RESULTS OF THE INVESTIGATIONS OF THE NATURE OF THE LONG-TERM INSTABILITY IN QUANTUM FREQUENCY STANDARDS AND MAGNETOMETERS

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The problem analysis results made the author to draw a conclusion that the nature of the resonance frequency long-term instability and drift at harmonic excitation is related to the phase dynamics of the "atom + field" system in the small ε - vicinity of the resonance. The investigation is based on the strictly substantiated asymptotic Krýlov-Bogolyubov perturbation theory. A time-dependent ( drift) first-order correction δω(1) of the perturbing field amplitude E1 (H1) to the resonance frequency ω0 was disclosed. It was found that this correction is always present and is responsible for the frequency drift and long-term instability. The necessary and sufficient conditions of accurate resonance, as well as the conditions of realization of a stable (stationary, steady-state) drift-free oscillation regime in a quantum system, are obtained.

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I. INTRODUCTORY REMARKS

Intensive investigations carried out by different research centers with the aim of development of new physical principles of designing frequency (time) standards of different applications resulted in the high absolute accuracy and short-term instability of frequency 10^−14τ^−1/2 over a time averaging period < 10^4 s. It is well known that the long-term instability of precision and reference atomic frequency standards over a time period of a day is an order of magnitude worse than the short-term instability. The difference in these parameters exists also in new devices, such as ion trap and atomic fountain and in active and passive hydrogen frequency standards. The attainment of a long-term instability comparable with the short-term instability remains a problem, i.e. frequency drift remains nonremovable, its sign and value are unpredictable, and its nature is still unknown.

Some authors tried to attribute the atomic standard, frequency drifts, perceptible even within over time periods τ > 10^4 s, to possible variability of the physical constants - the gravity constant G and the fine structure constant α. Though, another author based on the experimental data analysis, adduces arguments showing the groundlessness of this approach to explain the standard frequency drifts, at least, within time periods under consideration - a month, ...a year, which are extremely short with respect to cosmological measures.

In evaluating standard frequency stability, the "two-selectivity variation” model, proposed by D. Allan, is preferred. This model eliminates the slow drift. Such an evaluation gives a more pleasing result, but doesn’t show the true state of affairs. Theoretical investigations of the interaction of a two-level system with a harmonic field for the special case of the weak field E1/E0(H1/H0) ≪ 1, are described in many papers. This situation is realized in high-accuracy quantum devices under consideration. The papers point out one important result the resonance occurs at the frequency ω = ω0 + δω(2) which differs from the frequency of the unperturbed transition ω0. The arising constant correction δω(2) = (1/4)γH1^2/H0, is of the second order of smallness of the disturbing field H1 (or E1) amplitude, and is known as the Bloch-Siegert shift. The value of the correction is smaller than 10^-6% and in practice it is neglected in most cases.

However, in spite of the progress made towards the understanding of the two-level system dynamics, particularly, of the resonance phenomena in spin-systems, not a single of the theoretical papers, known to us, provides any information on the existence of the slow drift of the resonance center frequency in quantum systems.

A. The problem analysis results

The analysis of the problem from different view points has led us to a number of conclusions.

The generally accepted condition for the resonance Δω = |ω - ω0| = 0 is incomplete. On theory and in practice, it is customary to assume that the condition for the exact resonance is the equality of the difference in tuning frequencies Δω to zero. In practice, this condition is tried to be fulfilled with the highest accuracy through the use of up-to-date facilities of computerized tracking of the resonance center. However, when the resonance phase-frequency characteristic Δφ is taken into account, the equality of detuning Δω = |ω - ω0| = 0 only allows for the first condition of resonance (coherence). On the second resonance condition the theory shows that at zero detuning the oscillation phase difference Δφ between the
field $\vec{H}$, and the atom should be equal to $(-\pi/2)$. The investigations that rigorously prove practical fulfillment of the second conditions of coherence in a quantum system are lacking. The verification of practicability of this condition calls for investigations of the phase state dynamics of the "atom+field" system in the infinitesimal vicinity of resonance.

The basis for the resonance frequency drift is not a technical reason but an obscure physical effect, which brings about the instability of resonance regime of oscillations, and the instability cannot be obviated with a technical means.

B. Statement and solution of the problem

This paper shows the solutions of the following problems 1, 2:

1). Investigation of a phase state dynamics of the two-level system "atom + field" in a weak variable field in a small vicinity of a resonance.

2). The necessary and sufficient conditions for the exact resonance.

3). Existence of the stationary resonant oscillation regime and its steadiness.

The research was carried out using the strictly substantiated asymptotic Krylov-Bogolyubov theory of perturbations 3, 4 which allows studying the system over any time intervals $t \sim 1/\varepsilon$ ($0 < \varepsilon \ll 1$).

The results of the researches allow to explain the nature of long-term frequency (time) instability in reference quantum devices.

II. EQUATION OF THE EXACT FREQUENCY OF A PERTURBED QUANTUM SYSTEM

A. Equation with a small parameter

The typical equations for a density matrix of two-level system interacting with an external weak variable magnetic or electrical field are 3, 4:

\[
\begin{align*}
\dot{\rho}_{22} & = \Lambda_2 - \Gamma_2 \rho_{22} - iV (\rho_{21} - \rho_{12}) \\
\dot{\rho}_{11} & = \Lambda_1 - \Gamma_1 \rho_{11} + iV (\rho_{12} - \rho_{21}) \\
\dot{\rho}_{21} & = - (\Gamma_{21} + i\omega_0) \rho_{21} - iV (\rho_{22} - \rho_{11})
\end{align*}
\]

$\Lambda_1, \Lambda_2$ - rates of non-coherent pumping on the appropriate level; $\Gamma_1, \Gamma_2, \Gamma_{21}$ - relaxation rates;

\[V = \left| 1 \right| \vec{\mu} \vec{V}_1 2 / \hbar\]

- matrix element of interaction; $\vec{\mu}$ - operator of the dipole moment; $\vec{V}_1 (t) = \vec{V}_1 v (t)$ - intensity of a variable magnetic (or electrical) field in dipole approach. Let’s enter dimensionless time $t \rightarrow \omega t$, the Bloch variables $R_1, R_2, R_3$

\[
\begin{align*}
R_1 & = \rho_{12} + \rho_{21} \\
R_2 & = -i(\rho_{12} - \rho_{21}) \\
R_3 & = \rho_{22} - \rho_{11}
\end{align*}
\]

and, differentiating system (1) with respect to dimensionless time, we shall write down as

\[
\begin{align*}
\dot{R}_1 & = -\gamma_1 R_1 - \nu R_2 \\
\dot{R}_2 & = -\gamma_1 R_2 + \nu R_1 + 2\omega_1 v(t) R_3 \\
\dot{R}_3 & = \lambda - \gamma_2 R_3 - 2\omega_1 v(t) R_2
\end{align*}
\]

The designations normalized to $[\omega]$ are used in the system 2: $\nu = \omega_0 / [\omega]$, $\gamma_1 = \Gamma_1 / [\omega]$, $\gamma_2 = \Gamma_2 / [\omega]$ , $\lambda = (\Lambda_2 - \Lambda_1) / [\omega]$, $\omega_1 = \left| 1 \right| \vec{\mu} \vec{V}_1 2 / \hbar [\omega]$. The basic parameter of the problem $\omega_1 \ll 1$ (approach of a "weak" field). In radio spectroscopy $\omega_1 \sim 10^{-4}$, in optics $\omega_1 \sim 10^{-8}$. Parameters $\gamma_1, \gamma_2, \lambda$ are considered as small as $\omega_1$. Let’s specify it obviously by using the system 2, a formal small parameter. Then disturbed system will be written as

\[
\begin{align*}
\dot{R}_1 & = -\varepsilon \gamma_1 R_1 - \nu R_2 \\
\dot{R}_2 & = -\varepsilon \gamma_1 R_2 + \nu R_1 + 2\varepsilon \omega_1 v(t) R_3 \\
\dot{R}_3 & = \varepsilon \lambda - \varepsilon \gamma_2 R_3 - 2\varepsilon \omega_1 v(t) R_2
\end{align*}
\]

For investigations of systems 3 and the "action-angle" variables are usually used 4. However, instead of the Bloch variables $R_1, R_2, R_3$ we shall use the new variables actually observed $a, \psi, z: a = (R_1^2 + R_2^2) \varepsilon^{1/2}$ - amplitude of oscillations, $z = R_3$ - difference of population, $\psi = \text{arctg} (R_2 / R_3)$ - current phase.

In calculating derivatives for new variables, we shall obtain the system

\[
\begin{align*}
\dot{a} & = -\varepsilon \gamma_1 a + 2\varepsilon \omega_1 z v(t) \sin \psi \\
\dot{z} & = \varepsilon \lambda - \varepsilon \gamma_2 z - 2\varepsilon \omega_1 a v(t) \sin \psi \\
\dot{\psi} & = \omega (t, \varepsilon) = \nu + \varepsilon (2\omega_1 z / \alpha) v(t) \cos \psi
\end{align*}
\]

B. The exact resonant frequency

The last equation is the exact frequency in the perturbed system. First term is the transition eigenfrequency in the unperturbed system; shows that the oscillations occur with a constant frequency $\omega (t, \varepsilon = 0) = \nu$.

Second term is the generalized correction for a change of the eigenfrequency under influence of a variable field. This component, as will be shown below, is responsible for occurrence of the time constant second order corrections (known as the Bloch-Siegert shift), third, fourth, orders and the time variable correction of the first order of the field.
III. RESONANCE IN TWO-LEVEL SYSTEM

The perturbing field is a periodic field \( v(t) = \cos \omega t = \cos t \), where the last \( t \) is a dimensionless value and \( [\omega] = \omega \). The system [4] contains the equations with two fast phases: one phase (nonsynchronous) is \( \psi \), a role of the second phase (isochronal) is carried out by \( t \). The general research methods of type [4] systems are developed in [3, 10]. The case of the main resonance with corresponding equality \( 1 - \nu = 0 \) is most important. The experience shows that even a very accurate equality of the frequencies, \( \omega = \omega_0 \) (i.e. \( \Delta \omega = 0 \)), there is a slow drift of the centre of the resonance which is not eliminated technically. This makes it necessary to study the second coherence condition, i.e. the system phase state in the vicinity of resonance.

A. Dynamics of the system phase state in the \( \varepsilon \) - vicinity of resonance

For studying the phase state dynamics of the system [4] in the vicinity of the resonance we shall use a new variable \( \tilde{\vartheta} = t - \psi \), which is a difference between oscillation phases of the field and the atom and is a slow variable in the \( \varepsilon \) - vicinity of resonance. We shall obtain the equations in the standard Krylov-Bogolyubov form [4, 10]

\[
\dot{\bar{\alpha}} = -\varepsilon \gamma_1 \alpha - \varepsilon \omega_1 z \sin \bar{\vartheta} + \varepsilon \omega_1 \zeta \sin(2t - \bar{\vartheta})
\]

\[
\dot{\bar{z}} = \varepsilon \lambda - \varepsilon \gamma_2 \zeta + \varepsilon \omega_1 \alpha \sin \bar{\vartheta} - \varepsilon \omega_1 \alpha \sin(2t - \bar{\vartheta})
\]

\[
\dot{\bar{\vartheta}} = \varepsilon \Delta - \varepsilon \omega_1 \zeta / \alpha \cos \bar{\vartheta} - \varepsilon (\omega_1 z / \alpha) \cos(2t - \bar{\vartheta})
\]

(5)

where a small frequency detuning is \( \varepsilon \Delta = 1 - \nu = (\omega - \omega_0) / \omega \).

A method of averaging is applied to system [4, 9, 10]. Following this method, we shall use evolitional (drift) components \( \bar{\alpha}(\tau) \), \( \bar{z}(\tau) \), \( \bar{\vartheta}(\tau) \) (\( \tau = \varepsilon t \)-slow time) in variables \( \alpha(t) \), \( z(t) \), \( \vartheta(t) \):

\[
\alpha(t) = \bar{\alpha}(\tau) + \varepsilon u_1(\bar{\alpha}, \bar{z}, \bar{\vartheta}, t) + ...
\]

\[
z(t) = \bar{z}(\tau) + \varepsilon v_1(\bar{\alpha}, \bar{z}, \bar{\vartheta}, t) + ...
\]

\[
\bar{\vartheta}(t) = \bar{\vartheta}(\tau) + \varepsilon g_1(\bar{\alpha}, \bar{z}, \bar{\vartheta}, t) + ...
\]

(6)

which satisfy to system of the evolitional (averaged) equations of a form

\[
\dot{\bar{\alpha}} = \varepsilon A_1(\bar{\alpha}, \bar{z}, \bar{\vartheta}) + \varepsilon^2 A_2(\bar{\alpha}, \bar{z}, \bar{\vartheta}) + ...
\]

\[
\dot{\bar{z}} = \varepsilon Z_1(\bar{\alpha}, \bar{z}, \bar{\vartheta}) + \varepsilon^2 Z_2(\bar{\alpha}, \bar{z}, \bar{\vartheta}) + ...
\]

\[
\dot{\bar{\vartheta}} = \varepsilon H_1(\bar{\alpha}, \bar{z}, \bar{\vartheta}) + \varepsilon^2 H_2(\bar{\alpha}, \bar{z}, \bar{\vartheta}) + ...
\]

(7)

With the use of averaging, we shall define the oscillating corrections of the first order

\[
u_1 = - (\omega_1 / 2) \bar{z} \cos(2t \cos \bar{\vartheta} + \sin 2t \sin \bar{\vartheta})
\]

\[
g_1 = (\omega_1 \bar{z} / 2 \bar{\alpha}) \sin 2t \cos \bar{\vartheta} - \cos 2t \sin \bar{\vartheta}
\]

(8)

and equations for evolitional (drift) components \( \tilde{\alpha}(\tau) \), \( \tilde{z}(\tau) \), \( \tilde{\vartheta}(\tau) \) in the second order approximations

\[
\dot{\tilde{\alpha}} = -\varepsilon \gamma_1 \tilde{\alpha} - \varepsilon \zeta \omega_1 \sin \tilde{\vartheta}
\]

\[
\dot{\tilde{z}} = \varepsilon \lambda - \varepsilon \gamma_2 \zeta + \varepsilon \omega_1 \tilde{\alpha} \sin \tilde{\vartheta}
\]

\[
\dot{\tilde{\vartheta}} = \varepsilon \Delta - \varepsilon \omega_1 \zeta / \alpha \cos \tilde{\vartheta}
\]

(9)

B. Exact resonance condition – nessesary and sufficient

The third equation in system [4] represents an analytical form of a condition of strictly coherent interaction of two-level system with a resonant field. This condition consists in constancy in time of a difference of the current phases between a field and atom, i.e. \( \dot{\vartheta} = 0 \). At the same time this condition \( \dot{\vartheta} = 0 \) is a necessary and sufficient condition for the exact resonance.

Let’s pursue the brief analysis of the third equation. First term is equality of detuning to zero (\( \Delta = 0 \)), i.e. equality of frequencies \( \omega = \omega_1 \) (which is sought in practice), and only partially characterizes a resonance condition and is a necessary condition of the resonance. Third term \( \delta \omega = \omega_1^2 / 4 \), the constant correction, is the Bloch-Siegert shift. Second term \( \delta \omega = -\varepsilon (\omega_1 \zeta / \bar{\alpha}) \cos \bar{\vartheta} \) - the variable (drift) correction to resonance frequency in the first order. This correction significant effects the resonance condition, i.e. a two-level system can make a long-time drift of a resonant frequency being in the state of zero detuning \( \Delta = 0 \). Let’s write down a general form of equation for the resonance frequency of a two-level system interacting with a weak harmonic field

\[
\omega = \omega_0 + \delta \omega = \omega_0 + \delta \omega + \delta \omega + ... 
\]

IV. STEADINESS OF A STATIONARY RESONANT REGIME

The consideration of a problem of existence and steadiness of the stationary oscillation regimes in the system of equations [4] allows to make clear a possibility of practical realization of sufficient conditions of resonance at \( \dot{\vartheta} = 0 \) (for performance of sufficient conditions it is necessary to determine the stationary values of \( \tilde{\vartheta}_s \), \( \bar{\vartheta}_s \), \( \bar{\vartheta}_s \) and accuracy of their maintenance in time). The most important question is, in what degree the equality \( \bar{\vartheta}_s(\Delta = 0) = \pm \pi / 2 \) should be realized in practice.

A. Existence of stationary resonant oscillation regimes

Stationary regime, as is known [10], is characterized by an invariance in time of the all output signal parame-
ters - amplitude, phase and frequency. Stationary values of variables, designated as $\alpha_s$, $\bar{\varepsilon}_s$, $\bar{\vartheta}_s$, are defined from system $[\text{10}]$. This system of equations has the unambiguous solution

$$
\begin{align*}
\bar{\alpha}_s &= -\omega_1 \gamma_1^{-1} \bar{\varepsilon}_s \sin \bar{\vartheta}_s \\
\bar{\varepsilon}_s &= \lambda/ \left[ \gamma_2 + \gamma \omega_1^2 \left( \gamma_1^2 + \lambda^2 \right)^{-1} \right] \\
\bar{\vartheta}_s &= -\arctan \left( \bar{\lambda}/\gamma_1 \right)
\end{align*}
$$

From system $[\text{10}]$ we shall find the stationary values of variables for a resonance:

$$
\begin{align*}
\bar{\vartheta}_s (\bar{\lambda} = 0) &= \pm \pi/2 & \text{- difference of phase} \\
\bar{\varepsilon}_s (\bar{\lambda} = 0) &= \lambda/2 \gamma_2 & \text{- difference of population} \\
\bar{\alpha}_s (\bar{\lambda} = 0) &= \lambda/2 (\gamma_1 \gamma_2)^{1/2} & \text{- oscillation amplitude}
\end{align*}
$$

The obtained result means that in system $[\text{10}]$ there is a single stationary regime of sustained oscillations at the resonant frequency.

**B. Steadiness of resonant oscillation regimes**

The stationary values for $\bar{\alpha}_s$ and $\bar{\varepsilon}_s$ always exist because of the relaxation’s terms ($\gamma_1, \gamma_2$) in system of the equations $[\text{14}]$. The similar conclusion for the difference of phases $\bar{\vartheta}_s$ unequivocally cannot be made. There is a question arising: with what accuracy must the equality $\bar{\vartheta}_s = \pm \pi/2$ take place to provide the conditions of existing of the stationary regime $[\text{11}]$ and its steadiness at which the slow drift of the frequency is excluded. The quantitative estimation of an accuracy with which the equality $\bar{\vartheta}_s (\bar{\lambda} = 0) = \pm \pi/2$ would be executed the V. Volosov theorem about stability of stationary resonant regimes of oscillations is given $[\text{17}]$.

In order to attain steadiness of a stationary resonance regime of the "atom+field" system, initial conditions should be simultaneously set for the three parameters, i.e. oscillation amplitude, population difference, and the phase difference between the field and the atom in the vicinity of their stationary values. In so doing, the applied field frequency is assumed to be constant, $\omega = \text{const}$ ($\varepsilon$ is the Rabi frequency-to-applied field frequency ratio).

When $\omega \neq \text{const}$, as is usually the case, the range of the stationary regime steadiness decreases down to the $\varepsilon^2$-vicinity.

The impracticability of these conditions, in principle ($\varepsilon \sim 10^{-4}$ in radio spectroscopy and $\varepsilon \sim 10^{-9}$ in optics) results in solely non-steadiness operating regimes in spite of a great variety of frequency stabilization systems in quantum standards. As a consequence of the non-steadiness oscillation regime, a slow resonance center frequency drift, unpredictable both in sign and magnitude, appears in the system. This drift radically limits the long-term stability of quantum metrology instruments.

The presence of non-steadiness even in a simple two-level system should be taken into account in designing practical precise quantum devices with high long-term stability.

**V. SUMMARY**

The following results are achieved:

1). For the first time the existence of a variable component of the first order is stated as regards the $H_1 (E_1)$ disturbance field amplitude in the resonance frequency of the two level system interacting with a weak variable field.

2). As a result, necessary and sufficient conditions for the accurate resonance are obtained being distinct from the commonly accepted ones and topical for frequency standards and quantum magnetometers. In particular, the most important sufficient condition is a necessity of the coherence excitation with the specified phase at the initial time instant.

3). When the sufficient conditions are not satisfied (a situation which always takes place in practice), the oscillation regime with a principally non-removable unsteadiness is performed in the quantum system. It is expressed in drift and in found by the author long-term variations of all output signal parameters.

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