

COHERENT PHENOMENA IN SYSTEMS INTERACTING WITH RESONANT RADIATION

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1. INTRODUCTION

AN investigation of quantum transitions occurring following the interaction of an atomic system with the field of optical or radio-frequency resonant radiation is a well-known method of investigating the structure of atomic states and serves as a basis of optical and radio spectroscopy.

The great variety of the phenomena that occur in these processes makes it possible, on the one hand, to reveal fine details of the interaction, and on the other to understand the highly practically important mechanisms of transfer of coherence of the states of the atomic electrons to the radiation field.

Optical orientation of atoms and double radio optical resonance constitute a new type of phenomena, in which the features of both optical and radio spectroscopy become manifest. The physical gist of these phenomena was considered in detail in a number of review articles published both in the domestic^[1, 2] and in the foreign^[3-5] literature. It consists in the fact that during the process of resonant scattering of light by atoms there can be obtained a new population distribution among the different energy sublevels of the atoms, differing greatly from thermal equilibrium. In particular, if such a redistribution is produced in the ground state, then one speaks of optical orientation of the atoms. On the other hand, if it is produced in the excited state and its change is caused by the radio-frequency field, then a phenomenon of this kind is customarily called double radio optical resonance.

The development of optical methods of registration of magnetic resonance in gases^[6-9] and in solids^[10, 11] in the ground and excited states^[12-14] of atomic systems has led to the need for investigating in detail all the aspects of the interaction of the atoms with the field of the optical and radio-frequency bands, making it possible to refine its nature and to uncover a whole series of new properties of this interaction. For example, it was predicted theoretically and confirmed experimentally that there exist real and virtual optical and radio frequency transitions^[8, 17], the "optical" shifts of energy levels were observed and investigated,^[18] effects of "intersection" and "anti-intersection" of levels were discovered.^[19, 20]

Many of these phenomena have immediately found practical applications, particularly in quantum magnetometry, having led to new ways of development of this practically important region of physics and technology.

Interference phenomena in an atomic system, being a direct consequence of the quantum-mechanical principle of superposition under the condition for existence of definite phase relations between the wave functions of the atomic state (coherent superposition of states), were thoroughly reviewed anew.^[8, 13, 15, 16, 34]

In essence, all the known types of resonant phenom-

ena (NMR, EPR, ENDOR, radio-acoustic resonance, etc.), are connected in one way or another with the creation of a coherent superposition of states in an atomic ensemble. With the aid of optical methods of radio spectroscopy it is possible to obtain the most complete information on the character of atomic transitions, not only because of their high sensitivity, but also because of the features that are introduced by the very character of the interaction with the optical photons.

The investigation of the coherent has been the subject of an exceedingly large number of theoretical and experimental papers, the mere listing of which is in itself a laborious task. The present review therefore has only one purpose, to attempt to consider from unified points of view the main effects in which the principal role is assigned to interference of atomic states, and which presently are widely used for spectroscopic research.

The bibliography at the end of the paper does not claim to be complete. It contains references only to a few papers which as a rule are easily accessible. It lists also papers significantly supplementing the problems touched upon here.

II. SUPERPOSITION PRINCIPLE AND INTERFERENCE OF ATOMIC STATES

The effects considered in the present paper are a manifestation of one of the fundamental laws of quantum mechanics—the principle of superposition of states. It is therefore advantageous to dwell briefly on some of its consequences as applied to the group of questions considered here.

In accordance with the superposition principle, the state $|\psi(t)\rangle$ of an atom at an arbitrary instant of time can be represented in the form of an expansion in terms of a complete set of eigenvectors $|\varphi_m\rangle$ of a certain Hermitian operator \hat{f}

$$|\psi(t)\rangle = \sum_m a_m(t) |\varphi_m\rangle. \quad (1)$$

The probability amplitude $a_m(t)$, being in the general case a complex number, is characterized by a modulus and by a phase. Depending on the experimental conditions, it is possible not only to measure the weights $|a_m|^2$ of the individual basis states $|\varphi_m\rangle$, but also to investigate the phase relations with which these states enter in the superposition $|\psi(t)\rangle$.

The mean value of a physical quantity defined by an operator \hat{g} in a state $|\psi(t)\rangle$ is given by

$$\bar{g} = \sum_m |a_m(t)|^2 g_{mm} + \sum_{\substack{m, n \\ m \neq n}} a_m(t) a_n^*(t) g_{mn}, \quad (2)$$

where $g_{mn} = \langle \varphi_n | \hat{g} | \varphi_m \rangle$ is the matrix element of the operator \hat{g} . If the operator \hat{g} commutes with \hat{f} , then the mean value \bar{g} is determined only by the first term

of (2), and the information concerning the phase relations of the expansion coefficients drops out completely. On the other hand, if the operators \hat{g} and \hat{f} do not commute, then the expression for \bar{g} contains besides the first term also an interference term, the magnitude of which is determined by the phase difference between the states $|\varphi_m\rangle$ and $|\varphi_n\rangle$, a difference that depends in the general case on the time. Choosing as the basis the eigenvectors of the energy operator, we can write the phase difference in the form $(E_m - E_n)t/\hbar$. Then the mean value of \bar{g} is a periodic function of the time, varying with the frequency of the atomic transition between the interfering states.

Since the measurement of a physical quantity is carried out on an ensemble of particles, expression (2) must be averaged over the statistical ensemble consisting of N independent systems, and the result must be identified with the macroscopically observable quantity

$$\langle \bar{g} \rangle = \frac{1}{N} \sum_{i=1}^N \bar{g}_i = \text{Sp}(\rho \hat{g}), \quad (3)$$

where the index i numbers the atoms and ρ is the density matrix of the ensemble of atoms. In accordance with the general definition, its elements are given by

$$\rho_{mn}(t) = \frac{1}{N} \sum_{i=1}^N \rho_{mn}^{(i)}(t) = \frac{1}{N} \sum_{i=1}^N a_m^{(i)}(t) a_n^{(i)*}(t). \quad (4)$$

The appearance of the interference term in expression (3) depends on the result of the averaging of the nondiagonal elements of the density matrix over the statistical ensemble. Inasmuch as the matrix elements $\rho_{mn}^{(i)}$ are complex numbers whose phases φ_i are determined by the phase differences of the coefficients $a_m^{(i)}$ and $a_n^{(i)}$, representing the corresponding states in the expansion (1), two physically real cases can be encountered upon averaging. The ensemble of atoms is prepared in such a way that all the phases φ_i are equally probable. For sufficiently large N , this leads to vanishing of the resultant matrix elements ρ_{mn} , and accordingly to the interference term in (3).

If the phase distribution is not isotropic, then the resultant matrix elements ρ_{mn} differ from zero, and the observable quantity $\langle \bar{g} \rangle$ experiences beats at a frequency determined by the energy difference between the interfering states. In this case it is said that coherence exists between the states $|\varphi_m\rangle$ and $|\varphi_n\rangle$; this coherence leads to a macroscopically observable interference effect.

Thus, for an experimental observation of the interference of atomic states it is necessary to prepare the atomic system in such a way that the phase difference between the interfering states is the same for all the atoms of the ensemble.

Depending on the energy interval between the levels m and n , the set of quantities ρ_{mn} characterizes the radio-frequency or the hyperfine coherence. In the former case the sublevels $|\varphi_m\rangle$ and $|\varphi_n\rangle$, belong to one level of the hyperfine structure, and the frequency $(E_m - E_n)/\hbar$ lies in the radio band; in the latter case the sublevels $|\varphi_m\rangle$ and $|\varphi_n\rangle$ belong to different hyperfine states and the frequency $(E_m - E_n)/\hbar$ lies in the microwave band. In principle it is also possible for the level $|\varphi_m\rangle$ to belong to the ground state of the atom and

for the level $|\varphi_n\rangle$ to the excited state. The quantities ρ_{mn} are then measures of the optical coherence. They can be different from zero only if the atoms are excited by optically-coherent (laser) radiation.

In the present paper we shall discuss radio-frequency coherence in a system of sublevels of either the ground or the excited state of an atomic ensemble.

There are many methods of producing radio-frequency coherence in a system of atomic states, and most of them are connected with the use of an external perturbation. The most commonly used is a radio-frequency field whose frequency is close to the frequency of Zeeman splitting of the atoms in an external constant magnetic field H_0 (magnetic resonance). The interaction of the atoms with the radio-frequency field under resonance conditions leads to induced oscillations of the atomic states with the phase of this field. The interference of the states becomes manifest in this case in the occurrence of a component of macroscopic magnetization transverse to H_0 ; this component varies with the frequency of the radio-frequency field.

The described method of producing coherence is not the only one. Thus, for example, a coherent superposition of states can be obtained by exciting an atomic system with a perturbation that depends periodically on the time and has a frequency equal to the frequency difference between the interfering states. Such a perturbation can be produced by resonant optical radiation^[14, 21, 22] or by an intensity-modulated electron beam.^[23, 24] The perturbations should have characteristics such as to be able to excite atoms in the superposition state.

For optical excitation, this is possible if it is represented by a mixture of components with different polarizations (σ^\pm and π). Polarization of light with such a composition is customarily used coherent, and the excitation of atoms by such light is sometimes called coherent excitation.^[8] When atoms are excited by radiation with σ^+ , σ^- , or π polarization, they go over into pure states. Such excitation will henceforth be called incoherent.

III. DETECTION OF RADIO-FREQUENCY COHERENCE

We consider in this paper methods of detecting radio-frequency coherence, based only on the use of the phenomena of absorption and emission of light by atoms. We stipulate from the outset that these methods are not the only ones. Recently, coherence was also registered by using phenomena of birefringence of nonresonant light in an optically oriented medium,^[75-77] due to the anomalous dispersion near the resonant-absorption line. However, the extensive use of dispersion methods is hindered by the complexity, and in some cases by the unavailability of nonresonant-light sources whose emission line is at the same time sufficiently close to the absorption line of the investigated atoms. The fact that the intensity and polarization of the light absorbed or emitted by an ensemble of atoms depend on the angular momenta of the atoms in the states coupled by the optical transition determines the possibility of using optical emission for the registration of the interference effects.

The presently developed theory of interaction of resonant optical radiation with atoms makes it possible to obtain a quantitative estimate the coherent phenomena.

If $\rho_{\mu\mu'}$ is the density matrix of the atoms in the ground state and is situated in a constant magnetic field H_0 , then the intensity of the light absorbed by the atoms is described by the expression^[8]

$$S_A = \frac{1}{T_p} \sum_{\mu\mu'} F_{\mu\mu'} \rho_{\mu\mu'}(t), \quad (5)$$

where

$$F_{\mu\mu'} = \sum_m \langle J_e m | e_{q_0} D | J_g \mu' \rangle \langle J_e m | e_{q_0} D | J_g \mu \rangle^* \quad (6)$$

is the light-absorption matrix and depends on its propagation direction and polarization, defined by a unit vector e_{q_0} ; J_g and J_e are the angular momenta of the atoms in the ground and excited states, D is the electric dipole moment, T_p is the lifetime of the atom in the ground state, determined by the intensity of the optical excitation, and $|J_g \mu\rangle$ and $|J_e m\rangle$ are the vectors of the operators \hat{J}_{gz} and $\hat{J}_{ez}(H_0 \parallel Oz)$.

According to (5), from the character of absorption of light of definite polarization it is possible to obtain information both concerning the populations of the magnetic sublevels and the phase relations of the corresponding states. Indeed, in the case of incoherent polarization of the exciting light (for example, σ^+), the excitation matrix $F_{\mu\mu'}$ is diagonal, and the magnitude of the absorbed light depends only on the distribution of the populations of the magnetic sublevels or, in other words, on the longitudinal, $\langle J_z \rangle$, component of the total angular momentum of the atoms in the ground state. In the case of excitation with light with coherent polarization, there appear in (5) terms proportional to the non-diagonal density matrix elements. Since $\rho_{\mu\mu'}(t)$ depends harmonically on the time with frequency $\omega_g(\mu - \mu')$, where $\omega_g = \gamma_g H_0$, the intensity of the absorbed light will be modulated with the same frequency, and these beats offer evidence of the presence of coherence in the super position of the states $|J_g \mu\rangle$.

The intensity of the fluorescence light emitted by the atoms in the spontaneous decay of the excited state is determined in terms of the density matrix $\rho_{mm'}(t)$ of this state:

$$S_F = \frac{3\Gamma}{8\pi} \sum_{mm'} G_{mm'} \rho_{m'm}(t), \quad (7)$$

where

$$G_{mm'} = \sum_{\mu} \langle J_e m | e_q D | J_g \mu \rangle \langle J_e m' | e_q D | J_g \mu \rangle^* \quad (8)$$

is the emission matrix, e_q is the unit polarization vector of the emitted photon, and Γ is the natural width of the excited state. The average intensity of the fluorescence light when $m = m'$ in expression (7) characterizes the change of the populations of the sublevels of the excited state. When $m \neq m'$, the fluorescence light experiences beats with frequency $\omega_e(m - m')$, if radio-frequency coherence exists in the excited state.

It must be borne in mind that the degree of orientation of the atoms is characterized at least by two quantities, polarization and alignment. In the case of field alignment, the numbers of the atoms in states differing only in the sign of the magnetic quantum number are equal, and there is no macroscopic angular momentum (polarization).

In experiments on optical orientation one deals both with polarized and aligned systems. The existence of coherence in polarized systems is uniquely connected with the presence of a summary transverse angular momentum of the atoms. Therefore the appearance of harmonic components in the light absorbed or re-emitted by the atomic system is associated with the transverse components of the magnetization of the ground and excited states. In subsequent sections, where we shall use an interpretation in terms of magnetization, we have in mind a polarized system.

IV. EQUATIONS OF MOTION FOR THE DENSITY MATRIX IN THE INTERACTION OF ATOMS WITH OPTICAL RADIATION

The quantum theory of interaction of radiation with an atomic system in a magnetic field H_0 leads to the following equation of motion for the density matrix of atoms in the ground state:

$$\begin{aligned} \frac{d\rho_{\mu\mu'}}{dt} = & -\frac{1}{2T_p} \sum_{\mu''} F_{\mu\mu''} \rho_{\mu''\mu'} - \frac{1}{2T_p} \sum_{\mu''} F_{\mu''\mu'} \rho_{\mu\mu''} \\ & + \frac{\Gamma}{T_p} \sum \frac{B_{\mu''\mu\mu'}^{\mu''\mu''}}{\Gamma + i[\omega_e(\mu - \mu') - \omega_g(\mu'' - \mu'')]} - i\omega_g(\mu - \mu') \rho_{\mu\mu'}, \end{aligned} \quad (9)$$

where

$$B_{\mu''\mu\mu'}^{\mu''\mu''} = \sum_{mm'} \langle J_e m | e_q D | J_g \mu'' \rangle \langle J_e m | e_q D | J_g \mu \rangle^* \times (J_g 1 \mu' q / J_e m') (J_g 1 \mu q / J_e m), \quad (10)$$

$(J_g 1 \mu' q / J_e m')$ is the Clebsch-Gordan coefficient, J_e , m , and ω_e are the angular momentum, the magnetic quantum number, and the frequency of the Zeeman splitting of the excited state. Equation (9) has been written out for resonant optical excitation under the assumption that there are no optical shifts of the magnetic sublevels, and describes the motion of the density matrix of the ground state with allowance for polarization and spontaneous emission of the light quanta.

The first two terms of the right side of (9) describe the loss of coherence and the change of the populations of the magnetic sublevels upon absorption of light. The third term is the contribution of the spontaneous transitions to the change of the density matrix of the ground state, and in the case when $\mu = \mu'$ and $\mu'' = \mu'''$ it describes the rate of population of the magnetic sublevels.

Inasmuch as the probabilities of the spontaneous transitions to different sublevels are not equal, the rate of population of certain sublevels may turn out under certain conditions to be larger than the rate of decrease of population as a result of absorption of the photons, this being the physical basis for the phenomenon of optical orientation of the atoms.

If $\mu \neq \mu'$, then the third term of expression (9) describes the restoration of the coherence in the ground state under the condition that $|\omega_e - \omega_g| \ll \Gamma$. In this case one can speak of partial conservation of the phase memory for atoms undergoing the transition from the ground state to the excited state and back to the ground state.

The change of the density matrix of the excited state $\rho_{mm'}$ can be described by the equation^[8]

$$\frac{d\rho_{mm'}}{dt} = [\Gamma + i(m - m')\omega_e] \rho_{mm'}$$

$$= \frac{1}{T_p} \sum_{\mu\mu'} \langle J_{em} | e_{\gamma 0} \mathbf{D} | J_{g\mu} \rangle \langle J_{em'} | e_{\gamma 0} \mathbf{D} | J_{g\mu'} \rangle^* \rho_{\mu\mu'}. \quad (11)$$

This equation connects the density matrix of the excited states with the density matrix of the ground state. Therefore, even if at the initial instant there is no coherence in the excited state, subsequently the coherence can be transferred from the ground to the excited state, this being connected with the conservation of the coherence in the process of the optical excitation.

V. MAGNETIC RESONANCE INDUCED BY A RADIO FREQUENCY FIELD

1. Double Radio Optical Resonance

In 1952, Brossel and Bitter first realized in practice the idea of registering magnetic resonance in the excited state of mercury atoms by determining the change of the polarization of the emitted resonant radiation.^[12] Vapor of even isotopes of mercury (zero nuclear spin) were excited in the state 6^3P_1 by the light of the 2537 Å line, polarized linearly along the direction of the external magnetic field H_0 (incoherent π excitation). So long as the transitions between the magnetic sublevels in the 6^3P_1 state were not excited, the fluorescence light did not contain the σ^\pm polarization components. A weak radio-frequency field $H_1(t)$ with frequency $\omega \sim \omega_e$, applied perpendicular to H_0 , induced transitions between the sublevels of this state, as a result of which circularly-polarized components of resonant radiation appeared in the fluorescence light.

Brossel and Bitter registered the average intensity of the scattered light with specified polarization; this intensity is connected with the populations of the magnetic sublevels of the excited state. However, as a result of the action of the field $H_1(t)$, not only do the populations of the sublevels change, but a radio-frequency coherence sets in between the Zeeman states, since the perturbation $H_1(t)$ common to all the atoms sets the same instantaneous phase difference of the states for the entire statistical ensembles. In accordance with (7), the intensity of the light scattered by the atomic system should in this case change harmonically in time at the frequency ω_e . This effect, called in the literature "light beats," was first observed experimentally in 1959.^[25] In^[25] there were registered harmonic components of resonant fluorescence of mercury atoms in spontaneous transitions $6^3P_1 \rightarrow 6^1S_0$. The modulation of the intensity of the light emitted at the angle $\pi/2$ to the propagation direction of the exciting beam was observed not only at the frequencies ω_e and $2\omega_e$, as was proposed on the basis of a qualitative analysis, but also at the frequencies $3\omega_e$ and $4\omega_e$, i.e., the real picture of this phenomenon turned out to be much more complicated.

The harmonic dependence of the intensity of the resonant fluorescence on the time follows directly from expression (7)^[26]

$$S_F = \frac{3\Gamma}{8\pi T_p} \sum_{\substack{mm' \\ nn'}} \frac{F_{nn'} \langle m/M \rangle \langle M/n \rangle \langle n'/M' \rangle \langle M'/m' \rangle G_{m'm}}{\Gamma - i[\omega(n-n') + \omega_{\text{eff}}(M'-M)]} \times \exp[-i\omega(m-m' + n'-n)t], \quad (12)$$

where $\omega_{\text{eff}} = \gamma H_{\text{eff}}$, and $H_{\text{eff}} = [(H_0 - \omega/\gamma)^2 + H_1^2]^{1/2}$ is the effective field in a coordinate system rotating

with frequency ω . To determine the magnetic sublevels in the field H_{eff} , the magnetic quantum numbers M and M' were used, whereas the indices n and m determine the magnetic sublevels of the excited state in the laboratory frame.

In the case of incoherent optical excitation ($n = n'$), the excitation matrix $F_{nn'}$ turns out to be diagonal and modulation of the light S_F becomes possible at the frequencies ω and 2ω . In coherent excitation in a superposition state ($n \neq n'$), the modulation frequency is equal to $(m - m' + n' - n)\omega$, and consequently beats can be observed at the frequencies $0, \omega, 2\omega, 3\omega$, and 4ω . From (12) follows also a resonant dependence of the depth of modulation of the light on the magnetic field near $H = \omega_e/\gamma$. An interesting feature of double radio optical resonance in general and of the method of "light beats" in particular is that the thermal motion of the atoms, which shifts the average frequency of the radiation of the atom as a result of the Doppler effect and is responsible for the broadening of the resonant fluorescence line, does not influence the line width of the double radio optical resonance, which under these conditions remains equal to the natural width Γ (if one neglects the radio-frequency broadening). Indeed, the Doppler effect shifts the frequencies of the optical transitions from different magnetic sublevels of the excited state of the atom to the ground state by equal amounts, and the frequency of the "beat" between these optical harmonics remains unchanged and equal to or a multiple of the frequency of the radio frequency field $H_1(t)$ but induces the transitions between the magnetic sublevels. Thus, it becomes possible to observe paramagnetic resonance in excited states at frequencies much smaller than the Doppler line width of the radiation, and to obtain for the structure of the excited states information that is not available by the usual methods of optical spectroscopy.

The "light beats" were investigated in detail theoretically^[26] and experimentally,^[15] and their use as a reliable method for the study of the structure of energy levels of atoms was discussed in^[27a] and was confirmed by the results of an investigation of the hyperfine structure of the $5^2P_{1/2}$ state of the K^{39} atom.^[27b]

The high sensitivity attainable in optical detection of the change of the populations or of the appearance of coherence in atomic states, has made double radio optical resonance one of the most important methods of radio spectroscopy. Thus, for example, it was used successfully for the investigation of the state $6^2P_{3/2}$ of Rb atoms (see^[28]). It was also used to determine the g factors of the excited states of the atoms Ca (see^[29]), Ba (see^[30]), and Cs (see^[31]), and also for the measurement of the quadrupole moments of certain states of Ba¹³⁵, Ba¹³⁷,^[32] and Sr.⁸⁷,^[33]

2. Magnetic Resonance of Atoms in the Ground State

In the case when the frequency ω of a radio-frequency field $H_1(t)$ rotating around H_0 is close to the frequency ω_g , coherence is produced in the system of sublevels of the ground state. If under the conditions of the double radio optical resonance the radio-frequency transitions in the excited state were accompanied by changes of the characteristics of the fluorescent light,

then the excitation of the magnetic resonance in the ground state, by virtue of the symmetry of the processes of the resonant absorption and spontaneous emission, should affect the characteristics of the light absorbed by the atoms. The existence of radio-frequency coherence in the ground state will become manifest in modulation S_A at a frequency equal to or a multiple of the frequency of the field H_1 .

In order for the interference of the states to become observable in this case, in accordance with the general principles, it is necessary to satisfy at least two conditions: 1) to produce a population difference between interfering states, i.e., to prepare the atomic system predominantly in one of the pure states, 2) to ensure an identical phase difference between the states for all the atoms of the ensemble. The first requirement is realized because of the phenomenon of optical orientation, and the second is realized when the oriented atomic system interacts with the radio-frequency field H_1 .

To observe the modulation of the absorption, it is possible to use a second light beam, having a polarization and orientation relative to the field H_0 such as to be able to cause transitions from any pair of interfering sublevels of the ground state to the common sublevel of the excited state (i.e., light with coherent polarization). In most experiments one chooses such a beam to be circularly polarized and directed perpendicular to the field H_0 , the intensity of which is much smaller than the intensity of the main beam producing the orientation in the system. After passing through the sample, its intensity is modulated and the depth of modulation is varied resonantly near $\omega = \omega_g$.

All these properties of the phenomenon are reflected in expression (5) for the intensity of the light absorbed by the atomic system per unit time. In coherent polarization of the detecting beam ($\mu \neq \mu'$) the expression for S_A contains the time-dependent terms

$$S_A = \frac{1}{T_p} \sum_{\mu\mu'} F_{\mu\mu'} \rho_{\mu'\mu} \exp[-i(\mu - \mu')\omega t], \quad (13)$$

where the periodic dependence on the time is separated in explicit form. In analogy with the beats of the fluorescent light under conditions of double radio optical resonance, the harmonic signal S_A can be called absorption beats.

For a system with angular momentum 1/2 (for example Hg^{199} , Cd^{111} , Cd^{113}), the equations of motion of the density matrix in the ground state are quite simple, and in a coordinate system rotating with frequency ω they have a form analogous to the Bloch equation:^[8]

$$\left. \begin{aligned} dM_z/dt &= (M'_0 - M_z)\tau_1^{-1} + \omega_1 v, \\ du/dt &= -u\tau_2^{-1} + \Delta\omega v, \\ dv/dt &= -\omega_1 M_z - \Delta\omega u - v\tau_2^{-1}, \end{aligned} \right\} \quad (14)$$

where the following notation is used:

$$\begin{aligned} M_z &= \rho_{1/2, 1/2} - \rho_{-1/2, -1/2}, & \Delta\omega &= \omega - \omega_g, \\ u &= \rho_{1/2, -1/2} + \rho_{-1/2, 1/2}, & \omega_1 &= \gamma_g H_1, \\ v &= i(\rho_{1/2, -1/2} - \rho_{-1/2, 1/2}), \end{aligned}$$

$M'_0 = M_0 \tau_1 / \eta$ is the maximum attainable value of the longitudinal component of the magnetization, and the effective relaxation times τ_1 and τ_2 are determined by the expression $1/\tau = (1/T) + (1/\eta)$, where T and η are the times of thermal and optical relaxation.

The corresponding results of the experiment^[8] in

Hg^{199} vapor are given in Fig. 1. The solid curves in the same figure represent the stationary solution of Eqs. (14) for $|M_z| = (u^2 + v^2)^{1/2}$, u , and v , demonstrating the practically complete agreement between theory and experiment. Figure 2 shows the transient process of free damping of coherence in the Hg^{199} system following sudden turning off of the field H_1 , as revealed by modulation of the absorption of a transverse light beam.

It must be emphasized that a signal of this type is perfectly analogous, in the character of the observed phenomenon, to a signal obtained in nuclear magnetic resonance by radio electric methods. This has made it possible to use for the investigation of optically oriented systems all the previously developed stationary and nonstationary methods of radio spectroscopy (slow passage,^[7, 8] adiabatically fast passage,^[35] pulse radio frequency excitation,^[8, 36] spin echo,^[37] etc.).

Naturally, for spins $J_g > 1/2$ the equations of motion of the density matrix and the corresponding expressions for S_A become much more complicated and cannot be interpreted in such a simple manner. The observed signal of absorption modulation reflects in this case the entire aggregate of the interference phenomena in a complicated superposition of states. Nonetheless, in certain particular cases the modulation of the absorption of the transverse beam is a manifestation of the character of the variation of the transverse components of the electronic or nuclear magnetization, averaged over the ensemble.^[38]

Just as in the case of double radio optical resonance, interaction with the field H_1 leads to a broadening of the magnetic-resonance lines, this being a feature inherent in the method of producing coherence by radio frequency fields perpendicular to H_0 .

3. Concepts of an Atom "Dressed" by a Radio-frequency Field

Quite recently, for a unified interpretation of a number of interference effects that take place in the interaction of atoms with radio-frequency field, Cohen-Tan-

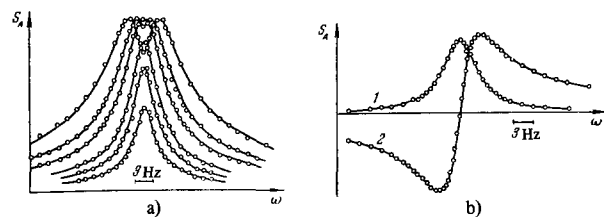


FIG. 1. a) Dependence of the amplitude of the modulation of the intensity of light on the frequency ω of the field $H_1(t)$ near the resonance of the nuclei Hg^{199} at different values of H_1 [8]. b) Analogous dependence for the components of the modulation in quadrature (1) and in phase (2) with the radio-frequency field. The signal amplitude S_A is given in relative units.



FIG. 2. Oscillogram of free damping of coherence in a system of Hg^{199} nuclei after turning off the radio frequency field [8].

noudji^[78, 79] proposed a new theory, based on the concept of an atom "dressed" by an electromagnetic field. This theory is in essence applicable for the solution of all problems in which one considers the interaction of an atom with two electromagnetic fields, one of which is sufficiently intense and the other weak. No limitations whatever are imposed on the nature of these fields, and they can be either of optical or radio frequency.*

In particular, when double radio optical resonance is considered, one field is optical and the second is a radio field. For concreteness, we denote by C_1 the strong radio-frequency field, and by C_2 the weak optical field. The interaction of the field C_2 with the combined "atom plus C_1 " system[†] is described from the point of view of quantum theory of radiation.

Let us consider for simplicity an atom that has in the excited state an angular momentum $J = 1/2$ and is located in a constant field H_0 and in a radio-frequency field $H_1(t)$ perpendicular to H_0 ; the radio frequency is close to $\omega_e = \gamma_e H_0$. Then the Hamiltonian of the combined system can be written in the form

$$\mathcal{H} = \omega_e J_z + \omega a^\dagger a + \lambda (a + a^\dagger) J_x \dots,$$

where the first term characterizes the Zeeman energy of the atom, the second the energy of the field C_1 (a^\dagger and a are the operators of creation and annihilation of the radio-frequency photon), and the third the binding energy of the atom with the field C_1 (λ is the coupling constant). If $\langle n \rangle$ and $\langle H_1 \rangle$ are the average number of the photons and the average amplitude of the field C_1 , then

$$\gamma \langle H_1 \rangle = 2\lambda \langle n \rangle^{1/2}.$$

The eigenstates of the operator $\mathcal{H}_0 = \omega_0 J_z + \omega a^\dagger a$ are states with energies $(\pm \omega_e/2) + n\omega$, where n is the number of photons of the radio-frequency field. A diagram of the levels corresponding to these states is shown by the dashed lines in Fig. 3. The binding energy $V = \lambda J_x (a + a^\dagger)$ is usually smaller than \mathcal{H}_0 , as a result of which the influence of the term V on the system is appreciable only at the points of intersection of the levels of the Hamiltonian \mathcal{H}_0 , i.e., at $\omega_e = p\omega$. The levels of the total Hamiltonian \mathcal{H} are shown by the solid lines in Fig. 3.

An analysis, which will not be presented here and which can be carried out with the aid of perturbation theory or by using Feynman diagrams, as was done in^[78], leads to the following conclusions:

If the optical excitation has an incoherent polarization (for example σ^-), then the absorption of the optical photon changes the system into pure state of the Hamiltonian \mathcal{H} , and near the point $\omega_0 = 3\omega$ there is a definite probability of the transition of the atom from the state $| -1/2 \rangle$ into the state $| +1/2 \rangle$, with a real absorption of three radio-frequency photons and with further emission of an optical σ^+ photon. This phenomenon, investigated earlier by Winter^[80] and called "multiquantum" transitions, can thus be interpreted as an anti-intersec-

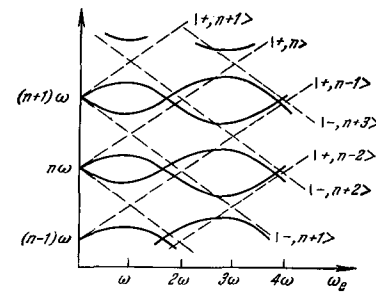


FIG. 3. Diagram of energy levels of the "atom plus radio frequency field" system. Ordinates—energy in relative units.

tion of the levels of the atoms "dressed" by the radio-frequency field.

We can interpret analogously the "even" resonances $\omega_0 + 2q\omega$ under conditions of "transverse pumping"^[74, 81], the only difference being that the transitions between the sublevels of the "atom plus field C_1 " system are virtual. In both cases the theory makes it possible to calculate quantitatively both the form of the resonance curves and their positions with allowance for the radiation shift. An interesting feature of this theory is the existence of different possibilities for the description of coherent effects both in the excited and in the ground state. If the field C_1 is applied to atoms in the ground state, then the reasoning remains unchanged, if we regard the optical field C_2 not as photons absorbed by the atoms, but as "holes" that are left optically excited in the ground state.

From the same point of view it is possible to describe completely the optical detection of parametric resonance when the field $H_1(t)$ and H_0 are parallel. We note only that in this case the energy diagram has a form different from that shown in Fig. 3, and contains only "intersections" of the levels.

If the radio frequency $H_1(t)$ is so strong that the interaction V cannot be regarded as weak, the analysis can be carried out in closed form only near $\omega_0 \sim 0$. Then the Zeeman term $\gamma H_0 J_z$ can be regarded as a weak excitation also with respect to the unperturbed Hamiltonian $\mathcal{H}'_0 = \omega a^\dagger a + \lambda J_x (a + a^\dagger)$. It is shown in^[82] that the magnetic moment of such a "dressed" atom can differ strongly from the magnetic moment of a free atom, and the dependence of their ratio on the amplitude of the radio-frequency field is described by a Bessel function $J_0(\omega_1/\omega)$. An experimental investigation in the ground state of Hg^{199} atoms has fully confirmed the theoretical calculations.

Finally, the concept of the "dressed" atom has made it possible to predict theoretically the possibility of transferring coherence in exchange collisions between atoms having strongly differing gyromagnetic ratios, provided the Lande factors of these atoms are altered by the additional radio-frequency field. At the present time, attempts are being made to confirm this effect experimentally.

VI. CONSERVATION OF RADIO FREQUENCY COHERENCE IN THE CYCLE OF OPTICAL ORIENTATION

The cycle of optical orientation of the atoms consists of three elementary stages: 1) absorption of a resonant photon and a transition of the atom to the excited state,

*This method is less general and in many cases less convenient in applications than the method of quasistationary states of Ya. B. Zel'dovich^[83].

†The combining of the atom and of the field C_1 into one system interacting with the field C_2 has served as a basis for the naming of this theory.

2) the evolution of the atomic system in the excited state during its lifetime, and 3) the spontaneous emission of the photon and the return of the atom to the ground state. Partial conservation of the radio-frequency coherence during the entire cycle is one of the most important features of interference phenomena in optically oriented systems. Under certain conditions, the phase relations established between the states of the different atoms with the aid of the external perturbation can be conserved in the processes of absorption and spontaneous emission of optical photons, as a result of which intensity beats having the frequency of the magnetic resonance in the ground state appear in the light of the resonant fluorescence.

The occurrence of these beats can be explained theoretically by using expression (7) for the fluorescence light and by using the equations of motion (11) of the density matrix $\rho_{mm'}$, from which it follows that the time evolution of the excited state is connected with the evolution of the ground state. In order to integrate (11), it is necessary to know the density-matrix elements $\rho_{\mu\mu'}$, which are written in the general form

$$\rho_{\mu\mu'} = \exp[-i(\mu - \mu')\omega_g t] \rho_{\mu\mu'}(t). \quad (15)$$

The dependence of $\rho_{\mu\mu'}'(t)$ on the time is exponential with a time constant T_p determined only by the intensity of the optical excitation. Remaining within the framework of the assumption of the smallness of the probability of the induced optical transitions compared with the probability of the spontaneous transitions, i.e., under the condition $T_p \gg 1/T$, we obtain

$$\rho_{mm'}(t) = \frac{1}{T_p} \sum_{\mu\mu'} \frac{\langle J_e^m | e_{q_0} D | J_g \mu \rangle \langle J_e^{m'} | e_{q_0} D | J_g \mu' \rangle^*}{\Gamma + i[(m - m')\omega_e - (\mu - \mu')\omega_g]} \rho_{\mu\mu'}(t). \quad (16)$$

It follows from (16) that the dependence of the density matrix of the excited state on the time contains harmonic components with frequencies $\omega_g(\mu - \mu')$ of the ground state. This result should not be surprising, in spite of the fact that the natural frequency of the evolution of the sublevels of the excited state differs from ω_g . For a qualitative explanation of the effect we can use the analogy with coupled oscillators having different damping constants T_p and $1/\Gamma$ and different eigenfrequencies. If the frequency ω_g of the natural oscillations of the first oscillator (ground state) lies within the bandwidth of the second oscillator (excited state) and is shifted from its central frequency ω_e by an interval smaller than Γ , then excitation of induced oscillations of the second oscillator at the frequency ω_g becomes possible. Since, however, this frequency is not an eigenfrequency of the excited state, a definite phase shift will occur between the motion of $\rho_{\mu\mu'}$ and $\rho_{mm'}$, and this shift should be a function of the difference between the frequencies ω_e and ω_g , i.e., it should depend on the magnetic field H_0 . Thus, if $\rho_{\mu\mu'} \neq 0$ (coherence takes place in the ground state), then $\rho_{mm'}$ experiences induced motion at the frequency of the ground state (transfer of coherence from the ground state to the excited state), and consequently, in accordance with (7), beats with frequency $\omega_g(\mu - \mu')$ can be observed in the fluorescence light.

Let us determine the conditions necessary for the observation of this phenomenon. Let $\mu \neq \mu'$ and $m = m'$,

i.e., let a coupling exist between the population ρ_{mm} of the excited state and the coherence $\rho_{\mu\mu'}$ of the ground state. It is seen from (7) and (8) that to this end it is necessary to produce optical excitation with light having coherent polarization e_{q_0} . Thus, the longitudinal angular momentum of the excited state turns out to be coupled with the transverse momentum of the ground state and oscillates with frequency $\omega_g(\mu - \mu')$. In order to observe these oscillations, it is necessary to register fluorescence light with incoherent polarization $e_g(\pi, \sigma^+, \text{ or } \sigma^-)$.

The presence of coherence $\rho_{\mu\mu'}$ in the ground state can also cause a stimulated change in the nondiagonal elements $\rho_{mm'}$ ($m \neq m'$) of the excited state. If we assume, to simplify the interpretation, that the polarization of the optical excitation is incoherent, then the matrix elements $\rho_{\mu\mu'}$ can no longer be connected with $\rho_{mm'}$, i.e., the longitudinal momentum of the excited state does not experience stimulated motion. However, as shown above, such a motion is experienced by the transverse components of the momentum, i.e., $\rho_{mm'}$. It can be observed if the polarization of the registered fluorescence light is coherent.

In both cases, the interference of the excited state, revealed by the beats of the fluorescence light at the frequency of the ground state, is induced. It is important only when the difference between the natural frequencies $\omega_e - \omega_g$ is not too large compared with Γ , as a result of which the amplitude and phase of the beats are functions of the external magnetic field H_0 .

Conservation of coherence in optical transitions between the ground and excited states was demonstrated by experiments in vapor of Hg^{199} , Ne^{23} and Rb (see [40]). Modulation of the fluorescence light was observed both in the case of its coherent polarization (coupling of $\rho_{\mu\mu'}$ with $\rho_{mm'}$) and in the case of incoherent polarization (coupling of $\rho_{\mu\mu'}$ with ρ_{mm}). The dependences of the beat amplitudes S_F (Fig. 4) and of their phases (Fig. 5) on the constant magnetic field at different polarizations of the exciting light (e_{q_0}) and

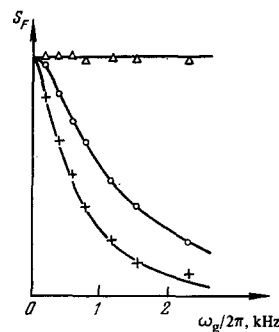


FIG. 4

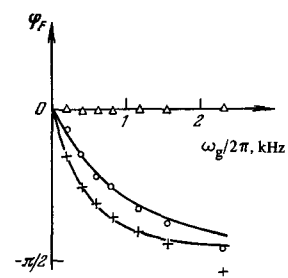


FIG. 5

FIG. 4. Dependence of the amplitude of modulation of fluorescence light of Hg^{199} at the frequency ω_g on the Zeeman splitting in the ground state [8]. Excitation by the hfs component $F = 1/2$: Δ —polarization e_{q_0} coherent, e_q incoherent; $+$ —polarization e_{q_0} incoherent, e_q coherent; excitation by hfs components $F = 3/2$. \circ —polarization e_{q_0} incoherent, e_q coherent.

FIG. 5. Dependence of the phase of the modulation of the fluorescence light of Hg^{199} at the frequency ω_g on the Zeeman splitting in the ground state [8]. The notation is the same as in Fig. 4.

of the fluorescence light (e_q) show splendid agreement between the experimental results and the theoretical curves shown by the solid lines. In Na^{23} vapor, using the beats of the fluorescence at the frequency of the magnetic resonance in the ground state, it was possible to resolve distinctly the Zeeman structure of the resonance line (Fig. 6),* and also to observe beats at double the frequency $2\omega_g$, which were predicted theoretically.

VII. TRANSFER OF RADIO-FREQUENCY COHERENCE IN SPIN EXCHANGE

The beats of fluorescence light at the frequency of the Zeeman transitions in the ground state are connected with the conservation of radio-frequency coherence within the limits of one atom in optical transitions between the ground and excited states. A very similar phenomenon can be observed also when different atoms interact, including when atoms of different elements collide, if these collisions are accompanied by spin exchange.

The spin-exchange phenomenon itself is well known.^[41, 42] The use of optical orientation of the atoms and nuclei has added greatly to the procedures of experimental investigation of spin exchange^[43, 44] and has led to the development of a detailed theory of this phenomenon.^[45] In the overwhelming majority of known experiments using spin exchange, interest was focused on the mechanism of transfer of only the longitudinal angular momentum from one system of atoms to the other, and the procedure for observing this phenomenon was accordingly based on measurement of the average intensity of the light passing through the system. It was tacitly assumed that the probability of the transfer of the transverse component of the angular momentum is negligibly small because of the large difference between the frequencies of the transitions in the interacting atomic systems.

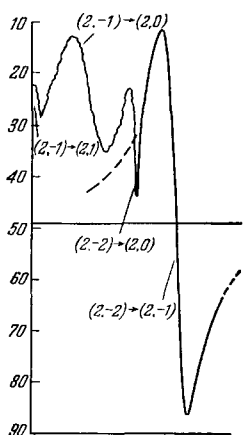


FIG. 6. Spectrum of the beats of the fluorescence light of Na^{23} atoms at the magnetic-resonance frequency in the ground state [39]. The plot was obtained after synchronous detection.

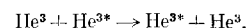
*The plots of signals of different coherent effects, which will be presented below, were obtained using different experimental methods. Since the description of these methods is beyond the scope of the article, the methods of obtaining the experimental curves will not be indicated, and examples of the signal plots will be given in order to demonstrate the existence of the effect and the possibility of its observation with the necessary reference to the original paper.

Further progress in the experimental technique has led to observation of modulation of 10 830-Å light passing through a cell filled with the isotope He^3 ,^[46] at the frequency of the nuclear resonance of He^3 in the ground state 1^1S_0 . On the other hand, the light was absorbed by atoms in the metastable 2^3S_1 state. Ruff and Carver^[47] described a similar experiment in the Na-H system, where the intensity of the resonant radiation of Na was modulated at the frequency of the magnetic resonance of the hydrogen atoms.

As already noted, the presence of beats in the light absorbed or re-emitted by an atomic system is evidence of the existence of a coherent superposition of atomic states of the absorbing or emitting system. However, if the characteristic beat frequency is equal to the frequency of another system that is under magnetic resonance conditions, then this is inevitably the consequence of the transfer of coherence between the systems in the process of the spin-exchange interaction.

The quantum theory of the phenomenon of conservation of radio frequency coherence in spin exchange in^[48] is based on the general theory of spin-exchange collision^[45] with allowance for the interaction between one of the colliding subsystems with the optical field of the resonant radiation. In spite of the fact that a theoretical analysis was carried out in^[48] for the particular case of the collision of He^3 atoms, its conclusion can be generalized to include other atomic systems.

It is assumed that the collision is strong (the cross section for spin exchange is $\sim 10^{-14}$ cm²) and can lead to the exchange of electron spins as well as of excitation energy:



Owing to the short duration of the collision compared with the reciprocal of the hyperfine-structure constant in the ground state ΔW and compared with the period of the precession in the external field H_0 , the nuclear spin of each atom retains its instantaneous orientation after the exchange process and re-establishes the coupling with the "new" electron that enters in the metastable atom. As a result, the precession motion at the nuclear frequency is transferred to the He^{3*} atoms, which leads to modulation of the light absorbed by them. Inasmuch as the precession frequencies of He^3 and He^{3*} differ noticeably, the magnitude of the effective conservation of the coherence should be determined by the relation between the difference of these two frequencies and the damping parameter.

To construct a theory of the transfer of coherence in the process of spin exchange, one uses the equations of motion for the density matrix $\rho_{\mu\mu'}$ of metastable atoms in the presence of an optical field of resonant radiation, similar to (9), but supplemented with allowance for the alteration of $\rho_{\mu\mu'}$ by exchange collisions. Without giving the details of the mathematical calculations, which can be found in the original paper,^[48] we shall stop to discuss only some of the results of the theoretical analysis.

Mathematically, the influence of the spin exchange on the density matrix $\rho_{\mu\mu'}(t)$ consists in the fact that after the collision the matrix is expressed in terms of the density matrix of the ground state with its inherent time dependence $\exp[(g - g')\omega_g t]$. Since the light absorbed

by the He^{3*} atom is proportional to $\rho_{\mu\mu'}(t)$, its intensity should vary harmonically with the frequency $\omega \approx \omega_g$ of the magnetic resonance in the ground state. In [48] they obtained the explicit form of the absorption signal S_A of the hyperfine component $F = 1/2$ of the resonant line of orthohelium

$$S_A \approx \frac{\text{const}}{\Gamma_\mu \Gamma_g} \left\{ \text{const} + \frac{\omega_1 \Delta\omega}{\Delta\omega^2 + \omega_1^2 + \Gamma_g^2} \times \cos \omega t + \frac{\Gamma_g \omega_1}{\Delta\omega^2 + \omega_1^2 + \Gamma_g^2} \sin \omega t \right\}, \quad (17)$$

where $\omega_1 = \gamma H_1$, $\Delta\omega = \gamma(H - H_0)$, and Γ_μ and Γ_g are the damping constants of the metastable and ground states. Expression (17) was obtained under the condition that for metastable atoms $\gamma_\mu H_0 > \Gamma_\mu$. If this condition is not satisfied, then in the limit when $\gamma_\mu H_0 > \Gamma_\mu$, the amplitude of the modulation at the frequency ω_g decreases by a factor $\gamma_\mu H_0 / \Gamma_\mu$.

A signal of this type was revealed [48] by the modulation of circularly polarized light with wavelength 10 830 Å (Fig. 7). The resonance curves had the characteristic form of absorption curves (a) and dispersion curves (b), coinciding with the theoretical form (17). The dependence of the effect on the ratio of the detuning $\Delta\omega$ to the width Γ_μ of the metastable was also confirmed by the experimental results. In spite of the fact that in the cited paper the ratio $\gamma_\mu H_0 / \Gamma_\mu$ was ~ 8 , the coherence-transfer signal in spin exchange was still observed, thus indicating that this process is an effective one.

VIII. INTERFERENCE EFFECTS ARISING WHEN THE CONSTANT FIELD IS MODULATED

The phase φ_i of the nondiagonal elements of the density matrix $\rho_{mn}^{(i)}$ of an individual atom, by definition, is

$$\varphi_i = \varphi_0 + \omega_{mn}(t - t_0). \quad (18)$$

In this expression the initial instant of time t_0 and the initial phase φ_0 are, generally speaking, random quantities, and if special measures are not taken to establish the correlation of these quantities for different atoms, they vanish when averaged over the ensemble. The phase equality over the ensemble can be established by fixing the instants of time t_0 at which the atom goes over into the superposition state; this was considered in detail in [49, 50]. Another possibility is to fix the phase φ_0 of the excitation. [51, 52] Finally, there is one more method of acting on the phase φ_i in order to make the phase of the elements $\rho_{mn}^{(i)}$ equal, namely to modulate the frequency ω_{mn} by an alternating mag-

netic field directed along the field H_0 under corresponding optical excitation. [53]

1. Parametric Resonance in the Excited State

A radio-frequency field oscillating in the direction of H_0 cannot by itself transfer the atom to the superposition state, since atomic transitions induced by such a field are forbidden. However, the action of the field, in conjunction with optical excitation, transfers the atoms into a coherent superposition of interfering states. In this process there is coherence over the ensemble, whereas the optical radiation excites atoms in the superposition state, as a result of which it should of necessity have a coherent admixture of σ^\pm and π polarizations.

For simplicity let us consider an atomic system having one sublevel in the ground state ($J_g = 0$) and three sublevels in the excited state ($J_e = 1$). If the width of the excitation spectrum is much larger than the Zeeman splitting of the excited state, then in the absence of an oscillating field the atoms are excited at all three sublevels $m = 0, \pm 1$ by independent harmonics, and the phases of the corresponding states remain arbitrary.*

If the field H_0 is modulated with frequency Ω , then a situation is possible wherein the magnetic sublevels are excited by one optical harmonic with participation of one or several radio-frequency quanta. [17, 54] Coherence over the ensemble of the atoms is introduced by the fact that the phase of the optical wave enters in the same manner when all sublevels are excited, and the phase difference of the states is determined only by the phase of the coherent radio-frequency field. If the field frequency is an integer multiple of the frequency of the transitions between the sublevels, then coherence is also introduced into the system, but following absorption of an appropriate number of radio-frequency quanta. It must be remembered, however, that the indicated radio frequency transitions, by virtue of the energy conservation law, should be completed without a change of the state of the radio-frequency field, at any rate within the time on the order of the lifetime of the atom in the excited state (virtual transition). This means that after a time $\Delta t \sim 1/(\Delta E)$ the virtually-absorbed radio-frequency quanta should be returned to the field. [54] When the atoms return to the ground state, the number of quanta released is such as to be able to replenish the virtually-absorbed energy, but this number need not necessarily be equal to the number of absorbed quanta. This mechanism leads to the appearance of beats in the fluorescence light not only at the modulation frequency but also at multiples of this frequency.

A simple theoretical analysis confirms these qualitative conclusions. We add to the right side of Eq. (11) for the density matrix of the excited state an expression describing the influence of the radio-frequency field H_1 :

$$-i[\omega_1 J_z \rho] \cos \Omega t, \quad \text{where } \omega_1 = \gamma_e H_1. \quad (19)$$

*Generally speaking, one cannot deny the existence of a correlation between the different harmonics of the optical radiation within the limits of the width of the spectrum of the exciting line. Recent studies of natural coherence in the emission of Xe confirms this fact [55]. But this correlation is so weak that its observation calls for a special experiment.

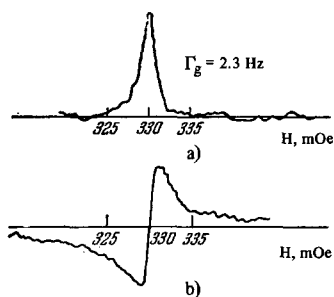


FIG. 7. Magnetic resonance signal of He^3 nuclei, registered by means of modulation of the light absorbed by the metastable He^{3*} atoms: a) in quadrature with the field H_1 and b) in phase with H_1 [48].

The equation for $\rho_{mm'}$ is then

$$\frac{d\rho_{mm'}}{dt} = -\Gamma\rho_{mm'} + NT_p^{-1}F_{mm'} - i[\mathcal{H}, \rho]_{mm'}, \quad (20)$$

where

$$\mathcal{H} = (\omega_e + \omega_1 \cos \Omega t) J_z.$$

Solving this equation and substituting the result in (7), we obtain^[53]

$$S_F = \frac{3\Gamma N}{8\pi T_p} \sum_{mm'} \sum_{nh} G_{mm'} F_{mm'} \frac{J_n(z) J_{n+h}(z)}{\Gamma + i[\omega_e(m-m') - n\Omega]} \times \exp(ik\Omega t), \quad (21)$$

where $J_n(z)$ is a Bessel function of first order of the argument $z = \omega_1(m-m')/\Omega$.

It is seen from expression (21) that the fluorescence light contains harmonics with frequencies $k\Omega$. Their amplitudes depend in resonant manner on the detuning, and are maximal at $\omega_e(m-m') - n\Omega = 0$.

At $m = m'$, the beats vanish, thus indicating that it is necessary to employ optical excitation with coherent polarization, for only in this case is the excitation matrix $F_{mm'}$ different from zero at $m \neq m'$. The denominator of expression (21) does not contain the amplitudes of the radio-frequency field (radio-frequency broadening), since there is no real absorption of radio-frequency photons.

The existence of parametric resonance was demonstrated experimentally in Cd vapor,^[53] where interference of the sublevels $m = \pm 1$ of the 5^3P_1 state was observed. The optical excitation was by means of linearly polarized resonant cadmium-lamp radiation propagating in the direction of the constant magnetic field. In that investigation, a study was made of the fluorescence-light component modulated at the modulation frequency of the magnetic field near $2\omega_e = \Omega$. The form of the resonance signal is shown in Fig. 8. The width of the parametric resonance was determined entirely by the lifetime of the atom in the excited state and was equal to $4.2 \times 10^6 \text{ sec}^{-1}$. No broadening of the resonant-signal line by the radio-frequency field was observed.

Resonances of higher orders, shown in Fig. 9, were also investigated. The position of the maxima of the beats agrees well with the theoretical predictions. However, the amplitude of the second resonance is somewhat overestimated compared with the calculated value, this being due to the contribution made to the observed signal by the double radio optical resonance due to inexact parallelism of the fields H_0 and $H_1(t)$.

Analogous investigations were carried out in^[17] on Hg^{199} atoms. An investigation was made of the dependence of the amplitude of signals of five harmonics of the fluorescence light on the depth of modulation of the

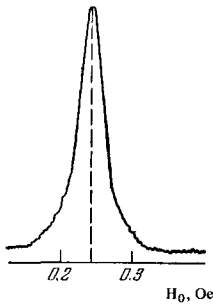


FIG. 8. Plot of the signal of the fluorescence beats of Cd atoms near resonance in the excited $3P_1$ state at resonant modulation of the magnetic field^[53].

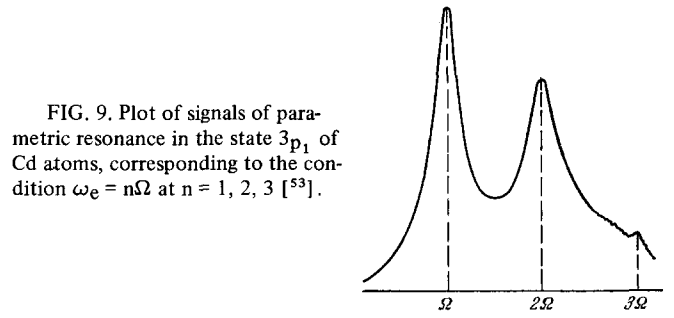


FIG. 9. Plot of signals of parametric resonance in the state $3P_1$ of Cd atoms, corresponding to the condition $\omega_e = n\Omega$ at $n = 1, 2, 3$ ^[53].

constant magnetic field. The experimental results are in good agreement with the theoretical predictions.

2. Parametric Resonance in the Ground State

It is possible to explain in similar fashion the occurrence of coherent superpositions of atomic states in modulation of the Zeeman splitting of the excited atoms. If the optical emission has coherent polarization, then, in the absence of a radio-frequency field oscillating in the direction of H_0 , the excitation is effected by its uncorrelated components. In the presence of $H_1(t)$, the atom can be excited by one optical harmonic with simultaneous virtual absorption of n radio-frequency quanta. The phase relations established in this manner between the magnetic sublevels, owing to the circulation of the coherence, do not vanish completely when the atom goes over into the excited state if $\Gamma \gg \omega_g$. After the end of the lifetime the atoms return to the ground state, introducing in it the coherence produced by the excitation.

If the modulation of the fluorescence light is a consequence of the uncertainty of the spontaneous-emission channel, then the uncertainty of the excitation channel leads in this case to modulation of the absorption. An essential factor in the phenomenon of parametric resonance in the ground state is the need for satisfying the conditions for the conservation of coherence in optical transitions.

The theoretical analysis can be based on Eq. (9) for the density matrix of the ground state:

$$\frac{d\rho_{\mu\mu'}}{dt} = -i[\mathcal{H}, \rho]_{\mu\mu'} + \frac{1}{T_p} \sum_{\mu'', \mu'''} B_{\mu''\mu'''}^{\mu\mu'} \rho_{\mu''\mu'''} - \frac{1}{T_p} F_{\mu\mu} \rho_{\mu\mu} + \beta \delta_{\mu\mu'} - \frac{1}{T} \rho_{\mu\mu'}, \quad (22)$$

where

$$\mathcal{H} = (\omega_g + \omega_1 \cos \Omega t) J_z.$$

We have omitted here terms connected with the attenuation of the coherence as a result of optical excitation; $1/T$ is the relaxation time, defined in the standard manner,^[56] and β is the rate of regeneration of the sublevels, determined by the mechanism of the thermal relaxation. Equation (22) is an equation with variable coefficients, and can be solved by successive approximations in terms of the perturbation. Confining ourselves to the first-order correction in the optical perturbation, we obtain

$$\rho_{\mu\mu'}(t) = \frac{\beta T}{T_p} \sum_{\mu'', \mu'''} \sum_{n, h} \frac{J_{n+h}(z) J_n(z) B_{\mu''\mu'''}^{\mu\mu'}}{T-1 + i[\omega_g(\mu-\mu') - n\Omega]} \exp(ik\Omega t). \quad (23)$$

This expression confirms our statement that the coherence in the ground state is produced only if the coherence is conserved in the optical transitions, i.e., at non-

zero coefficients $B_{\mu\mu'}^{\mu\mu'}$. In principle, it is impossible to detect this coherence by observing the change of the intensity of the light producing the population difference of the magnetic sublevels of the system, for in this case $\mu = \mu'$ and the time-dependent terms vanish. It is necessary to use for this purpose an additional beam with coherent polarization. Then

$$S_A = \frac{\beta T}{T_p T_p} \sum_{\mu\mu', \mu''\mu'''} \sum_{n, k} \frac{J_{n+k}(z) J_n(z)}{T^{-1} + i\{\omega_g(\mu - \mu') - n\Omega\}} B_{\mu''\mu'''}^{\mu\mu'} F_{\mu\mu'}^\alpha \exp(ik\Omega t), \quad (24)$$

where the index α pertains to the additional beam. Thus, the absorption signal S_A for this beam contains harmonics with frequencies $k\Omega$ and exhibits properties analogous to the properties of parametric resonance in the excited state.

Parametric resonance in the ground state was investigated in detail experimentally on cesium vapor^[57] and mercury vapor,^[74] and perfectly satisfactory agreement was obtained between the experimental and theoretical results.

IX. PRODUCTION OF RADIO-FREQUENCY COHERENCE BY INTENSITY-MODULATED OPTICAL EXCITATION

It is known that in radiation whose intensity is modulated at a frequency Ω , the harmonics of the fundamental frequency ω' and of the sideband frequencies $\omega' \pm \Omega$ are correlated. Thus, the excitation of the atomic system by modulated light can establish definite phase relations between different sublevels of the excited state.

A theoretical description of this phenomenon can also be based on the equations of motion (11). Since orientation of the atoms in the ground state is not obligatory in this case, it is convenient to assume all the populations $\rho_{\mu\mu'}$ to be identical and equal to $N/(2J_g + 1)$. The only quantity that depends on the excitation intensity is $1/T_p$. We shall regard it as a function of the time, varying at the modulation frequency Ω . Then

$$\frac{d\rho_{mm'}}{dt} = -\Gamma\rho_{mm'} - i\{\omega_{\alpha}\rho\}_{mm'} + \frac{N}{2J_g + 1} \frac{F_{mm'}}{T_p}, \quad (25)$$

where $1/T_p = K_0(1 + \epsilon \cos \Omega t)$, and ϵ is the index of light modulation. The solution of this equation is

$$\rho_{mm'} = \frac{NK_0}{2J_g + 1} F_{mm'} \left\{ \frac{1}{\Gamma + i\omega_e(m - m')} + \frac{\epsilon}{2} \frac{\exp(i\Omega t)}{\Gamma + i\{\omega_e(m - m') + \Omega\}} + \frac{\epsilon}{2} \frac{\exp(-i\Omega t)}{\Gamma + i\{\omega_e(m - m') - \Omega\}} \right\} \quad (26)$$

Substituting (26) in (7), we note that the intensity of the fluorescence light

$$S_F = \frac{3}{8\pi} \frac{\Gamma NK_0}{2J_g + 1} \sum_{mm'} G_{mm'} F_{mm'} \left\{ \frac{1}{\Gamma + i\omega_e(m - m')} + \frac{\epsilon}{2} \frac{\exp(i\Omega t)}{\Gamma + i\{\omega_e(m - m') + \Omega\}} + \frac{\epsilon}{2} \frac{\exp(-i\Omega t)}{\Gamma + i\{\omega_e(m - m') - \Omega\}} \right\} \quad (27)$$

is subject to beats with frequency Ω ; the amplitude of the signal is a resonant function of the detuning $\omega_e - \Omega$. Interference phenomena of this type will be observed in the fluorescence light only under the condition $m \neq m'$, i.e., in coherent excitation and detection of the optical signal.

The first experimental observation of the beats at the frequency of magnetic resonance in the excited state, using modulated optical excitation, was realized

in cadmium vapor.^[14] As a result of the optical transitions $5^1S_0 \rightarrow 5^3P_1$, induced by linearly polarized light propagating in the direction of a constant magnetic field, the sublevels $m = \pm 1$ were excited, and their interference was registered by means of the beats of the light scattered at 90° . Figure 10 shows a plot of the resonant signal in the case when the modulation frequency Ω of the light intensity is equal to the frequency of the transitions between the sublevels with $m = \pm 1$. In accord with (27), the depth of modulation of the fluorescence lines changes in resonant fashion as a function of the magnetic field intensity. In this case, just as under conditions of parametric resonance, the width of the resonance line does not depend on the modulation index of the exciting light and is equal to the natural width of the excited state.

Modulated optical excitation can produce radio-frequency coherence also in the ground state, owing to the conservation of coherence in the cycle of the optical orientation. If the modulation frequency Ω is close to the frequency of the Zeeman splitting in the ground state and the atoms retain the phase memory during the lifetime in the excited state, then upon their return to the ground state they will bring with them coherence, which can be observed, for example, by modulating the absorption of an auxiliary light beam.^[58]

A theoretical analysis of this phenomenon on the basis of the equations of motion for the density matrix in the ground states leads to the following expression for $\rho_{\mu\mu'}$ in first approximation in the optical perturbation:^[59]

$$\rho_{\mu\mu'} = \frac{K_0\beta}{\Gamma_0} \sum_{\mu''} B_{\mu''\mu'''}^{\mu\mu'} \left\{ \frac{1}{\Gamma_0 + i\omega_g(\mu - \mu')} + \frac{\epsilon}{2} \frac{\exp(i\Omega t)}{\Gamma_0 + i\{\omega_g(\mu - \mu') + \Omega\}} + \frac{\epsilon}{2} \frac{\exp(-i\Omega t)}{\Gamma_0 + i\{\omega_g(\mu - \mu') - \Omega\}} \right\} \quad (28)$$

In accordance with (5), the intensity of the auxiliary light beam with coherent polarization will be modulated at a frequency Ω .

The possible occurrence of the described effect was first demonstrated experimentally in^[21] in vapor of Cs, Rb, and He. An interesting result of this method of producing coherence is the possibility of stimulating forbidden transitions between Zeeman sublevels, since this method is not connected with direct transitions between the sublevels of the ground state, viz., the transition from one sublevel to another occurs via an excited state and is accompanied by establishment of correlation or relations between them. It should also be noted that in experiments of this type the line width of the magnetic resonance in the ground state is determined only by processes of thermal and optical relaxation.

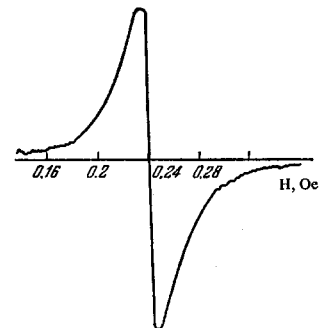


FIG. 10. Dependence of the amplitude of the beat of the fluorescence light of Cd atoms on the constant field H in the case of intensity-modulated optical excitation^[14].

X. INTERFERENCE OF STATES ARISING UPON INTERSECTION AND ANTI-INTERSECTION OF ENERGY LEVELS

In the preceding sections we have dealt exclusively with interference of the nondegenerate atomic states. Nonetheless, interference phenomena can take place also in superposition of degenerate states.

Interference of degenerate states was first observed in experiments on magnetic depolarization of resonant radiation,^[60] known as the Hanle effect.^[61] The qualitative picture consists in the following. When an atomic system is excited by linearly polarized resonant radiation in the absence of an external magnetic field, the fluorescence light has the same polarization as the exciting light. If a magnetic field H_0 perpendicular to the polarization vector of the exciting light is superimposed on the system of atoms, then depolarization of the fluorescence light is observed in the direction of the field H_0 . This depolarization depends on the value of H_0 . This example is only a particular case of a large class of phenomena, called intersection and anti-intersection of the energy levels, and observed as a rule in strong magnetic fields.

The intersection effect was first revealed in He by the resonant increase of the intensity of the scattered light of the 10 830 Å line ($2^3P_2 \rightarrow 2^3S_1$) at the points of intersection of the fine-structure levels 3P_1 and 3P_2 , as noted by the circles in Fig. 11. The physical picture in the intersection of the levels is perfectly analogous to the Hanle effect. When the distance between levels becomes smaller than their width, their coherent excitation by one optical harmonic becomes possible. The probability of spontaneous emission of the proton is determined now already by the wave functions of both sub-levels, and this leads to a change in the intensity of the scattered light.

To describe the effective intersection one can use the interference-phenomena theory described in the preceding sections. Turning to expression (27) and assuming that there is no modulation of the excitation ($\epsilon = 0$), we obtain Breit's well known result^[62]

$$S_p = \frac{3\Gamma}{8\pi T_p} \frac{N}{2J_g + 1} \sum_{mm'} \frac{F_{mm'} G_{m'm}}{\Gamma - i(\omega_m - \omega_{m'})}, \quad (29)$$

describing the change of the average intensity of the fluorescence light with polarization \mathbf{e}_q . Here ω_m is the energy of the m -th level. This expression describes

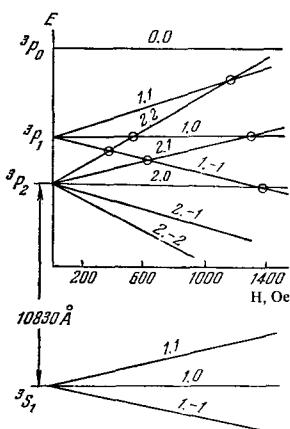


FIG. 11. Diagram of energy levels of helium atoms. The intersection points of the fine-structure levels are marked by circles.

both the effective intersection of the levels in strong fields ($\omega_m = \omega_{m'} \neq 0$), and the Hanle effect ($\omega_m = \omega_{m'} = 0$).

The use of expression (29) for concrete particular cases^[63] has shown that it not only explains satisfactorily the general character of the observed phenomena, but makes it possible to calculate the line shape of the level-intersection signal, which in the general case is a superposition of the Lorentz and dispersion curves with width Γ . A characteristic feature of the intersection signal (Fig. 12) is the practically complete absence of sources of line broadening, as a result of which the experimentally observed line width coincides with the natural width.^[34] The only possible sources of broadening are inelastic collisions and the reorientation of the atoms during the lifetime in the excited state. However, these interactions are usually of low probability, since the density of the atoms in the sample is quite small ($\sim 10^{10} - 10^{11}$ atoms/cm³).

In an investigation^[65] of the fine splitting of the 2P state of the Li atoms by the level-intersection method, it was observed that the form of the observed signal differs noticeably from the theoretical one. A detailed analysis has shown that the effect of anti-intersections of the levels was observed, for the first time, in this experiment.

Anti-intersection takes place when the distance between two energy levels is of the order of their natural width. However, if they are coupled by some interaction (for example, hyperfine interaction), then the levels may not intersect at all. A most complete treatment of the theory of the anti-intersection effect is found in^[64-66]. We therefore present here only some of the most general arguments essential for a qualitative interpretation of the phenomenon.

Let $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{V}$ be the Hamiltonian of the system, where \hat{V} is the operator of the perturbation that couples the states $|1\rangle$ and $|2\rangle$. Then the eigenvalues of $\hat{\mathcal{H}}$ are

$$E_{1,2} = \{(\mathcal{H}_{11} + \mathcal{H}_{22}) \pm [(\mathcal{H}_{11} - \mathcal{H}_{22})^2 + 4\mathcal{H}_{12}^2]^{1/2}\} / 2, \quad (30)$$

where $\mathcal{H}_{ik} = \langle i | \hat{\mathcal{H}}_0 | k \rangle \delta_{ik} + \langle i | \hat{V} | k \rangle$. It is seen from this equation that if $\langle i | \hat{V} | k \rangle \neq 0$, then the energy difference ($E_1 - E_2$) always differs from zero, i.e., the levels cannot intersect. The states corresponding to the energies E_1 and E_2 are determined by the expressions

$$\begin{aligned} |\psi_1\rangle &= \cos(\beta/2) |1\rangle + \sin(\beta/2) |2\rangle, \\ |\psi_2\rangle &= -\sin(\beta/2) |1\rangle + \cos(\beta/2) |2\rangle, \end{aligned} \quad (31)$$

where $\tan \beta = 2H_{12}/(H_{11} - H_{22})$, and $|1\rangle$ and $|2\rangle$ are the eigenvectors of the unperturbed Hamiltonian $\hat{\mathcal{H}}_0$. Figure 13 shows a diagram of anti-intersecting energy levels satisfying Eq. (30). In the vicinity of the anti-intersection point, the states of the $|\psi_1\rangle$ and $|\psi_2\rangle$ at-

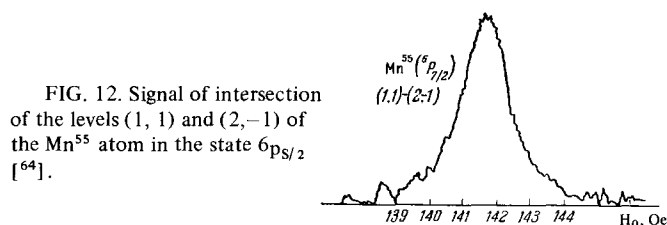


FIG. 12. Signal of intersection of the levels (1, 1) and (2, -1) of the Mn^{55} atom in the state $6P_{5/2}$ ^[64].

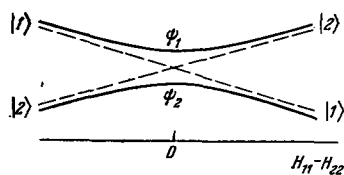


FIG. 13. Position of the energy levels of a perturbed system as a function of the splitting of the levels of the unperturbed system.

oms become superpositions of the pure states $|1\rangle$ and $|2\rangle$, a fact accompanied by a change of the characteristics of the scattered light registered in the experiment.

Even incoherent excitation of the pure states $|1\rangle$ and $|2\rangle$ leads to the occurrence of the superposition states $|\psi_1\rangle$ and $|\psi_2\rangle$ in the anti-intersection region. This indeed is the main reason why anti-intersection signals can be observed for such levels whose coherent excitation is forbidden by the selection rules and for which the intersection method cannot be used in principle.

The anti-intersection signal was observed experimentally in a number of investigations.^[20, 65, 67] An important difference between it and the intersection signal is the dependence of the line width on the perturbation energy V_{12} . This is precisely the circumstance that makes it possible to use the method of anti-intersection to determine the perturbation energy from the known width Γ of the transition.

In the original experiment of Dupont-Roc, et al.,^[68] they succeeded in observing the occurrence of a coherent superposition of the states of Hg^{199} in a zero magnetic field upon lifting of the degeneracy of the Zeeman sublevels of the ground state of the mercury atom by means of an additional optical nonresonant excitation (Fig. 14). The authors not only demonstrated the equivalence of the beam of nonresonant circularly polarized light to a certain magnetic field H_S that lifts the degeneracy in the system of sublevels $|\pm 1/2\rangle$, but also measured experimentally the energy of the sublevels of the ground state at $H_0 \rightarrow 0$. Figure 15 shows a diagram of the sublevels of Hg^{199} with allowance for the energy of the optical interaction. When the nonresonant light beam and the field H_0 are parallel, intersection of the sublevels $\mu = \pm 1/2$ takes place when $H_0 = -H_S$. On the other hand, if the field H_0 is perpendicular to the direction of this beam, then the coherent excitation of both sublevels couples them and makes the eigenvalues of the states $|\pm 1/2\rangle$ equal to $\pm \gamma(H_0^2 + H_S^2)^{1/2}$, i.e., the levels are anti-intersecting in this case.

The foregoing features of the anti-intersection effect make it a very valuable method of investigating states that cannot be studied by other methods, and makes it possible to determine important atomic constants. Thus, in^[67] there was investigated the spectrum of the anti-intersection signal of Yb^{171} and Yb^{173} in an atomic beam,

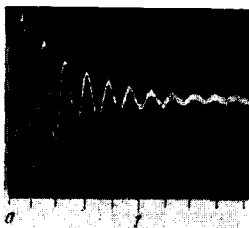


FIG. 14. Signal of precession of Hg^{199} nuclei in the fictitious magnetic field produced by a nonresonant light beam^[67]. The oscilloscope sweep scale is in seconds.

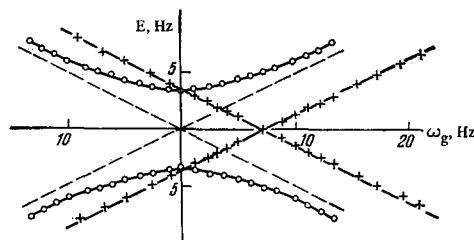


FIG. 15. Diagram of energy levels of the ground state of Hg^{199} following the lifting of the degeneracy by an optical perturbation. Experimental data of^[67]: +—perturbing beam parallel to the field H_0 , O—perturbing beam perpendicular to H_0 .

and the constant of the hyperfine was determined. In addition, in that investigation they observed for the first time the "coherent" anti-intersection signal, which differs from the usual one in the fact that the excitation of the states and the registration of the fluorescence light were coherent.

XI. COHERENT PHENOMENA IN EFFECTIVE FIELD

We have considered above various methods of producing and observing radio-frequency coherence in a system of states determined by the Zeeman splitting in a magnetic field H_0 . The creation of coherence over the ensemble presupposes the presence of a corresponding perturbation common to all the atoms. This perturbation, however, not only gives rise to transitions between the interfering sublevels, but in the general case changes also the energy state of the atomic system. Radio-frequency coherence can be produced also in this new aggregate of perturbed states if the second perturbing action is suitably chosen.

Such a situation is encountered in the interaction between atoms and a radio-frequency field $H_1(t)$ that rotates around H_0 . In a coordinate rotating at a frequency ω , the atoms are acted upon by a constant effective field $H_{\text{eff}} = [H_0 - \omega\gamma^{-1}]^2 + H_1^2]^{1/2}$, the direction of which makes an angle $\varphi = \sin^{-1}(H_1/H_{\text{eff}})$ with the field H_0 . In this coordinate system, the atomic moments precess around the direction of the effective field, and the precession phases of the individual moments are not correlated, as in the case of precession around the constant field H_0 in the absence of a radio frequency field $H_1(t)$.

It is possible to phase-in the precession of the moments in the rotating coordinate system, or in other words to produce coherence in the states determined by the effective field H_{eff} , for example, with the aid of a second radio-frequency field having a frequency $\Omega = \gamma H_{\text{eff}}$. This additional action on the atomic ensemble can be ensured by amplitude modulation of the radio frequency field $H_1(t) = H_1(1 + \epsilon \cos \Omega t) \cos \omega t$ at a frequency Ω , or else by superimposing on the field H_0 a parallel modulating field $H_2 \cos \Omega t$.

In either case, the atomic system is acted upon in the rotating coordinate frame by an alternating field with frequency Ω . This field has two components, $H_{\perp}(t)$ perpendicular to the field H_{eff} and inducing magnetic resonance of the ordinary type in the system of sublevels determined by the field H_{eff} , and b) $H_{\parallel}(t)$ parallel

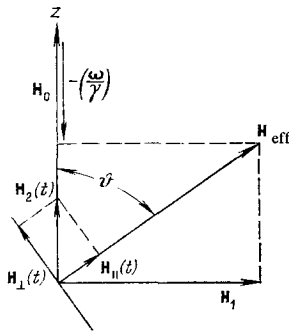


FIG. 16. Arrangement of fields in the rotating coordinate system.

to the field \mathbf{H}_{eff} , inducing parametric resonance (the arrangement of the fields in the rotating coordinate system is shown in Fig. 16).

The influence of the component $H_{\perp}(t)$ was considered in a number of investigations^[69-71] and consists in the occurrence of "sideband" resonance lines, whose distance from the fundamental line is a multiple of the frequency Ω . The characteristics of these additional lines are in the main analogous to the characteristics of magnetic resonance of ordinary type in the laboratory coordinate system, and the interaction with the field $H_{\perp}(t)$ leads to a broadening of the "sideband" line of the resonance.

The second component $H_{\parallel}(t)$ of the field acting in the rotating coordinating system, in the sense of its action on the atomic system, produces a situation similar to parametric resonance in the laboratory frame, which was considered above. This component also introduces coherence in the states determined by the effective field, but it does not lead to saturation of the resonance line.

In the general case, the system of atoms is acted upon by both components, and the observed signal is a superposition of the signals of the ordinary resonance and of the parametric resonance, their relative contributions being determined completely by the angle φ between the fields \mathbf{H}_0 and \mathbf{H}_{eff} .

A theoretical analysis of the coherent phenomena in the effective field^[72, 73] can be based on Eqs. (14), which under certain (usually realizable) experimental conditions are suitable for the description of the angular-momentum components averaged over the ensemble.^[38] The solution of Eqs. (14) under the assumption $T_1 = T_2 = T$ near the k -th resonance in the effective field ($\omega_{\text{eff}} + k\Omega = 0$) induced by the field $H_2 \cos \Omega t$ is given by

$$M_z^{(k)} = M_0 \cos^2 \vartheta \frac{T^{-2} + (k\Omega + \omega_{\text{eff}})^2}{T^{-2} + (k\Omega)^2 \text{tg}^2 \vartheta J_k^2(z) + (k\Omega + \omega_{\text{eff}})^2} + M_0 \sin^2 \vartheta \sum_{n=-\infty}^{\infty} (A_n^{(k)} \cos n\Omega t + B_n^{(k)} \sin n\Omega t), \quad (32)$$

where

$$A_n^{(k)} = \frac{J_k(z) J_{k+n}(z)}{T^{-2} + (k\Omega + \omega_{\text{eff}})^2} \left[\frac{1}{T^2} + k\Omega (k\Omega + \omega_{\text{eff}}) \times \frac{T^{-2} + (k\Omega + \omega_{\text{eff}})^2}{T^{-2} + (k\Omega)^2 \text{tg}^2 \vartheta J_k^2(z) + (k\Omega + \omega_{\text{eff}})^2} \right], \quad (33)$$

$$B_n^{(k)} = \frac{J_k(z) J_{k+n}(z)}{T^{-2} + (k\Omega + \omega_{\text{eff}})^2} \left[k\Omega + \omega_{\text{eff}} - k\Omega \frac{T^{-2} + (k\Omega + \omega_{\text{eff}})^2}{T^{-2} + (k\Omega)^2 \text{tg}^2 \vartheta J_k^2(z) + (k\Omega + \omega_{\text{eff}})^2} \right]; \quad (34)$$

here $J_k(z)$ is a Bessel function of first order of the ar-

gument $z = \gamma H_2 \cos \varphi / \Omega$, the solution being obtained under the condition $\gamma H_2 \sin \varphi / \Omega < 1$, which is usually satisfied in the experiment.

As follows from (32), the signal proportional to the longitudinal component of the magnetization $M_z^{(k)}$ contains a dc component as well as harmonic components whose amplitudes depend in resonant fashion on the frequency Ω of the field $H_2(t)$. Resonance takes place in the effective field if $\Omega = \omega_{\text{eff}} / |k|$, where $k = -1, -2, \dots$, at arbitrary values of the angle φ . The term $(k\Omega)^2 \text{tg}^2 \vartheta J_k^2(z)$ in the denominator of (32) describes the broadening of the resonance line due to the influence of the field components $H_{\perp}(t)$. This broadening becomes negligibly small as $\varphi \rightarrow 0$, i.e., under conditions when parametric resonance is predominantly excited.

These conclusions were experimentally confirmed in^[72]. The change of intensity of circularly polarized resonant radiation passing through an ensemble of Cs^{133} atoms in the direction of \mathbf{H}_0 , was registered. As seen from Fig. 17, a resonance signal in the effective field was obtained at the frequencies $\Omega = \omega_{\text{eff}} / |k|$ at $k = -1, -2, -3$, and -4 .

The main result obtained in^[72] was the experimental proof of the existence of parametric resonance in the effective field. The measurements of the dependence of the signal on the field amplitude H_2 at different angles φ , performed for this purpose, have demonstrated good agreement with theory. Figure 18a shows this dependence for the angle $\varphi = 45^\circ$, when the contribution of the resonance of ordinary types predominate, while Fig. 18b shows the same dependence obtained at $\varphi \sim 0.5^\circ$. In the former case the saturation of the resonance sets in already at $z < 0.01$, whereas in the latter saturation is not reached in practice at values of the field amplitude H_2 corresponding to $z > 1$. Thus, in the latter case the main contribution to the signal is made by parametric resonance in the effective field.

Finally, it can be noted that the coherence in the system of states determined by the effective field \mathbf{H}_{eff} can be produced also by intensity-modulated optical excitation,^[59] in which case the observed effect is similar to that described in Ch. IX.

XII. USE OF COHERENT PHENOMENA FOR THE INVESTIGATION OF ATOMIC STATES

All the effects considered in the present article and connected with the creation of a coherent superposition of states are extensively used as methods for the study of the structure and the lifetimes of atomic states.

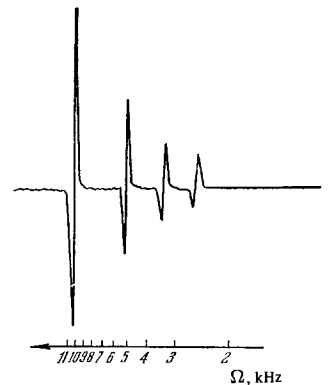


FIG. 17. Magnetic-resonance signal of Cs^{133} in the effective field \mathbf{H}_{eff} ^[73].

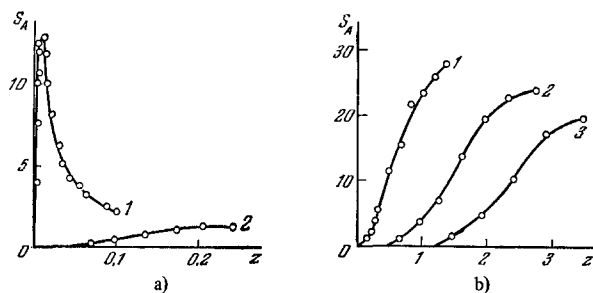


FIG. 18. Cs^{133} resonance signal in a field H_{eff} vs. Z in the following cases [73]: a) $\vartheta = 45^\circ$, curve 1—resonance at $\Omega = \omega_{\text{eff}}$, 2—resonance at $\Omega = \omega_{\text{eff}}/2$; b) $\vartheta' \sim 0.5^\circ$, 1—resonance at $\Omega = \omega_{\text{eff}}$, 2— $\Omega = \omega_{\text{eff}}/2$, 3— $\Omega = \omega_{\text{eff}}/3$.

These methods can be used for the investigation of the Zeeman and Stark effects, for the determination of the hyperfine-structure constants and electric quadrupole moments, for a detailed investigation of interatomic collision processes, and also for a direct study of the mechanisms of interaction between the electromagnetic field and matter.

Excitation of magnetic resonance by a radio-frequency field is at present the most widely used method of radio spectroscopic research. Its characteristic feature is that the radio-frequency field, by inducing real transitions between the investigated sublevels, introduces into the system an additional perturbation that changes the effective lifetime of the atoms on the magnetic sublevels. This leads to a broadening of the magnetic-resonance line and to a distortion of its shape, a distortion undesirable for atomic-spectroscopy purposes. From this point of view, interference between the atomic states, not accompanied by real transitions between the sublevels of the system, can be used as a much more promising research method. The possibility of registering interference of states by means of the beats of the intensity of the light absorbed or re-emitted by the atoms greatly increases its sensitivity. An important advantage of the "beat" method is the fact that it can be used for an investigation of sublevels between which radiative transitions are forbidden. The most interesting and highly promising in this respect is the method of "zero beats" under conditions of intersection and anti-intersection of the levels, which is already used extensively for the study of excited states that are not accessible to the ordinary methods of double radio optical resonance.

It should be noted that interference effects are of definite interest from the applied point of view. Thus, for example, a conservation of coherence at the resonance frequency He^3 in the ground state under exchange collisions with metastable He^3 atoms has made it possible to produce highly sensitive magnetometers, which are in essence spin generators operating at the precession frequency of the helium nuclei.

Magnetic resonance of alkali-metal atoms in the ground state is the basis of the operating principle of the majority of modern quantum magnetometers operating in a very wide range of magnetic fields.

Narrow parametric-resonance lines, with widths practically independent of the amplitude of the radio-frequency field, can be used for highly accurate stabili-

zation of magnetic fields. Parametric resonance in an effective field is a very exact method of measuring the amplitude of a radio-frequency magnetic field.

The possibility of actively controlling the coherent properties of emission from atomic ensembles is of great scientific and practical interest.

An investigation of the interference phenomena in atomic systems not only greatly extends the limits of our knowledge on the mechanism of interaction of the atoms with the radiation field, but has also laid the ground work for new methods in fundamental and applied physics.

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