the spins opposite the orientation of the magnetic field. Because in the considered process the temperature T is not constant, we are choosing it separately in Eq. (6). Then the main Eq. (8) obtains the form:

$$\frac{\partial \tilde{M}_z}{\partial \tilde{t}} = \frac{T_r}{T}(1-\tilde{M}^2)\frac{\partial^2}{\partial \tilde{y}^2}\left(\tilde{M}_z - th(\tilde{M}_z T_c/T) - \tilde{H} - \frac{\partial^2 \tilde{M}_z}{\partial \tilde{y}^2}\right) - q\left(\tilde{M}_z - th(\tilde{M}_z T_c/T) - \tilde{H} - \frac{\partial^2 \tilde{M}_z}{\partial \tilde{y}^2}\right) - \tilde{G}_o \tilde{M}_z. \quad (13)$$

Upon this form of the dynamics equation, the dimensionless parameters were left the same as determined by Eqs. (9) and (10) at $T = T_r = 300$ K.

Thus, we consider the following scenario: The illuminated magnetic crystal has been at the temperature T_2, the pumping is turned off at $t = 0$, and the temperature decreases. We assume that the cooling of the sample does not depend on the magnetic system and approximate the dependence of temperature on time by the following formula:

$$T(t) = T_r + (T_2 - T_r)\exp\left(-\frac{t}{\tau_{\rm ph}}\right), \quad (14)$$

where T_2 and T_r are, respectively, the sample temperatures at the time of switching off the pumping and after cooling, and $\tau_{\rm ph}$ is the cooling rate of the phonon system.

We investigate the magnetic crystal using Eq. (13) starting from the moment $t = -t_1 < 0$. The pumping with some value $\tilde{G}_o$ is turned on at the moment of time $t = -t_1$, and the formation of a domain is considered by introducing initial fluctuations just as it has been done in the previous paragraph finding the stationary states. The time $-t_1$ is chosen so that, at the moment when the pumping is turned off at $t = 0$, the domain has already formed in the magnetic crystal, and a

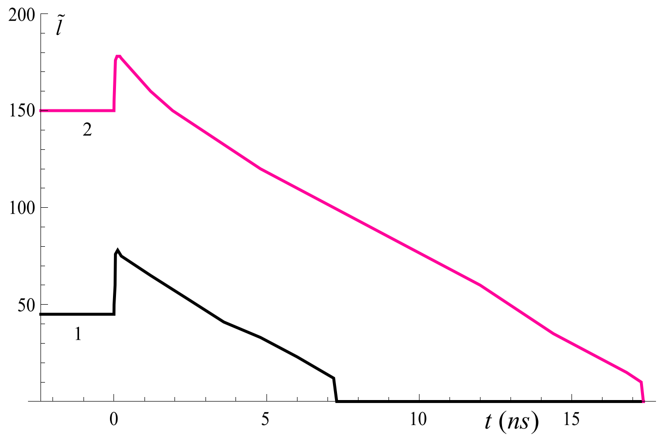

FIG. 8. Dependences of the domain thickness $\tilde{l} = l/d$ on time after switch off at $t = 0$; the pumping for $\tilde{T}_r = 0.5357$, $\tilde{T}_2 = 0.6$, given at the values of pumping: $\tilde{G}_o = 2 \times 10^{-10}$ in black points (curve 1), $\tilde{G}_o = 5 \times 10^{-10}$ in red points (curve 2).

stationary state was established at constant pumping $\tilde{G}_o$ with temperature T_2. The dependences of pumping and temperature on time were suggested in the following way: $\tilde{G}_o$ is a constant value, and $T = T_2$ at $t < 0$; at $t > 0$, the pumping is $\tilde{G}_o = 0$, and the temperature is determined by Eq. (14). With increasing time, a domain of the type shown in Fig. 4 appears in the solution of Eq. (13); the magnetic moment of the domain is opposite to the orientation of the magnetic field. At first, the domain width rises, then the expansion slows down, and then the width of the domain drops as the system thermalizes to the stationary state. After the pumping is turned off at $t = 0$, a sharp jump in the domain width occurs due to the excess density of flipped spins in the sample compared with the equilibrium value, and then the domain width gradually decreases, completely disappearing when the equilibrium state of the magnetization is reached. Figure 8 shows the dependence of the domain width on time for two cases of pumping values.

Thus, when pumping is turned off, the domain size increases significantly, and domains exist for some time. The increasing of the domain size should lead to the rise of BLS. Therefore, the effects observed in Ref. [9] may be explained by the existence of condensed phases of magnons in space after a decrease in temperature without involvement of BEC of magnons. Calculations show that, with increasing pumping, domains with a larger width grow, and their lifetime increases after pumping is turned off. The sharp break in the dependence of the domain width on time in Fig. 8 is because the destruction of a domain, a size of which is smaller than the critical one, occurs very quickly in the absence of pumping. In the experiment, this break should be washed out due to other BLS mechanisms (scattering by single spins, for example) as well as the presence of differently shaped structures and the domains with different widths.

VI. STIMULATION OF THE CONDENSED PHASE FORMATION DURING RAPID COOLING OF A SAMPLE

The previous consideration shows that, during rapid cooling, the magnon density increases for some time in comparison with their equilibrium value, and this contributes to the spatial density of magnons.

Let us consider the possibility of the formation of spatial structures upon cooling the sample from the temperature T_2 to a lower temperature T_r in the case when the pumping is less than the critical value. Consequently, at constant temperatures T_r or T_2, the structure does not appear, and the magnetization in the sample is uniform. To present a concrete instance, we will consider the pumping rate $\tilde{G}_o$ to be equal to 1.5×10^{-10}, which is a subthreshold value for the parameters of the magnetic crystal given by Eq. (10). The evolution of the initial fluctuation at such a value of pumping is illustrated in Fig. 9, which shows the spatial distribution of the magnetization obtained from the solution of Eq. (13) at different times for the sample temperature $\tilde{T}_r = 0.5357$. The damping of fluctuations confirms the fact that no structures appear at the selected pumping and constant temperature. At the temperature $\tilde{T}_2 = 0.88$, fluctuations behave similarly. Now let us consider the transition when the temperature changes and the system moves from point B to point A during cooling (Fig. 10). The pumping-induced shift of the magnetization at points B and A is not visible in the figure due to the relative smallness of its magnitude. During extremely fast cooling, when the equilibration of the phonon system takes much less time than the time necessary for establishing the equilibrium of the magnetization, the motion of the system is described by the broken curves which go first from point B to point B1 in Fig. 10, and then from point B1 to point A. When moving from point B1 to point A, the system is in a state in which the temperature is close to the temperature T_r, and the magnetization is lower than the magnetization in the equilibrium state for this temperature. In this case, there is an excess concentration of flipped spins, and in the presence of fluctuations, a structure should appear in the system. The structure disappears when the system goes into equilibrium. If the cooling rate of phonon system τ_{ph} increases, the curve of the transition from point B to point A rises upward. In this case, the widths of the emerging domains decrease, and with a certain increase in the cooling rate, the structure is not formed. The results of magnetization calculations obtained from the solutions of Eqs. (13) and (14), with the temperature changing over time, are presented in Figs. 11 and 12. Upon calculations, the pumping and initial fluctuation are the same as those used when considering Fig. 9 for a fixed temperature. Figure 11 demonstrates the formation of spatial structures of magnetization at different times at transition from temperature $\tilde{T}_2 = 0.88$ (492.8 K) to temperature $\tilde{T}_r = 0.5357$ (300 K) for the value of the cooling rate parameter equal to $\tau_{ph} = 2.4$ ns.

In Fig. 12, the dependence of the domain thickness on time is presented for three values of the high temperature T_2. Parameters of the system are given by Eq. (10). As seen from the figure, a structure emerges despite the fact that the value of the pumping is subthreshold. The largest value of the domain thickness grows with increasing the higher temperature T_2, beginning from which the rapid cooling occurs. The time of its existence grows with increasing its maximal size. The curves of the dependence of the width on time are asymmetric, and the width has an order of 20 ns. Such a value is close to the width of experimental dependence of BLS intensity on

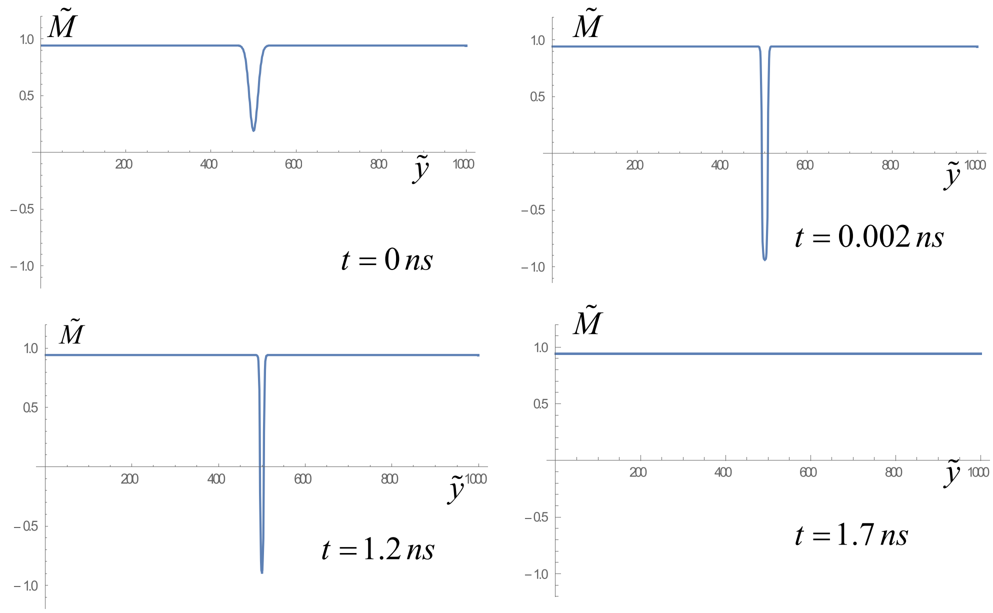

FIG. 9. Temporal behavior of fluctuations of the magnetization at a subthreshold value of the pumping magnitude $\tilde{G}_o = 1.5 \times 10^{-10}$ at $T_r = 0.5357$ (300 K).

time obtained in yttrium iron garnet in the condition of rapid cooling (see Fig. 2(b) in Ref. [23]).

There is also stimulation of the nucleation of structures during rapid cooling of the sample. The nucleation of structures, even with an excess number of flipped spins, occurs in the presence of seed fluctuations of the magnetization, which have some critical parameter values. Calculations like those carried out above when considering subcritical pumping show that, with rapid cooling, the critical fluctuation parameters become less rigid. In experiments [25], the stabilization of bullets, which are excited states in magnets, was observed upon rapid cooling.

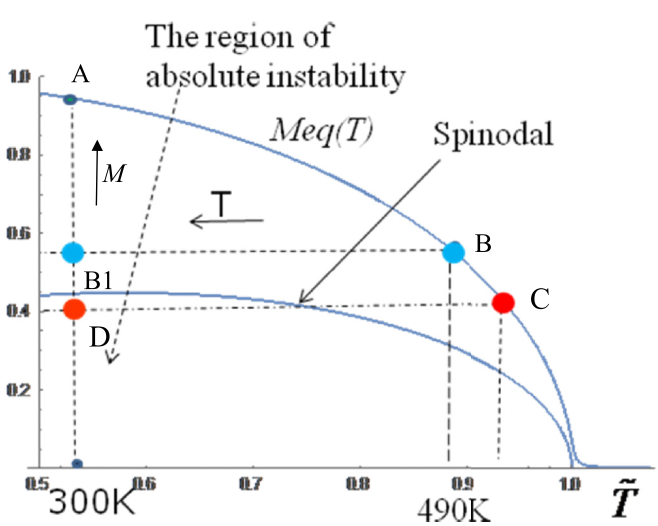

FIG. 10. Temperature dependence of magnetization and spinodal curve in the equilibrium state. Horizontal dotted lines correspond to processes with a rapid temperature change.

Thus, during rapid cooling, the appearance of structures occurs at pumping values lower than the threshold value. The phenomenon occurs due to increasing the number of inverted spins during cooling the crystal that stimulates the phase transition in the spin system. In Refs. [23–25], such a phenomenon was observed and explained by BEC.

VII. FORMATION OF PHASES WITH INVERTED SPINS BY RAPID COOLING WITHOUT MAGNON PUMPING

The results presented in the previous paragraph show that, during rapid cooling of the sample, the higher the temperature of the sample in the initial state, the more intense the stimulation of the formation of structures in the magnetic solid. From the dependence of magnetization on temperature (Fig. 10), with an increase in the initial temperature at a certain magnitude, the value of the equilibrium magnetization falls into the region of magnetization values located below the spinodal curve. As we saw in Sec. III, states with uniform magnetization in the region below the spinodal curve are unstable with respect to creation of nonhomogeneous structures. Hence, it becomes possible by quickly cooling the sample (for example, by transferring its state from point C to point D in Fig. 10) to create an inhomogeneous magnetization structure both without magnon pumping and without initial fluctuations. To study the possibility of implementing such processes, numerical solutions of Eq. (13) were carried out with the temperature dependence on time according to Eq. (14) at different values of the cooling rate $\tau_{\rm ph}$. We assumed that the sample is cooled down from temperature T_2 to temperature T_r in the setup with no pumping ($\tilde{G}_o = 0$), no initial fluctuation at $t = 0$, and that the value of the equilibrium magnetization in the initial state is in the range of the magnetization values below the spinodal curve. The movement of the system from point C to point A in Fig. 10 is considered. With instantaneous cooling, the

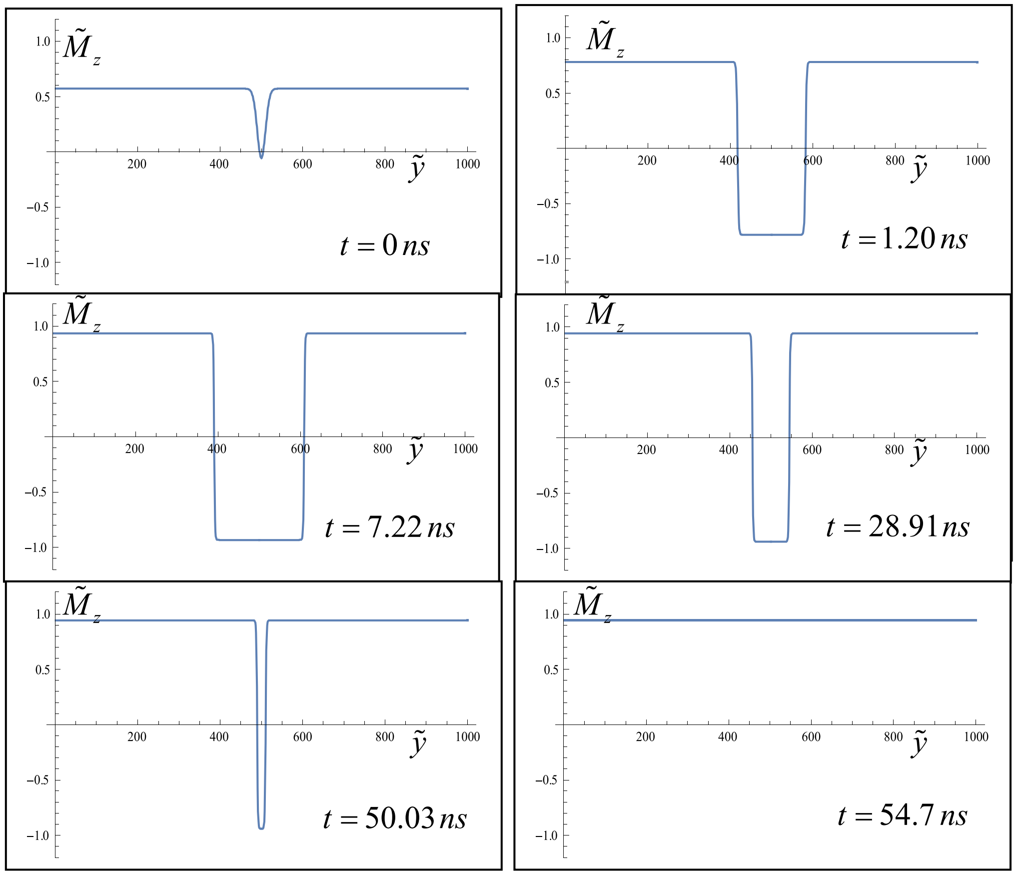

FIG. 11. Spatial structure of the magnetization distribution at different times after turning on the cooling from temperature $\tilde{T}_2 = 0.88$ to temperature $\tilde{T}_r = 0.5357$ in the presence of subthreshold pumping $\tilde{G}_o = 1.5 \times 10^{-10}$. The cooling rate of phonon system equals $\tau_{\rm ph} = 2.4$ ns.

trajectory must pass through point D. As the value of the parameter $\tau_{\rm ph}$ increases, the trajectory C-A in Fig. 10 rises above the spinodal curve. The result of the influence of cooling on the formation of structures depends on which part of the C-A

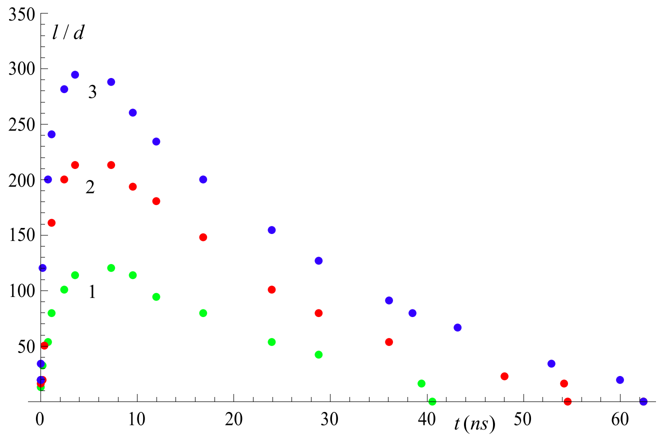

FIG. 12. The time dependence of the thicknesses of domains which arise in the presence of subthreshold pumping $\tilde{G}_o = 1.5 \times 10^{-10}$ under rapid cooling from the temperature T_2 to $\tilde{T}_r = 0.5357$ (300 K). Results are given for $\tilde{T}_2 = 0.75$ (420 K) in green (curve 1), for $\tilde{T}_2 = 0.88$ (492.8 K) in red (curve 2), for $\tilde{T}_2 = 0.95$ (532 K) in blue-green (curve 3).

movement trajectory, and for how long, is under the spinodal curve.

Since in the considered case a homogeneous solution of Eq. (13) for the magnetization is unstable, the slightest inhomogeneity will lead to violation of the uniformity. Any infinitely small perturbation should bring the system out of the uniform state. The presence of the boundaries and the defects in a sample may be such perturbations. In the case under consideration, such inhomogeneities are the presence of boundaries of the samples. Also, when solving this problem, the finite difference method can be such a small perturbation since, in the method, the continuous function of magnetization is given at discrete points.

Figure 13 shows the dependence of the magnetization obtained from the solution of Eq. (13) at $\tilde{G}_o = 0$ and without fluctuations at $t = 0$ at various points in time. At a small value of time, the nonhomogeneities arise near the surface of the sample. Henceforth, a structure in the shape of numerous domains appears with very small gaps between them. Subsequently, the domains merge, the narrow ones vanish, and large regions with uniform magnetization grow and expand. In the end, the domains disappear completely, and the magnetization becomes equal to the equilibrium uniform magnetization at T= 300 K.

The phonon cooling rate used in calculations of Fig. 13 equals $\tau_{\rm ph} = 0.24$ ns and at the initial temperature $\tilde{T}_2 = 0.98$ (548.8 K), which is near the temperature of phase transition 1

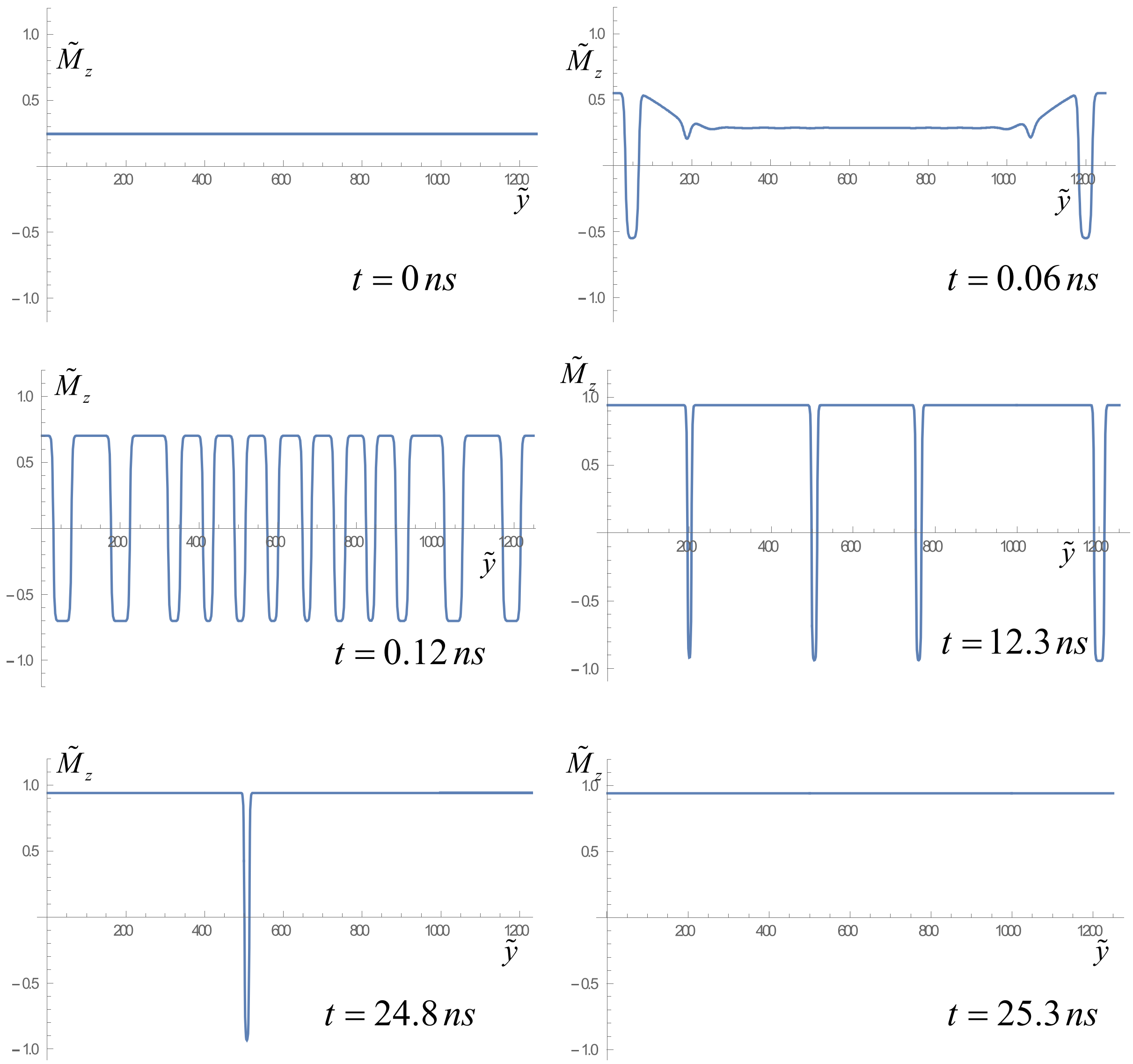

FIG. 13. Spatial distribution of sample magnetization during rapid cooling from temperature $\tilde{T}_2 = 0.98$ (548.8 K) to temperature $\tilde{T}_r = 0.5357$ (300 K) without external pumping and without the initial inhomogeneous fluctuation. The parameters of the system are from Eq. (10) and $\tau_{\rm ph} = 0.24$ ns.

(560 K). As the phonon parameter $\tau_{\rm ph}$ decreases (more rapid cooling), the phenomenon can be observed in a larger distant temperature from the phase transition temperature. Thus, at $\tau_{\rm ph} = 0.25$ ns, appearance of structure of the type shown in Fig. 13 can also be observed at $T_2 = 536$ K.

The temperature fluctuations of various modes of magnetization exist in a magnetic crystal. With rapid cooling, some of them will survive and grow. The answer to the question of which one will survive depends on the mode spectrum of the sample, its geometry, and dimensions. In this paper, we do not consider these issues. We study only the effects by appearance of domain shape structures. However, in any case, the appearance of regions of macroscopic dimensions should manifest itself in BLS as a phenomenon of the formation of new phases.

Note that the considered phenomenon can be observed both in ferromagnets and in ferrimagnets since the mean field method describes well the dependence of magnetization on temperature in both a ferromagnet and a ferrimagnetic [38,39]. However, in a ferrimagnet, the position of the spinodal may differ from that in a ferromagnet, which may affect the temperature regions where the effects are manifested. However, the qualitative picture of the manifestation of effects should be the same.

VIII. DISCUSSION

In this paper, the formation of structures is considered in the macroscopic description of phase transition which corresponds to the Cahn-Hillert model [19,20] of the spinodal decomposition. In magnetic crystals, it is convenient to consider many phenomena using magnons as elementary excitations. Although magnons are rather approximate formations under strong excitations of the system, the existence of structures and the formation of phases in a nonequilibrium magnetic crystal with the help of magnons can be qualitatively described by the Lifshiz-Slyozov model [40] of the nucleation and growth. Let us assume that an inclusion has formed in the magnetic crystal, where magnetic moments of spins are oriented opposite the orientation of the magnetic field and the orientation of the moments in the rest of the sample. Since the orientations of the spins of atoms of the inclusion and the inverted spins outside the inclusion (magnons) are the same, there are attractive exchange interactions between them. Therefore, magnons from the surrounding space will be captured by the inclusion during collisions, giving energy to phonons. Of course, the reverse processes of the transition of spins into the surrounding space with the excitation of magnons there also occur. If there is an excess of magnons above their equilibrium value in the surrounding space, then the number of captured magnons will exceed the number of lost ones. Within the region itself, the disappearance of magnons will occur due to weak spin-orbital and magnon-magnon interactions. Thus, the new phase will grow due to the strong exchange interaction, and the growth rate will be proportional to the area of the inclusion edge, while the decrease in the inclusion size would be due to the weak relativistic interactions and would have a probability proportional to the volume of the region. Therefore, if an excess concentration of magnons as compared with equilibrium is created in a magnetic system at a certain level, the resulting phase with the inverse direction of spins will be stable, and its linear dimensions will be limited and determined by the amount of magnon excess.

The observation of the magnetic lattice in Ref. [8] is direct experimental evidence in favor of the existence of regions with the inverse direction of magnetization. Theoretically, the periodic distribution of magnetization was obtained in Ref. [12] and in this paper. Comparison with the experiment requires some discussion. It is shown from Ref. [8] that the thickness of the magnetic wall in the lattice is of the order of 1 μm, and the value of the magnetization in the wall is 3.5% of the magnetization in the crystal (see Fig. 2 of Ref. [8]). At the same time, according to the results in Sec. IV, it follows that the width of the domains is much narrower than this value and is of the order of $10^{-2} \div 10^{-1}$ μm, and the ratio of the absolute value of the magnetization to the magnetization of the sample is much larger than this value and is of the order of a unit. In our opinion, the discrepancy is because the accuracy of the BLS method used in Ref. [8] cannot determine the spatial dimensions for objects $<0.5\ \mu$m; therefore, the widths of the domains are overestimated, and the overestimation of the size of an object leads to an underestimated value of the magnetization. At the same time, authors of Ref. [8] remark that the modulation depth increases quickly at powers above threshold and then stays nearly constant. Such behavior has the process in our model of phase formation with increasing the pumping: The value of magnetization grows very quickly and then becomes almost constant.

The resulting magnetic structures with magnetization orientation opposite the orientation of the magnetic field should have their own excitation spectrum, determined by the exchange, dipole-dipole spin-spin interaction, and interaction of spin with the magnetic field. In this paper, this spectrum is not investigated. However, let us make important remarks about its manifestation. In the considered structures, the spins are oriented opposite the spins of the magnet, and the system is in nonequilibrium. If the spins come to equilibrium in the structure (inclusion) very faster than they fall into inclusion from the environment and disappear at relaxation, we may assume that the spin system in inclusion is in a quasiequilibrium state, in which the spins are oriented opposite the orientation of spins in the matrix. The traditional methods [41,42] may be applied for the calculation of spectra of such systems. In this case, the ground state is the state in which all spins in the inclusion are flipped. The excited state is the state in which the magnetic moment of one spin is oriented along the magnetic field. In our case of the small-sized systems, the methods of investigation of the magnon spectra in the thin films [43,44] should be applied. The excitation spectra should depend on the geometry and the sizes of sample and on orientations of the domain and magnetic field. If we consider only the exchange interaction and the interaction of spin with magnetic field. the dispersion law for excitation in the domain equals $\varepsilon(\vec{k}) = I(0) - I(\vec{k}) - 2\mu_o H$, where $\overleftarrow{k}$ is a wave vector parallel to domain surface, and $I(\overleftarrow{k})$ is the Fourier transformation of the exchange interaction. It is seen from the formula that, at small values of vector $\overleftarrow{k}$, the energy of excitation is negative [$\varepsilon(\vec{k}) < 0$] at $\vec{k} \to 0$. The spin-flipped structures under consideration are systems with an inverse-level population of states. In such a nonequilibrium system, the negative energy excited state of inclusion has a spin orientation such as that of the matrix spins. Therefore, the transition to an excited state of inclusion corresponds to a relaxation transition in the system. For a small number of such excited states, they can be described by the Bose creation and annihilation operators. An operator describing relaxation processes, i.e., transitions to a lower-energy state, should be the Bose creation operator, and the transition probability during relaxation should be proportional to $w_i(1 + n_i)$, where the value w_i depends on the relaxation mechanism, and n_i is the number of excited states of the type i in the inclusion. Thus, this is the number of states created during relaxation processes in the inclusion. The probability of relaxation grows with increasing n_i. That may explain the increase in the intensities of the BLS spectrum regions and their growth and narrowing with time based on the proposed phase formation model. The stimulated thermalization of a parametrically driven magnon gas was observed in yttrium iron garnet film [45].

Note that, in this paper, we consider the simplest but often implemented problem of the formation of new domain-shaped phases in a magnetic crystal supersaturated with magnons. Realistically, in a crystal oversaturated with magnons, new phases having a different geometric shape may be formed: cylindrical domains, in the form of disks, balls, etc. Excited states of crystal may be considered as fluctuations. They may be the nucleus of new phase formation. Stabilization of the states of nonlinear bullets at rapid cooling was observed in Ref. [25]. To study new phase properties quantitatively, one should use relaxation terms accounting for the excess of magnetization as compared with its equilibrium value and Baryakhtar's additional term associated with the spin diffusion.

In this paper, we did not study the processes of spin currents in magnetic materials. The study of the influence of the formation of inclusions with the inverse direction of spins requires a separate consideration of the transfer processes because the strong excited magnetic systems are prospective for the development of spintronics and magnonics [46–48]. Phase inclusions with flipped spins of any shape, not only domains, must interact strongly with an inhomogeneous magnetic field that sets the spins in motion. In the inclusion, the spins are rigidly interconnected, and their movement is coherent. The spin current will be determined by the movement of the aggregate of inclusions (phases) and not by the aggregate of individual spins.

IX. CONCLUSIONS

The model of the spatial formation of phases with the orientation of spins inverted with respect to the orientation of the matrix spins in the supersaturated magnon gas is considered in this paper. The model is an alternative to the Bose-Einstein model of magnon condensation in the momentum space. The results obtained in this paper in the model of spatial formation of phases for the case of rapid cooling of the sample are consistent with the experimental data and confirm the validity of the model. The model can be used to obtain new effects. Thus, it predicts the appearance of structures at rapid cooling in the absence of the external pumping of magnons for the parameters in a certain range as well as the formation of phases stimulated by the excited states of the crystal serving as nuclei of the phases. Systems with the considered phases can be extremely diverse. They can be controlled by various external sources and, therefore, are promising for use in spintronics and magnonics.

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