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Electromagnetic superradiance from single-molecule magnets in the presence of a classical driving magnetic field

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The electromagnetic superradiance of a nanomagnet crystal (magnetic molecules) subjected to a resonant driving magnetic field and a dc magnetic field has been theoretically investigated. The driving magnetic field is supposed to be strong enough so that it essentially influences the spectrum of the magnetic molecules. The conditions favorable for observing the superradiance have been formulated.

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I. INTRODUCTION

Among the papers devoted to quantum coherence and quantum tunneling of magnetization of high-spin molecules and magnetic ions,[1–3] the works which deal with their behavior in alternating magnetic fields are of interest because they enable one to obtain important information about the spectrum and the relaxation times of magnetic molecules and ions. Therefore, a stationary high-frequency susceptibility connected with transitions between states of the fundamental doublet of a high-spin molecule has been investigated in Refs. 4–7. The measurements that have been carried out show a nonlinear effect, which consists of a considerable decrease in the imaginary part of the susceptibility with an increase in the alternating field amplitude.[4,5] The temperature dependence of the imaginary part of the stationary susceptibility has a nontrivial nature, which has been investigated in Refs. 6 and 7. The high-spin molecule, which is subject to a bifrequency ac magnetic field, and rare-earth ion behavior, have been theoretically studied[8] longer than the relaxation times that characterize the transitions between three lower levels of a molecule (a stationary process). In our opinion, some interesting papers should be noted, e.g., those devoted to the fine spectroscopic research of the molecular clusters of Fe_8 and Mn_{12} that allow one to determine the spectrum particulars of the molecular magnets.[9–12]

The works mentioned above dealt with stationary processes, i.e., the processes that occur at times much longer than the typical relaxation times. It is also of interest to investigate the nonstationary processes of the interaction between magnetic molecules and ions and ac magnetic fields. These processes proceed at times much shorter than the typical relaxation times. In their work, Vernier *et al.*[13] observed an echo of magnetic tunneling states in Kramers-ion doped glasses, and they discussed the states of the fundamental doublet that are formed as a result of the quantum tunneling of magnetization. The influence of an ac magnetic field at the frequency corresponding to the transition between the lower states of the fundamental and the first excited doublets on the tunneling rate of the magnetization of a high-spin molecule has been theoretically studied.[14] The nonstationary behavior (at times much shorter than the relaxation times) of a magnetic molecule subject to a bifrequency ac magnetic field is theoretically considered in Ref. 15: it has been shown that the molecule can absorb or emit energy (stimulated processes).

Since Dicke's original paper,[16] the well-known phenomenon of superradiance still attracts a great deal of attention even today. Superradiance is essentially a nonstationary effect when the system dipole radiation intensity is proportional to N^2, where N is the number of particles in the system. Therefore, an interesting theoretical paper[17] should be noted, where it has been assumed that the magnetization resonant tunneling (in the context of the Landau–Ziner effect[18–20]) can cause the superradiance of high-spin molecular magnets (see also the discussion in Refs. 21–23).

The bursts of microwave pulses have actually been detected in a number of experimental investigations,[24–26] where it is the author's opinion that the superradiance can be the physical mechanism responsible for this phenomenon. Therefore, the experiment discussed in Ref. 27 is of interest.

The theoretical research in Ref. 28 is also very interesting, where the attention therein is focused on the role of a resonator in enhancing the collective radiation when the microwave radiation in the crystal of molecular magnets is considered. The importance of the resonator, as is mentioned in Ref. 28, was emphasized in Ref. 23 and earlier in Ref. 29.

From our point of view, it is interesting to mention Ref. 30, which further considers the role of the dipole-dipole interactions as the basic dephasing factor in the phenomenon of superradiance in spin systems.

In this work, we consider nonstationary processes driven by a strong ac magnetic field (the driving field) that occur in an ensemble of high-spin molecules and develop in the following "scenario."

Originally (in the absence of the driving field), the energy spectrum of the magnetic molecules is a consequence of the splitting of doublets by a dc magnetic field that is perpendicular to the easy anisotropy axis.[1–3] The temperature is assumed to be low enough so that only the fundamental doublet levels are populated in the initial state (thermodynamic equilibrium state).

Under the influence of the driving field, which causes the transitions between lower levels of the fundamental and the first excited doublets, the initial system of the energy levels undergoes essential changes. In this situation, it turns out to be productive to introduce the concept of quasienergy states.[31]

As a result of this modification, the system begins to radiate. The frequencies of the radiated field are a combination of the frequencies corresponding to the initial energy structure and the Rabi frequencies corresponding to the driving field. By using a resonator of a defined quality factor, one is able to choose concrete modes in the radiated spectrum. In our case, the frequency of such radiation is much smaller than the frequency of the driving field. Note that such behavior (spontaneous radiation) of a three-level system under the influence of a strong coherent field was considered in detail (see, for example, Refs. 32 and 33).

Finally, and it is the main idea in our work, we show that this radiation can be realized in the form of the superradiance. The formation of a superradiance pulse in the considered case as in the traditional problem should be realized for times much longer than the lifetime of the radiated photon, but much shorter than the dephasing time.

Thus, this paper is a continuation of theoretical investigations started in Ref. 15, where, in the same problem formulation, the stimulated radiation and the absorption of a low-frequency field have been considered. To be specific, we confine our investigation to magnetic molecules with an easy anisotropy axis subject to a strong enough dc transverse magnetic field. Moreover, it should be noted that, in fact, three levels of a magnetic molecule (out of the four levels that belong to the fundamental and the first excited doublets) are involved in the process discussed below. The permissibility of such simplifying assumptions that do not disturb the essence of the phenomenon was examined in detail in Ref. 15.

The paper is organized as follows: In Sec. II, the formation of the quasienergy spectrum of magnetic molecules constituting a crystal under the influence of the driving field is considered. Section III deals with the relaxation times that approach the system of equations that describes the kinetics of the process. We have also examined the initial conditions in detail. The numerical solutions describing the superradiance for various values of the number of molecules and the dephasing time are given in Sec. IV. The conclusions are summarized in Sec. V.

II. ENERGY SPECTRUM OF THE MAGNETIC MOLECULES IN THE PRESENCE OF THE DRIVING FIELD: INTERACTION OF MAGNETIC MOLECULES WITH THE FIELD OF RADIATION

Let us consider a crystal of molecular magnets subject to the dc magnetic field and the driving field. The dc field and the driving field are perpendicular to the easy anisotropy axis of the molecule. We also take into account the interaction of the magnetic molecules to the magnetic field of the resulting radiation. The magnetic field of the radiation is parallel to the easy axis in our scheme. Such polarization of the radiation field is connected with the selection rules that occur for the transitions between the molecular levels. It will be explained in detail below.

We assume that the dc magnetic field and the driving field are strong enough that they determine the resulting quasienergy structure. In that way we neglect the dipole-dipole interactions between the molecules. In other words, we suppose that the molecules do not directly interact with each other. Note that the dipole-dipole interaction, in general, determines the dephasing time (see Refs. 22 and 30). This aspect will be considered in Secs. III and IV.

In this case, the molecule Hamiltonian reads

$$\hat{H} = \sum_{\alpha=1}^{N} (\hat{H}_{\alpha;\mathrm{mol}} + \hat{H}_{\alpha}^{dr} + \hat{H}_{\alpha;\mathrm{int}}^{\mathrm{rad}}) + \hat{H}_f. \tag{1}$$

Here,

$$\hat{H}_{\alpha;\mathrm{mol}} = - D\hat{S}_z^2 + \hat{H}_{tr} + g\mu_B H_0 \hat{S}_{\alpha;x}, \tag{2}$$

$$\hat{H}_{\alpha}^{dr} = g\mu_B H_x(t)\hat{S}_{\alpha;x}, \tag{3}$$

$$\hat{H}_{\alpha;\mathrm{int}}^{\mathrm{rad}} = g\mu_B H_z(t)\hat{S}_{\alpha;z}, \tag{4}$$

and index α enumerates the molecules $(1 \leq \alpha \leq N)$. Here, z is the easy axis of anisotropy; $\hat{S}_x$ and $\hat{S}_z$ are the x and z projections of the spin operator, respectively. D, g, and μ_B are the anisotropy energy constant, the Landé factor, and the Bohr magneton, respectively. H_0 is the dc magnetic field (we suppose the direction of this field to coincide with the x axis), $H_x(t)$ is the driving field that is parallel to the x axis, $H_z(t)$ is the magnetic field of the radiation that is parallel to the z axis, and $\hat{H}_{tr}$ describes the transverse anisotropy. The total spin of the molecule is equal to S.

In the absence of magnetic fields $(H_0 = H_x = H_z = 0)$ and by neglecting the transverse anisotropy, the energy spectrum of the molecule $E_m = -Dm^2$ $(m = S, S-1, \ldots, -S)$ represents $S + 1/2$ degenerate doublets if S is a half-integer, and S degenerate doublets and one nondegenerate level if S is an integer. The dc magnetic field H_0 directed along the x axis and the transverse anisotropy induce spin tunneling between two states of any doublet and split the latter to the levels $E_m^{\pm} \simeq E_m \pm \frac{1}{2}\Delta E_m$ (here, $m > 0$).[34,35] The quantity ΔE_m is much smaller than the spacing $E_{m-1}^{-} - E_m^{-} \simeq D(2m-1)$ between the nearest doublets and essentially increases by decreasing the number m. The eigenfunctions of the Hamiltonian (2) in the absence of magnetic fields and transverse anisotropy can be chosen from the following form:

$$\psi_m^{(s)} = \frac{\psi_m + \psi_{-m}}{\sqrt{2}}, \qquad \psi_m^{(a)} = \frac{\psi_m - \psi_{-m}}{\sqrt{2}}. \tag{5}$$

Here, $\psi_{\pm m}$ are the eigenfunctions of the operator $\hat{S}_z$ $(\hat{S}_z \psi_{\pm m} = \pm m \psi_{\pm m})$. As follows from the symmetry of the Hamiltonian (2), the eigenfunctions corresponding to the lower and upper levels of the split doublet are combinations of the symmetric functions $\widetilde{\psi}_m^{(s)}$ (5) and combinations of the antisymmetric functions $\widetilde{\psi}_m^{(a)}$ (5), respectively.

As we have mentioned in the Introduction, only two lower doublets are of interest. For brevity, we designate $\varepsilon_0 = E_S^-$, $\varepsilon_1 = E_S^+$, $\varepsilon_2 = E_{S-1}^-$, $\varepsilon_3 = E_{S-1}^+$, $\varphi_0 = \widetilde{\psi}_S^{(s)}$, $\varphi_1 = \widetilde{\psi}_S^{(a)}$, $\varphi_2 = \widetilde{\psi}_{S-1}^{(s)}$, and

$\varphi_3=\tilde{\psi}_{S-1}^{(a)}$, and we introduce the natural frequencies of transitions $\omega_{jk}=(\varepsilon_j-\varepsilon_k)/\hbar$. The states φ_0 and φ_1 form the fundamental doublet, and the states φ_2 and φ_3 form the first excited doublet.

Due to the symmetry of the wave functions of the Hamiltonian (2), the magnetic field polarized along the x axis produces the transitions between states of the same symmetry: $\varphi_0 \leftrightarrow \varphi_2$ and $\varphi_1 \leftrightarrow \varphi_3$, whereas the magnetic field polarized along the z axis produces the transitions between the symmetric and antisymmetric states: $\varphi_0 \leftrightarrow \varphi_1$ and $\varphi_2 \leftrightarrow \varphi_3$ (see Refs. 36 and 37). The driving field couples the lower levels of the fundamental and the first excited doublets (the states of the same symmetry), which is why we consider the driving field to be polarized along the x axis, while for the transition discussed below, the magnetic field of the radiation has to be polarized along the z axis. Note that the transition frequencies under discussion are well segregated,[9] because this allows one to choose the transitions.

The driving field is considered as a classical one, i.e., as a prescribed function of time as follows:

$$H_x(t) = H_x \sin \omega_x t. \tag{6}$$

The interaction of the molecule with this field is described by the operator $\hat{H}_\alpha^{dr}$.

The ac magnetic field polarized along the z axis corresponds to the radiation field. This field will be regarded in a single-mode approach. For the sake of convenience and clarity, we will describe this field as a quantum object as follows:

$$\hat{H}_z(\mathbf{r},t) = -H_{\nu z}(\mathbf{r})\sqrt{\frac{\hbar\omega_{\nu z}}{2}}(\hat{a}_{\nu z}^\dagger + \hat{a}_{\nu z}). \tag{7}$$

Here, $H_{\nu z}(\mathbf{r})$ is a coordinate dependence of the magnetic field of the mode, $\omega_{\nu z}$ is the mode frequency, and $\hat{a}_{\nu z}^\dagger$ and $\hat{a}_{\nu z}$ are the creation and annihilation photon operators, respectively, of the corresponding mode.

The Hamiltonian of this field in Eq. (1) is as follows:

$$\hat{H}_f = \hbar\omega_{\nu z}\left(\hat{a}_{\nu z}^\dagger \hat{a}_{\nu z} + \frac{1}{2}\right). \tag{8}$$

The interaction of the molecules with the radiation field is described by the operator $\hat{H}_{\alpha;\mathrm{int}}^{\mathrm{rad}}$ as follows:

$$\hat{H}_{\alpha;\mathrm{int}}^{\mathrm{rad}} = -g\mu_B H_{\nu z}\sqrt{\frac{\hbar\omega_{\nu z}}{2}}\hat{S}_{\alpha;z}[\hat{a}_{\nu z}^\dagger(t) + \hat{a}_{\nu z}(t)]. \tag{9}$$

The interaction representation will be used below, where the operator $\hat{H}_{\mathrm{int}}^{\mathrm{rad}}$ plays the role of the interaction operator. Therefore, we have $\hat{a}_{\nu z}^\dagger(t)=\hat{a}_{\nu z}^\dagger \exp(i\omega_{\nu z}t)$ and $\hat{a}_{\nu z}(t)=\hat{a}_{\nu z}\exp(-i\omega_{\nu z}t)$. It is convenient to describe the molecule's states using the wave functions of Shrödinger's evolution equation as a basis as follows:

$$i\hbar\frac{\partial\psi_\alpha(t)}{\partial t} = [\hat{H}_{\alpha;\mathrm{mol}} + \hat{H}_\alpha^{dr}(t)]\psi_\alpha(t). \tag{10}$$

These functions, which are obtained as the solution of Eq. (10) in the resonance approaching $\omega=\omega_{20}$, have the following form:

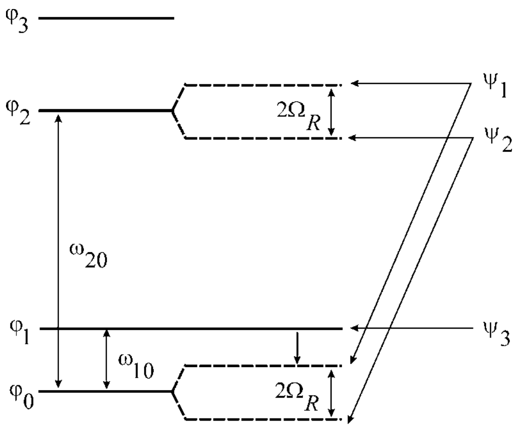

FIG. 1. Formation of the quasienergy levels (dotted lines). Two lower doublets of a nonperturbed magnetic molecule are shown by solid lines. A long vertical line with two arrowheads depicts the transitions caused by the driving field, with the frequency close to ω_{20}. The vertical arrow with one arrowhead demonstrates the transitions considered in this study.

$$\psi_{1,\alpha} = \frac{1}{\sqrt{2}}\left\{\varphi_{0,\alpha}\exp\left[i\left(-\Omega_R - \frac{\varepsilon_0}{\hbar}\right)t\right] + i\varphi_{2,\alpha}\exp\left[i\left(-\Omega_R - \frac{\varepsilon_2}{\hbar}\right)t\right]\right\}, \tag{11}$$

$$\psi_{2,\alpha} = \frac{1}{\sqrt{2}}\left\{\varphi_{0,\alpha}\exp\left[i\left(\Omega_R - \frac{\varepsilon_0}{\hbar}\right)t\right] - i\varphi_{2,\alpha}\exp\left[i\left(\Omega_R - \frac{\varepsilon_2}{\hbar}\right)t\right]\right\}, \tag{12}$$

$$\psi_{3,\alpha} = \varphi_{1,\alpha}\exp\left(-i\frac{\varepsilon_1}{\hbar}t\right), \tag{13}$$

where

$$\Omega_R = \frac{g\mu_B H_x}{2\hbar}\langle\varphi_2|\hat{S}_x|\varphi_0\rangle \tag{14}$$

is the Rabi frequency.

Using the functions (11)–(13) as a basis for calculating the matrix elements of molecule operators is a key to the interaction representation. Note that the functions $\psi_{1,\alpha}$ and $\psi_{2,\alpha}$ describe the quasienergy states of magnetic molecules (see, for example, Ref. 31). The initial structure of the molecule's energy levels and the structure that appears under the influence of the driving field are sketched in Fig. 1.

In the basis (11)–(13), the starting Hamiltonian (1) takes the following form:

$$\hat{H} = \frac{\varepsilon_0 + \varepsilon_2 + 2\Omega_R\hbar}{3}\sum_{\alpha=1}^{N}(R_{\alpha;12} + R_{\alpha;13}) + \frac{\varepsilon_0 + \varepsilon_2 - 2\Omega_R\hbar}{3}\sum_{\alpha=1}^{N}(R_{\alpha;23} - R_{\alpha;12}) - \frac{2\varepsilon_1}{3}\sum_{\alpha=1}^{N}(R_{\alpha;23} + R_{\alpha;13}) + \frac{\varepsilon_0 - \varepsilon_2}{2}\sum_{\alpha=1}^{N}(e^{i2\Omega_R t}R_{\alpha;12(+)} + e^{-i2\Omega_R t}R_{\alpha;12(-)}) + \hbar\omega_{\nu z}\left(\hat{a}^{+}_{\nu z}\hat{a}_{\nu z} + \frac{1}{2}\right) + \frac{C(\hat{a}^{+}_{\nu z}e^{i\omega_{\nu z}t} + \hat{a}_{\nu z}e^{-i\omega_{\nu z}t})}{\sqrt{2}} \times \sum_{\alpha=1}^{N}(e^{i(\Omega_R - \omega_{10})t}R_{\alpha;13(+)} + e^{i(-\Omega_R + \omega_{10})t}R_{\alpha;13(-)} + e^{i(-\Omega_R - \omega_{10})t}R_{\alpha;23(+)} + e^{i(\Omega_R + \omega_{10})t}R_{\alpha;23(-)}). \qquad (15)$$

Here,

$$C = g\mu_B\langle\varphi_1|\hat{S}_z|\varphi_0\rangle\sqrt{\frac{\hbar\omega_{\nu z}}{2}}H_{\nu z} \qquad (16)$$

is a parameter of the magnetic molecule's interaction with the radiation field (9) and $H_{\nu z} \sim (\omega_{\nu z}/c)^{3/2}$ is the amplitude of the coordinate dependence of the magnetic field, which is produced by a resonator in the molecule's location. The operators $\hat{R}_\alpha$ are

$$\hat{R}_{\alpha;12(\pm)} = \frac{1}{2}\left[\begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}_\alpha \pm i\begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}_\alpha\right], \qquad (17)$$

$$\hat{R}_{\alpha;12} = \frac{1}{2}[\hat{R}_{\alpha;12(+)};\hat{R}_{\alpha;12(-)}] = \frac{1}{2}\begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}_\alpha, \qquad (18)$$

$$\hat{R}_{\alpha;13(\pm)} = \frac{1}{2}\left[\begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}_\alpha \pm i\begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}_\alpha\right], \qquad (19)$$

$$\hat{R}_{\alpha;13} = \frac{1}{2}[\hat{R}_{\alpha;13(+)};\hat{R}_{\alpha;13(-)}] = \frac{1}{2}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}_\alpha, \qquad (20)$$

$$\hat{R}_{\alpha;23(\pm)} = \frac{1}{2}\left[\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}_\alpha \pm i\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}_\alpha\right], \qquad (21)$$

$$\hat{R}_{\alpha;23} = \frac{1}{2}[\hat{R}_{\alpha;23(+)};\hat{R}_{\alpha;23(-)}] = \frac{1}{2}\begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}_\alpha. \qquad (22)$$

Here, the numeration of the columns of the matrices (top-down) and the rows of the matrices (from left to right) corresponds to the sequence of the functions $\psi_{1,\alpha}$, $\psi_{2,\alpha}$, and $\psi_{3,\alpha}$.

Thus, commutative relations for the operators related to one molecule are obvious. The operators related to the different molecules commute. These operators (17)–(22) are similar to the suitable operators for a two-level system.[38]

As follows from the expression for the Hamiltonian (15), the molecule's interaction with the radiation field has a resonance character. The frequency $\omega_{\nu z} = \omega_{10} - \Omega_R$ corresponds to the transition between the state $\psi_{3,\alpha}$ (13) and the quasienergy state $\psi_{1,\alpha}$ (11), and the frequency $\omega_{\nu z} = \omega_{10} + \Omega_R$ corresponds to the transition between the state $\psi_{3,\alpha}$ (13) and the quasienergy state $\psi_{2,\alpha}$ (12) (see Fig. 1).

As mentioned above, we will consider the single-mode radiation regime. Furthermore, the frequency of this mode will be assumed to be equal to one of the transition frequencies in the system of the formed quasienergy levels, and this can be achieved by using a resonator of good quality.

For distinctness, it is assumed that

$$\omega_{\nu z} = \omega_{10} - \Omega_R \qquad (23)$$

(the corresponding transition is shown by the vertical arrow with one arrowhead in Fig. 1) and instead of expression (15), we have the following:

$$\hat{H} = \frac{\varepsilon_0 + \varepsilon_2 + 2\Omega_R\hbar}{3}\sum_{\alpha=1}^{N}(\hat{R}_{\alpha;12} + \hat{R}_{\alpha;13}) + \frac{\varepsilon_0 + \varepsilon_2 - 2\Omega_R\hbar}{3}\sum_{\alpha=1}^{N}(\hat{R}_{\alpha;23} - \hat{R}_{\alpha;12}) - \frac{2\varepsilon_1}{3}\sum_{\alpha=1}^{N}(\hat{R}_{\alpha;23} + \hat{R}_{\alpha;13}) + \frac{\varepsilon_0 - \varepsilon_2}{2}\sum_{\alpha=1}^{N}(e^{i2\Omega_R t}\hat{R}_{\alpha;12(+)} + e^{-i2\Omega_R t}\hat{R}_{\alpha;12(-)}) + \hbar\omega_{\nu z}\left(\hat{a}^{+}_{\nu z}\hat{a}_{\nu z} + \frac{1}{2}\right) + \frac{C}{\sqrt{2}}\sum_{\alpha=1}^{N}(\hat{a}^{+}_{\nu z}\hat{R}_{\alpha;13(+)} + \hat{a}_{\nu z}\hat{R}_{\alpha;13(-)}). \qquad (24)$$

Note that the last term in Eq. (24) describes the molecule's interaction with the radiation field and corresponds to $\hat{H}^{\text{rad}}_{\text{int}}$. The others terms describe the molecules in the driving field and the energy of the radiation field.

III. SUPERRADIANCE KINETICS

According to the chosen interaction representation, the time evolution of a density matrix in the absence of relaxation is defined by the operator $\hat{H}^{\text{rad}}_{\text{int}}$ as follows:

$$i\hbar\frac{d\hat{\rho}}{dt} = [\hat{H}^{\text{rad}}_{\text{int}};\hat{\rho}]. \qquad (25)$$

The exact description of the time evolution requires taking into account different relaxation processes. For simplicity, we will use the relaxation time approach and take into consideration a defined lifetime of the radiated quantum τ. If the resonator has the quality factor $Q_{\nu z}$, then this time can be determined by the following expression:

$$\tau \sim \frac{Q_{\nu z}}{\omega_{\nu z}}. \tag{26}$$

Furthermore, we will take into account the relaxation time of the dipole moments of the molecules T_2. Such relaxation is caused generally by the dipole-dipole interactions between the molecules. We emphasize that using the relaxation time for considering the dephasing influence of the dipole-dipole interaction is nothing but an approximation. Here, it is essential to point to Refs. 30 and 39, where the authors have used computer calculations. However, we assume that using the relaxation time approach allows one to qualitatively describe the phenomenon under consideration.

By taking into consideration this relaxation process and using a semiclassical approximation, one can find a system of equations for ensemble-averaged variables and correlators[40–43] as follows:

$$\frac{dn}{dt} + \frac{n}{\tau} = \frac{F}{T_0}, \tag{27}$$

$$\frac{dF}{dt} + \frac{F}{2\tau} + \frac{F}{2T_2} = \frac{2}{T_0}(S + S_0 + 2Rn), \tag{28}$$

$$\frac{dS}{dt} + \frac{S}{T_2} = \frac{2FR}{T_0}, \tag{29}$$

$$\frac{dR}{dt} = -\frac{F}{T_0}. \tag{30}$$

Here,

$$n = \langle \hat{a}^{\dagger}_{\nu z}\hat{a}_{\nu z}\rangle \tag{31}$$

is the number of quanta of the radiation field,

$$F = i\left\langle \sum_{\alpha=1}^{N} (\hat{R}_{\alpha,13(-)}\hat{a}_{\nu z} - \hat{R}_{\alpha,13(+)}\hat{a}^{\dagger}_{\nu z})\right\rangle \tag{32}$$

is a correlator that describes the effects of the correlation between the magnetic dipole moments (corresponding to $\psi_{1,\alpha} \leftrightarrow \psi_{3,\alpha}$ transitions) and the radiation field,

$$S = \left\langle \sum_{\alpha;\alpha'}^{N;N} \hat{R}_{\alpha,13(-)}\hat{R}_{\alpha',13(+)} \right\rangle_{\alpha \neq \alpha'}, \tag{33}$$

is a correlator that describes the effects of the correlation between the magnetic dipole moments (corresponding to $\psi_{1,\alpha} \leftrightarrow \psi_{3,\alpha}$ transitions) of the different molecules,

$$S_0 = \left\langle \sum_{\alpha}^{N} \hat{R}_{\alpha,13(-)}\hat{R}_{\alpha,13(+)} \right\rangle = R + \frac{1}{2}(N_1 + N_3), \tag{34}$$

$$R = -\left\langle \sum_{\alpha}^{N} \hat{R}_{\alpha,13} \right\rangle \tag{35}$$

is the half-difference of the energy level populations of the state $\psi_{3,\alpha}$ and the quasienergy level of the state $\psi_{1,\alpha}$. In Eq. (34), N_1 and N_3 are the populations of the states ψ_1 and ψ_3, respectively. Under our conditions $N_1 + N_3 = \text{const}$,

$$T_0 = \frac{\hbar\sqrt{2}}{C} \tag{36}$$

is a characteristic time corresponding to the parameter C.

The magnitudes in Eqs. (31)–(35) are averaged with the density matrix, which yields Eq. (25). Since the relaxation time of populations is considered to be much longer than T_2, the relaxation term is neglected in Eq. (30).

Note that Eqs. (27)–(30) are identical to the equations describing the well-known model: The system of directly noninteracting two-level atoms, with the atoms being in resonance with a single mode of the radiation field (see, for example, Refs. 40–43). The specificity of our situation is that the single radiation mode corresponds to the transitions between the energy state $\psi_{3,\alpha}$ and the quasienergy state $\psi_{1,\alpha}$ rather than between the initial energy states.

To solve the system of Eqs. (27)–(30), it is necessary to define the initial conditions at the moment $t=0$: $n(0)$, $F(0)$, $S(0)$, $R(0)$, N_1, and N_3. The initial point of time ($t=0$) is thought to be the switching moment onto the driving field. We suppose that up to this moment ($t<0$), the molecules are in thermodynamic equilibrium and are statistically independent (keep in mind that the molecules do not interact with each other directly) and that there are no quanta of radiation field in the system and, therefore, no correlation between the state of the molecules and the radiation field. It is evident that at the moment $t=0$ (as for $t<0$), there is no correlation between the states of the molecules and between the state of the molecules and the radiation field. That is,

$$n(0) = 0, \tag{37}$$

$$F(0) = 0, \tag{38}$$

$$S(0) = 0. \tag{39}$$

As mentioned above, the temperature T is small enough ($\hbar\omega_{10}/k_B \ll T \ll \hbar\omega_{20}/k_B$) so that for $t<0$, all the molecules are in the states of the fundamental doublet φ_0, φ_1 only, and these states are equally populated. Thus, the probability to find the molecule in the state φ_0 or φ_1 is equal to 1/2, while the probability to find the molecule in the states φ_2 and φ_3 is equal to zero. A switch to the driving field at the moment $t=0$ does not disturb the φ_1 state. Therefore, according to Eq. (13), the probability of being a molecule in the $\psi_{3,\alpha}$ state is equal to 1/2. By using Eqs. (11) and (12), it is easy to show that for the moment $t=0$, the molecule will be in the $\psi_{1,\alpha}$ and $\psi_{2,\alpha}$ states, respectively, with probabilities being equal to 1/4. Therefore, for $R(0)$, we have the following from Eqs. (35) and (20):

$$R(0) = -\langle \hat{R}_{13}\rangle_{t=0} = -\left\langle \frac{1}{2}\sum_{\alpha}\begin{pmatrix}1 & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & -1\end{pmatrix}_{\alpha}\right\rangle_{t=0}$$

$$= -\frac{N}{2}\left\langle\begin{pmatrix}1 & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & -1\end{pmatrix}\right\rangle_{t=0} = \frac{N}{8}. \qquad (40)$$

By using Eq. (34) and the fact that the total probability of being a molecule in the states ψ_1 and ψ_3 is equal to 3/4, we have the following:

$$\frac{1}{2}(N_1 + N_3) = \left\langle \frac{1}{2}\sum_{\alpha}\begin{pmatrix}1 & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & 1\end{pmatrix}_{\alpha}\right\rangle = \frac{N}{2}\left\langle\begin{pmatrix}1 & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & 1\end{pmatrix}\right\rangle = \frac{3}{8}N. \qquad (41)$$

The combined Eqs. (27)–(30) contain the time scales τ, T_2, and T_0. However, the solution of this system is susceptible to the initial conditions (37)–(41); therefore, another time scale comes up as follows:

$$\tau_c = \frac{T_0^2}{\tau N}. \qquad (42)$$

It is well known that if the time scales are tied together by the following condition:

$$\tau \ll \tau_c \ll T_2 \qquad (43)$$

when the regime of superradiance is realized,[40–43] in particular, in the extreme case ($T_2 \to \infty$), the solution of the system of Eqs. (27)–(30) with the initial conditions (37)–(41) will give the following simple expressions for the population difference ($2R$) and intensity ($I = n/\tau$):

$$2R = -\frac{N}{4}\tanh\left(\frac{t - t_0}{2\tau_c}\right), \qquad (44)$$

$$I = \frac{n}{\tau} = \frac{N}{16\tau_c}\operatorname{sec}^2\left(\frac{t - t_0}{2\tau_c}\right). \qquad (45)$$

Here,

$$t_0 \simeq 2\tau_c \ln N. \qquad (46)$$

One can see from Eqs. (44) and (45) that these equations describe a superradiance pulse with the duration $\tau_c \sim N^{-1}$ at the delay time of a superradiance pulse t_0, and the upper bound of the intensity is proportional to N^2 as follows:

$$I_{\max} \simeq \left(\frac{n}{\tau}\right)_{\max} = \frac{N^2\tau}{16T_0^2}. \qquad (47)$$

Numerical solutions for different values of the time scales are of interest. In particular, in contrast to Eq. (43), we do not suppose that $\tau_c \ll T_2$, but rather $\tau_c \leq T_2$. The numerical calculations are given in the next section.

IV. NUMERICAL CALCULATIONS AND ESTIMATIONS

First of all, we estimate the values of the dc magnetic field, the amplitude of the driving field, and the quality factor of the resonator that should allow us to realize the problem under consideration.

To use the quasienergy approach, one has to take into account that the time corresponding to the Rabi frequency of the driving field is much shorter than the characteristic times of all other processes considered in the system (as the relaxation time T_2 and the characteristic time of the collective radiation or the superradiance time τ_c). Therefore, the Rabi frequency is supposed to be of the order of 10^8 s^{-1}, which, according to Eq. (14), fits the value of the driving field amplitude $H_x \sim 5$ Oe [for Fe$_8$, $D = 0.23$ K, $g = 2$, and $\hat{H}_{tr} = K\hat{S}_y^2$, where $K/D = 0.4$ (see Ref. 9)]: it corresponds to the intensity of the order of 10^4 W/cm^2.

To calculate the molecule's spectrum, we use the experimental results,[44] where the transitions between the states of fundamental doublet of Fe$_8$ are observed. The form of the Hamiltonian of the anisotropy, the constants of the anisotropy, and the total spin S of the magnetic molecule determine the transition frequencies. Using the anisotropy constants that satisfy the experimental data (namely, the transition frequencies of the magnetic molecule) allows one to find out the frequencies $\omega_{10} \simeq 10^{10}$ s^{-1} and $\omega_{20} \simeq 5 \times 10^{11}$ s^{-1} for $H_0 = 2 \times 10^4$ Oe.

So as to separate the frequency $\omega_{10} - \Omega_R$ from the frequency $\omega_{10} + \Omega_R$, the resonator has to have a width $\Delta\omega \leq \Omega_R \simeq 10^8$ s^{-1} (with the quality factor being $Q \sim \frac{\omega_{10}}{\Delta\omega} \sim 10^2$). This allows one to estimate the photon lifetime $\tau \sim 10^{-8}$ s according to Eq. (26).

Since the pulse duration τ_c is supposed to exceed τ to estimate the orders of N and τ_c, we have

$$\tau \ll \tau_c \simeq \frac{T_0^2}{\tau N}. \qquad (48)$$

The value of the characteristic time T_0 (36) can be found by using Eq. (16). Assuming that $\omega_{\nu z} \leq 10^{10}$ s^{-1}, we have $T_0 \sim 10$ s.

Thus, as follows from Eqs. (42) and (46), for $N \sim 0.5 \times 10^{17}$ (which corresponds to the volume $V \sim 10^{-4}$ cm^3 for Fe$_8$ according to Ref. 44), the superradiance regime is realized with the pulse duration $\tau_c \sim 10^{-7}$ s and the delay time of the superradiance pulse $t_0 \sim 6 \times 10^{-6}$ s. Note that this estimation corresponds to idealizing $T_2 \to \infty$. Further, it is important to carry out the estimations to find the value T_2 since the observation of a superradiance requires the condition

$$\tau \ll \tau_c < T_2. \qquad (49)$$

By this, we mean a high-quality sample having an ellipsoidal shape and a highly homogeneous magnetic field applied to the sample.[17] Therefore, the relaxation time T_2 is determined by the variation of the local dipole field[22] and at a low temperature $(T_2)_{\text{low}} \sim T^{-1}$. It can be thought[22] that for $T \leq 0.1$ K, $T_2 > 10^{-6}$ s (see also Ref. 28).

In our model, we note that in the absence of the driving field, the molecules have to be distributed into equal parts in the states of the fundamental doublet. The number of molecules in the first excited doublet is very small. The temperatures ensuring such distribution (for $\omega_{10} \simeq 10^{10}$ s^{-1}) are of the order of $T \sim 0.1$ K.

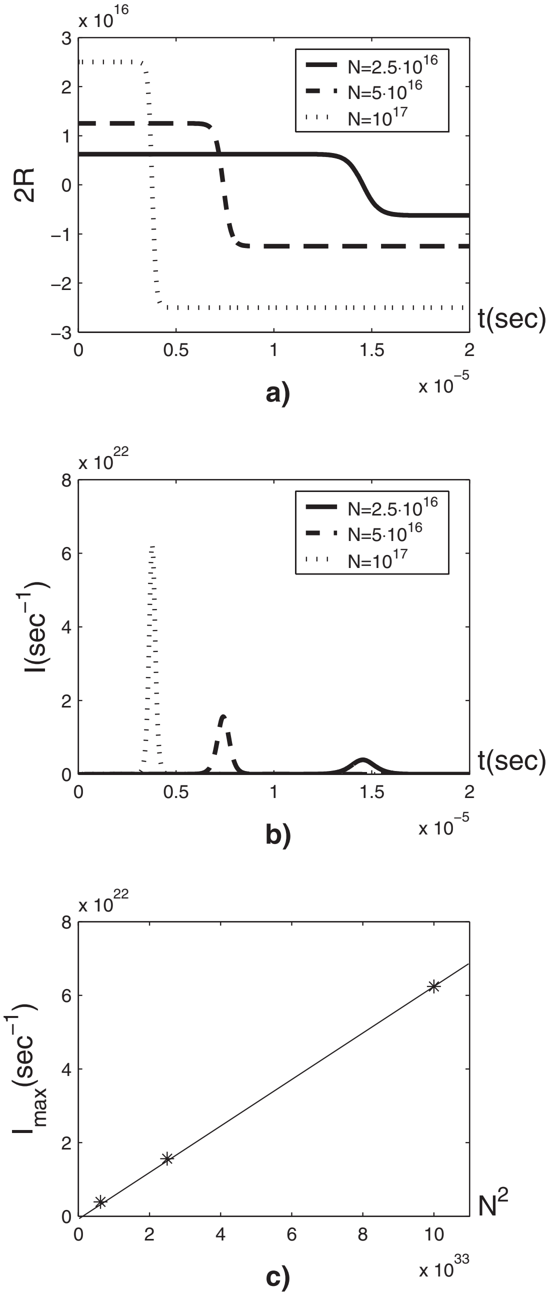

FIG. 2. Time dependences of (a) the population difference $2R$, (b) the intensity I, and (c) corresponding dependence of $I_{\max}$ on N^2 for $T_2 \to \infty$. The calculations were carried out using formulas (42) and (44)–(46) for three values of N ($N=2.5\times10^{16} \Rightarrow \tau_c=4\times10^{-7}$ s, $t_0 \simeq 1.5\times10^{-5}$ s; $N=5\times10^{16} \Rightarrow \tau_c=2\times10^{-7}$ s, $t_0 \simeq 7.4\times10^{-6}$ s; and $N=10^{17} \Rightarrow \tau_c=10^{-7}$ s, $t_0 \simeq 3.8\times10^{-6}$ s).

Therefore, the numerical computations will correspond to the solution of the system of Eqs. (27)–(30) for the chosen parameters $\tau=10^{-8}$ s and $T_0=10$ s and for different values of N ($N \leq 10^{17}$) and T_2.

By comparing with a realistic situation ($T_2^{-1} \neq 0$), Fig. 2 shows the population difference $2R$, the intensity $I=\frac{n}{\tau}$, and the corresponding dependence of $I_{\max}$ on N^2, which are calculated according to Eqs. (42) and (44)–(46), for the idealized situation $T_2 \to \infty$.

Figure 3 shows the magnitudes $2R$, $I=n/\tau$, and the corresponding dependence of $I_{\max}$ on N^2, which are numerically calculated using the combined Eqs. (27)–(30), for the idealized situation $T_2 \to \infty$. The same magnitudes, which are numerically calculated using the combined Eqs. (27)–(30), for the different values $T_2^{-1} \neq 0$ (the realistic situation) are shown in Figs. 4 and 5.

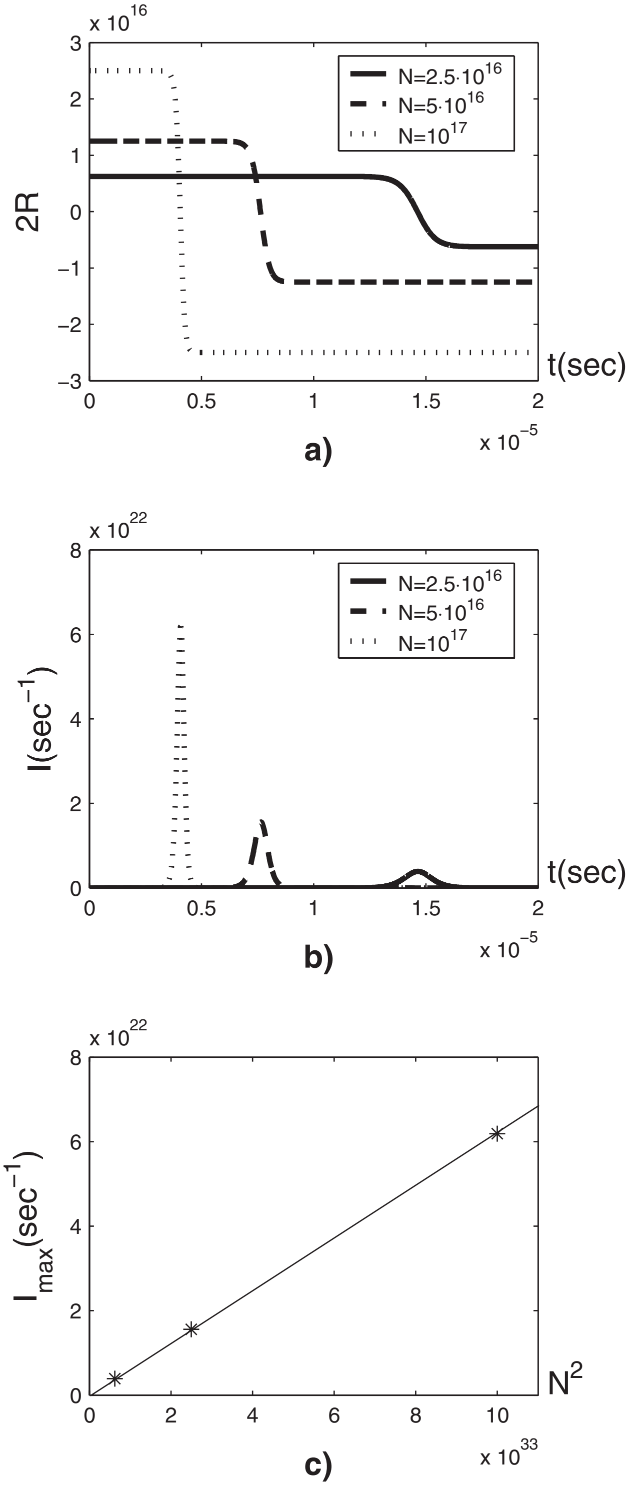

FIG. 3. Time dependences of (a) the population difference $2R$, (b) the intensity I, and (c) the corresponding dependence of $I_{\max}$ on N^2 for $T_2 \to \infty$. Numerical solution.

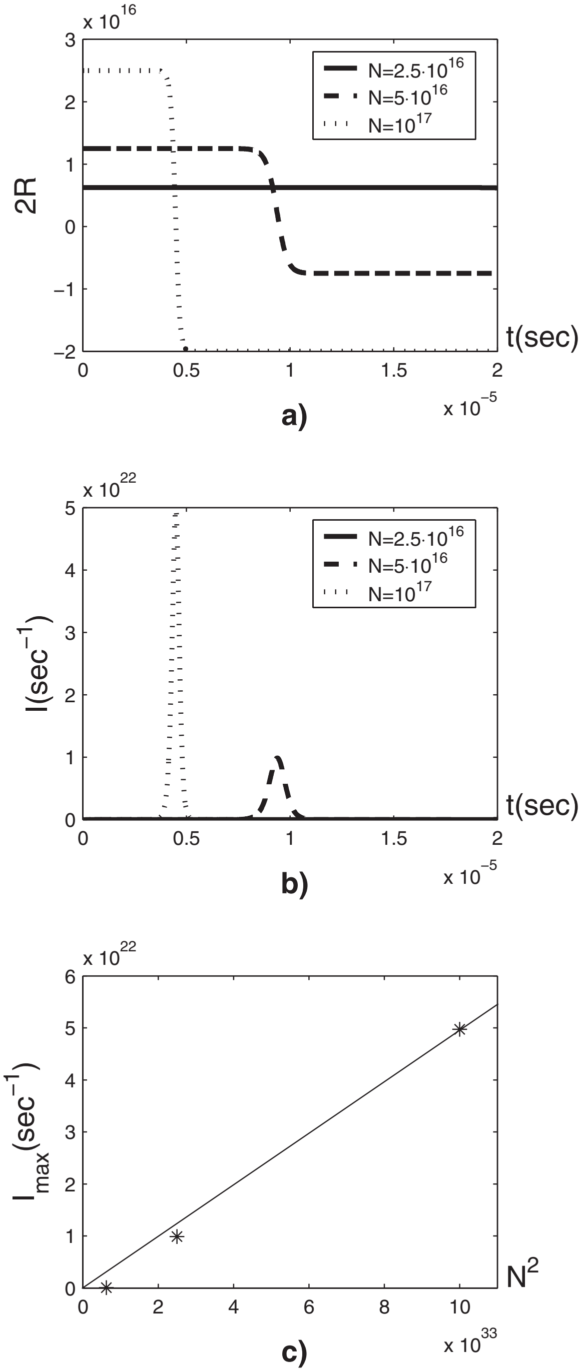

FIG. 4. Time dependences of (a) the population difference $2R$, (b) the intensity I, and (c) the corresponding dependence of $I_{\max}$ on N^2 for $T_2=10^{-6}$ s. Numerical solution. For (c), the straight line is drawn through the origin of the coordinates and the point corresponding to the value $N=10^{17}$.

It is clear from the obtained results that for the chosen region of the parameters and for the idealized case $T_2 \rightarrow \infty$, the calculations using the formulas (42) and (44)–(46) (see Fig. 2) and the numerical solution of the combined Eqs. (27)–(30) (see Fig. 3) give almost identical results. The characteristic superradiance dependence $I_{\max} \sim N^2$ takes place here [Figs. 2(c) and 3(c)]. On the other hand, for the same region of the parameters but at $T_2^{-1} \neq 0$, the decrease in T_2 causes the increase in the superradiance pulse width and the increase in the delay time of the superradiance pulse t_0 [Figs. 4(b) and 5(b)]. The decrease in T_2 leads to the fact that the radiation ceases to have a character of the superradiance, i.e.,

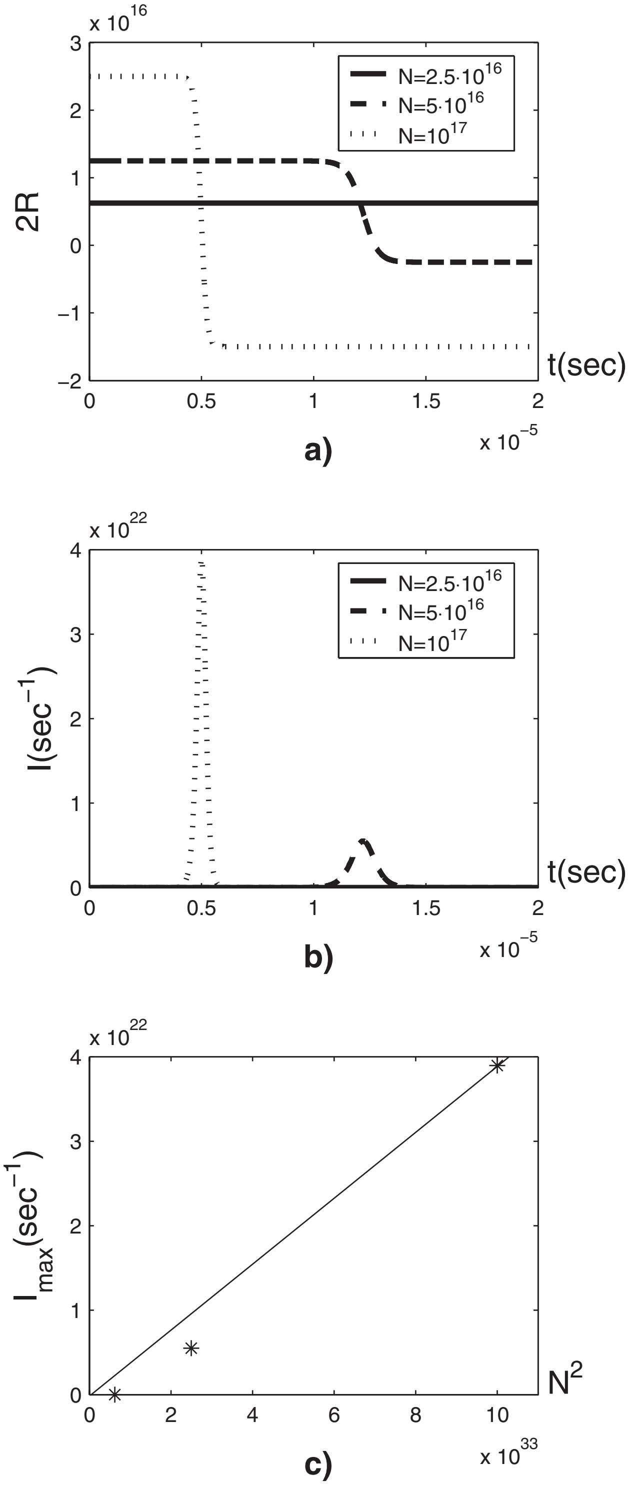

FIG. 5. Time dependences of (a) the population difference $2R$, (b) the intensity I, and (c) the corresponding dependence of $I_{\max}$ on N^2 for $T_2=5\times10^{-7}$ s. Numerical solution. For (c), the straight line is drawn through the origin of the coordinates and the point corresponding to the value $N=10^{17}$.

$I_{\max}$ is not proportional to N^2 although it has a pulse nature. We note that, as follows from the results shown in Figs. 4 and 5, the delay time of the superradiance pulse exceeds the relaxation time T_2 and the superradiance pulse width is a few times less than T_2 only. Note that, as is mentioned earlier,[41] the condition $T_2 > t_0$ is not necessary to observe superradiance.

V. CONCLUSIONS

We have theoretically shown that a crystal of molecular magnets subject to a dc magnetic field under the influence of a strong ac magnetic field can become the source of an electromagnetic superradiance. The superradiance at times shorter than the relaxation times appears as the effect of the radiation process correlation of single molecules, the spectrum of which is essentially modified by a strong ac magnetic field. The estimations pointing to the possibility of realistically observing the effect have been carried out.

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