NEW RESULTS OF THE INVESTIGATIONS OF THE NATURE OF THE LONG-TERM INSTABILITY IN QUANTUM FREQUENCY STANDARDS AND MAGNETOMETERS. NON-REMOVABLE RESONANCE DRIFT.

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ABSTRACT

The problem analysis results made the authors 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 infinitesimal ε - vicinity of the resonance. The investigation is based on the strictly substantiated asymptotic Krylov-Bogolyubov perturbation theory. A time-dependent (drift) first-order correction $\delta\omega^{(1)}$ of the perturbing field amplitude H_1 to the resonance frequency ω_0 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.

long-term instability, frequency drift, resonance conditions, unsteadiness, frequency standard, quantum magnetometer

1. INTRODUCTION

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 $10^{-14}\cdot\tau^{-1/2}$ 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 $\tau > 10^4$ s, to possible variability of the physical constants – the gravity constant G [Ref. 1] and the fine structure constant α [Ref. 2]. Though, another author [Ref. 3], 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 $H_1/H_0 << 1$, 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 $\omega = \omega_0 + \delta\omega^{(2)}$, which differs from the frequency of the unperturbed transition ω_0. The arising constant correction $\delta\omega^{(2)} \cong ¼\, \gamma\, H_1^2/ H_0$ is of the second order of smallness of the disturbing field H_1 (or E_1) amplitude, and is known as the Bloch-Siegert shift. The value of the correction is $< 10^{-6}$ %, and in practice it is neglected in most cases.

A general solution of the problem in the dynamics of two-level spin-systems is given in [Ref. 5]. In this paper a fundamental system of solutions for any relationships H_1/ H_0 and a harmonic field frequency ω is derived without any simplifying assumptions and approximations. The system of solutions not only generalized all kinds of observed ″periodic″ resonances – longitudinal and transversal (diagonal and nondiagonal), but also indicates the presence of new, nonperiodic resonances in the ″atom + field″ system.
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 ω in quantum systems.

1.1 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 $\Delta\boldsymbol{\omega} = |\ \boldsymbol{\omega}_0 - \boldsymbol{\omega}\ | = 0$ 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 $\Delta\boldsymbol{\omega}$ to zero, (i.e. $\Delta\boldsymbol{\omega} = |\boldsymbol{\omega}_0 - \boldsymbol{\omega}| = 0$).
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 $\Delta\varphi\ (\Delta\boldsymbol{\omega})$ is taken into account, the equality of detuning $\Delta\boldsymbol{\omega} = 0$ only allows for the 1st condition of resonance (coherence). On the 2nd resonance condition the theory shows that at zero detuning ($\Delta\boldsymbol{\omega} = 0$) the oscillation phase difference $\Delta\varphi$ between the field H_1 and the atom should be equal to $-\pi/2$ ($\Delta\varphi = -\pi/2$) which allows for the 2nd condition of resonance (coherence).
The investigations that rigorously prove practical fulfilment 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.

1.2 Statement and solution of the problem [Refs 14, 15]

This paper shows the solutions of the following problems:
a) Investigation of a phase state dynamics of the two-level system ″atom + field″ in a *weak variable field,* H_1 (or E_1), in a *small vicinity* of a resonance.
The research was carried out using the strictly substantiated asymptotic Krylov-Bogolyubov theory of perturbations [Refs 6, 7] which allows to study the system over any time intervals $t \sim 1/\varepsilon$ $(0 < \varepsilon << 1)$.
b) *Existence of the stationary resonant oscillation regime* and its *steadiness*.
The results of the researches allow to explain the nature of long-term frequency (time) instability in reference quantum devices.
c) The necessary and sufficient conditions for the exact resonance.

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

2.1 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 [Refs 8, 9]

$$\dot{\rho}_{11} = \Lambda_1 - \Gamma_1 \rho_{11} - i V (\rho_{21} - \rho_{12})$$
$$\dot{\rho}_{22} = \Lambda_2 - \Gamma_2 \rho_{22} - i V (\rho_{21} - \rho_{12}) \quad (1)$$
$$\dot{\rho}_{21} = -(\Gamma_{21} + i\omega_0)\rho_{21} - i V (\rho_{22} - \rho_{11})$$

where Λ_1, Λ_2 – rates of non-coherent pumping on the appropriate level; Γ_1, Γ_2, $\Gamma_{12} = \Gamma_{21}$ – relaxation rates;
$\hat{V} = < 1 | \hat{\mu}\, \mathbf{V}_1 | 2 > / \hbar$ - matrix element of interaction, $\hat{\mu}$ - operator of the dipole moment, $\mathbf{V}_1(t) = V_1 \cdot \upsilon(t)$ – intensity of a variable magnetic (or electrical) field in dipole approach.
Let's enter dimensionless time $t = \omega t$, the Bloch variables R_1, R_2, R_3 [Ref. 8]

$$R_1 = \rho_{12} + \rho_{21} = \mathrm{Re}\,\rho_{12} = |\rho_{12}| \cos\psi$$
$$R_2 = -i(\rho_{12} - \rho_{21}) = \mathrm{Jm}\,\rho_{12} = |\rho_{12}| \sin\psi$$
$$R_3 = \rho_{22} - \rho_{11}$$

and, differentiating system (1) with respect to dimension-less time t, we shall write down as

$$\dot{R}_1 = -\gamma_2 R_1 - \nu R_2$$
$$\dot{R}_2 = -\gamma_2 R_2 + \nu R_1 + 2\omega_1 \cdot \upsilon(t) R_3 \quad (2)$$
$$\dot{R}_3 = \lambda - \gamma_1 R_3 - 2\omega_1 \cdot \upsilon(t) R_2$$

The designations normalized to ω are used in the system (2) :

$\gamma_1 = \Gamma_1 = \Gamma_2/\omega$, $\gamma_2 = \Gamma_{12} = \Gamma_{21}/\omega$, $\nu = \omega_0/\omega$, $\lambda = (\Lambda_2 - \Lambda_1)/\omega$,
$\omega_1 = < 1 | \hat{\mu}\, \mathbf{V}_1 | 2 > / \hbar\, \omega$ (or $\omega_1 = \gamma H_1/\omega$).
The basic parameter of the problem $\omega_1 \ll 1$ (approach of a ″weak″ field). In radiospectroscopy $\omega_1 \sim 10^{-4}$, in optics $\omega_1 \sim 10^{-8}$. Parameters γ_1, γ_2, λ are considered as small as ω_1. Let's specify it obviously by using the system (2) a formal small parameter ε ($0 < \varepsilon << 1$). Then disturbed system will be written as

$$\dot{R}_1 = -\varepsilon\gamma_2 R_1 - \nu R_2$$
$$\dot{R}_2 = -\varepsilon\gamma_2 R_2 + \nu R_1 + 2\varepsilon\omega_1 \cdot \upsilon(t) R_3 \quad (3)$$
$$\dot{R}_3 = \varepsilon\lambda - \varepsilon\gamma_1 R_3 - 2\varepsilon\omega_1 \cdot \upsilon(t) R_2$$

For investigations of systems (3) the ″action-corner″ variables are usually used [Ref. 10].
However, instead of the Bloch variables R_1, R_2, R_3 we shall use *the new variables actually observed* a, z, ψ :
$a = (R_1^2 + R_2^2)^{1/2} = |\rho_{12}|$ - amplitude of oscillations,
$z = R_3$ - difference of population ,
$\psi = \mathrm{arctg}\,(R_2/R_1)$ - current phase.
In calculating derivatives for new variables, we shall *obtain the system*

$$\dot{a} = -\varepsilon\gamma_2 \cdot a + 2\varepsilon\omega_1 z \cdot \upsilon(t) \sin\psi$$
$$\dot{z} = \varepsilon\lambda - \varepsilon\gamma_1 \cdot z - 2\varepsilon\omega_1 a \cdot \upsilon(t) \sin\psi \quad (4)$$
$$\dot{\psi} = \nu + \varepsilon(2\omega_1 z/a) \cdot \upsilon(t) \cos\psi$$

2.2 The exact resonant frequency

The last equation is the **exact frequency** in the perturbed system.
First term is the transition eigenfrequency, ν, in the unperturbed system; shows that the oscillations occur with a constant frequency, $\dot{\psi} = \nu$.
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 corrections;* corrections of the **second order** (known as the Bloch-Siegert shift), third, fourth,.., orders and the *time variable* ***correction*** of the **first order of the field H_1 amplitude**.

3. RESONANCE IN TWO-LEVEL SYSTEM

The perturbing field is a periodic field $\upsilon(t) = \cos\omega t = \cos t$, where the last t is a dimensionless value. The system (4) contains the equations with two fast phases: one phase (non-isochronal) is ψ, a role of the second phase (isochronal) is carried out by t. The general research methods of type (4) systems are developed in [Refs 6, 7]. 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, $\omega_0 = \omega$ (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.

3.1 Dynamics of the system phase state in the ε - vicinity of resonance

For studying the phase state dynamics of the system (4) in the ε – vicinity of the resonance we shall *use a new variable* $\vartheta = t - \psi$, which is a difference between oscillation phases of the field and the atom and is a slow variable

in the vicinity of resonance.
We shall obtain the equations in the standard Krylov-Bogolyubov form [Ref. 6]

$$\dot{a} = -\varepsilon\gamma_{12}\, a - \varepsilon\,\boldsymbol{\omega}_1\, z \sin\vartheta + \varepsilon\,\boldsymbol{\omega}_1\, z\,(\sin 2t \cos\vartheta - \cos 2t \sin\vartheta)$$
$$\dot{z} = \varepsilon\lambda - \varepsilon\gamma_1 z + \varepsilon\,\boldsymbol{\omega}_1\, a \sin\vartheta - \varepsilon\,\boldsymbol{\omega}_1\, a(\sin 2t\cos\vartheta - \cos 2t \sin\vartheta) \quad \textbf{(5)}$$
$$\dot{\vartheta} = \varepsilon\Delta - \varepsilon\,(\boldsymbol{\omega}_1\, z/a)\cos\vartheta - \varepsilon\,(\boldsymbol{\omega}_1\, z/a)\,(\cos 2t\cos\vartheta + \sin 2t \sin\vartheta)$$

where a small frequency detuning is $\varepsilon\Delta = 1-\nu = (\omega-\omega_0)/\omega$.
A method of averaging is applied to system (5) [Refs 6, 11]. Following this method, we shall use evolutional (drift) components $\bar{a}(\tau)$, $\bar{z}(\tau)$, $\bar{\vartheta}(\tau)$ in variables $a(t)$, $z(t)$, $\vartheta(t)$, where $\tau = \varepsilon t$ – slow time,

$$a(t) = \bar{a}(\tau) + \varepsilon u_1(\bar{a}, \bar{z}, \bar{\vartheta}, t) + \varepsilon^2 u_2(\ldots) + \varepsilon^3\ldots$$
$$z(t) = \bar{z}(\tau) + \varepsilon \upsilon_1(\bar{a}, \bar{z}, \bar{\vartheta}, t) + \varepsilon^2 \upsilon_2(\ldots) + \varepsilon^3\ldots \quad (6)$$
$$\vartheta(t) = \bar{\vartheta}(\tau) + \varepsilon g_1(\bar{a}, \bar{z}, \bar{\vartheta}, t) + \varepsilon^2 g_2(\ldots) + \varepsilon^3\ldots$$

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

$$\dot{\bar{a}}(\tau) = \varepsilon A_1(\bar{a}, \bar{z}, \bar{\vartheta}) + \varepsilon^2 A_2(\bar{a}, \bar{z}, \bar{\vartheta}) + \varepsilon^3\ldots$$
$$\dot{\bar{z}}(\tau) = \varepsilon Z_1(\bar{a}, \bar{z}, \bar{\vartheta}) + \varepsilon^2 Z_2(\bar{a}, \bar{z}, \bar{\vartheta}) + \varepsilon^3\ldots \quad (7)$$
$$\dot{\bar{\vartheta}}(\tau) = \varepsilon H_1(\bar{a}, \bar{z}, \bar{\vartheta}) + \varepsilon^2 H_2(\bar{a}, \bar{z}, \bar{\vartheta}) + \varepsilon^3\ldots$$

With the use of averaging, we shall define the oscillative corrections of the 1st order u_1, υ_1, g_1, unknown coefficients A_1, A_2, .., Z_1, Z_2, .., H_1, H_2, … in system (7) and *derive the system of the evolutional equations* (8) *for dynamics of the resonance* in its ε – vicinity over long period of time τ.
Let's write down these equations in the first and second approximations

$$\dot{\bar{a}}(\tau) = -\varepsilon\gamma_2\cdot\bar{a} - \varepsilon\,\bar{z}\,\boldsymbol{\omega}_1 \sin\bar{\vartheta} + \varepsilon^3\ldots$$
$$\dot{\bar{z}}(\tau) = \varepsilon\lambda - \varepsilon\gamma_1\,\bar{z} + \varepsilon\,\bar{a}\,\boldsymbol{\omega}_1 \sin\bar{\vartheta} + \varepsilon^3\ldots \quad (8)$$
$$\dot{\bar{\vartheta}}(\tau) = \varepsilon\Delta - \varepsilon\,(\boldsymbol{\omega}_1\,\bar{z}/\bar{a})\cos\bar{\vartheta} - \varepsilon^2\,\boldsymbol{\omega}_1^{\,2}/4 + \varepsilon^3\ldots$$

3.2 Exact resonance conditions – necessary and sufficient

The *third equation* in system (8) represents *an analytical form* of a condition of ***strictly coherent interaction*** of two-level system with a resonant field. This condition consists in a constancy in time of a difference of the current phases between a field and atom, i.e. $\dot{\bar{\vartheta}} = \mathbf{0}$. At the same time this condition $\dot{\bar{\vartheta}} = \mathbf{0}$ 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 $\omega_0 = \omega$ (which is sought in practice), and only partially characterizes a resonance condition and is *a necessary condition of the resonance*.
Third term $\delta\omega^{(2)} = \omega_1^2/4$, the constant correction, is the Bloch-Siegert shift.
Second term $\boldsymbol{\delta}\omega^{(1)} = (\omega_1\bar{z}/\bar{a})\cos\bar{\vartheta}$ - the variable (drift) correction to resonance frequency ω_0 in the first order of H_1. This correction significant effects the resonance condition, i.e. a *two-level system can make a long-period 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 ω of a two-level system interacting with a weak harmonic field

$$\omega = \omega_0 + \boldsymbol{\delta}\omega^{(1)} + \delta\omega^{(2)} + \ldots \quad (9)$$

4. 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 (8) allows to make clear a **possibility of practical realization of** *sufficient* conditions of resonance at $\dot{\bar{\vartheta}} = 0$ (for performance of *sufficient* conditions it is necessary to determine the stationary values of $\bar{a}$, $\bar{z}$, $\cos\bar{\vartheta}$ and accuracy of their maintenance in time).
The most important question is, in what degree the equality $|\bar{\vartheta}| = \pi/2$ should be realized in practice. [It is generally accepted that at the resonant frequency ($\Delta\omega = 0$) the equality $|\bar{\vartheta}| = \pi/2$ is a priori].

4.1 Existence of stationary resonant oscillation regimes

Stationary regime, as is known [Refs 7, 11], is characterized by an invariance in time of the all output signal parameters - amplitude, phase and frequency. Stationary values of variables, designated as $\bar{a}_s$, $\bar{z}_s$, $\bar{\vartheta}_s$, are defined from system (8). This system of equations has the an unambiguous solution

$$\bar{a}_s = -\,\omega_1\,\gamma_2^{-1}\,\bar{z}_s \sin\bar{\vartheta}_s$$
$$\bar{z}_s = \lambda/[\gamma_1 + \gamma_2\,\omega_1^2\,(\gamma_2^2 + \Delta^2)^{-1}] \quad (10)$$
$$\bar{\vartheta}_s = -\,\mathrm{arcctg}\,(\Delta/\gamma_2)$$

From system (10) we shall find the stationary values of variables for a resonance:

$\bar{a}_s(\Delta=0) = \lambda/2\cdot(\gamma_1\gamma_2)^{1/2}$ – oscillation amplitude

$\bar{z}_s(\Delta=0) = \lambda/2\cdot\gamma_1$ - difference of population (11)

$\bar{\vartheta}_s(\Delta=0) = -\pi/2$ - difference of phases

The obtained result means that in system (8) there is a *single stationary regime of sustained oscillations at the resonant frequency*.

4.2 Steadiness of stationary resonant oscillation regimes

The stationary values for $\bar{a}_s$ and $\bar{z}_s$ always exist because of the relaxation's terms (γ_1, γ_2) in system of the equations (11).
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 $\bar{\vartheta}_s = -\pi/2$ take place to provide the conditions of existing of the stationary regime (11) and its steadiness at which the slow drift of the frequency is excluded. The quantitative estimation of an accuracy with which the equality $\bar{\vartheta}_s(\Delta=0) = -\pi/2$ would be executed the V.Volosov theorem about stability of stationary resonant regimes of oscillations is given [Ref. 11].

According to this theorem a stationary regime of oscillations at zero detuning, $\Delta = 0$, will be steady, if at the initial time the stationary values of *variables* correspond to system (11), and the most important value of a phase difference $|\overline{\vartheta}_s| = \pi/2$ should be maintained in time with an accuracy of ε^2 ($\varepsilon \sim \omega_1 \sim 10^{-4}$ in radiospectroscopy and $\varepsilon \sim 10^{-8}$ in optics). So the condition for $\overline{\vartheta}_s$ is difficult to obtain in a magnetic resonance at low and medium frequencies, practically impossible – at microwave frequencies and the more so in an optical range.
It results in a *non-steadiness* of oscillation regimes and *non-removable slow drift of a resonance* in precise quantum devices.

Thus, **the very excitation method of a coherence signal by harmonical resonant field is a source of drift and long-term frequency instability of quantum systems.** *The system, tuned to a resonance, makes slow oscillations.*

The radical improvement of long-term stability and complete eliminate of the frequency drift are possible with the use of a **principally other method of coherence excitation** in quantum systems presented in [Refs 12, 13].

5. CONCLUSION

1. The existence of the *first-order correction* $\delta\omega^{(1)}$ due to the perturbing field amplitude H_1 (or E_1) *of the resonance frequency* ω_0 in a two-level system interacting with a weak variable field *was found for the first time.*

2. The first-order correction is *time-variable* and characterizes a *regular component* of the resonance frequency ω_0 *drift.*

3.The *necessary and sufficient conditions for exact resonance,* different from conventional conditions and essential for precision frequency standards and alkali vapor magnetometers, are obtained.

4. At zero detuning, $\Delta = 0$, the *sufficient conditions of realization of a stationary oscillation regime* are defined and which are difficult to realized in magnetic resonance even in the field of low and medium frequencies, practically impossible - at microwave frequencies and the more so in an optical range.

5. When the sufficient conditions for the accurate resonance are not satisfied (the usual situation in practice), the oscillation regime with a principally non-removable non-steadiness is performed in the quantum frequency standards and magnetometers. It is expressed in **drift and long-period variations in time** of resonant frequency.

6. The **harmonical resonant field** used for excitation of the coherence signal **is a source of drift** and **medium**- and **long-term instability** of quantum systems.

7. The results presented in this paper enable the evaluation and prediction of drift and long-period variation values in metrological standards and quantum magnetometers (and also in other quantum devices, for example, ring lasers).

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