

Towards a microscopic understanding of the phonon bottleneck

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The problem of the phonon bottleneck in the relaxation of two-level systems (spins) to a narrow group of resonant phonons via emission-absorption processes is investigated from first principles. It is shown that the kinetic approach based on the Pauli master equation is invalid because of the narrow distribution of the phonons exchanging their energy with the spins. This results in a long-memory effect that can be best taken into account by introducing an additional dynamical variable corresponding to the nondiagonal matrix elements responsible for spin-phonon correlation. The resulting system of dynamical equations describes the phonon-bottleneck plateau in the spin excitation, as well as a gap in the spin-phonon spectrum, for any finite concentration of spins. On the other hand, it does not accurately render the line shape of emitted phonons and still needs improving.

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I. INTRODUCTION

The problem of the phonon bottleneck (PB) was recognized in 1941 by Van Vleck who started the abstract of his article¹ with the sentence “The present paper is rather negative in character.” Analyzing different ways for the phonons emitted by magnetic impurities (henceforth spins) to relax, Van Vleck found that in many typical cases the phonon rates are insufficient to keep the phonon subsystem at equilibrium. The phonons emitted by *direct processes*, forming a narrow resonant group in the energy space, should be absorbed by spins again, which drastically throttles the net relaxation rate of the latter. This happens, of course, if the concentration of spins is large enough. There is no PB for a single spin interacting with phonons in a macroscopic crystal, no matter what the phonon relaxation rate is.

Van Vleck’s “negative finding” excited other researchers for an activity that has not ebbed until now. Subsequent publications^{2,3} explored the analogy with trapping of resonant radiation in gases, considered earlier by Holstein.⁴ The corresponding theory deals with the spatial diffusion of emitted and reabsorbed photons or phonons from the body of the specimen towards its boundaries where they escape (see also the recent Ref. 5). Giordmaine and Nash³ cite a number of early experiments where the PB was observed. Recently indications of the phonon bottleneck were seen in molecular magnets.^{6,7} The usual “fingerprint” of the PB is the decrease of the net spin relaxation rate if the thermal contact between the crystal and the holder is bad. Then the emitted phonons cannot efficiently escape from the crystal, and the only way to their relaxation are nonlinear phonon processes. The dependence of the PB on the thermal contact becomes pronounced at low temperatures, where direct processes dominate. At higher temperatures *Raman processes* strongly come into play. It was shown^{8–10} that a kind of phonon bottleneck takes place for Raman processes as well, but the effect is much weaker than that for direct processes.

Existing theories of the phonon bottleneck for direct processes operate with diffusion, kinetic, or rate equations that are not derived from first principles. These equations are set

up using the balance of the excitation numbers and the energy between the spins and phonons, as well as the Fermi golden rule. The quasicontinuum of resonant phonons is considered in a simplified way as a single dynamical variable. One group of publications^{4,3,5} takes into account the effects of spatial inhomogeneity in the sample. Another group^{8,11–14} further simplifies the problem by ignoring spatial effects and modeling the relaxation of phonons by a single rate of approaching the equilibrium at some bath temperature.

A related class of problems deals with the influence of spins on the phonon spectrum¹⁵ and with the resonance scattering of acoustic waves on spins (see Ref. 16 and references therein). Jacobsen and Stevens¹⁵ showed that the hybridization between spins and phonons in a crystal with *regularly spaced* spins leads to a *gap* or *stop band* in the phonon spectrum around the spin-phonon resonance. This can be considered as an extreme case of the phonon bottleneck, as, obviously, one cannot speak about a unidirectional energy transfer from spins to phonons, at least if the phonons are undamped. If spins are diluted, the resonance phonon gap should persist, in a reduced form. This effect has never been discussed in existing theories of the PB.

Since Van Vleck’s original work, the main stress in the PB problem was put on the sufficiency or insufficiency of the phonon relaxation processes to transport the energy away from the spins. A common idea is that the phonon bottleneck is something that happens if the phonons are not relaxing fast enough. One can find in the literature a dimensionless *bottleneck factor* that becomes large if the phonon relaxation rate is small [see, e.g., Eq. (36) of Ref. 8]. However, numerous publications on the *single-spin-plus-phonon-bath* model do not care for the phonon relaxation at all, which is certainly correct. The problem arises only if the number of spins is macroscopic and their concentration is large enough. Then the PB occurs and, as a consequence, one has to take into account the relaxation of phonons. This means that there should be another dimensionless *bottleneck parameter* that is independent of the phonon relaxation or escape rate.

The important component of the PB problem dealing exclusively with the energy transfer between the spins and resonant phonons has not received a due attention until now.

Starting, for simplicity, with undamped harmonic phonons, one can ask what will be the accurate dynamics of the system, the large-time asymptote of the evolution, and the energy distribution of emitted phonons under the bottleneck condition. One can expect that the system of equations^{8,11-14} describing resonant phonons as a single dynamical variable follows from a more *detailed* energy-resolved theoretical framework via integration over the phonon energies.

Such a detailed description is likely to include an equation of motion for the spin averages coupled to the system of kinetic equations for phonons of all possible modes, considered separately. Are there any difficulties that prevent the derivation of such detailed spin-phonon equations from first principles? It is puzzling why it has not been done yet, given that quantum kinetic equations result from many-body quantum mechanics, under certain conditions. The procedure includes at first the derivation of the Pauli master equation,^{17,18} which breaks the reversibility of the quantum mechanics. Then kinetic equations for particular observables can be obtained by averaging appropriate operators with the Pauli master equation.

Studying the mechanism of the energy exchange between the spins and the resonant group of phonons is the aim of this work. Identical spins $S=1/2$ (two-level systems) randomly placed in the crystal are considered. One starts with undamped harmonic phonons and adds their relaxation at the very end in the same simplest way as was done in Refs. 8,11-14. The main finding is that the narrow distribution of emitted phonons violates the condition for the Pauli master equation to be valid. Hence no standard kinetic equations for the resonant phonons can be derived. Instead, one obtains in this way equations with memory that are of little practical use. In this situation it is better to step back and use a more basic system of dynamic equations including nondiagonal elements of the spin-phonon density matrix. Numerical solution of this system of equations shows a phonon bottleneck for the concentration of spins large enough. There is a non-trivial asymptotic plateau for the spin excitation, if the phonons are undamped. For a low level of the initial spin excitation the resulting system of equations can be solved analytically. The analytical solution shows a resonance gap that is similar to that found by Jacobsen and Stevens¹⁵ for nondiluted spins. For the model with damped phonons, the system eventually relaxes to thermal equilibrium. However, in the case of a strong bottleneck, the effective relaxation rate for the spins is much less than the phonon relaxation rate, contrary to the first-glance expectation.

The following parts of the article are organized as follows. Section II sets up the Hamiltonian of the spin-phonon system, its quantum states, and the Schrödinger equation. In Sec. III the derivation of the Pauli master equation is reviewed and its applicability conditions are discussed. It is argued that for a narrow resonant group of phonons the standard kinetic formalism based on the master equation should fail because of the long-memory effect. Section IV contains a discussion of the conditions for the phonon bottleneck, and the dimensionless *bottleneck parameter* is introduced. Section V explores the short-memory approach to the PB based on the master equation. One obtains a system of coupled kinetic equations for spins and phonons that contain powers

of the energy δ function and is thus incorrect. It is shown how one can obtain previously published bottleneck equations^{8,11-14} by mathematically nonrigorous manipulations with δ functions. In Sec. VI one steps back to the master equation with memory and obtains a system of *dynamical* equations describing the emission and absorption of phonons by spins and the bottleneck. These equations are enhanced by including terms responsible for the relaxation of phonons in a simple way. Numerical solutions are presented and discussed. Further the analytical solution of the system of dynamical spin-phonon equations in the case of low spin excitation is presented, and analytical results for the bottleneck plateau in the spin excitation and the effective relaxation rate of the spin-phonon system under the bottleneck condition are obtained. Discussion is done throughout the article. Section VII contains a summary of the results obtained, as well as a discussion of the inhomogeneous broadening and the interplay between the phonon bottleneck and phonon superradiance.¹⁹

II. THE HAMILTONIAN AND SCHRÖDINGER EQUATION

Consider a spin-phonon Hamiltonian for N_S two-level systems (spins) within an elastic body of N cells:

$$\hat{H} = \hat{H}_0 + \hat{V}, \quad (1)$$

where

$$\hat{H}_0 = -\frac{\hbar\omega_0}{2} \sum_i \sigma_{iz} + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \quad (2)$$

describes spins and harmonic phonons, σ being the Pauli matrix. Neglecting processes that do not conserve energy, one can write the spin-phonon interaction \hat{V} in the rotating-wave approximation as

$$\hat{V} = -\frac{\hbar}{\sqrt{N}} \sum_i \sum_{\mathbf{k}} (A_{i\mathbf{k}}^* X_i^{01} a_{\mathbf{k}}^\dagger + A_{i\mathbf{k}} X_i^{10} a_{\mathbf{k}}), \quad (3)$$

where $A_{i\mathbf{k}} \equiv V_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{r}_i}$. Below \hat{V} will be treated as a perturbation. The operator $X^{10} \equiv \sigma_-$ brings the spin from the ground state $|\uparrow\rangle \equiv |0\rangle \equiv \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ to the excited state $|\downarrow\rangle \equiv |1\rangle \equiv \begin{pmatrix} 0 \\ 1 \end{pmatrix}$ while $X^{01} \equiv \sigma_+$ does the opposite.

To describe quantum states of the system of phonons and spins, we use a basis that is a direct product of the states of all phonon modes and of the spins. The wave function can be written in the form

$$\Psi = \sum_{\{\mu_i\}\{\nu_{\mathbf{k}}\}} c_{\{\mu_i\}\{\nu_{\mathbf{k}}\}} \prod_i |\mu_i\rangle \prod_{\mathbf{k}} |\nu_{\mathbf{k}}\rangle, \quad (4)$$

where $\mu_i=0,1$ correspond to the ground and excited states of the spins, respectively, and $\nu_{\mathbf{k}}=0,1,2,\dots$ are the occupation numbers of the phonon modes. We will use the shortcuts for different basis states:

$$\{\mu_i\} \equiv S, \quad \{\nu_{\mathbf{k}}\} \equiv \mathcal{P}, \quad \{\mu_i\}\{\nu_{\mathbf{k}}\} = (S\mathcal{P}) \equiv \mathcal{W}. \quad (5)$$

The Schrödinger equation (SE) for our spin-phonon system $i\hbar d\Psi/dt = \hat{H}\Psi$ can be written as a system of equations for the coefficients $c_{\mathcal{W}}$, which can be obtained by acting on Ψ with the Hamiltonian of Eq. (1). The general form of the SE for the coefficients is

$$i\hbar \frac{d}{dt} c_{\mathcal{W}} = \sum_{\mathcal{W}_1} c_{\mathcal{W}_1} \langle \mathcal{W} | \hat{H} | \mathcal{W}_1 \rangle = \varepsilon_{\mathcal{W}} c_{\mathcal{W}} + \sum_{\mathcal{W}_1} c_{\mathcal{W}_1} \langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle, \quad (6)$$

where

$$\varepsilon_{\mathcal{W}} = \langle \mathcal{W} | \hat{H}_0 | \mathcal{W} \rangle = \hbar \omega_{\mathcal{W}} \quad (7)$$

is the unperturbed energy in the state \mathcal{W} . It is convenient to introduce the slow amplitudes $\tilde{c}_{\mathcal{W}}$ via

$$c_{\mathcal{W}}(t) = \tilde{c}_{\mathcal{W}}(t) e^{-i\omega_{\mathcal{W}} t}; \quad (8)$$

then, the SE becomes

$$\frac{d}{dt} \tilde{c}_{\mathcal{W}} = - \frac{i}{\hbar} \sum_{\mathcal{W}_1} e^{i(\omega_{\mathcal{W}} - \omega_{\mathcal{W}_1}) t} \tilde{c}_{\mathcal{W}_1} \langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle. \quad (9)$$

Let us work out the concrete form of the SE for the spin-phonon interaction \hat{V} given by Eq. (3). In the matrix element $\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle$ the state \mathcal{W}_1 differs from \mathcal{W} by one spin flip and creation or annihilation of one phonon. Thus it is convenient to write \mathcal{W}_1 in incremental form. In particular, $\mathcal{W}_1 = \mathcal{W}, 1_i, -1_{\mathbf{k}}$ means that in \mathcal{W}_1 the spin on site i is excited and one phonon in the \mathbf{k} mode is annihilated, relative to \mathcal{W} . This can only happen if $\mu_i = 0$ in \mathcal{W} . In the state $\mathcal{W}_1 = \mathcal{W}, 0_i, +1_{\mathbf{k}}$ the spin on site i is deexcited and one phonon in the \mathbf{k} mode is created, relative to \mathcal{W} . This can only happen if $\mu_i = 1$ in \mathcal{W} . One obtains

$$\begin{aligned} \langle \mathcal{W} | \hat{V} | \mathcal{W}, 0_i, +1_{\mathbf{k}} \rangle &= - \frac{\hbar}{\sqrt{N}} A_{i\mathbf{k}} \mu_i \sqrt{\nu_{\mathbf{k}} + 1}, \\ \langle \mathcal{W} | \hat{V} | \mathcal{W}, 1_i, -1_{\mathbf{k}} \rangle &= - \frac{\hbar}{\sqrt{N}} A_{i\mathbf{k}}^* (1 - \mu_i) \sqrt{\nu_{\mathbf{k}}}, \end{aligned} \quad (10)$$

where μ_i and $\nu_{\mathbf{k}}$ refer to the state \mathcal{W} . With the help of this, Eq. (9) can be written in the form

$$\begin{aligned} \frac{d\tilde{c}_{\mathcal{W}}}{dt} &= \frac{i}{\sqrt{N}} \sum_i \sum_{\mathbf{k}} e^{-i(\omega_{\mathbf{k}} - \omega_0) t} A_{i\mathbf{k}} \mu_i \sqrt{\nu_{\mathbf{k}} + 1} \tilde{c}_{\mathcal{W}, 0_i, +1_{\mathbf{k}}} \\ &+ \frac{i}{\sqrt{N}} \sum_i \sum_{\mathbf{k}} e^{i(\omega_{\mathbf{k}} - \omega_0) t} A_{i\mathbf{k}}^* (1 - \mu_i) \sqrt{\nu_{\mathbf{k}}} \tilde{c}_{\mathcal{W}, 1_i, -1_{\mathbf{k}}}. \end{aligned} \quad (11)$$

III. PAULI MASTER EQUATION

Let us construct now the elements of the slow density matrix for our closed system of spins and phonons by multiplying coefficients of the wave function

$$\tilde{\rho}_{\mathcal{W}|\mathcal{W}'} = \tilde{c}_{\mathcal{W}} \tilde{c}_{\mathcal{W}'}^*. \quad (12)$$

The equation of motion for them follows from Eq. (9) and reads

$$\begin{aligned} \frac{d}{dt} \tilde{\rho}_{\mathcal{W}|\mathcal{W}'} &= - \frac{i}{\hbar} \sum_{\mathcal{W}_1} \langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle e^{i(\omega_{\mathcal{W}} - \omega_{\mathcal{W}_1}) t} \tilde{\rho}_{\mathcal{W}_1|\mathcal{W}'} \\ &+ \frac{i}{\hbar} \sum_{\mathcal{W}'_1} \langle \mathcal{W}' | \hat{V} | \mathcal{W}'_1 \rangle^* e^{-i(\omega_{\mathcal{W}'} - \omega_{\mathcal{W}'_1}) t} \tilde{\rho}_{\mathcal{W}|\mathcal{W}'_1}. \end{aligned} \quad (13)$$

We restrict our consideration to the case of a nearly diagonal density matrix of the spin-phonon system. This will be the case if the spins are prepared in the initial state with random phases, so that the averages of the transverse spin components are zero, $\langle S_x \rangle = \langle S_y \rangle = 0$, and the phonons are at thermal equilibrium. We are going to derive equations for the populations of the quantum states

$$P_{\mathcal{W}} = \tilde{\rho}_{\mathcal{W}|\mathcal{W}}. \quad (14)$$

One can see from Eqs. (13) with $\mathcal{W}' = \mathcal{W}$ that diagonal elements $\tilde{\rho}_{\mathcal{W}_1|\mathcal{W}}$ are dynamically coupled to nondiagonal elements such as $\tilde{\rho}_{\mathcal{W}_1|\mathcal{W}}$. The latter are generated by \hat{V} and thus are small. One can integrate them out using equations similar to Eq. (13). This yields

$$\begin{aligned} \tilde{\rho}_{\mathcal{W}_1|\mathcal{W}}(t) &= \tilde{\rho}_{\mathcal{W}_1|\mathcal{W}}(t_0) \\ &- \frac{i}{\hbar} \sum_{\mathcal{W}_2} \langle \mathcal{W}_1 | \hat{V} | \mathcal{W}_2 \rangle \int_{t_0}^t dt' e^{i(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}_2}) t'} \tilde{\rho}_{\mathcal{W}_2|\mathcal{W}}(t') \\ &+ \frac{i}{\hbar} \sum_{\mathcal{W}_2} \langle \mathcal{W} | \hat{V} | \mathcal{W}_2 \rangle^* e^{-i(\omega_{\mathcal{W}} - \omega_{\mathcal{W}_2}) t} \tilde{\rho}_{\mathcal{W}_1|\mathcal{W}_2} \end{aligned} \quad (15)$$

and $\tilde{\rho}_{\mathcal{W}_1|\mathcal{W}}(t) = (\tilde{\rho}_{\mathcal{W}_1|\mathcal{W}}(t))^*$. Remaining within second order in the perturbation \hat{V} , it is sufficient to drop the nondiagonal terms on the right-hand sides of the above equation. Plugging the resulting expressions for $\tilde{\rho}_{\mathcal{W}_1|\mathcal{W}}(t)$ and $\tilde{\rho}_{\mathcal{W}|\mathcal{W}_1}(t)$ into Eq. (13) yields the master equation with memory

$$\begin{aligned} \frac{d}{dt} P_{\mathcal{W}}(t) &= \frac{2}{\hbar^2} \sum_{\mathcal{W}_1} |\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle|^2 \int_{t_0}^t dt' \cos[(\omega_{\mathcal{W}} - \omega_{\mathcal{W}_1})(t - t')] \\ &\times [P_{\mathcal{W}_1}(t') - P_{\mathcal{W}}(t')]. \end{aligned} \quad (16)$$

In the usual case where the kinetic theory is applicable, $|\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle|^2$ and $P_{\mathcal{W}_1}(t')$ are smooth functions of \mathcal{W}_1 . The summation over \mathcal{W}_1 in Eq. (16) goes over a wide energy interval limited by the maximal energy of the phonon reservoir $\hbar \omega_{\max}$ in some terms and limited by the thermal energy $k_B T$ in other terms. The resulting expression is then peaked at $t - t' \lesssim 1/\omega_{\max}$ or $t - t' \lesssim \hbar/(k_B T)$. Both of these characteristic times are much shorter than the relaxation time $1/\Gamma$ for $P_{\mathcal{W}}(t)$ since the spin-phonon relaxation rate $\Gamma \propto |V_{\mathbf{k}}|^2$ is small. Thus one can make the short-memory approximation $P_{\mathcal{W}}(t') \Rightarrow P_{\mathcal{W}}(t)$ and $P_{\mathcal{W}_1}(t') \Rightarrow P_{\mathcal{W}_1}(t)$ in Eq. (16). After that the time integration can be done easily,

$$\int_{t_0}^t dt' \cos[(\omega_{\mathcal{W}} - \omega_{\mathcal{W}_1})(t - t')] = \frac{\sin[(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}})(t - t_0)]}{\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}}}. \quad (17)$$

In the kinetic time range

$$t - t_0 \sim \frac{1}{\Gamma} \gg \frac{1}{\omega_{\max}}, \quad \frac{\hbar}{k_B T}, \quad (18)$$

one can replace

$$\frac{\sin[(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}})(t - t_0)]}{\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}}} \Rightarrow \pi \delta(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}}), \quad (19)$$

which breaks the time reversibility of the quantum mechanics and leads to the famous Pauli master equation¹⁷

$$\frac{d}{dt} P_{\mathcal{W}} = \sum_{\mathcal{W}_1} \tilde{\Gamma}_{\mathcal{W}\mathcal{W}_1} (P_{\mathcal{W}_1} - P_{\mathcal{W}}), \quad (20)$$

with the Fermi-golden-rule detailed transition rate

$$\tilde{\Gamma}_{\mathcal{W}\mathcal{W}_1} = \frac{2\pi}{\hbar^2} |\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle|^2 \delta(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}}). \quad (21)$$

The sum of the latter over \mathcal{W}_1 ,

$$\Gamma_{\mathcal{W}} = \sum_{\mathcal{W}_1} \tilde{\Gamma}_{\mathcal{W}\mathcal{W}_1} = \frac{2\pi}{\hbar^2} \sum_{\mathcal{W}_1} |\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle|^2 \delta(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}}), \quad (22)$$

is the decay rate of the state \mathcal{W} . One can write $\Gamma_{\mathcal{W}}$ as

$$\Gamma_{\mathcal{W}} = \frac{2\pi}{\hbar^2} N \langle |\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle|^2 \rangle \rho(\omega_{\mathcal{W}}), \quad (23)$$

where N is the total number of phonon modes in the system, which that is proportional to the number of atoms in it, $\langle |\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle|^2 \rangle$ is the average over the resonant states \mathcal{W}_1 , and

$$\rho(\omega_{\mathcal{W}}) = \frac{1}{N} \sum_{\mathcal{W}_1} \delta(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}}) \quad (24)$$

is the density of states, which satisfies $\int d\omega_{\mathcal{W}} \rho(\omega_{\mathcal{W}}) = 1$. The name ‘‘master equation’’ says that one can generate kinetic equations for different physical quantities from it by averaging appropriate operators over the quantum states \mathcal{W} with $P_{\mathcal{W}}$.

The reader can find a more extensive derivation and analysis of the Pauli master equation in Ref. 18. The authors argue that, as Eq. (20) indeed suggests, a system initially in the quantum state \mathcal{W} will spread over all mutually accessible resonant states \mathcal{W}_1 in a nonoscillative way, the final result of the relaxation being the microcanonical distribution $P_{\mathcal{W}_1} \propto \delta(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}})$. However plausible these arguments might appear, there is a problem since one obtains a square of the energy δ function in Eq. (20) in the case of the decay of an initial fully occupied state \mathcal{W} : One δ function is contained in the transition probability $\tilde{\Gamma}_{\mathcal{W}\mathcal{W}_1}$ of Eq. (21), and another one is carried by $P_{\mathcal{W}_1}$. In the derivation of the master equation

above [see discussion below Eq. (16)] it was stressed that $P_{\mathcal{W}_1}(t')$ should be a smooth functions of \mathcal{W}_1 for the short-memory approximation leading to Eq. (20) to be valid. Physically it means that the probability of the quantum system should be not sharp but distributed over many states with different energies. Only in this case can one rigorously derive kinetic equations. In the case of the spin-phonon relaxation via direct processes, the energy of the system is fixed and the emitted phonons build a narrow resonant group in the energy space that is as sharp as the energy δ function describing the probability of this process. It will be demonstrated below that this results in the inapplicability of the kinetic approach to the description of the phonon bottleneck and that effective bottleneck equations of Refs. 8 and 11–14 can only be obtained by mathematically incorrect manipulations with δ functions.

IV. THE ORIGIN OF THE PHONON BOTTLENECK

As we have seen above, the Pauli master equation does not resolve line shapes, which is the origin of the difficulties of applying it to the PB problem. Finite linewidths, however, follow from the time-energy uncertainty principle. In the simple case of one spin in a macroscopic crystal (or one atom in a free space or a large cavity) one can neglect the incoming term in Eq. (20); then, the solution for the decay of the initially prepared state is $P_{\mathcal{W}}(t) = e^{-\Gamma_{\mathcal{W}} t}$. The finite lifetime of the decaying state leads to a finite linewidth of the emitted phonons. At $t \rightarrow \infty$ from the Schrödinger equation one obtains^{20,21}

$$P_{\mathcal{W}_1} = \frac{1}{\hbar^2} |\langle \mathcal{W} | \hat{V} | \mathcal{W}_1 \rangle|^2 \frac{1}{(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}})^2 + \Gamma_{\mathcal{W}}^2/4}, \quad (25)$$

which satisfies $\sum_{\mathcal{W}_1} P_{\mathcal{W}_1} = 1$.

Apart from the Lorentzian dependence on the energy, $P_{\mathcal{W}_1}$ also depends on the matrix element that can differ for different directions of the phonon wave vector \mathbf{k} . One can average $P_{\mathcal{W}_1}$ keeping the frequency $\omega_{\mathcal{W}_1}$ constant. Then using Eq. (23) one can rewrite the result in the form

$$\langle P_{\mathcal{W}_1} \rangle = \frac{2}{\pi N \rho(\omega_{\mathcal{W}}) \Gamma_{\mathcal{W}}} \frac{\Gamma_{\mathcal{W}}^2/4}{(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}})^2 + \Gamma_{\mathcal{W}}^2/4}, \quad (26)$$

which allows us to estimate the probabilities of the states with emitted phonons. At resonance the Lorentzian factor here equals 1, so that the estimation is $\langle P_{\mathcal{W}_1} \rangle \sim 1/N_{\Gamma}$, where

$$N_{\Gamma} = \pi N \rho(\omega_{\mathcal{W}}) \Gamma_{\mathcal{W}} \quad (27)$$

is the effective number of states in resonance with the state \mathcal{W} , taking into account its finite linewidth. $N_{\Gamma} \gg 1$ is the condition for using a quasicontinuum approximation for the states of the system and replacing the sums over \mathcal{W} by integrals. Only if $N_{\Gamma} \gg 1$ is fulfilled can one write $\delta(\omega_{\mathcal{W}_1} - \omega_{\mathcal{W}})$ in Eqs. (21) and (24).

For macroscopic bodies the factor N in Eq. (27) makes N_{Γ} very large indeed. In this case $\langle P_{\mathcal{W}_1} \rangle$ of Eq. (26) is very small. This validates neglecting the incoming terms in the master equation, which leads to $P_{\mathcal{W}}(t) = e^{-\Gamma_{\mathcal{W}} t}$ and Eq. (25). The de-

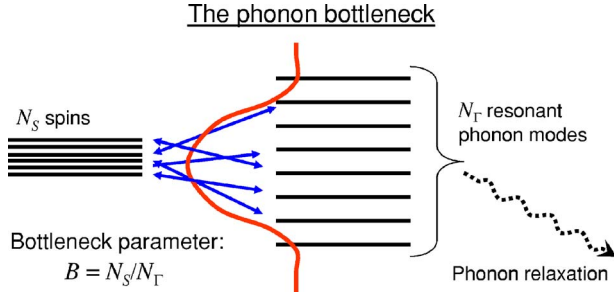


FIG. 1. (Color online) The phonon bottleneck happens for $B \geq 1$, and it results in the reabsorption of the initially emitted phonons by spins. Fast phonon relaxation suppresses the bottleneck.

decay of an initially prepared state into the continuum can be understood from a very simple statistical argument. The excitation initially localized at the state \mathcal{W}_0 becomes equidistributed between the $N_{\Gamma} + 1 \cong N_{\Gamma}$ states as a result of the relaxation. Then the probability to find the system in any of the states $1/N_{\Gamma}$ is extremely small. Once the excited state of the spin has decayed, the excitation never comes back because it is statistically improbable.

For very small crystals the phonon modes can be so sparse that N_{Γ} is of order 1 or even smaller. If the spin is at resonance with only few phonon modes, the dynamics of the system is complicated and the spin cannot relax completely since it is being excited by phonons again. This is the case of the phonon bottleneck. The simplest case of the PB is that of the resonance between the spin and a single phonon mode that is described in quantum-mechanics textbooks as the resonance between two general quantum mechanical states. It is well known that the probability to stay in the initially prepared state is harmonically oscillating and there is no relaxation.

The consideration above in this section pertains to the case of one two-level system (spin) relaxing to a phonon bath at zero temperature. Below we will consider, in particular, relaxation of N_S spins $1/2$ prepared in their excited states and relaxing to a phonon bath at $T=0$ (see Fig. 1). The initial state \mathcal{W} (all spins excited, no phonons) decays, initially, into the states with one spin flipped and one phonon created. All arguments above are still valid for this process. However, the resulting states will further decay into states with more spins flipped and more phonons created. The number of emitted phonons can be by a factor N_S greater than that in the case of one spin. Then, in turn, these phonons will begin to excite the spin subsystem. In this situation one cannot neglect incoming terms in the master equation. A criterion for this can be obtained by the generalization of the statistical argument above: For N_S spins at resonance with N_{Γ} phonon modes one can construct a dimensionless parameter

$$B \equiv \frac{N_S}{N_{\Gamma}} = \frac{N_S}{\pi N \rho(\omega_{\mathcal{W}}) \Gamma_{\mathcal{W}}}, \quad (28)$$

which controls the phonon bottleneck and can be called the *bottleneck parameter*. Note that here $n_s \equiv N_S/N$ is the number of spins per unit cell, so that B is independent of the system size. If $B \lesssim 1$, then the most energy goes to phonons.

If $B \geq 1$, then the energy mainly remains in the spin subsystem, if the resonant phonons do not transfer their energy elsewhere. The latter case corresponds to the PB. It is clear that in most situations the inequality $B \gg 1$ should be fulfilled. Indeed, if one roughly replaces $\rho(\omega_{\mathcal{W}}) \Rightarrow 1/\omega_{\max}$, then $B \gg 1$ is equivalent to $\Gamma_{\mathcal{W}}/\omega_{\max} \ll n_s$. The left part of this inequality is very small, so that the inequality holds for any not too low concentrations of spins.

It is tempting to take into account that the state resulting from the decay of the spin, \mathcal{W}_1 , has its own relaxation rate $\Gamma_{\mathcal{W}_1}$ due to the damping of the emitted phonons. This would lead to the replacement $\Gamma_{\mathcal{W}} \Rightarrow \Gamma_{\mathcal{W}} + \Gamma_{\mathcal{W}_1}$ in Eq. (28), where $\Gamma_{\mathcal{W}} = \Gamma$, the spin relaxation rate due to the phonon emission, and $\Gamma_{\mathcal{W}_1} = \Gamma_{\text{ph}}$, the phonon relaxation rate. Then one can assume that the bottleneck should disappear in the case $\Gamma_{\text{ph}} \gg \Gamma$. Below the reader will see that the sum $\Gamma + \Gamma_{\text{ph}}$ does not arise in the theory. The PB, indeed, is suppressed for large Γ_{ph} , but the condition for this is more subtle and given by Eq. (67).

Inhomogeneous broadening that typically exceeds relaxation rates tends to suppress the PB. Although it would be important for experiments, appropriate extension of the theory will not be explicitly made here, for the sake of transparency. This extension is discussed in the concluding section of the paper.

V. SHORT-MEMORY APPROACH TO THE PHONON BOTTLENECK

In this section we try to derive equations describing the phonon bottleneck from the Pauli master equation (20)—i.e., within the short-memory approximation. We will see that in this way one has to deal with powers of the energy δ function, which forces one to choose a better approach.

A. Spin-phonon master equation

Using the expressions for the matrix elements of Eq. (10), for the detailed transition probabilities of Eq. (21) one obtains

$$\begin{aligned} \tilde{\Gamma}_{\mathcal{W}; \mathcal{W}, 0, +1_{\mathbf{k}}} &= \frac{2\pi}{N} |V_{\mathbf{k}}|^2 \mu_i(\nu_{\mathbf{k}} + 1) \delta(\omega_{\mathbf{k}} - \omega_0), \\ \tilde{\Gamma}_{\mathcal{W}; \mathcal{W}, 1, -1_{\mathbf{k}}} &= \frac{2\pi}{N} |V_{\mathbf{k}}|^2 (1 - \mu_i) \nu_{\mathbf{k}} \delta(\omega_{\mathbf{k}} - \omega_0). \end{aligned} \quad (29)$$

The master equation (20) for the spin-phonon system then becomes

$$\begin{aligned} \frac{d}{dt} P_{\mathcal{W}} &= \frac{2\pi}{N} \sum_i \mu_i \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 (\nu_{\mathbf{k}} + 1) \delta(\omega_{\mathbf{k}} - \omega_0) \\ &\quad \times (P_{\mathcal{W}, 0, +1_{\mathbf{k}}} - P_{\mathcal{W}}) \\ &\quad + \frac{2\pi}{N} \sum_i (1 - \mu_i) \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \nu_{\mathbf{k}} \delta(\omega_{\mathbf{k}} - \omega_0) \\ &\quad \times (P_{\mathcal{W}, 1, -1_{\mathbf{k}}} - P_{\mathcal{W}}). \end{aligned} \quad (30)$$

Let us now redefine $\nu_{\mathbf{k}}$ so that they refer to the state in front of which they stand, instead of referring to the state \mathcal{W} as initially defined. Then the master equation takes the form

$$\begin{aligned} \frac{d}{dt}P_{\mathcal{W}} &= \frac{2\pi}{N} \sum_i \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \mu_i \delta(\omega_{\mathbf{k}} - \omega_0) \\ &\quad \times [\nu_{\mathbf{k}} P_{\mathcal{W}, 0, \nu_{\mathbf{k}}+1_{\mathbf{k}}} - (\nu_{\mathbf{k}} + 1) P_{\mathcal{W}}] \\ &\quad + \frac{2\pi}{N} \sum_i \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 (1 - \mu_i) \delta(\omega_{\mathbf{k}} - \omega_0) \\ &\quad \times [(\nu_{\mathbf{k}} + 1) P_{\mathcal{W}, 1, \nu_{\mathbf{k}}-1_{\mathbf{k}}} - \nu_{\mathbf{k}} P_{\mathcal{W}}]. \end{aligned} \quad (31)$$

This master equation can be simplified by taking into account that all N_S spins are identical and the state of the system depends on the global spin excitation number

$$\mu = \sum_i \mu_i, \quad 0 \leq \mu \leq N_S, \quad (32)$$

rather than on the particular spin configuration $\{\mu_i\}$. Recalling the definition of the shortcuts for the quantum states in Eq. (5) one can replace $P_{\mathcal{W}} \Rightarrow P_{(SP)} \Rightarrow P_{\mu, (P)}$ and rewrite Eq. (31) as

$$\begin{aligned} \frac{d}{dt}P_{\mu, (P)} &= \frac{2\pi}{N} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \mu \delta(\omega_{\mathbf{k}} - \omega_0) \\ &\quad \times [\nu_{\mathbf{k}} P_{\mu-1, (P, +1_{\mathbf{k}})} - (\nu_{\mathbf{k}} + 1) P_{\mu, (P)}] \\ &\quad + \frac{2\pi}{N} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 (N_S - \mu) \delta(\omega_{\mathbf{k}} - \omega_0) \\ &\quad \times [(\nu_{\mathbf{k}} + 1) P_{\mu+1, (P, -1_{\mathbf{k}})} - \nu_{\mathbf{k}} P_{\mu, (P)}]. \end{aligned} \quad (33)$$

B. Observables

The average excitation per spin is defined by

$$p = \frac{1}{N_S} \bar{\mu} = \frac{1}{N_S} \sum_{\mu=0}^{N_S} \frac{N_S!}{\mu! (N_S - \mu)!} \mu \prod_{\mathbf{k}} \sum_{\nu_{\mathbf{k}}=0}^{\infty} P_{\mu, (P)}, \quad (34)$$

where the binomial factor accounts for the degeneracy of the microscopic spin states. One has $p=0$ if all spins are in the ground state and $p=1$ if all spins are in the excited state. The average population of the phonon mode \mathbf{k} is defined in a similar way as

$$n_{\mathbf{k}} = \sum_{\mu=0}^{N_S} \frac{N_S!}{\mu! (N_S - \mu)!} \sum_{\nu_{\mathbf{k}}=0}^{\infty} \nu_{\mathbf{k}} \prod_{\mathbf{q} \neq \mathbf{k}} \sum_{\nu_{\mathbf{q}}=0}^{\infty} P_{\mu, (P)}. \quad (35)$$

It is never assumed that the spin and phonon parts of the density matrix factorize.

It is convenient to introduce the distribution function for the spins,

$$f_{\mu} \equiv \frac{N_S!}{\mu! (N_S - \mu)!} \prod_{\mathbf{k}} \sum_{\nu_{\mathbf{k}}=0}^{\infty} P_{\mu, (P)}, \quad (36)$$

which is normalized as $\sum_{\mu=0}^{N_S} f_{\mu} = 1$. Then Eq. (34) takes the form

$$p = \frac{1}{N_S} \bar{\mu} = \frac{1}{N_S} \sum_{\mu=0}^{N_S} \mu f_{\mu}. \quad (37)$$

For a macroscopic number of spins, $N_S \gg 1$, the distribution function f_{μ} is sharply peaked at $\mu = \bar{\mu}$. For instance, for uncorrelated spins one has

$$f_{\mu} = \frac{N_S!}{\mu! (N_S - \mu)!} p^{\mu} (1-p)^{N_S - \mu}, \quad (38)$$

from which follows $\bar{\mu} \cong \mu_{\max} \cong p N_S$. Clearly f_{μ} remains sharp if there is a correlation between the spins via the emitted and absorbed phonons.

One can also introduce the conditional probability $n_{\mu, \mathbf{k}}$ as the total number of phonons in the \mathbf{k} mode in the spin state μ . It is defined by

$$n_{\mu, \mathbf{k}} f_{\mu} = \frac{N_S!}{\mu! (N_S - \mu)!} \sum_{\nu_{\mathbf{k}}=0}^{\infty} \nu_{\mathbf{k}} \prod_{\mathbf{q} \neq \mathbf{k}} \sum_{\nu_{\mathbf{q}}=0}^{\infty} P_{\mu, (P)}, \quad (39)$$

so that, evidently, $n_{\mathbf{k}}$ of Eq. (35) can be written as

$$n_{\mathbf{k}} = \sum_{\mu=0}^{N_S} n_{\mu, \mathbf{k}} f_{\mu}. \quad (40)$$

Since f_{μ} is sharply peaked, we will use $n_{\mathbf{k}} \cong n_{\bar{\mu}, \mathbf{k}}$ below.

C. Kinetic equations for spins and phonons

The time derivative of p defined by Eq. (34) can be calculated with the help of master equation (33). The right-hand side of the resulting equation can be simplified by introducing f_{μ} and $n_{\mu, \mathbf{k}}$ with the help of Eq. (39). After some algebra one arrives at the equation

$$\begin{aligned} \frac{dp}{dt} &= \frac{\Gamma}{N_S} \sum_{\mu=0}^{N_S} \mu [(\mu + 1) f_{\mu+1} - \mu f_{\mu}] \\ &\quad + \frac{2\pi}{N N_S} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \delta(\omega_{\mathbf{k}} - \omega_0) \sum_{\mu=0}^{N_S} \mu \\ &\quad \times [n_{\mu-1, \mathbf{k}} (N_S - \mu + 1) f_{\mu-1} - n_{\mu, \mathbf{k}} \mu f_{\mu}] \\ &\quad + \frac{2\pi}{N N_S} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \delta(\omega_{\mathbf{k}} - \omega_0) \sum_{\mu=0}^{N_S} \mu \\ &\quad \times [n_{\mu+1, \mathbf{k}} (\mu + 1) f_{\mu+1} - n_{\mu, \mathbf{k}} (N_S - \mu) f_{\mu}]. \end{aligned} \quad (41)$$

The first line of this equation does not contain the phonon occupation numbers, and it describes the spontaneous emission of phonons. Here

$$\Gamma = \frac{2\pi}{N} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \delta(\omega_{\mathbf{k}} - \omega_0) = 2\pi \langle |V_{\mathbf{k}}|^2 \rangle \rho(\omega_0) \quad (42)$$

is the single-spin decay rate,

$$\rho(\omega_0) = \frac{1}{N} \sum_{\mathbf{k}} \delta(\omega_{\mathbf{k}} - \omega_0) \quad (43)$$

is the phonon density of states at the transition frequency of the spins, and $\langle |V_{\mathbf{k}}|^2 \rangle$ is the angular average:

$$\langle |V_{\mathbf{k}}|^2 \rangle \equiv \int \frac{dO_{\mathbf{k}}}{4\pi} |V_{\mathbf{k}}|^2. \quad (44)$$

Equation (41) can be drastically simplified by shifting the μ index under the sum over μ , so that only f_{μ} enters. As a result one obtains

$$\begin{aligned} \frac{dp}{dt} = & -\Gamma p + \frac{2\pi}{NN_S} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \delta(\omega_{\mathbf{k}} - \omega_0) \\ & \times \sum_{\mu=0}^{N_S} f_{\mu} n_{\mu,\mathbf{k}} (N_S - 2\mu). \end{aligned} \quad (45)$$

Here the sharpness of f_{μ} [see Eq. (38) and the comment below] leads to the final simplification. In the sum over μ one can replace $\mu \Rightarrow \bar{\mu} = pN_S$ and $n_{\mu,\mathbf{k}} \Rightarrow n_{\mathbf{k}}$, according to Eq. (40), and then use the normalization condition for f_{μ} [see Eq. (36)]. This leads to the final result

$$\frac{dp}{dt} = -\Gamma p + (1-2p) \frac{2\pi}{N} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \delta(\omega_{\mathbf{k}} - \omega_0) n_{\mathbf{k}}. \quad (46)$$

The kinetic equation for the phonons can be derived in a similar way. The result reads

$$\frac{dn_{\mathbf{k}}}{dt} = N_S \frac{2\pi}{N} |V_{\mathbf{k}}|^2 \delta(\omega_{\mathbf{k}} - \omega_0) [p - (1-2p)n_{\mathbf{k}}]. \quad (47)$$

In fact, Eq. (47) could be guessed since, together with Eq. (46), it satisfies the excitation conservation

$$pN_S + \sum_{\mathbf{k}} n_{\mathbf{k}} = \text{const.} \quad (48)$$

A disappointing feature of Eqs. (46) and (47) is that powers of $\delta(\omega_{\mathbf{k}} - \omega_0)$ enter both of them. If, say, there are no phonons in the initial state, then at short times $n_{\mathbf{k}}$ will grow accordingly to Eq. (47) as $n_{\mathbf{k}} \propto t \delta(\omega_{\mathbf{k}} - \omega_0)$. Then multiplication of this by $\delta(\omega_{\mathbf{k}} - \omega_0)$ leads to a mathematically incorrect expression. This problem was mentioned already in the discussion of the applicability of the Pauli master equation at the end of Sec. III. The short-memory approach leading to the master equation is inapplicable for a description of the decay of an initially prepared state if the reabsorption processes have to be taken into account, as in the case of the phonon bottleneck.

D. *Ad hoc* broadening of the δ function

Instead of stepping back to correct the error made in the derivation of Eqs. (46) and (47), one can choose a simple solution using the regularization of the δ functions by ascribing them a finite linewidth. One can, say, assume that the line shapes are Lorentzian with a linewidth Γ , such as radiational decay line shape given by Eq. (25). In this case the square of the δ function regularizes as

$$\delta^2(\omega_{\mathbf{k}} - \omega_0) \Rightarrow \frac{1}{\pi\Gamma} \delta(\omega_{\mathbf{k}} - \omega_0). \quad (49)$$

Practically one can replace the energy δ function in Eq. (46) as

$$\delta(\omega_{\mathbf{k}} - \omega_0) \Rightarrow \frac{1}{\pi\Gamma}, \quad (50)$$

because there is one more delta function in $n_{\mathbf{k}}$. In the simplest case of $|V_{\mathbf{k}}|^2 = |V|^2$ independent of the direction of \mathbf{k} and no phonons in the initial state one obtains

$$\begin{aligned} \frac{dp}{dt} = & -\Gamma p + (1-2p) \frac{2\pi}{N} \frac{1}{\pi\Gamma} |V|^2 \sum_{\mathbf{k}} n_{\mathbf{k}} \\ = & -\Gamma p + (1-2p) \frac{\Gamma}{N_{\Gamma}} N_S (p_0 - p), \end{aligned} \quad (51)$$

where we used Eqs. (42) and (27) to transform

$$\frac{2|V|^2}{N\Gamma} = \frac{1}{N\pi\rho(\omega_0)} = \frac{\Gamma}{N_{\Gamma}} \quad (52)$$

and used the conservation of the excitation, Eq. (48), p_0 being the spin excitation in the initial state. Finally one obtains

$$\frac{d}{dt} p = -\Gamma p + B\Gamma(p_0 - p)(1-2p), \quad (53)$$

where the bottleneck parameter B is defined by Eq. (28). This equation is a particular case of the bottleneck equation that can be found in the Abragam and Bleaney's book,¹³ and it is similar to all other bottleneck equations^{8,11,12,14} published earlier and later. None of these publications provides a derivation of the bottleneck equations so that it is impossible to judge whether these equations have been written *ad hoc* or derived in an incorrect way similar to that described above.

VI. DYNAMICAL EQUATIONS FOR THE PHONON BOTTLENECK

A. Derivation and analysis of the equations

Having seen the origin of the breakdown of the kinetic description of the PB, one can easily correct the error by stepping back to the master equation with memory, Eq. (16). Then calculations similar to those of Sec. V yield the equations with memory for spins and phonons,

$$\begin{aligned} \frac{dp}{dt} = & -\frac{2}{N} \sum_{\mathbf{k}} |V_{\mathbf{k}}|^2 \int_{t_0}^t dt' \cos[(\omega_{\mathbf{k}} - \omega_0)(t - t')] \\ & \times \{p(t') + [2p(t') - 1]n_{\mathbf{k}}(t')\} \end{aligned} \quad (54)$$

and

$$\begin{aligned} \frac{dn_{\mathbf{k}}}{dt} = & \frac{2N_S}{N} |V_{\mathbf{k}}|^2 \int_{t_0}^t dt' \cos[(\omega_{\mathbf{k}} - \omega_0)(t - t')] \\ & \times \{p(t') + [2p(t') - 1]n_{\mathbf{k}}(t')\}, \end{aligned} \quad (55)$$

instead of Eqs. (46) and (47). As there is no more energy δ function in these equations, one has to deal with all possible phonon modes. This means that in general the system of equations above should be solved numerically.

As equations with memory are inconvenient for numerical solutions, it is better to remove the memory by introducing the additional dimensionless variable

$$r_{\mathbf{k}}(t) = \Gamma \int_0^t dt' e^{i(\omega_{\mathbf{k}} - \omega_0)(t-t')} \times \{p(t') + [2p(t') - 1]n_{\mathbf{k}}(t')\}, \quad (56)$$

where Γ is the spin-phonon decay rate given by Eq. (42). Evidently $r_{\mathbf{k}}(t)$ is related to the nondiagonal element of the density matrix that was integrated out in Sec. III. One can see now that integrating out this nondiagonal element was unnecessary. The same results could be obtained directly from the Schrödinger equation (11). The resulting system of dynamical equations describing the direct spin-phonon processes has the form

$$\begin{aligned} \frac{dp}{dt} &= -\frac{1}{N} \sum_{\mathbf{k}} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \text{Re } r_{\mathbf{k}}, \\ \frac{dr_{\mathbf{k}}}{dt} &= i(\omega_{\mathbf{k}} - \omega_0)r_{\mathbf{k}} + \Gamma[p + (2p - 1)n_{\mathbf{k}}], \\ \frac{dn_{\mathbf{k}}}{dt} &= \frac{N_S}{N} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \text{Re } r_{\mathbf{k}}. \end{aligned} \quad (57)$$

Note that this system of equations is *time reversible*, as the underlying SE, Eq. (11). However, Eqs. (57) are *nonlinear* since they provide a reduced description of a many-body quantum-mechanical system in terms of a few variables. We suppose that there is no spin-phonon correlation in the initial state and use the initial condition

$$r_{\mathbf{k}}(0) = 0. \quad (58)$$

If $|V_{\mathbf{k}}|^2 = |V|^2$ independently of the direction of the emitted phonons, then with the help of Eqs. (42) and (28) one can represent the coefficient in the third equation as

$$\frac{N_S}{N} \frac{2|V|^2}{\Gamma} = \Gamma B. \quad (59)$$

This gives an idea of the strength of the PB in Eqs. (57). In the case $B \ll 1$ and no phonons in the initial state, the generated phonon populations $n_{\mathbf{k}}$ are small and can be neglected in the second equation of Eqs. (57). After that the latter can be integrated and the result can be plugged into the first equation. Here one can make the short-memory approximation $\text{Re } r_{\mathbf{k}} \cong \Gamma p \pi \delta(\omega_{\mathbf{k}} - \omega_0)$, which is justified. This results the well-known pure decay equation $dp/dt = -\Gamma p$.

At this point one can include the phonon dissipation in Eqs. (57) in the simplest possible way, generalizing the method of Refs. 8–14:

$$\frac{dp}{dt} = -\frac{1}{N} \sum_{\mathbf{k}} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \text{Re } r_{\mathbf{k}},$$

$$\frac{dr_{\mathbf{k}}}{dt} = \left[i(\omega_{\mathbf{k}} - \omega_0) - \frac{1}{2}\Gamma_{\text{ph}} \right] r_{\mathbf{k}} + \Gamma[p + (2p - 1)n_{\mathbf{k}}],$$

$$\frac{dn_{\mathbf{k}}}{dt} = \Gamma_{\text{ph}}(n_{\text{eq}} - n_{\mathbf{k}}) + \frac{N_S}{N} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \text{Re } r_{\mathbf{k}}. \quad (60)$$

Here Γ_{ph} is the phonon damping and n_{eq} is the thermally equilibrium phonon population near the resonance,

$$n_{\text{eq}} = \frac{1}{\exp\left(\frac{\hbar\omega_0}{k_B T}\right) - 1}. \quad (61)$$

At equilibrium one has $n_{\mathbf{k}} = n_{\text{eq}}$, $r_{\mathbf{k}} = 0$, and p is obtained from the equation $p + (2p - 1)n_{\text{eq}} = 0$, which yields

$$p_{\text{eq}} = \frac{n_{\text{eq}}}{1 + 2n_{\text{eq}}} = \frac{1}{\exp\left(\frac{\hbar\omega_0}{k_B T}\right) + 1}, \quad (62)$$

an expected result.

In the case $\Gamma_{\text{ph}} \gg \Gamma$ the variables $r_{\mathbf{k}}$ and $n_{\mathbf{k}}$ can be expected to relax much faster than p . Thus after some time they should adiabatically adjust to the instantaneous value of p . The same will happen in the case of a strong bottleneck at asymptotically large times. Neglecting $\dot{r}_{\mathbf{k}}$ and $\dot{n}_{\mathbf{k}}$ in Eq. (60) one obtains

$$n_{\mathbf{k}} = n_{\text{eq}} + \frac{\frac{N_S}{N} |V_{\mathbf{k}}|^2 [p + (2p - 1)n_{\text{eq}}]}{(\omega_{\mathbf{k}} - \omega_0)^2 + \frac{\Gamma_{\text{ph}}^2}{4} - \frac{N_S}{N} |V_{\mathbf{k}}|^2 (2p - 1)} \quad (63)$$

for the adjusted value of $n_{\mathbf{k}}$. The second nonequilibrium term here is small, and it has a Lorentz line shape with phonon linewidth Γ_{ph} if the last term in the denominator can be neglected. Note that for large Γ_{ph} the distribution of emitted phonons becomes a smooth enough function to make the short-memory Pauli master equation applicable [see discussion below Eq. (16)]. In this case, however, the problem is trivialized and the bottleneck disappears. One can see that the linewidth of emitted phonons narrows in the case of the inverse spin population, $p > 1/2$. The reason is the stimulated phonon emission acting as phonon pumping, which competes with their natural damping Γ_{ph} . The strength of the effect depends on the bottleneck parameter B , as follows from Eq. (59).

Similarly to Eq. (63), one can find the adjusted value $\text{Re } r_{\mathbf{k}}$ and plug it into the first equation of Eqs. (60). If $|V_{\mathbf{k}}|^2 = |V|^2$ independently of the direction of the emitted phonons, integration over \mathbf{k} yields the equation

$$\frac{dp}{dt} = -\Gamma^*(p)[p + (2p - 1)n_{\text{eq}}], \quad (64)$$

where

$$\Gamma^*(p) = \Gamma \left[1 - \frac{2B(1-2p)\Gamma^2}{[\Gamma_{\text{ph}} + \Gamma_{\text{ph}}^*(p)]\Gamma_{\text{ph}}^*(p)} \right] \quad (65)$$

and

$$\Gamma_{\text{ph}}^*(p) \equiv \sqrt{\Gamma_{\text{ph}}^2 + 2B(1-2p)\Gamma^2}. \quad (66)$$

One can see that the condition

$$\Gamma_{\text{ph}} \geq \sqrt{B}\Gamma \quad (67)$$

is needed to ensure $\Gamma^*(p) \cong \Gamma$ and thus to suppress the bottleneck. For $B \gg 1$, this is a stronger condition than the first-glance expectation $\Gamma_{\text{ph}} \geq \Gamma$. If Eq. (67) is satisfied, then the well-known solution of this equation,

$$p(t) = p_{\text{eq}} + (p_0 - p_{\text{eq}})e^{-\Gamma_T t}, \quad \Gamma_T = \Gamma(1 + 2n_{\text{eq}}), \quad (68)$$

is recovered. In the case of a strong bottleneck, opposite to Eq. (67), simplification of Eq. (65) yields

$$\Gamma^*(p) \cong \frac{\Gamma_{\text{ph}}}{\sqrt{2B(1-2p)}} \quad (69)$$

for $p < 1/2$. One can see that for a strong bottleneck the effective spin relaxation rate does not depend on Γ at all. This means that the transfer of energy from the resonant phonons to the rest of the phonon bath or elsewhere is really the bottleneck of the whole process. On the other hand, the effective spin relaxation rate is not just Γ_{ph} , as one could assume.

It should be stressed that in the case $B \gg 1$ and Eq. (67) not satisfied, Eq. (64) becomes a reasonable approximation at asymptotically large times only. At small and intermediate times the full solution of Eq. (60) shows oscillations of p similar to probability oscillations in the solution for two coupled quantum-mechanical states. The heuristic condition $\Gamma_{\text{ph}} \geq \Gamma$ mentioned at the beginning of this analysis is insufficient to establish adiabatic adjustment of $r_{\mathbf{k}}$ and $n_{\mathbf{k}}$ to the instantaneous value of p because of the large term $\sim B$ in the third equation of Eqs. (60).

B. Numerical solution of the dynamical spin-phonon equations

It is not difficult to solve Eqs. (57) and (60) numerically. The results of the numerical solution of Eqs. (57) for macroscopic samples and no phonons in the initial state (i.e., $T=0$) are shown in Fig. 2. The direct spin-phonon relaxation rate was taken to be $\Gamma=0.01$ and the phonon cutoff frequency $\omega_{\text{max}} \sim 1$, so that the short-memory approximation is valid in the absence of the bottleneck. For simplicity, the phonon modes have been discretized equidistantly, ensuring $N_{\Gamma} \gg 1$. Here and in all other numerical calculations we have set $|V_{\mathbf{k}}|^2 = |V|^2$ for simplicity, to use Eq. (59). One can see that for $B=0$ the solution for $p(t)$ is the pure-decay exponential $p = p_0 e^{-\Gamma t}$ practically in the whole time domain. The initial quadratic dependence of $p(t)$ stemming from the time reversibility of Eqs. (57) is confined to very short times of order $1/\omega_{\text{max}}$, and it is not seen in Fig. 2. The results in Fig. 2(a) for the initial spin excitation $p_0=0.1$ show *oscillations* approaching a plateau that becomes higher with increasing B . These oscillations are a memory effect that is absent in ear-

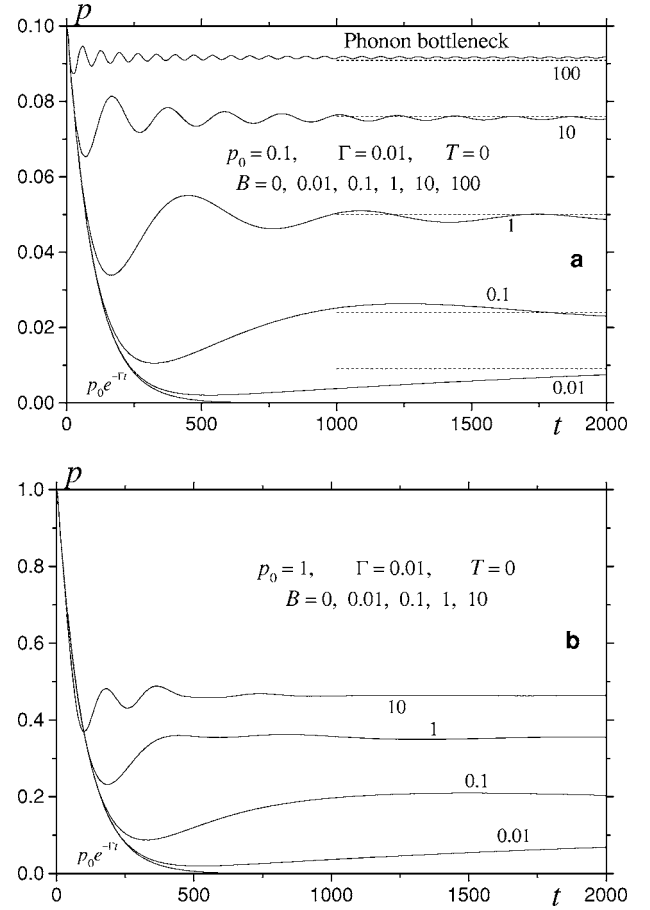


FIG. 2. Numerical solution of the dynamical equations for the phonon bottleneck with nonrelaxing phonons at $T=0$ for different values of the bottleneck parameter B . (a) Low initial excitation of spins, $p_0=0.1$. Asymptotes $p(\infty)$ given by Eq. (74) with $n_0=0$ are shown by dashed horizontal lines on the right. (b) High spin excitation, $p_0=1$. In the case (b) the solution diverges for $B \geq 100$. The bottleneck plateau $p(\infty)$ grows with B but it should not exceed $1/2$.

lier theories of the PB.^{8,11-14} For low spin excitations $p \ll 1$, the bottleneck equations can be solved analytically (see below). The analytically obtained asymptotic values of p are shown in Fig. 2(a) by dashed horizontal lines on the right.

Numerical results for the initially fully excited spin system, $p_0=1$, are represented in Fig. 2(b). The results are qualitatively similar to those in the case of the low spin excitation, Fig. 2(a), if the bottleneck parameter B is not too high. However, for $B=100$ the solution of Eqs. (57) shows an unphysical divergence with p going to infinity. This indicates that our bottleneck equations are not accurate enough to describe the high-excitation strong-bottleneck case. High excitation means the inverted population in the initial spin state, $p_0 > 1/2$, which formally corresponds to the *negative* spin temperature. Asymptotically the spin temperature should equilibrate with the temperature of the resonant phonon group. As the phonon energies are not bounded from above, phonons cannot have a negative temperature. Thus also spins cannot have a negative temperature in the asymptotic state. One can expect that for the initial spin inversion and huge values of B the asymptotic common spin-phonon temperature will be just

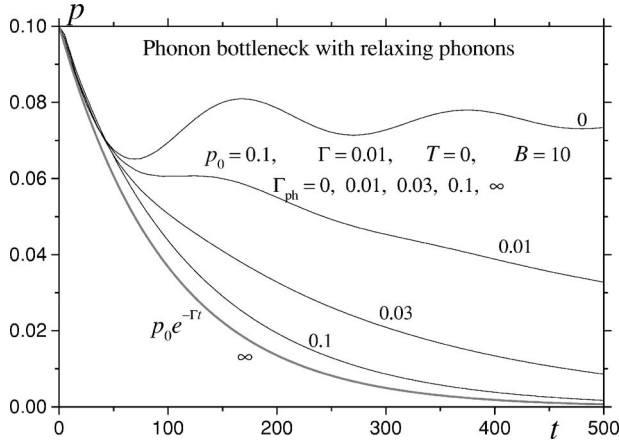


FIG. 3. Numerical solution for the spin excitation $p(t)$ of the dynamical equations with relaxing phonons, Eqs. (60), at $T=0$ for $B=10$ and different values of the phonon relaxation rate Γ_{ph} . Note that $\Gamma_{\text{ph}} \gg \Gamma$ is needed to suppress the bottleneck if $B \gg 1$.

very high; i.e., the asymptotic value of p will be close to $1/2$. This means that the asymptotic value of p should saturate at $1/2$ with increasing B . This tendency is seen in Fig. 2(b); only instead of the saturation at $p=1/2$, one obtains a divergence.

Figures 3 and 4 show the numerical solution of Eqs. (60) for $B=10$ and different values of the phonon relaxation rate Γ_{ph} . In Fig. 3 the temperature of the phonon bath is $T=0$ and the starting spin excitation is $p_0=0.1$. One can see that for all $\Gamma_{\text{ph}} > 0$ the spin excitation p relaxes to zero. The effective spin relaxation rate increases with Γ_{ph} and asymptotically approaches Γ , as the PB is gradually suppressed by the phonon relaxation. However, the values of Γ_{ph} needed to achieve the effective rate close to Γ are substantially greater than Γ , in accordance with the estimation in Eq. (67).

Figure 4 shows the PB with the opposite direction of the relaxation, also with $B=10$ for different values of Γ_{ph} . Here $p_0=0$ and the temperature is nonzero, $k_B T / (\hbar \omega_0) = 2$. Thus

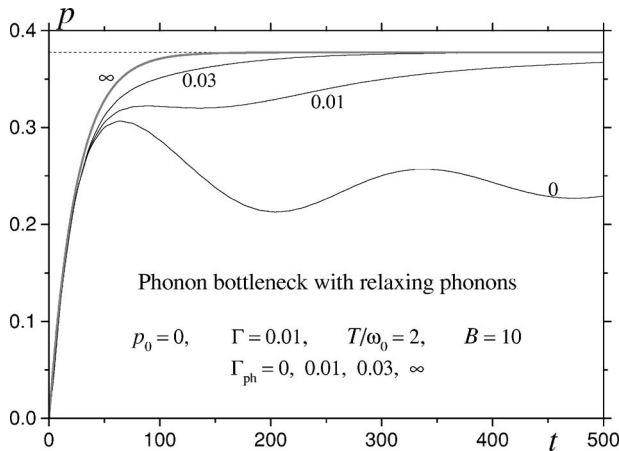


FIG. 4. Numerical solution for $p(t)$ with relaxing phonons and $k_B T / (\hbar \omega_0) = 2$. Here the spins are initially in the ground state, $p_0 = 0$, and they absorb the energy from phonons. For $\Gamma_{\text{ph}} = 0$, depletion of the phonons prevents reaching the thermal equilibrium value of p (the dotted horizontal line).

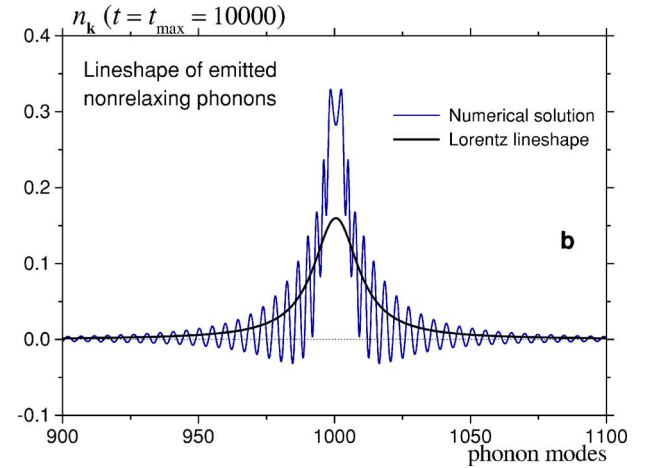
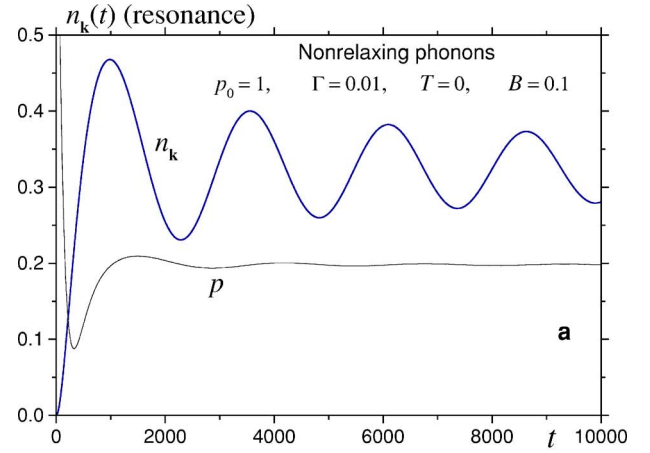


FIG. 5. (Color online) Emitted phonons in the absence of the phonon relaxation at $T=0$ for a moderate bottleneck, $B=0.1$. (a) Time dependence $n_{\mathbf{k}}(t)$ at resonance remains oscillating long after the spin excitation $p(t)$ reached the bottleneck plateau. (b) Line shape of the emitted phonons (at the maximal time) is narrow but not everywhere positive, which indicates that Eqs. (57) needs improving.

the spins absorb the energy from the phonon bath. If $\Gamma_{\text{ph}} = 0$ and $B \geq 1$, there are too few phonons in resonance with the spins, so that the resonant phonons get depleted and the spins cannot reach the equilibrium value p_{eq} given by Eq. (62). However, for all $\Gamma_{\text{ph}} > 0$ the phonons are replenished and the system reaches equilibrium. Again, for Γ_{ph} satisfying the strong inequality of Eq. (67), the spin relaxation curve acquires its standard form without the PB, given by Eq. (68).

Studying the time evolution of the line shape of emitted phonons, in the absence of the phonon relaxation, gives additional insights into the dynamics described by Eqs. (57). Figure 5 shows the numerical results obtained with $p_0=1$ and $B=0.1$. The time dependence of $n_{\mathbf{k}}$ at resonance in Fig. 5(a) remains oscillating at very large times, where $p(t)$ is already a constant. The line shape $n_{\mathbf{k}}$ at the maximal time of the calculation plotted versus the phonon-mode number (1000 corresponds to the resonance) is shown in Fig. 5(b). It is strongly oscillating, and $n_{\mathbf{k}}$ becomes negative for some modes at some moments of time. That is, the line shape oscillates both as the function of the phonon mode number

and the time, as Fig. 5(a) indicates. At large times, when the excitation of the spins has already reached its asymptotic value, the energy is still being exchanged between different phonon modes of the resonant group. Even for smaller values of B , the line shape does not approach the radiational line shape of Eq. (25) which is also shown in Fig. 5(b). This implies that Eqs. (57) is still not accurate enough to correctly describe the line shape of the emitted phonons. Still it yields some line shape, whereas earlier theories^{8,11-14} did not consider the line shape at all.

C. Bottleneck at low excitation

In the case of low excitation of the spins throughout the process, $p(t) \ll 1$, Eqs. (60) can be linearized by setting $p + (2p-1)n_{\mathbf{k}} \Rightarrow p - n_{\mathbf{k}}$ and then they can be solved by the Fourier transformation. We define the functions on the whole time interval $(-\infty, \infty)$ and set them to zero at $t < 0$. Then one has to introduce the initial-condition terms $p_0 \delta(t)$ and $n_0 \delta(t)$ into the first and third equations. Note that the initial phonon occupation can differ from the equilibrium value n_{eq} . Fourier transforms are defined by

$$p(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \tilde{p}(\omega) e^{-i\omega t}, \quad \tilde{p}(\omega) = \int_{-\infty}^{\infty} dt p(t) e^{i\omega t}, \quad (70)$$

etc. With

$$r_{\mathbf{k}} = \xi_{\mathbf{k}} + i\psi_{\mathbf{k}}, \quad (71)$$

Eqs. (60) can be put into the form

$$\begin{aligned} \frac{dp}{dt} &= p_0 \delta(t) - \frac{1}{N} \sum_{\mathbf{k}} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \xi_{\mathbf{k}}, \\ \frac{dn_{\mathbf{k}}}{dt} &= n_0 \delta(t) + \Gamma_{\text{ph}} [n_{\text{eq}} \theta(t) - n_{\mathbf{k}}] + \frac{N_S}{N} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \xi_{\mathbf{k}}, \\ \frac{d\xi_{\mathbf{k}}}{dt} &= -(\omega_{\mathbf{k}} - \omega_0) \psi_{\mathbf{k}} - \frac{1}{2} \Gamma_{\text{ph}} \xi_{\mathbf{k}} + \Gamma [p - n_{\mathbf{k}}], \\ \frac{d\psi_{\mathbf{k}}}{dt} &= (\omega_{\mathbf{k}} - \omega_0) \xi_{\mathbf{k}} - \frac{1}{2} \Gamma_{\text{ph}} \psi_{\mathbf{k}}. \end{aligned} \quad (72)$$

After the Fourier transformation one obtains a system of linear algebraic equations

$$\begin{aligned} -i\omega \tilde{p} &= p_0 - \frac{1}{N} \sum_{\mathbf{k}} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \tilde{\xi}_{\mathbf{k}}, \\ -i\omega \tilde{n}_{\mathbf{k}} &= n_0 + \Gamma_{\text{ph}} \left\{ \frac{n_{\text{eq}}}{-i\omega + 0} - \tilde{n}_{\mathbf{k}} \right\} + \frac{N_S}{N} \frac{2|V_{\mathbf{k}}|^2}{\Gamma} \tilde{\xi}_{\mathbf{k}}, \\ -i\omega \tilde{\xi}_{\mathbf{k}} &= -(\omega_{\mathbf{k}} - \omega_0) \tilde{\psi}_{\mathbf{k}} - \frac{1}{2} \Gamma_{\text{ph}} \tilde{\xi}_{\mathbf{k}} + \Gamma [\tilde{p} - \tilde{n}_{\mathbf{k}}], \\ -i\omega \tilde{\psi}_{\mathbf{k}} &= (\omega_{\mathbf{k}} - \omega_0) \tilde{\xi}_{\mathbf{k}} - \frac{1}{2} \Gamma_{\text{ph}} \tilde{\psi}_{\mathbf{k}}. \end{aligned} \quad (73)$$

Here one eliminates $\tilde{n}_{\mathbf{k}}$ and $\tilde{\psi}_{\mathbf{k}}$ and expresses $\tilde{\xi}_{\mathbf{k}}$ through \tilde{p} . Then plugging $\tilde{\xi}_{\mathbf{k}}$ into the first equation yields an isolated equation for \tilde{p} that can be solved. In the resulting expression for \tilde{p} one sums over \mathbf{k} . Making then the inverse Fourier transformation one obtains, in particular, the asymptotic value

$$p(\infty) = \frac{p_0 \sqrt{B} + n_0}{1 + \sqrt{B}} \quad (74)$$

in the case $\Gamma_{\text{ph}} = 0$, describing the phonon-bottleneck plateau. Also for $\Gamma_{\text{ph}} = 0$ one obtains

$$\xi_{\mathbf{k}}(t) = (p_0 - n_0) \frac{\sqrt{B}}{1 + \sqrt{B}} \frac{\sin(\Omega_{\mathbf{k}} t)}{\Omega_{\mathbf{k}}}, \quad (75)$$

where

$$\Omega_{\mathbf{k}} \equiv \sqrt{(\omega_{\mathbf{k}} - \omega_0)^2 + B\Gamma^2}. \quad (76)$$

This result features the avoided level crossing between the energy levels of the spins and phonons, as $\xi_{\mathbf{k}}(t)$, which describes the dynamical spin-phonon correlations, continues to oscillate even at resonance, $\omega_{\mathbf{k}} - \omega_0$. The splitting $\Delta = B\Gamma^2$ in $\Omega_{\mathbf{k}}$ is proportional to the concentration of spins via B given by Eq. (28). One can see from Eq. (59) that in the case of nondiluted spins, $N_S = N$, one has $\Delta = B\Gamma^2 = 2|V|^2$. This result is similar to that of Ref. 15.

For the model with relaxing phonons, in the strong-bottleneck limit $B \gg 1$ one obtains the following asymptotic ($\Gamma_{\text{ph}} t \gg 1$) relaxation law

$$p(t) = \left(p_0 + \frac{\tilde{\Gamma}}{\Gamma_{\text{ph}}} n_0 \right) e^{-\tilde{\Gamma} t} + (1 - e^{-\tilde{\Gamma} t}) n_{\text{eq}}, \quad (77)$$

where

$$\tilde{\Gamma} \equiv \frac{\Gamma \Gamma_{\text{ph}}}{\sqrt{\Gamma_{\text{ph}}^2 + 2B\Gamma^2}} \quad (78)$$

is the effective relaxation rate of the spin-phonon system in the case of strong bottleneck, $B \gg 1$. One can see that for $\Gamma_{\text{ph}} \ll \sqrt{B}\Gamma$ the effective rate is $\tilde{\Gamma} \cong \Gamma_{\text{ph}} / \sqrt{2B} \ll \Gamma_{\text{ph}}$. The same result follows from Eq. (69) in the limit $p \ll 1$. For $B \ll 1$ in Eq. (67) the effective rate becomes $\tilde{\Gamma} \cong \Gamma$ and the bottleneck disappears.

D. Asymptotic stability of the bottleneck equations

With the methods of the previous section it is possible to analyze the asymptotic stability of the bottleneck equations with undamped phonons, Eqs. (57). Asymptotically $p = \text{const}$, so that one can substitute it into Eqs. (72) with $\Gamma_{\text{ph}} = 0$. Assuming, for simplicity, $|V_{\mathbf{k}}|^2 = |V|^2 = \text{const}$ and introducing a new variable $\phi_{\mathbf{k}}$ as

$$\psi_{\mathbf{k}} = \phi_{\mathbf{k}} + \frac{\Gamma p}{\omega_{\mathbf{k}} - \omega_0}, \quad (79)$$

one obtains the system of linear ordinary differential equations

$$\begin{aligned}\dot{\xi}_{\mathbf{k}} &= -(\omega_{\mathbf{k}} - \omega_0)\phi_{\mathbf{k}} + (2p - 1)\Gamma n_{\mathbf{k}}, \\ \dot{\phi}_{\mathbf{k}} &= (\omega_{\mathbf{k}} - \omega_0)\xi_{\mathbf{k}}, \\ \dot{n}_{\mathbf{k}} &= B\Gamma\xi_{\mathbf{k}}.\end{aligned}\quad (80)$$

The eigenfrequencies of this system of equations are $\Omega_{\mathbf{k}}=0$ and

$$\Omega_{\mathbf{k}} = \pm \sqrt{(\omega_{\mathbf{k}} - \omega_0)^2 + B\Gamma^2(1 - 2p)}, \quad (81)$$

cf. Eq. (76). For the inverse population, $p > 1/2$, the eigenfrequency $\Omega_{\mathbf{k}}$ becomes imaginary in the frequency region near the resonance. This leads to an instability that is seen in the numerical results as positive divergence of p and thus negative divergence of $n_{\mathbf{k}}$ near the resonance. The problem is that for the inversely populated initial spin states, $p_0 > 1/2$ and $B \gg 1$ the asymptotic value of p is close to $1/2$, as discussed above. Then even small inaccuracies of the dynamic equations can render p slightly exceeding $1/2$ which leads to the divergency. The fact that for $p_0=1$ the instability happens only starting from $B \sim 100$ [see Fig. 2(b)], where $p(\infty)$ should be already very close to $1/2$, indicates that the instability is driven by the inaccuracies of Eqs. (57).

VII. DISCUSSION

In the main part of this paper it was shown that the dynamics of two-level systems (spins) interacting with a continuum of resonant phonons via direct emission and absorption processes, including the phonon-bottleneck effect, is much more complicated than generally accepted. Since the emitted and absorbed phonon packet is narrow in the energy space, the usual *kinetic* description based on the short-memory Pauli master equation is invalid. Taking into account memory amounts to stepping back to a *dynamic* description including nondiagonal elements of the density matrix. This cures the problem of the powers of the energy δ functions in the standard formalism while being capable of describing the PB. The resulting system of dynamical equations (57) can be enhanced to include the relaxation of phonons in a simple way, resulting in Eqs. (60). For the low spin excitation $p \ll 1$, these equations linearize and can be solved analytically. In a number of particular cases, such as small bottleneck parameter B of Eq. (28) or fast phonon relaxation rate Γ_{ph} [see Eq. (67)], simple results without a bottleneck are reproduced.

The approach formulated here is not a final solution of the long-standing PB problem but rather a next step in improving the existing theories. A deficiency of the present approach is its inability to provide an accurate line shape for emitted phonons, even the well-known Lorentzian shape of Eq. (28) for the weak bottleneck, $B \ll 1$. This should be the reason for the instability of Eqs. (57) in the case of the strong

bottleneck, $B \gg 1$, with spin inversion in the initial state. As the dynamical bottleneck equations have been obtained within the same method as the Pauli master equation—that is, by cutting the infinite chain of coupled equations for the nondiagonal elements of the density matrix at the lowest possible level—an evident idea of improvement is to take into account the next-generation nondiagonal matrix elements. This would result in a more complicated formalism, however, that deserves to be dealt with in a separate work.

In real systems the spin transition frequency ω_0 is inhomogeneously broadened. That is, there are different transition frequencies ω_i for different spins, distributed around the average value ω_0 with a width $\Delta\omega_0 \gg \Gamma$. Obviously this leads to the reduction of the PB effect since more phonon modes can exchange excitation with the spin system, or, in other words, fewer spins can exchange excitation with a given phonon mode. Thus one can expect that the situation will depend on the effective bottleneck parameter $\tilde{B} \sim (\Gamma/\Delta\omega_0)B \ll B$.

Incorporating inhomogeneous broadening in the theory seems to be straightforward but it would result in a serious complication of the formalism. The problem is that spins cannot be described by a single variable p any longer. Spin with each particular ω_i should exhibit its own dynamics, so that one has to search for the distribution $p(\omega_i)$. It is not clear if any analytical solutions for the corresponding system of equations are available, and the numerical solution should be significantly more difficult than the one presented above. Note that inhomogeneous broadening was mentioned in previous publications on the phonon bottleneck (see, e.g., Ref. 13) but it was treated as part of the “phenomenological” linewidth of the transition, without any frequency resolution of the type suggested in this paragraph.

Spin-spin interactions make the problem even more complicated. Although the effects of inhomogeneous broadening and spin-spin interactions are important for comparison with experiment, it does not make sense to consider them in the present paper for the reason stressed above. In principle, to be on safe grounds, one has to find a satisfactory solution for the core problem of the PB and obtain a correct line shape of the emitted phonons, before adding inhomogeneous broadening and other real-life effects to the model.

Of course, one can argue that in the presence of inhomogeneous broadening the phonon line shape should be dominated by the latter and one obtains a good-looking everywhere positive line shape by a kind of averaging over the spin transition frequency ω_i , instead of the unsatisfactory line shape in Fig. 5(b). According to this logic, including inhomogeneous broadening would have more sense, as the next step, than struggling for an accurate line shape in the bare model. Practically this might be true, although one cannot easily find a microscopic justification for such an approach.

It should be stressed that the whole consideration in this paper pertains to magnetically diluted systems, in which the phases of phonons emitted by one spin and reaching another spin are random. This excludes magnetically dense systems such as molecular magnets. In the latter, emission of phonons by different spins is correlated, which in the ideal case leads to the phonon superradiance.¹⁹ The interplay of the phonon

bottleneck and phonon superradiance is an exciting and challenging problem, and its theoretical description requires including further correlators that have been ignored above. Obviously, for magnetically dense systems applicability of the *ad hoc* bottleneck equations^{8,11-14} is even less justified than for diluted systems.

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