Superslow Self-Organized Motions in a Multimode Microwave Phonon Laser (Phaser) under Resonant Destabilization of Stationary Acoustic Stimulated Emission

D. N. Makovetski
Usikov Institute of Radiophysics and Electronics of National Academy of Sciences, 12, Ac. Proskura street, Kharkov, 61085, Ukraine
(Dated: February 6, 2004)

Two qualitatively different kinds of resonant destabilization of phonon stimulated emission (SE) are experimentally revealed for periodically forced multimode ruby phaser (photon laser) operating at microwave acoustic frequencies \( \Omega_N \approx 9 \text{ GHz} \) (acoustic SE wavelength \( \approx 1 \mu\text{m} \)). The inversion state of \( \text{Cr}^{3+} \) electronic spin-system in ruby was created by electromagnetic pump with frequency \( \Omega_P = 23 \text{ GHz} \). Under deep modulation of pump power at low frequencies \( (\omega_m = 70 - 200 \text{ Hz}) \), where \( \omega_m \) is the modulation frequency, a fast deterministic chaotic alternation of the microwave phonon SE modes with different indexes \( N \) is observed. This range of SE destabilization corresponds to the relaxational resonance that is well known for optical class-B lasers. Outside the relaxational resonance range, namely at ultra-low (infrasonic) frequencies \( \omega_m \approx 10 \text{ Hz} \) of electromagnetic pump modulation, the other kind of resonant destabilization of stationary phonon SE is observed for the first time. This new nonlinear resonance (we call it \( \lambda \)-resonance) manifests itself as very slow and periodically repeated self-reconfiguration of the acoustic microwave power spectra (AMPS) of a phaser generation. The period of such self-organized motions depends significantly on \( \omega_m \) and changes by several orders of magnitude when \( \omega_m \) varies within several percent. Near the vertex of \( \lambda \)-resonance the period of AMPS self-reconfigurations takes giant values of several hours (at \( T = 1.8 \text{ K} \)). The second kind of SE resonant destabilization is explained in terms of antiphase energy exchange between acoustic SE modes in a modulated phaser. The role of polarized nuclear spin-system (formed by \( \text{Al}^{27} \) nuclei of the ruby crystalline matrix \( \text{Al}_2\text{O}_3 \)) in these superslow self-organized motions is discussed.

PACS numbers: 05.65.+b, 42.65.Sf, 43.35.+d

I. INTRODUCTION

The possibility of phonon stimulated emission (SE) in activated crystals was considered as early as 1960s [1, 2]. Yet, speculation about various mechanisms of phonon SE persists even today (see, e.g., [3]). In experiments, phonon SE was first observed and studied [4, 5, 6, 7, 8] on dielectric crystals doped by Fe-group paramagnetic ions. The SE effect manifests itself as the quantum paramagnetic amplification of a coherent microwave phonon flux (hypersound) when spin levels that may take part in spin-phonon interaction are inversely populated. This effect may be viewed as an acoustic analogue [11, 12, 13, 14] of maser amplification of electromagnetic waves [10] (not counting a number of features of nonlinear processes in the signal and pump channels [11, 12, 13, 14]).

At the same time, the mechanism of quantum generation of microwave acoustic oscillations, which was discovered experimentally in [15, 16], has long remained unclear. The reasons were the attempts to draw analogy (see, e.g., [17]) between acoustic quantum generators (phasers) and usual electromagnetic quantum generators of maser type, which does exist between related quantum amplifiers.

In experimental studies [12, 18, 19] of microwave acoustic SE in \( \text{Ni}^{2+}:\text{Al}_2\text{O}_3 \) and \( \text{Cr}^{3+}:\text{Al}_2\text{O}_3 \) crystals, it was shown that operation of microwave phaser generator is physically much closer to optical or near-infrared (near-IR) lasing than to operation of microwave maser generator. In fact, the hypersound wavelength in a Fabry-Perot acoustic resonator (FPAR) is roughly 1–3 \( \mu\text{m} \) (i.e., falls into the near-IR range). The quality factor \( Q_C \) of an FPAR, as well as the quality factor of electromagnetic cavities in many lasers, is high: \( Q_C \approx 10^5 - 10^6 \) [12, 18, 19] (certainly, this value is reached at liquid helium temperatures, when the non-resonant absorption of hypersound is low). Therefore, experimental SE spectra of phasers operating in the autonomous regime [12, 18, 19] sometimes are similar to those observed for class-B multimode solid-state lasers (for which \( \tau_1 \gg \tau_C \gg \tau_2 \), where \( \tau_1 \) and \( \tau_2 \) are the longitudinal and transverse relaxation times for active centers, \( \tau_C \) is the lifetime of field excitations in the cavity).

However, phasers differ radically from lasers in regard to the intrinsic quantum noise (spontaneous emission) intensity \( J_{\text{spont}} \). Since the velocity of hypersound \( v_n \) is five orders of magnitude lower than the velocity of light, the SE frequency \( \Omega \) in a phaser with a hypersound wavelength of \( 1-3 \mu\text{m} \) lies in the range \( \Omega \approx 3 - 10 \text{ GHz} \) [12, 18, 19], i.e., is five orders of magnitude lower than in a laser. Accordingly, the relative spontaneous emission intensity in a phaser is \( \approx 15 \) orders of magnitude lower than in a laser (because \( J_{\text{spont}} \) grows as \( \Omega^2 \)). In essence, a phaser may be

*Electronic address: makov@ire.kharkov.ua
considered as a deterministic dynamic system throughout the SE intensity range available. This is of crucial importance for studying motion in systems with a complex stratified phase space. It is known that multiplicative noise (including spontaneous emission in a nonlinear active medium) affects the behavior of dynamic systems in a very intriguing manner \cite{24}, causing coarsening of the phase space topology \cite{21}, etc.

Earlier \cite{22}, we experimentally revealed strong dynamic narrowing of the SE spectra in a nonlinear FPAR with modulated electromagnetic pump of paramagnetic centers. This narrowing was attributed to the resonant destabilization of energy exchange between FPAR hypersonic (microwave acoustic) modes under ultra-low-frequency (infrasonic) forcing of ruby (Cr$^{3+}$:Al$_2$O$_3$) spin-phonon system. Further investigation of this microwave deterministic system as a part of nonautonomous phaser allowed us to reveal its still more unexpected property: superslow large-scale laminar self-reconfigurations of the SE spectra akin to autowave motions \cite{19, 23}. Below, we report these experimental studies in details and discuss the nature of the phenomena observed.

II. PHASER SYSTEM

A. Microwave Fabry-Perot Acoustic Resonator, Active Centers, and Hypersonic Transducer

Experiments were carried out by using a ruby phaser \cite{12, 22, 24} with the electromagnetic pump power $P$ periodically modulated at ultra-low (infrasonic) and low (usual sonic) frequencies: $\omega_m/2\pi = 1$ Hz – 3 kHz (hereafter, the factor $2\pi$ will be omitted). A solid-state FPAR, which was made of synthetic single-crystal pink ruby, had the form of a cylinder with a diameter $d_C = 2.6$ mm and length $L_C = 17.6$ mm. The end faces of the cylinder are parallel to each other and optically flat: they serve as acoustic mirrors for hypersonic waves. The 3-rd order crystallographic axis $\vec{O}_3$ of the ruby coincides with the geometrical axis $\vec{O}_C$ of the FPAR. The concentration of Cr$^{3+}$ ions is $C_n = 1.3 \cdot 10^{19}$ cm$^{-3}$ (i.e. $\approx 0.03\%$). All measurements were made in the interval of temperatures $T = 1.8 - 4.2$ K.

For a hypersonic frequency near $\Omega = 9.1$ GHz and $L_C$ mentioned above, the separation between longitudinal acoustic modes of the FPAR is $\Delta\Omega_N^{(0)} \equiv \Omega_N^{(0)} - \Omega_{N-1}^{(0)} = 310$ kHz. Here, $\Omega_N^{(0)}$ is the frequency of an $N$-th mode of the FPAR in the “cold” regime, i.e., at $P = 0$. The frequencies of the hypersound emitted, i.e., the frequencies of phonon SE modes $\Omega_N \approx \Omega_N^{(p)}$ in the “hot” regime ($P > P_g$), where $P_g$ is the pump power at which phaser generation starts, lie near 9.12 GHz according to the frequency $\Omega_S = \Omega_{32} \equiv h^{-1} [E_3(H) - E_2(H)]$ of the inverted spin transition $E_3 \leftrightarrow E_2$ of active centers in a static magnetic field $H \approx H_0$. Thus, the frequency $\Omega_S$ corresponds to the vertex of the acoustic paramagnetic resonance (APR) line coinciding by magnetic field with two electron spin resonance (ESR) lines allowed for the electromagnetic pump (Fig. 1).

The value of $H_0$ depends on the frequency $\Omega_P$ of the pump microwave field, which saturates the spin transitions $E_1 \leftrightarrow E_3$ and $E_2 \leftrightarrow E_4$, with $\Omega_P = \Omega_{31} = \Omega_{42} \gg \Omega_S$, where $\Omega_{31} \equiv h^{-1} [E_3(H_0) - E_1(H_0)]$; $\Omega_{42} \equiv h^{-1} [E_4(H_0) - E_2(H_0)]$ (see. Fig. 1). The symbols $\langle \psi_i \rangle$ denote wave functions that belong to the energy levels $E_i$ of the ground spin quadruplet (spin $S = 3/2$, orbital quantum number $L = 0$) of a Cr$^{3+}$ ion in the crystal field of ruby. Since this field is of trigonal symmetry, all $E_i$ and $\langle \psi_i \rangle$ depend only on $H = |\vec{H}|$ and azimuthal angle $\vartheta$ between $\vec{H}$ and $\vec{O}_3$ \cite{10}.

One of the FPAR mirrors was covered by a thin (approximately 0.5 µm thick) textured ZnO piezoelectric film with a 0.1 µm-thick Al sublayer (both layers were applied by evaporation in vacuum). The texture axis runs normally to the FPAR mirror. The ZnO film is the basic component of a bidirectional hypersonic transducer designed for converting a microwave phonon field to an electromagnetic field and vice versa. The phonon SE arising in the FPAR excites electromagnetic oscillation in the ZnO film, and the electromagnetic signal may be detected by standard microwave techniques. On the
other hand, exciting the ZnO film from the outside by electromagnetic waves with a frequency \( \Omega \), we inject hypersonic waves with the same frequency into the FPAR, with \( \lambda_0(S) \approx 3.3 \cdot 10^{-4} \lambda_e(S) \), where \( \lambda_e(S) \) is the wavelength of longitudinal hypersonic in our system, \( \lambda_0(S) \approx 1 \mu m; \) and \( \lambda_0(S) \approx 3 \) cm is the wavelength of an electromagnetic wave of the same frequency as the hypersonic wave).

**B. Inversion States of Active Centers and Conditions of Phaser Generation**

As it was noted earlier, inverted spin states of \( E_3 \leftrightarrow E_2 \) transition of \( Cr^{3+} \) active centers are formed by the pump microwave electromagnetic field. The frequency \( \Omega_P \) of the pump source may be tuned within 22 – 25 GHz; that is, the pump wavelength \( \lambda'(P) \) belongs to 1.25 cm range of microwaves. Maximal pump power at resonant frequency \( \Omega_P \approx 23 \) GHz reaches \( P = P^{(\text{full})} = 12 \) mW. Through a diffraction coupler, the pump field is excited in the cylindrical electromagnetic pump cavity of type \( H_{011} \), which has an eigenfrequency 23.0 GHz, quality factor \( Q_{C,P} \approx 8 \cdot 10^3 \), and geometrical length coinciding with the length \( L_C \) of the FPAR.

The ruby FPAR is placed in the pump cavity along its axis. If \( P = 0 \) and the magnitude and direction of \( \vec{H} \) are beyond the APR range, the absorption of the hypersonic injected into the FPAR depends on such the two parameters:

(i) the nonresonant volume attenuation \( \eta_{\text{vol}} \) (including losses on the lateral surfaces of the FPAR) and

(ii) losses on the FPAR mirrors \( \eta_{\text{mirr}} \).

If \( P = 0 \) and the magnitude and direction of \( \vec{H} \) fall into the APR range (i.e., \( \vec{H} \approx \vec{H}_0 \)), the third absorption mechanism comes into play:

(iii) the resonant paramagnetic absorption of the hypersonic, which depends considerably on the frequency of the signal injected and on the offset of the magnetic field from the APR line vertex.

Finally, \( \vec{H} \approx \vec{H}_0 \) and the pump power is applied. Then, the resonant paramagnetic absorption of the hypersonic decreases. If, as \( P \) rises, one succeeds in passing into the range where the paramagnetic absorption becomes negative (i.e., the inversion ratio \( K(P, \vec{H}) \) becomes positive), nonparamagnetic losses of the hypersonic in the FPAR are compensated for partially or completely. The complete compensation of the losses (i.e., the onset of phaser generation) takes place first at that mode (let its frequency be \( \Omega_1 \)) closest to the center of the APR inverted line, for which the condition

\[
\frac{1}{Q^{(1)}_{\text{vol}}} + \frac{1}{Q^{(1)}_{\text{mirr}}} + \frac{1}{Q^{(1)}_{\text{magn}}} < 0, \tag{1}
\]

is met prior to other modes. In (1), \( Q^{(1)}_{\text{vol}} = k_1/\eta_{\text{vol}} \), \( Q^{(1)}_{\text{mirr}} = k_1/\eta_{\text{mirr}} \), \( k_1 = \Omega_1/\nu_u \), and \( Q^{(1)}_{\text{magn}} \) is the negative (at \( K > 0 \)) magnetic “quality factor” of this mode (for which phaser generation starts first). This magnetic “quality factor” is given by

\[
Q^{(1)}_{\text{magn}} = \frac{-k_1}{\alpha_1(P, \vec{H}, \Omega_1)} = \frac{-k_1}{K(P, \vec{H})\sigma(\vec{H}, \Omega_1)} \tag{2}
\]

where \( \alpha_1 \) is the nonresonant positive (at \( K(P) > 0 \)) quantum gain (increment of amplification) for the mode under consideration and \( \sigma \) is the hypersonic paramagnetic absorption at \( P = 0 \) (i.e at \( K(0) = -1 \)).

The expression for \( \sigma \) has the form (see, e.g., page 283 in book [27]):

\[
\sigma_{mn} = \frac{2\pi^2C_0\nu^2g(\nu)|\Phi_{mn}|^2}{(2S + 1)^{3/2}k_B\nu}\tag{3}
\]

where \( \nu = \Omega/2\pi \), \( g(\nu) \) is the form factor of the APR line, \( \rho' \) is the crystal density, \( k_B \) is the Boltzmann constant, and the matrix element \( \Phi_{mn} \) characterizes coupling of an \( E_m \leftrightarrow E_n \) spin transition with a resonant hypersonic wave of given propagation direction and polarization.

The form factor of the APR line is normalized to unity,

\[
\int_{-\infty}^{\infty} g(\nu)d\nu = 1, \tag{4}
\]

and the matrix element \( \Phi_{mn} = \frac{\partial\langle\psi_m|\hat{H}|\psi_n\rangle}{\partial\varepsilon_{zz}} \) for the longitudinal hypersonic wave traveling along the ruby crystallographic axis \( \hat{z} \) (the axis \( \hat{z} \) is aligned with the \( z \) coordinate axis) is given by:

\[
\Phi_{mn} = \frac{G_{33}}{2} \left( 3(\langle\psi_m|S_z^2|\psi_n\rangle - S(S + 1)\langle\psi_m|\psi_n\rangle) \right), \tag{5}
\]

where \( \varepsilon_{zz} \) is the component of the elastic strain tensor, \( \hat{H} \) is the Hamiltonian of spin-phonon interaction, \( G_{33} \) is the component of the spin-phonon interaction tensor (in the Voigt pair-index form), \( S_z \) is the projection of the vectorial spin operator on the \( z \) axis.

To estimate \( \Phi_{mn} \) we use the value \( G_{33} = 5.8 \text{ cm}^{-1} = 1.16 \cdot 10^{-15} \text{ erg} \) (found by us experimentally by means of direct APR measurements at \( T = 1.8 - 4.2 \) K and the wave functions \( |\psi_i\rangle \) for \( Cr^{3+} \) ion in ruby (calculated in [10] from experimental ESR data). With \( H = 3.92 \text{ kOe} \) and \( \vec{H} \) directed at the angle \( \vartheta = \theta_{\text{symm}} \) to the \( z \) axis, where \( \theta_{\text{symm}} \approx \arccos(1/\sqrt{3}) = 54.73^\circ \), we find from (5) that \( \Phi_{mn} \approx 10^{-15} \text{ erg} \). The choice \( \vartheta = \theta_{\text{symm}} \) refers to the so-called symmetric (or push-pull) pump conditions.

Such conditions are set up owing to the equality \( E_4 - E_2 = E_3 - E_1 \) (Fig.1), which takes place at \( \vartheta = \theta_{\text{symm}} \) and provides the most efficient inversion at the signal transition \( E_3 \leftrightarrow E_2 \) in the spin system. Eventually, with \( \nu_S = 9.1 \text{ GHz} \), \( g(\nu_S) = 10^{-8} \text{ s} \), \( C_n = 1.3 \cdot 10^{19} \text{ cm}^{-3} \), \( \rho' = 4 \text{ g/cm}^3 \), \( \nu_u = 1.1 \cdot 10^6 \text{ cm/s} \), \( T = 1.8 \) K we find from (3) that \( \sigma \approx 0.04 \text{ cm}^{-1} \).

The loaded acoustic quality factor \( Q_C^{(0)} \) of the ruby FPAR (with the piezoelectric film on one of the acoustic
mirrors) was measured at $T = 1.8 \text{ K}$ by the pulsed echo method at frequencies $\Omega = 9.0 - 9.2 \text{ GHz}$. With $\vec{H} = 0$ and $P = 0$, it was found that $Q_C^{(0)} \approx (5.2 \pm 0.4) \cdot 10^5$ for all longitudinal acoustic modes falling into this frequency interval. Hence, $\eta \equiv \eta_{\text{vol}} + \eta_{\text{mirr}} = \Omega Q_C^{(0)} \bar{v}_u^{-1} \approx 0.1 \text{ cm}^{-1}$.

The parameters $\sigma$, $\eta$, and $\alpha = \alpha_g$ (here $\alpha = \alpha_g$ is the value of $\alpha$ at which phaser begins generation) are obviously related as

$$\alpha_g = \eta = K_g \sigma,$$

where $K_g$ is the critical value of the inversion ratio $K$ for the transition $E_3 \leftrightarrow E_2$. Substituting $\sigma \approx 0.04 \text{ cm}^{-1}$ and $\eta \approx 0.1 \text{ cm}^{-1}$ into (6) yields $K_g \approx 2.5$. This value is readily attained in the case of push-pull pump, which provides $K_{\text{max}} \approx 3.3$ under the conditions of our experiments.

### III. EXPERIMENTAL RESULTS

#### A. Regime of Free Generation in Ruby Phaser

Since the frequency width $\Gamma_{32}$ of the APR line at the spin transition $E_3 \leftrightarrow E_2$ is $\approx 100 \text{ MHz}$ and the mode separation is as small as $\approx 300 \text{ kHz}$, single-mode SE changes to multimode one even if the pump threshold is exceeded slightly. For $\Omega_P = \Omega_{CP}^{(0)} = 23.0 \text{ GHz}$ and $H = H_0 = 3.92 \text{ kOe}$, free multimode phaser generation is observed even at $P \geq 50 \mu\text{W}$. If $\Delta H \equiv H - H_0 \neq 0$, the pump power must be much higher (by one to two orders of magnitude) for the condition $K > K_g$ to be fulfilled.

With pump switched on stepwise, the free phaser generation begins in a damped oscillatory regime. For our system, the frequency $\omega_R$ of these damped oscillations (the so-called relaxation frequency) lies in the low-frequency range, $\omega_R \approx 130 \text{ Hz}$ at $H = H_0$. In a free-running multimode phaser, the number of acoustic modes in acoustic microwave power spectra (AMPS) does not exceed about thirty even if $P = P_{\text{full}} \gg P_g$. That is, the maximal width of AMPS for autonomous phaser generator ($30 \times 310 \text{ kHz} \approx 10 \text{ MHz}$) is one order of magnitude smaller than $\Gamma_{32}$, which is explained by the well-known Tang-Statz-deMars mechanism (exhaustion of power supplies for spatially-overlapping competing modes of SE). If magnetic field offset is absent, free phaser generation proceeds under near-steady-state conditions (the integral intensity $J_3$ of multimode SE, which is registered by the hypersonic transducer on one of the FPAR mirrors, is virtually time independent).

If the offset is small, $|\Delta H| \leq 3 \text{ Oe}$, the value of $J_3$ also remains practically time independent. Only with $|\Delta H|$ increasing up to $\approx 30 \text{ Oe}$, the integral intensity $J_3$ of phonon SE oscillates weakly because few of the individual SE modes demonstrate small-scale moving along the frequency axis (with correspondent changing of their SE intensities) or decay at all. At $|\Delta H| \geq 30 \text{ Oe}$, some of the free generation modes in AMPS becomes split.

These splittings are usually of order of several kilohertz (more rarely several tens of kilohertz), which is much less than $\Delta \Omega \approx 300 \text{ kHz}$; the number of splitted modes is one or two (at most) even if $|\Delta H| \approx 100 \text{ Oe}$. The peak intensities of splitted components are much lower than that of unsplitted SE modes. This fine structure of SE modes under free generation regime is of oscillating character, with not only the spectral component amplitudes but also their frequency positions varying smoothly (the latter within 10 kHz). The only exceptions are narrow windows in sets of control parameters $\{P, \Delta H, \ldots\}$, where the spontaneous breaking of SE coherence takes place for one or two modes, causing small-scale phonon turbulence. In most cases, however, free microwave acoustic generation in a ruby phaser proceeds under near-steady-state regime even if the offset by static magnetic field is as large as $|\Delta H| \approx 100 \text{ Oe}$.

#### B. Resonant Destabilization of Phonon Stimulated Emission under Relaxational (Low-Frequency) Resonance

The situation changes when the pump modulation frequency lies near $\omega_m \approx \omega_R \approx 10^2 \text{ Hz}$, where pronounced nonlinear low-frequency resonance is observed. If the depth of periodic modulation is small, the phonon SE integral intensity $J_S(t)$ oscillates synchronously with the external force period: $J_S(t) = J_S(t + \tau_m)$, where $\tau_m = 2\pi/\omega_m$. As the pump modulation depth $k_m$ increases, the period of $J_S(t)$ doubles according to the Feigenbaum scenario $J_S(t) = J_S(t + 2t_m)$, where $f = f(k_m)$ successively takes values $f = 1, 2, 3, \ldots$, which condense $(f \to \infty)$ in the vicinity of a critical point $k_m = k_m^{(c)}$.

A further increase in the depth of modulation $(k_m > k_m^{(c)})$ switches the phaser into the state of deterministic chaos. In the case of hard excitation (for example, by a pulse of hypersound injected into the FPAR from the outside), a phaser with periodically modulated pump exhibits multistability of microwave acoustic SE (branching of periodic and/or deterministic chaotic regimes of the phaser generation, which may accompanied by hysteresis). Finally, the collision of a strange attractor with an unstable manifold that separates the upper periodic SE branch leads to so-called crises (sharp changes of the basin of a strange attractor, which are accompanied by attractor reconfiguration).

However, all the above phenomena were detected by measuring $J_3(t)$. More detailed information about phaser destabilization by a periodic force can be extracted from the microwave spectral characteristics of phonon SE. It has been found (Fig.2) that, when the depth of modulation increases, modes of AMPS alternate in a stepwise manner and cover the entire spectrum of phaser generation (for the Fig.2 in the journal variants of this paper see 24).
A panoramic view of the AMPS shows that the position of each individual SE mode is apparently the same at all the stages of the AMPS alternations. In other words, an individual AMPS mode may generate or not (at this or that stage of SE evolution), but the position of every generating phonon mode is practically same, as the position of corresponding “cold” (unpumped) FPAR absorptive mode. Strictly speaking, one can see small amplitude and frequency motions of SE modes if the sweep range of the spectrum analyzer is decreased by two or three orders of magnitude, but these small-scale motions are insufficient at the background of global chaotic reconfigurations of the AMPS under relaxational resonance.

The described picture of resonant destabilization of the microwave phonon SE is typical for the whole range of the broad-band relaxational resonance at $\omega_m = 70 - 200$ Hz. In a number of cases, the intensity of the most powerful components of nonautonomous SE exceeds the intensity of the central SE component of an autonomous phaser by two orders of magnitude. The evolution of the SE spectrum shown in Fig. 2 (in discrete time) illuminates the actual complicated motions in the spin-phonon system of an acoustic quantum generator, which is shaded by the integral value $J_N(t)$. A similar evolution of the AMPS was also observed in experiments, where pump was stationary, but magnetic field $H$ was modulated at relaxational resonance frequencies $\omega_m = 70 - 200$ Hz.

The mean lifetime of microwave phonon modes under the conditions shown in Fig. 2 is several tenths of a second. The modes pattern changes in an irregular manner, the mode distribution in AMPS is not repeated, etc. Such a chaotic evolution of the phonon SE spectra took place over the entire frequency range $\omega_m = 70 - 200$ Hz, where resonant low-frequency destabilization of phaser generation and, accordingly, chaotic oscillations of $J_N(t)$ were previously observed \cite{27, 28}.

C. Superslow Self-Organized Motions in Phaser under Fundamental $\lambda$-Resonance

Under the infrasonic-frequency modulation ($\omega_m \approx 10$ Hz) of pump or magnetic field, the destabilization of microwave phonon SE assumes another, laminar, character. Unlike the case of relaxational resonance ($\omega_m = 70 - 200$ Hz), the resonance at infrasonic frequencies $\omega_m \approx 10$ Hz is characterized by an extremely high correlation of spectral motions. If the modulation frequency $\omega_m$ is precisely tuned to the vertex $\omega_{\lambda}$ of $\lambda$-resonance and the depth of modulation is high (close to 100%), the AMPS narrows roughly fourfold and has no greater than six or seven modes of microwave phonon SE.

With a small detuning $\Delta_\lambda \equiv \omega_m - \omega_{\lambda}$ of pump modulation frequency $\omega_m$ with respect to the vertex $\omega_{\lambda}$ of the $\lambda$-resonance, these narrow SE spectra demonstrate regular self-reconfigurations with intriguing features. It was found experimentally that the period of the self-reconfigurations $\tau_d^{(\lambda)}$ changes by several orders of magnitude when $\Delta_\lambda$ varies by no more than 1 Hz. In addition, the period $\tau_d^{(\lambda)}$ turned out to be incommensurate with the period $\tau_m \equiv 2\pi/\omega_m$ of external force (that is, the frequency $\omega_d^{(\lambda)} \equiv 2\pi/\tau_d^{(\lambda)}$ generally is not a harmonic or subharmonic of the driving force frequency $\omega_m$). In experiments, this shows up as the instability of states that have rational values $\tau_d^{(\lambda)}/\tau_m$.

The essence of the revealed self-reconfigurations of the phonon SE spectra is the $\tau_d^{(\lambda)}$-periodic unidirectional moving of the range of active (i.e. generating) modes along the SE-frequency axis $\Omega$ if $\omega_m \approx \omega_{\lambda}$. This range typically comprises from three to seven microwave phonon SE modes.

It is noteworthy that the frequency position of each of the SE modes remains nearly unchanged (if higher order dynamic effects due to the nonstationary fine structure of the SE spectra \cite{24} are disregarded). Only the position of the spectral part with active modes changes. Thus, the ignition of new FPAR modes at one side of the AMPS (beginning from some discrete starting position $\Omega_N^{(\text{start})}$) is accompanied by the extinguishing of the same number of modes at the opposite side of the AMPS. Such a motion lasts until the phaser generation ceases completely (at some discrete finishing position $\Omega_N^{(\text{finish})}$ of the frequency axis $\Omega$). After a relatively short period of complete absence of phaser generation (time of refractority), the process of AMPS global self-reconfiguration is repeated, starting from the same position $\Omega_N^{(\text{start})}$ on the frequency axis $\Omega$. 

![FIG. 2: Evolution of the phonon SE spectra near the low-frequency relaxational resonance at $\omega_m = 137$ Hz. The step between sequential registered stages of AMPS evolution $E_K$ is roughly equal to 1 s.](image)
FIG. 3: Evolution of the phonon SE spectra near the ultralow frequency $\lambda$-resonance at $\omega_m = 9.56$ Hz. The step between sequential registered stages of AMPS evolution $E_K$ is about 2.5 s.

The period $\tau_d^{(\lambda)}$ of these unidirectional spectral motions remains the same if a set of control parameters does not change. On the screen of a spectrum analyzer, this evolution of the microwave phonon SE spectrum appears as the periodic motion of some mode cluster (a set of simultaneously generating modes, having fixed own frequencies). Typical sequences of microwave phonon SE spectra at the $\lambda$-resonance conditions ($\omega_\lambda = 9.79$ Hz, $\Delta_\lambda = -0.23$ Hz) in the absence of static magnetic field offset ($\Delta_H = 0$) are shown in Fig.3.

An instantaneous set of microwave phonon SE modes forms a cluster of certain width, and this width varies insignificantly during the spectral motion (Fig.3). At the same time, a set of individual SE modes (that form active cluster) permanently varies (see discrete positions $N$ of generating modes $\Omega_N$ for successive AMPS snapshots $E_K$ at Fig.3). As follows from Fig.3, such the self-reconfigurations of the AMPS are imposed on irregular oscillations of the SE mode intensity.

When the sweep range of the spectrum analyzer is decreased by two or three orders of magnitude, very weak irregular motions of SE modes along the frequency axis $\Omega$ and sometimes splittings of these modes are observed (Fig.4). The modes split when their intensity are very low, e.g. before extinguishing (because of this, the instrument noise is noticeable in Fig.4). Obviously, such fine effects cannot be seen on panoramic spectra in Fig.3, where the positions of $\Omega_N$ are apparently fixed (the fine structure of the AMPS in an autonomous ruby phaser was studied by us in [19, 24]). In general, it can be said that large-scale ordered (laminar) motions of SE spectra in a phaser with infrasonic-frequency pump modulation are imposed on small-scale irregular motions.

Similar large-scale laminar self-reconfigurations of phonon microwave spectra in a nonautonomous ruby phaser were observed in experiments at $\Delta_\lambda = +0.23$ Hz; however, the cluster moved in the opposite direction. Further investigations showed that the sign of the derivative $d\Omega_V/dt$ (here, $\Omega_V$ is the center frequency of a mode cluster) strictly correlates with the sign of frequency detuning of the external force from resonance: $\text{sgn}[d\Omega_V/dt] = -\text{sgn}\Delta_\lambda$. Going toward the exact $\lambda$-resonance discovered ($|\Delta_\lambda| \to 0$), the self-reconfigurations period $\tau_d^{(\lambda)}$ takes giant values. Direct measurements of $\omega_d^{(\lambda)}$ were performed with superfluid helium ($T = 1.8$ K) in order to avoid problems associated with cryogenic liquid boiling.

Such a character of AMPS self-reconfigurations was observed not only for zero-offset of static magnetic field ($\Delta_H = 0$); it persists over a wide range of $\Delta_H$. Moreover, at $|\Delta_H| < 10$ Oe, the value of $\omega_\lambda$ even remains almost constant (close to 9.8 Hz). Only when the magnetic field offset modulo $|\Delta_H|$ increases further, does the resonance frequency $\omega_\lambda$ decrease tangibly (about twofold for $|\Delta_H| \approx 60$ Oe). It is essential that the above dependence of the motion direction of an SE mode cluster on the modulation frequency detuning, $\text{sgn}[d\Omega_V/dt] = -\text{sgn}\Delta_\lambda$, remains valid.

D. Even and Odd Harmonics of $\lambda$-Resonance

Along with the discontinuous regimes of phaser generation (which include the time of refractority at each period of the AMPS self-reconfigurations), we also observed regimes without refractorities. For such the regimes at least one SE mode appears in the AMPS starting range before the last SE mode in the finishing range disappears. In other words, there are two narrow SE mode clusters (at the $\Omega$ axis) during some part $\delta \tau_d^{(\lambda)}$ of the AMPS self-reconfiguration period. The virtual vertici of these clusters $V_1$ and $V_2$ are moving simultaneously along the $\Omega$ axis in the same direction and with the same velocity: $d\Omega_V/\Omega_V = d\Omega_V/\Omega_V$ during $\delta \tau_d^{(\lambda)}$.

The same effect was observed at the first three even harmonics of $\lambda$-resonance, i.e. at $\omega_m \approx \omega_{2s\lambda} \equiv 2s\omega_\lambda$, where $s \in \{1, 2, 3\}$. As for the fundamental $\lambda$-resonance (for which $\omega_m = \omega_\lambda$), in the case of poined lowest $2s\lambda$-resonances correspondent periods of the AMPS self-reconfigurations $\tau_d^{(2s\lambda)}$ are incommensurate with the external force period $\tau_m \equiv 2\pi/\omega_m$. Experiments at $T = 1.8$ K show, that these periods $\tau_d^{(2s\lambda)}$ are increasing up to 100 s and more if the absolute values of detunings $|\Delta_L^{(2s\lambda)}| \equiv |\omega_m - 2s\omega_\lambda|$ are small (less than 0.05-0.1 Hz). The sign of $d\Omega_V/\Omega_V$ (or $\text{sgn}[d\Omega_V/\Omega_V]$, $\text{sgn}[d\Omega_V/\Omega_V]$ for
off-refractority regimes) was opposite to the sign of $\Delta L_{2\lambda}$ (as in the case of the fundamental $\lambda$-resonance).

With $4 \leq s \leq 11$, the driving force frequency $\omega_m$ falls into the range of broad-band relaxation resonance ($\omega_m = 70 – 200 \text{ Hz}$) described above. However, for $s > 11$ (when the driving force leaves this range of resonant destabilization) experiments again distinctly revealed narrow-band resonant responses of the generating phaser system to the pump modulation. These narrow-band resonances were observed not only at even harmonics $\omega_{2s\lambda}$ of the fundamental $\lambda$-resonance but also at odd harmonics $\omega_{(2s+1)\lambda}$. $s \approx (2s + 1)\omega_{\lambda}$.

Responses of the ruby phaser at the pointed high harmonics (both even and odd) somewhat differ from those in the case of fundamental resonance $\lambda$-resonance and its first even harmonics. For example, at $s > 11$, the deviation of $\Omega_N$ is, as a rule, no greater than one or two SE mode separations (i.e., no greater than 0.3–0.6 MHz). Besides of this, at $s > 11$, SE modes experience deep periodic automodulation (of depth 50% or more).

At $s > 11$ ($\omega_m > 215 \text{ Hz}$) and with detunings $\approx 1 \text{ Hz}$, this automodulation is fast (its period is about a second). As $\omega_{\lambda}$ approaches the top of each of the $2s\lambda$- or $(2s + 1)\lambda$-resonances (at the same $s > 11$), the automodulation period $\tau_{am}$, as well as the period $\tau_d^{(\lambda)}$ of the AMPS self-reconfigurations at $\omega_m \approx \omega_{\lambda}$ and lower even harmonics ($s < 4$), increases monotonically. The highest values of $\tau_{am}$, which were stably observed at $11 < s < 20$, reached several minutes for $T = 1.8 \text{ K}$. Note that the same “blinking” regimes of phonon SE (but with smaller automodulation depths) were also found for the first two odd harmonics ($\omega_m \approx 3\omega_{\lambda}$ and $\omega_m \approx 5\omega_{\lambda}$), where “great” self-reconfigurations of the AMPS are absent.

IV. DISCUSSION

Our experimental data for infrasonic-frequency-modulated phaser suggest spin-phonon self-organization caused by intermode energy exchange at hypersonic frequencies. It should be emphasized that under infrasonic $\lambda$-resonance highly organized collective motions in the spin-phonon system are observed not only for each of individual microwave acoustic modes (as in the case of autonomous multimode phaser generation) but also at the global level, where all SE modes obey the same rhythm (the frequency of which is not a harmonic/subharmonic of an external perturbation). In other words, if upon autonomous multimode phaser generation there exist $N$ virtually independent microwave oscillators (each corresponding to a specific SE mode), at a resonant infrasonic perturbation of the pump or magnetic field, these oscillators behave consistently (cooperatively). The same phenomenon of global-level self-organization in ruby phaser was observed by us at some higher harmonics of the fundamental $\lambda$-resonance (outside of the broad-band relaxation resonance, having low, — but not infrasonic, — fundamental frequency).

The features of collective motions in a phaser suggest that this effect is similar to antiphase dynamics processes $[30, 31, 32]$, which were discovered previously in multimode lasers. In the simple case of two-mode lasing $[30]$, antiphase dynamics appears as consistent oscillations of modes strictly in antiphase. In $N$-mode systems, antiphase motions may be much more complicated (see, e.g., $[31, 32]$); however, the general nontrivial tendency, namely, coherent unidirectional SE mode oscillations with a time delay $\tau_d^{(\lambda)}/N$ between nearest neighbors, still persists.

As was found in this work, energy exchange between microwave phonon SE modes in a phaser results in an additional characteristic frequency $\omega_{\lambda}$, which is much lower than the relaxation frequency $\omega_R$. Collective motions are excited when a control parameter (pump or magnetic field) of the active system is modulated at frequencies close to $\omega_{\lambda}$. The same is true for lasers exhibiting antiphase dynamics $[31, 32]$. Accordingly, phonon spectral self-reconfigurations may be treated as the occurrence of antiphase states in phaser system when the spatial distribution of stationary hypersonic SE modes is destabilized by an external force at infrasonic frequencies $\omega_m \approx \omega_{\lambda}$ (or its harmonics). Moreover, the value of $\omega_{\lambda}$ estimated by formulas given in $[30]$ (see Appendix 1) is one order of magnitude lower than $\omega_R$, which is also in agreement with our experimental data for nonautonomous phaser generation.

It should be noted, however, that the laser model of nonlinear dynamics of microwave phonon SE cannot describe adequately all features of self-organization in a phaser near $\lambda$-resonance and its harmonics, although it gives a satisfactory estimate of $\omega_{\lambda}$ and predicts more or less accurately the character of mode motions.

For a better understanding of self-organization in a ruby phaser, one should consider the unusual hierarchy of spin reservoirs $[33]$, which are responsible for many features of the quantum transitions saturation (both electromagnetic and acoustic) in the microwave range. Essentially, all nonlinearities showing up in microwave resonant
interactions of the signal acoustic field and the electromagnetic pump field with the electron Zeeman reservoir $\tilde{Z}_E$ of Cr$^{3+}$ ions in ruby phaser are sensitive to the presence of slowly relaxing Al$^{27}$ nuclei, as revealed in our early experiments on hypersound quantum amplification

The reason for this sensitivity is the direct thermal interaction between the nuclear Zeeman reservoir $\tilde{Z}_N$ and electronic dipole-dipole ($d-d$) reservoir $\tilde{D}_E$. This interaction leads to energy exchange between electronic and nuclear spin reservoirs: $\tilde{Z}_E \approx \tilde{D}_E \approx \tilde{Z}_N$. It is important that the heat capacities of the reservoirs $\tilde{Z}_N$ and $\tilde{Z}_E$ are comparable to each other, although the frequency of nuclear magnetic resonance (NMR) for Al$^{27}$ ($\approx 10$ MHz) is three orders of magnitude lower than the ESR and APR frequencies for Cr$^{3+}$ ions at $H \approx 4$ kOe. This is because the concentration of impurity Cr$^{3+}$ paramagnetic ions Cr$^{3+}$ in pink ruby is as low as several hundredths of a percent; that is, for one electron spin, there are several thousands of nuclear spins. As a result, the inertial nuclear system, while unseen in direct ESR and APR measurements, participates in all population redistributions over electron spin levels (in more exact terms, over a quasicontinuous set of sublevels due to dipole-dipole interactions).

Returning back to the $\lambda$-resonance, we may assume that the observed decrease in its frequency at static magnetic field mismatches is caused by Al$^{27}$ nuclear spin-system, which is thermally connected to Cr$^{3+}$ electronic spin-system. More precisely, $\tilde{Z}_N$ is involved (by the direct thermal contact to $\tilde{D}_E$ and by the indirect one to $\tilde{Z}_E$) in energy exchange between microwave acoustic SE modes. In fact, the most important feature of interaction between the saturated electron subsystem and polarized nuclear subsystem is the strong independence of the spin temperatures (in all the three reservoirs $\tilde{Z}_E$, $\tilde{D}_E$, $\tilde{Z}_N$) on static magnetic field mismatches and/or saturating microwave field detunings in ruby phaser (see our early experimental works).

It is also noteworthy that the effect of the low-energy (combined) reservoir $\tilde{Z}_N + \tilde{D}_E$ on the high-energy reservoir $\tilde{Z}_E$ in a phaser differs radically from the effect of similar low-energy reservoir in optical lasers (see, e.g., [24]). The matter is that, in our system, the relative heat capacity and inertia of combined low-energy reservoir are much greater than in CO$_2$ lasers [24]. First, in our system, as was noted above, for one active center Cr$^{3+}$ there are several thousands of magnetic nuclei Al$^{27}$, which make a low-energy reservoir “heavier”, while in CO$_2$ lasers, the low- and high-energy reservoirs are formed by the same CO$_2$ molecules (the low-energy reservoir is formed here by rotational-vibrational degrees of freedom of CO$_2$ molecules). So, in a CO$_2$ laser, there are analogues to the reservoirs $\tilde{Z}_E$ and $\tilde{D}_E$, but that laser does not have an analogue to the reservoir $\tilde{Z}_N$. Second (and most important), the relaxation time of the low-energy reservoir in ruby phaser is much longer than the relaxation time of the high-energy reservoir [11, 12, 18], while the reverse is true for CO$_2$ laser [24]. Therefore, in a ruby phaser, the combined inertial reservoir $\tilde{Z}_N + \tilde{D}_E$ is involved in the leading self-organization processes which always the slowest processes are.

V. CONCLUSIONS

We experimentally studied the influence of an external periodic force on the dynamics of microwave phonon stimulated emission in a ruby phaser at liquid helium temperatures. It is shown that the periodic modulation of pump (or static magnetic field) at low frequencies $\omega_m = 70 - 200$ Hz chaoticizes energy exchange in the spin-phonon system because of resonant destabilization of a phaser near its relaxation resonance frequency $\omega_R$. In this range of destabilization, the width of the phonon microwave power spectrum does not noticeably change. For ultra-low (infrasonic) frequencies of pump modulation ($\omega_m \approx \omega_\lambda \approx 10$ Hz), a qualitatively new kind of phonon SE destabilization is discovered. First, the AMPS narrows considerably (almost four times). Instead of fast chaotic SE modes alternations, the self-organized periodic slow motions of SE modes near the vertex of the $\lambda$-resonance are observed. Period of these motions may exceed the period of the driving force by several orders of magnitude. The phonon SE self-organization is of a global type: cooperative behaviour in the infrasonically-modulated phaser includes the whole spin-phonon system generating microwave phonons (unlike conventional “individualistic” intramode self-organization, which is typical of multimode lasers).

Self-organization shows up as slow consistent regular pulsations of each of microwave phonon SE modes with a time delay $\tau_\Delta / N$. This appears as autowave motion of a mode cluster in the spectral space. The total self-reconfiguration cycle of AMPS depends considerably on $\omega_m - \omega_\lambda$ and changes by several orders of magnitude when $|\omega_m - \omega_\lambda|$ changes by several percent. The same processes were observed for the first three even harmonics of the fundamental $\lambda$-resonance. For higher even harmonics, as well as for all odd harmonics, “blinking” periodic regimes are discovered (outside of wide-band low-frequency relaxational resonance). The results obtained are treated in terms of the antiphase dynamics of phonon SE. The role of the Al$^{27}$ magnetic nuclear subsystem (with MHz-range NMR-frequencies) in self-organization of GHz-range phonon SE is discussed.

Acknowledgments

The author is grateful to E. D. Makovetsky (Karazin National University, Kharkov) and S. D. Makovetskiy (Kharkov National University of Radiotechnics) for
their valuable help with computer processing of the experimental data, to A. P. Korolyuk (Institute of Radiophysics and Electronics of NASU) for interest in the study of phaser dynamics, and to P. Mandel (Université Libre de Bruxelles) for the kindly submitted publications on antiphase dynamics. Finally, the author is indebted to Academician V. M. Yakovenko and all participants of his seminar for discussion and valuable comments on nonlinear phenomena in phaser system.

This work was partially supported by the Scientific and Technology Center of Ukraine (STCU).

APPENDIX A: NONLINEAR RESONANCES IN A TWO-MODE CLASS-B QUANTUM GENERATOR

In this Appendix, we reproduce some results of the work [30] on antiphase dynamics, where the nature of additional nonlinear resonance in a simple two-mode system was considered basing on the Tang-Statz-DeMars approximation [26].

Equations of motion for a two-mode class-B quantum generator (with \( \tau_1 \gg \tau_C \gg \tau_2 \)) may be formulated using 5D vectorial order parameter \( \vec{B} \):

\[
\dot{\vec{B}}(t) = (J_1, J_2, D_0, D_1, D_2),
\]

where \( J_n(t) \) is the normalized dimensionless intensity of SE for \( n \)-th mode \( (n = 1; 2) \); \( D_j(t) \) is the spatial Fourier components for inversion in an active medium \( (j = 0; 1; 2) \):

\[
D_j(t) = \frac{1}{L_C} \int_0^{L_C} D(z, t) \cos(2k_jz) \, dz.
\]

Here \( D(z, t) \) is normalized spatio-temporal distribution of inversion along axis \( O_z \) of the Fabry-Perot resonator; \( k_0 = 0; k_1, 2 \) are the wave numbers for the SE modes \( (k_1, 2 \gg L_C^{-1}) \); \( L_C \) is the length of the Fabry-Perot resonator. The gain for the 2-nd SE mode \( \alpha_2 \) is assumed to be less than the gain of the 1-st SE mode \( \alpha_1 \).

Nonlinear dynamical model for a class-B quantum generator has such the form:

\[
\tau_1 \frac{d\vec{B}}{dt} = \tilde{\Psi}^{(L)}(\vec{B}) + \tilde{\Psi}^{(NL)}(\vec{B})
\]

where \( \tilde{\Psi}^{(L)} \) are \( \tilde{\Psi}^{(NL)} \) linear and nonlinear parts of the vector field with the following components:

\[
\begin{align*}
\Psi^{(L)}_{1,2} &= -2J_1,2/q_1; \\
\Psi^{(L)}_3 &= A - D_0;
\end{align*}
\]

\[
\begin{align*}
\Psi^{(NL)}_{1,2} &= (2D_0 - D_1,2)\mu_1,2J_1,2/q_1; \\
\Psi^{(NL)}_3 &= [(D_1 - 2D_0)\mu_1J_1 + (D_2 - 2D_0)\mu_2J_2]/2; \\
\Psi^{(NL)}_{4,5} &= \mu_1,2D_0J_1,2 - (\mu_1J_1 + \mu_2J_2)D_1,2
\end{align*}
\]

where \( q_1 \equiv 2\tau_C/\tau_1, \mu_n \equiv \alpha_n/\max(\alpha_n), \) i.e. \( \mu_1 = 1, \mu_2 = \alpha_2/\alpha_1 < 1 \).

Because of \( q_1 \ll 1 \) (in our experiments with acoustic quantum generator \( q_1 \approx 10^{-5} \div 10^{-6} \), see Section III, one can find from equations (A3), (A4) two resonances in our active system.

The first nonlinear resonance is usual relaxational resonance with \( \omega_R = |\text{Im}(\Lambda_{1,2})| \approx |(4 - D_0^{(st)})J_1^{(st)}|^{1/2}, J_1^{(st)} = J_1^{(st)}/2\tau_1\tau_C, M_2 = [4(1 + \mu_2) - 3\mu_2D_0^{(st)}] \), where \( \Lambda_{1,2} \) is the first pair of complex conjugate Liapunov exponents; index \( (st) \) corresponds to the stationary solutions of (A3).

Thus, such the resonance presents in single-mode class-B laser too.

The second nonlinear resonance is due to intermode energy exchange (it is obviously absent in a single-mode class-B laser). The frequency of second nonlinear resonance is [30]: \( \omega_\lambda = |\text{Im}(\Lambda_{3,4})| \), where

\[
|\text{Im}(\Lambda_{3,4})|^2 \approx J_2^2M_2^2D_0^{(st)} - 4(1 - \mu_2)^2 < |\text{Im}(\Lambda_{1,2})|^2.
\]

Here \( A \) is the pump parameter (normalized by the threshold of the single-mode generation), \( A > A^{(2m)} \), \( A^{(2m)} \) is the threshold of the two-mode generation:

\[
A^{(2m)} = \frac{4\mu_2 - 2\mu^2 - 1}{(2\mu_2 - 1)\mu_2}.
\]

Thus, intermode energy exchange in class-B quantum generator leads to appearance of additional resonant frequency \( \omega_\lambda = |\text{Im}(\Lambda_{3,4})| \), which is much less than \( \omega_R \) and may be attributed to infrasound-frequency \( \lambda \)-resonance in ruby phaser (Section III).

APPENDIX B: LIST OF ABBREVIATIONS

AMPS - Acoustic Microwave Power Spectrum
APR - Acoustic Paramagnetic Resonance
ESR - Electron Spin Resonance
FPAR - Fabry-Perot Acoustic Resonator
IR - InfraRed
NMR - Nuclear Magnetic Resonance
SE - Stimulated Emission
