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OPTICAL MANIFESTATIONS OF THE INTERFERENCE OF NONDEGENERATE ATOMIC STATES

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The research on optical phenomena connected with interference of nondegenerate atomic states is reviewed. The main types of phenomena are subdivided into four groups, called pulsed beats, beat resonance, parametric resonance, and phase resonance. Common to all these phenomena is the appearance of modulations in the spontaneous emission of the atoms or in the absorption coefficient of the atomic system. Certain nonlinear manifestations of interference of states are also considered. The possibility of using interference phenomena as methods for the investigation of subtle details of the energy structures of atoms and molecules is considered.

CONTENTS

1. INTRODUCTION

IN connection with the remarkable progress made in the interpretation of atomic spectra, the concepts of energy levels and their populations have become so firmly entrenched in atomic physics and spectroscopy that they became gradually independent concepts, losing the meaning attributed to them by quantum mechanics. Yet the statement commonly made in spectroscopy, that the atom is at a given (excited) level, is incorrect in the overwhelming majority of cases.

In order for this to be so, i.e., for the state vector of an atom to be directed along one of the basis vectors, the atom must be subjected to a strictly measured regular action of sufficient power, something that could be realized until recently only in radiospectroscopy (pulsed coherent inversion). In all ordinary cases in optics, the excitation raises the atom to a state of indefinite energy, with a vector that is not parallel to any of the basis direction (although it can be orthogonal to some or even to the majority of them). It is known that such a state Ψ can always be mathematically represented in the form of a superposition of states $|k\rangle$ with definite energy E_k, taken with definite weights (the superposition principle):

$$\Psi = \sum_k C_k |k\rangle e^{-iE_k/\hbar t}. \quad (1)$$

The mean squared moduli of the complex coefficients C_k have the meaning of level populations. The populations describe fully the distribution of the intensities of the spectral lines, and this causes the phases of the coefficients C_k to be forgotten, and at the same time it is forgotten that an atom described by relation (1) prior to the measuring operation did not have a definite energy and in a certain sense was situated simultaneously at all levels.

The habit of thinking in terms of the population concept in problems involving spectroscopy has resulted in the fact that it was not until the 60's of this century that an entire set of new optical phenomena was discovered, the theoretical basis for which was developed back in the 20's. We have in mind here the interference of atomic states. Until recently, there were in optics practically no known measuring procedures with which to distinguish a system of atoms in the mixed state (1) from a suitably chosen system of atoms with different but definite energies. Nonetheless, a system of atoms in the state (1) has distinct properties that can be observed.

It is easily seen that measurement of quantities whose operators do not commute with the energy lead in the case of mixed states (1) to a periodic time dependence of the measured quantities. This is well known in magnetic-resonance techniques, where one measured the transverse components of the angular momentum.

There exists also another possibility, more convenient for optical methods, of revealing mixed states, namely observation of the time dependence of the intensities of the transitions from the state (1) to some new state, under the action of some disturbance. Indeed, the probability P(t) of a transition from a state (1) to a certain eigenstate $|0\rangle$ under the influence of a perturbation $\hat{V}$, proportional to $|\langle 0|\hat{V}|\Psi\rangle|^2$, takes the form

$$P(t) \sim \sum_{k,k'} C_k C_{k'}^* \langle 0|\hat{V}|k\rangle \langle 0|\hat{V}|k'\rangle^* e^{-i(E_k - E_{k'})t/\hbar}. \quad (2)$$

As seen from (2), the transition probability has a complicated time dependence that can be expanded into

harmonics with frequencies corresponding to the distances between the sublevels contained in the superposition (1). If one deals with the radiative decay of the state (1), then these beats have a very lucid interpretation[1a], namely, spontaneous emission of the mixed state (1) occurs already in the elementary act with emission of an entire set of optical harmonics corresponding to transitions from the eigenstates making up the superposition into the common state. The interference of these harmonics leads to intensity beats. This concept of the intensity beats in the elementary radiative process turned out to be fruitful and made it possible to predict new effects correctly.

The history of research on optical phenomena in interference of states is quite inconsistent. The first observations of interference of degenerating states ("beats" with zero frequency) were made back in the 20's in Hanle's work on magnetic depolarization of luminescence, and were correctly explained (references can be found in the review[2]). However, this work was so little pursued that in 1959, when level crossing[3-5], in essence a variant of the Hanle effect, was observed, this phenomenon seemed puzzling at first[3]. This second birth of the Hanle effect led to a rapid development of research in this direction. The first observations of the beats proper—interferences of nondegenerate states—were started at that time. (It is interesting to note that in spite of being quite common in principle, these researches were carried out quite independently of the first ones, although they were unified by a common wave of interest in this field of atomic physics, stimulated by the remarkable work of a French group of physicists headed by A. Kastler and J. Brossel, who developed the technique of double resonance and optical orientation.) On the basis of the theory of magnetic resonances, Dehmelt[6] proposed an experiment, which was immediately realized in[7,8], with optically oriented alkali-metal vapor, in which magnetic resonance in the ground state was detected by the modulation of the resonant-radiation absorption. A decisive shift in the research on the interference of nondegenerate states, however, began with[8], where modulation of spontaneous emission was observed upon induction of magnetic resonance in the excited state. In this study, the phenomenon was treated directly in terms of interference of the states. Under the influence of work on level crossing[3,4] and on double resonance[8], several workers proposed independently and approximately at the same time to perform special experiments on the interference of nondegenerate states, not connected directly with any coherent perturbation (unlike the first experiments[8]). Indications to this effect can be found in the papers of Series[1a] (1959), Podgoretskiĭ[9] (1960), Dodd and Series[10] (1961) and Kastler[11] (1961). The new interference phenomena as applied to optics and nuclear physics were discussed and predicted most completely in unified form in[9]. All the hypotheses advanced were subsequently realized. A number of other experiments were also performed in this field, so that an entire group of new optical phenomena involving interference of nondegenerate states was discovered.

It is precisely to these phenomena, in which the properties of nonstationary quantum states are most clearly pronounced, that this review is devoted. It is related in its scope to an earlier review by Podgoretskiĭ and Khrustalev[12], who considered different prospective variants of interference experiments in atomic and nuclear physics, and to a recent review by Novikov, Pokazan'ev, and Skrotskiĭ[13], devoted mainly to coherent phenomena in radio-optical experiments. We point out also the review papers by Series[1b] and by Hanle and Pepperl[2].

The present review is limited to optical phenomena. This limitation, dictated by the scope, is in essence quite arbitrary, since interference of states is a common property of quantum systems and can be observed in any frequency band. In particular, the first manifestation of interference of nondegenerate noninteracting states must apparently considered to be the free spin precession observed in radio spectroscopy. A similar phenomenon was observed in nuclear physics[12] in 1955 with the aid of the gamma-quantum angular correlation technique. We note that the appearance and development of ideas of interference of states occurred in optics, practically independently of analogous work in nuclear physics (incidentally, a reverse influence did exist). The first paper in which it was indicated that all the state-interference phenomena are common and in which a connection was established between the already realized experiments in nuclear physics and the planned experiments in physical optics was that of Podgoretskiĭ[9].

The bibliography of the present review was compiled to provide maximum coverage of the experimental papers in the optical region. Only selected theoretical papers are cited.

2. QUALITATIVE DISCUSSION OF THE CONDITIONS FOR REALIZING COHERENT INTERFERENCE OF ATOMIC STATES AND PRINCIPLES OF ITS THEORETICAL DESCRIPTION

It is convenient to consider the conditions for the occurrence of interference phenomena for a simplest three-level system (Fig. 1a). For concreteness we assume that there is a single lower state $|0\rangle$, connected by an optical transition with two states $|1\rangle$ and $|2\rangle$ of close energy, so that $\omega_{01} \approx \omega_{02} \gg \omega_{12}$ (see Fig. 1a). Such a model corresponds exactly to the conditions in most experiments and can be directly generalized to include any more complicated case. Assume that as a result of some pulsed process of excitation at the instant of time $t = t_0$ the wave function describing the excited atom is given by a mixture of the states $|1\rangle$ and $|2\rangle$:

$$\Psi(t, t_0) = C_1(t, t_0)\, e^{-i\omega_{01}(t-t_0)}|1\rangle + C_2(t, t_0)\, e^{-i\omega_{02}(t-t_0)}|2\rangle. \quad (3)$$

The spatial distribution of the density of the electron cloud of such an atom, which is proportional to $|\Psi|^2$, oscillates in time with frequency $\omega_{12} = \omega_{01} - \omega_{02}$, this being a manifestation of the nonstationary character of the state (3). It is obvious therefore that any projection of the dipole-moment vector of the transition from the state Ψ to the state $|0\rangle$, and with it also the probability $P_\lambda(t, t_0)$ of emission of a proton with definite polarization λ will experience similar oscillations. In the simplest case, when the atom at the instant $t = t_0$ is subjected to a pulsed excitation, after which it experiences no perturbations whatever, the amplitudes $C_1(t, t_0)$

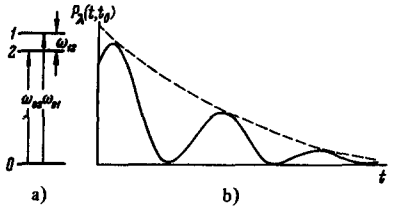

FIG. 1. a) Three-level scheme; b) decay kinetics of superposition state.

and $C_2(t, t_0)$ vary in time only as a result of radiative damping. If we assume that both sublevels 1 and 2 have the same radiative width Γ, then we can write

$$\Psi(t, t_0) = e^{-\Gamma(t-t_0)}[C_1 e^{-i\omega_{01}(t-t_0)}|1\rangle + C_2 e^{-i\omega_{02}(t-t_0)}|2\rangle],$$

so that

$$P_\lambda(t, t_0) \sim |\langle 0|\hat{d}\lambda|\Psi\rangle|^2 \tag{4}$$
$$= e^{-\Gamma(t-t_0)}[|C_1|^2|\lambda_{01}|^2 + |C_2|^2|\lambda_{02}|^2 + 2\,\mathrm{Re}\,C_1C_2^*\lambda_{01}\lambda_{20}e^{-i\omega_{12}(t-t_0)}]$$
$$= e^{-\Gamma(t-t_0)}\{A + B\cos[\omega_{12}(t-t_0)+\varphi_0]\},$$

where $\hat{\mathbf{d}}$ is the dipole-moment operator, $\lambda_{01} = \langle 0|\hat{\mathbf{d}}\lambda|1\rangle$, and $\lambda_{20} = \langle 2|\hat{\mathbf{d}}\lambda|0\rangle$. In a turbulent particular case we have A = B, and then the plot of $P_\lambda(t, t_0)$ has the form as shown in Fig. 1b. Expression (4) describes the time distribution of the probability of the elementary radiative process. It is possible to separate in it two components. The component with factor A depends only on the populations of the levels 1 and 2. The second component with factor B describes the beats connected with the interference of the states 1 and 2. If no special measures are taken, then the oscillations with the random phase distribution will vanish from the radiation of an ensemble of atoms as a result of the averaging. Obviously, in order for collective oscillations of the intensity to be produced, it is necessary to ensure in the radiation of the ensemble of the atoms a definite phase synchronism between the terms of the elementary beats.

Strict phase synchronism of elementary oscillatory processes corresponds to the equality (apart from a term $2\pi n$, where n is an integer) of the running phases $\chi = \omega_{12}(t - t_0) + \varphi_0$ of the elementary beats of the different atoms.

The initial phase of the beats $\varphi_0 = \arg C_1C_2^* + \arg\lambda_{01}\lambda_{20}$ depends on the parameters of the initial state and on the character of the polarization of the registered radiation. In all the hitherto realized cases, the interfering states had different angular-momentum projections. For such states, the averaging, in the course of observation, over the types of polarization leads to a vanishing of the beats. In other words, the radiation intensity integrated over space does not experience any oscillations, that is, such beats reduce to a periodic spatial redistribution of a constant total intensity. A model of such a radiator is a rotating dipole. In exactly the same manner, the beats vanish in the case of isotropic excitation of states with different angular momentum projections. These conclusions are proved with the aid of the Eckart-Wigner theorem with account taken of the orthogonality properties of the coefficients of the vector addition of the angular momentum [14a]. The physical meaning of these conclusions reduces to the statement that the total intensity of the emitted light does not depend on the orientation of the radiating system in space.

No beats corresponding to modulation of the integral intensity of spontaneous emission were observed in the case of isotropic excitation and observation, although they are possible in principle. Such beats assured results from the interference of states with identical projection of the angular momentum or states with indefinite value of the projection of the angular momentum. In particular, the integral modulation of the intensity could be observed in experiments on the deexcitation of the 2S metastable state of hydrogen in an electric field, when two close interfering states of hydrogen are produced, each of which is a mixture of 2S and 2P states [14b].

At a fixed phase φ_0, there remain two other factors that permit freedom of the phase χ, namely variations of the beat frequency ω_{12} and of the instant of excitation t_0. The frequency ω_{12} is under these conditions a parameter of the atom, and is constant for a system of light atoms. It should be noted that this frequency, under the condition $\omega_{01} \approx \omega_{02} \gg \omega_{12}$, is practically not subject to a Doppler spread, unlike the optical frequencies ω_{01} and ω_{02}, since both these frequencies shift in the same direction when the atom moves by an approximately the same amount, so that their difference remains constant[1)].

At fixed ω_{12} and φ_0, which is a characteristic condition for experiments on resonant fluorescence, the possible occurrence of interference of states is determined by the character of the distribution of the instants t_0 of excitation of different atoms. There is one particular case in which the interference is not averaged for any distribution of the instants t_0, namely the case $\omega_{12} = 0$, the case of beats with zero frequency. Depending on the phase φ_0, the interference increases or decreases of the intensity of the radiation registered in a given direction in the vicinity of the level degeneracy. An interference effect of this type is called level crossing. Let us return, however, to interference of nondegenerate states. The most obvious method of organizing collective beats is pulsed excitation—excitation of the entire ensemble of atoms at a fixed instant of time $t = t_0$. The pulse duration Δt should satisfy the obvious relation $\Delta t \ll \omega_{12}^{-1}$. Under these conditions, the phenomenon has the most lucid form, namely, after the brief excitation the intensity of the spontaneous emission decreases and exhibits damped oscillations.

Instead of pulsed excitation, collective beats can be produced by using harmonic modulation of the excitation intensity. The advantage of such a method is obvious from the following analogy. An atom whose emission kinetics is described by expression (4) is analogous to an oscillating system with natural frequency ω_{12} and damping Γ. It is known that in the case of a periodic driving force, a resonance is produced when the frequency of the action approaches the natural frequency of the system. A fully defined phase of the induced oscillations is established in this case, so that an arbitrary set of such independent identical systems will oscillate in phase. Thus, one can expect the luminescence of an ensemble of atoms to be modulated in amplitude

[1)] More accurately, the Doppler spread of the frequency ω_{12} exists, but it is smaller by a factor ω_{12}/ω_0 than the spread of the optical frequencies, and is therefore negligible.

when the modulation frequency of the excitation intensity approaches the frequency of the free beats.

A distinction should be made between the radiation modulation produced under resonance conditions and the trivial luminescence modulation connected with the oscillations of the populations of the radiating states in the case of intermittent excitation. This trivial modulation decreases with increasing frequency Ω of the interruptions of the excitation, owing to the inertia of the spontaneous emission, and vanishes almost completely if $\Omega \gg \Gamma$. To the contrary, the interference resonance of the beats should arise with equal intensity regardless of the lifetime of the system.

Both considered methods of organizing beats are based on introduction of synchronization into the process of excitation of different atoms. This does not exhaust all the possibilities of realizing collective interference. Phasing of the elementary beats can be attained with a uniform distribution of the instant t_0 of excitation, by acting on the initial phase φ_0 or on the frequency ω_{12} of the elementary oscillatory process. The first method is particularly obvious if one specifies a linear variation of the initial phase with time, $\varphi_0 = \Omega t_0$. Then the instant of excitation vanishes from the expression for the running phase of the oscillation χ at $\Omega = \omega_{12}$, and the phase becomes the same for all atoms. A linear variation of the phase with time is difficult to realize for purely methodological reasons, and one uses harmonic modulation of the phase, which also leads to the occurrence of partial phasing of the elementary beats.

The synchronizing action of the modulation of the frequency ω_{12} is less lucid, but it is clear that since the phase χ at a given instant t depends on the variation of the frequency in the past, $\chi = \int_{t_0}^{t} [\omega_{12}(t') + \varphi_0]dt'$, by properly acting on the instantaneous frequency ω_{12} it is possible to "pull in" the phase of the beats of the atoms excited at different instants t_0 to a certain common value.

These qualitative considerations served as a basis for a quantitative analysis that confirmed these predictions. We note that the foregoing methods of ensuring phasing of the beats coincide with the main known types of modulation of harmonic oscillations—amplitude, frequency, phase, and pulse modulation. This circumstance allows us to think that these methods exhaust the possibilities of organizing collective interference (there remain, of course, all possible combined actions; see, for example,[15a,16b]).

The employed qualitative interpretation of the formation of collective beats as a superposition of elementary excitations can be readily made quantitative[17]. For this purpose, it suffices to assume that the number of atoms excited per unit time is large. Then, replacing the summation of the elementary intensities by integration, we have for the summary intensity the expression

$$I(t) = k \int_{-\infty}^{t} P_\lambda(t, t_0)\, \mu(t_0)\, dt_0, \qquad (5)$$

where $\mu(t_0)$ is the density of the excitation acts in time, and k is a proportionality coefficient. The integration extends from $-\infty$ to the running instant of time t, meaning that a stationary solution is obtained. It is assumed that the form of the expression for the probability $P_\lambda(t, t_0)$ of the elementary beat was established beforehand.

In such an analysis, it is also assumed that the excitation process occurs instantaneously, or more accurately within a time $\Delta t \ll \omega_{12}^{-1}, \Gamma^{-1}$. This condition is certainly satisfied in the case of excitation with fast particles. For example, the excitation with electrons at resonant energy lasts less than 10^{-16} sec. In the case of optical excitation, the effect of time of realignment of the state of the atom is of the order of the reciprocal width of the exciting-radiation spectrum[5]. Therefore, if we confine ourselves to beats with frequencies less than 10^9 Hz, we can assume that the condition under which the excitation can be regarded as of short duration is satisfied also in the optical method of excitation, if ordinary (non-laser) light sources are used.

A consistent theory of interference phenomena of nondegenerate states was developed by Konstantinov and Perel'[18,19], Corney and Series[20a], Series[1c], and others[2)]. We present the main features of a description that makes use of the density-matrix formalism.

The intensity of spontaneous emission with a polarization vector $\boldsymbol{\lambda}$, connected with the decay of an excited state with sublevels $|m\rangle$, is given by

$$I_\lambda(t) = k \sum_{m, m'} \sigma_{mm'}(t)\, A_{mm'},$$

where $A_{mm'}$ is the so-called observation matrix

$$A_{mm'} = \sum_{\mu} \langle \mu | \hat{d}\lambda | m \rangle \langle m' | \hat{d}\lambda | \mu \rangle,$$

k is a proportionality coefficient, and the indices μ designate the sublevels of the lower state. To find the density matrix $\sigma_{mm'}(t)$ of the upper state, it usually suffices to solve the Schrödinger equation in first order of perturbation theory, the perturbation being the exciting action. In this approximation, the equation for $\sigma_{mm'}(t)$ takes the form

$$(d\sigma_{mm'}/dt) + \sigma_{mm'}(i\omega_{mm'} + \Gamma) = F_{mm'}(t); \qquad (6)$$

here $F_{mm'}(t)$ is an expression characterizing the method of excitation. For example, in optical excitation with a sufficiently broad spectral line and polarization e, from a state with sublevels μ, we have

$$F_{mm'}(t) = J(\omega_0, t)\, \hbar^{-2} \sum_{\mu, \mu'} \sigma_{\mu\mu'} \langle \mu | \hat{d}e | m \rangle \langle m' | \hat{d}e | \mu' \rangle, \qquad (7)$$

where $J(\omega_0, t)$ is the spectral density of the exciting radiation at the central frequency of the working transition, and $\sigma_{\mu\mu'}$ is the density matrix of the initial state. As a rule, in experiments on the interference of excited states, the initial state is characterized by equality of the populations of all the sublevels and, in addition, by the absence of phase coherence between them, so that the matrix $\sigma_{\mu\mu'}$ is diagonal. The condition for the onset of beats is that the off-diagonal matrix elements $\sigma_{mm'}$, obtained with the aid of (6), be different from zero.

[2)] The start of a theoretical description of these phenomena is contained also in an earlier paper. [9].

It is seen from (7) that the necessary natural condition for the onset of a superposition of the levels m and m′ is the simultaneous existence of a transition probability from the common state $|\mu\rangle$ to the states $|m\rangle$ and $|m'\rangle$, for which it is necessary to choose correctly the polarization of the exciting light. It is also easy to see that in the case of a sufficiently large separation of the levels, $\omega_{mm'} \gg \Gamma$, appreciable off-diagonal components of the density matrix can appear only if the coefficients $F_{mm'}$ or $\omega_{mm'}$ are definite functions of the time. The methods considered above for organizing collective interference of states correspond to modulation of the frequency $\omega_{mm'}$, and of the amplitude or phase of the complex quantity $F_{mm'}$.

3. PULSED BEATS

Beats obtained by pulsed excitation are attractive because of their faultless clarify. The luminescence oscillations are not accompanied by any external regular action and demonstrate the natural oscillations of the radiation intensity of atoms with characteristic frequencies. A particularly lucid beat picture is obtained in the interference of only two excited levels. The experimental technique is just as simple as the phenomenon: after a brief excitation of the atomic vapor it is necessary to take oscillograms of the kinetics of the luminescence quenching. In spite of the attractiveness of such an experiment, it was realized already after the interference of nondegenerate states was observed in other less direct experiments. This was due to the appreciable experimental difficulties in obtaining a sufficiently powerful short excitation pulse. These difficulties are clearly illustrated by the first two studies in which pulsed beats were observed almost simultaneously[21,22c].

The object in[21] was mercury vapor, and the transition $6^3P_1 - 6^1S_0$, 2537 Å was used. The triplet 6^3P_1 was split in the magnetic into three levels with m = 0 and ±1. If the polarization of the exciting light is suitably chosen, then the excitation of the level with m = 0 can be excluded (to this end, the electric field vector of the exciting light should have no components along the magnetic field). In this the system coincides with that in the three-level model. To obtain clearly pronounced beats, the distance between the split levels must exceed noticeably their width. The level width of the state 6^3P_1 is approximately 10^7 sec^{-1}. The levels were split to a frequency $\omega_{12}/2\pi = 10^7$ Hz. The vapor was excited by a 10^{-6}-sec light flash from a mercury lamp, shaped by an electron-optical shutter. Such a short excitation-pulse duration, together with a large loss of light in the shutter, led to an extremely low efficiency of luminescence registration. Only one photoelectron connected with the registration of the luminescence was registered in 50 pulses of excitation. Of course, there would be no thought of obtaining direct oscillograms under such conditions. Instead, a system of prolonged accumulation of the photopulses in 16 intervals of 25 nsec each, with different delays after the excitation pulse, was used. Altogether, some 3×10^6 excitation pulses were produced in 15 hours. Figure 2 shows the results. The experimental curve 2, with indication of the spread of the points, is superimposed on the theoretical curve 1 (the shaded rectangle marks the excitation interval). On the whole, as seen from Fig. 2, the experiment points to the existence of beats in the luminescence, although the spread of the points of the same order as the maximum amplitude of the registered oscillatory process. We note that the phase shift observed in Fig. 2 between the experimental and theoretical curves at the start of the process is a consequence of the appreciable background of unmodulated radiation, connected with a number of technical factors.

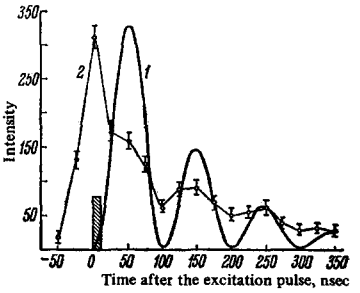

FIG. 2. Pulsed beats [21].

More definite results were obtained in[22c], where the object was cadmium vapor. An analogous transition, $5^3P_1 - 5^1S_0$, 3261 Å, was excited. The lifetime of the state 5^3P_1 of cadmium is 20 times larger than that of the mercury state 6^3P_1, so that a smaller splitting could be used and the exciting pulse could be lengthened to 10^{-7} sec. This has made it possible to use a shutter with relatively large transmission. As a result, the photocathode of the photomultiplier used to register the luminescence of the vapor emitted up to 100 photoelectrons per excitation pulse. Nonetheless, even this was not enough for direct oscillography of the process. Consequently, accumulation was used likewise, but by an entirely different method, which turned out to be at the same time very simple and effective. A pulsed modulator synchronized with the triggering of the oscilloscope sweep was turned on in a stroboscopic regime at a frequency 50 Hz. The oscillograms from the oscilloscope screen were photographed on a single frame of the film, so that ultimately there was obtained a synthetic picture of some 10,000 single oscillograms. The noise was averaged out, and the regular features of the process accumulated. It is essential that in such an accumulation method there is no loss due to the stepwise separation of the individual time intervals of the process[23a]. The information concerning each pulse is registered completely. This is due to the short duration of the employed accumulation—altogether several minutes—with a sufficiently reliable result. Figure 3 shows two copies of strongly contrasting synthetic oscillograms, one used as a control (a) and corresponding to level degeneracy when there are no beats, and the other for the case when the degeneracy is lifted and beats are present (b). The

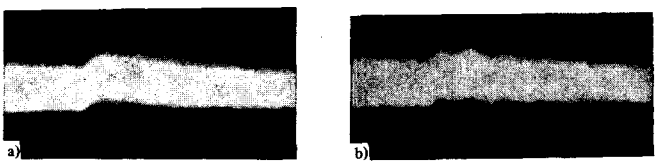

FIG. 3. Synthetic oscillograms of beats [22c].

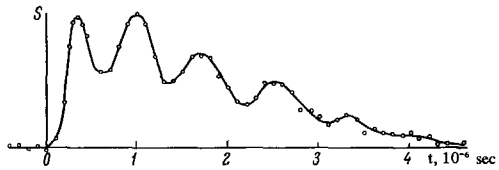

FIG. 4. Results of microphotometry of the original oscillogram of Fig. 3b.

first shows a broad light band—averaged noise bursts. The rapid rise of the band corresponds to the excitation. The envelope of the beam gives the kinetics of the process—a smooth decrease of the spontaneous radiation. This oscillogram was obtained in a zero magnetic field, when the sublevels of the 5^3P_1 state are degenerate. The lower oscillogram demonstrates beats with frequency 1 MHz. The original of this oscillogram was subjected to microphotometry, the result of which is shown in Fig. 4, which demonstrates beats in which the ratio of the maximum oscillation amplitude to the spread of the points is approximately 30. The fact that the depth of modulation is not 100% due to a number of purely technical causes, principal among which is the presence in the cadmium vapor of odd isotopes with different beat frequencies, and the finite time of the exciting pulse.

The investigation of[21] was repeated three years later by a partly reorganized group[23b]. The authors improved the experimental technique and changed the object of investigation, replacing the mercury by cadmium, as in[22c]. The beats were observed with approximately the same reliability as in[22c], but the results were processed much more thoroughly.

We mention one more perfectly analogous experiment[24], in which cadmium vapor was subjected to pulsed excitation by an electron beam. Excitation by an electron beam is also capable of producing a collective phase dependence of the states, as is evidenced by the presence of predominant polarization of luminescence in the absence of a magnetic field, with a direction that coincides with the direction of the electron beam when viewed in a direction perpendicular to the beam. The electron excitation is convenient because of the simplicity with which the beam can be modulated, but leads to a less distinct effect, since the coherence of the states is not complete in this case. The presence of beats in the luminescence was established in[24] by a delayed-coincidence technique.

Beats connected with interference of magnetic sublevels admit of an intuitive treatment based on the concepts of precession of dipoles and magnetic moments in a magnetic field. In all the described experiments, a linear dipole oriented transversely to the magnetic field was excited at a definite instant of time. Such a dipole precesses about the magnetic field, and when viewed, say, in a direction perpendicular to the magnetic field, its projection on the observation direction consequently changes periodically, and this leads to modulation of the intensity of the emitted light. In cases when the level splitting is not connected with the magnetic field, interpretations of this kind are difficult. For sublevels of Stark origin, an attempt of such an intuitive explanation is given in[25].

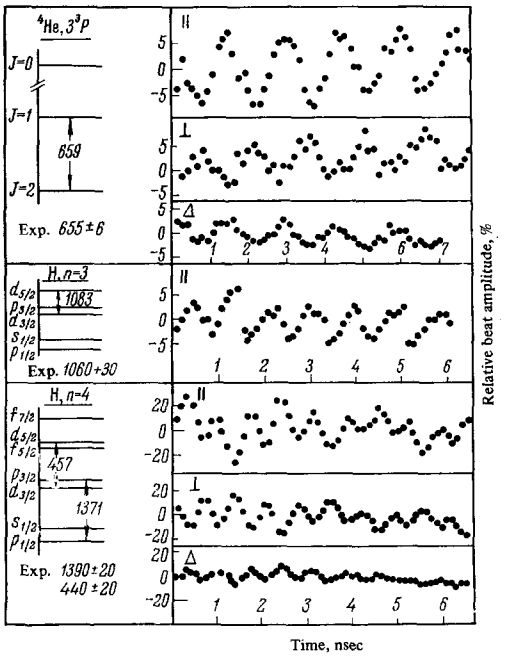

FIG. 5. Beats in a fine-structure system of hydrogen or helium [27].

Beats were observed in pulsed excitation of atoms also in a series of experiments of an entirely different kind, namely the study of the luminescence of fast excited atoms produced by charge exchange of high-energy ions passing through a thin film[26-28]. The most definite results were obtained in[27], where the emission intensity of a beam of atoms was measured as a function of the distance from a carbon film, in which charge exchange of hydrogen and helium atoms took place. The optical system made it possible to separate a section of a beam approximately 1 millimeter long, thus resulting in a very high time resolution, on the order of 10^{-10} sec. Thus, beats were observed at the frequencies of the fine splitting of the helium state $2^3P_{1,2}$ (655 MHz) and the hydrogen states $3D_{5/2,3/2}$ (1083 MHz), $4D_{5/2,3/2}$ (457 MHz) and $4P_{3/2,1/2}$ (1371 MHz), observed on the lines H_α and H_β. These are the highest frequencies on which free beats could be observed so far[3)]. The experimental points obtained in[27] are shown in Fig. 5 (the symbols ∥ and ⊥ denote the positions of the plane of polarization of the registered light relative to the beam, and Δ denotes registration in unpolarized light).

It should be noted that the possible onset of a collective effect of beats under such conditions is not obvious. To this end it is necessary that the phases of the fine states contained in the superpositions be unevenly distributed after the charge exchange (it is this which corresponds to the presence of a definite phase φ_0 in expression (4)). As shown in[14a,27], the occurrence of beats is connected with the alignment of the direction of the atomic beam with the orbital angular momentum of the atom following the charge exchange. As a result of

[3)] Beats connected with stimulated radio-frequency transitions in the hyperfine structure of alkali metals were observed in [29]. In nuclear physics, in the same frequency region, beats of the probability of observing K_0 and $\bar{K}_0$ mesons resulting from interference of the states K_1^0 and K_2^0 were observed in 1960 (see the reviews [12,30]).

further spin-orbit interaction, the spin and orbital components of the electron angular momentum begin to precess about the direction of the total angular momentum, and there is a preferred initial direction of the orbital angular momentum of the atoms. This is what leads to a superposition, coherent over the ensemble of the atoms, of the states of the fine splitting.

The foregoing examples, namely beats in a system of magnetic sublevels and fine-splitting levels, clearly demonstrate the common character of the interference of states, regardless of their nature or the cause of the splitting. (The same circumstance is illustrated splendidly in the nonlinear region by the beats of K_0 mesons; see the preceding footnote.) A very interesting variant of a pulsed experiment with beats is considered in [16a]. In that study, the mercury nuclei Hg^{199} in the ground state were optically oriented in a zero magnetic field. The process of optical orientation brings an appreciable fraction of the atoms (and in the limit all of them) into a state with definite projection of the angular momentum of the nucleus relative to the direction of the orienting light beam [31]. If we now turn on suddenly a magnetic field of sufficiently large intensity transverse to the direction of orientation of the nuclear moments, the latter begin to precess about the field direction, and this is a manifestation of the onset of superposition of energetically different states with different projections of the angular momentum on the direction of the magnetic field ($m = \pm 1/2$). The wave function of the system is given in the form

$$\Psi(t) = (\sqrt{2}/2)\,|\,1/2\rangle\, e^{+i\omega t/2} + (\sqrt{2}/2)\,|\,-1/2\rangle\, e^{-i\omega t/2}, \qquad (8)$$

where ω is the frequency of the splitting of the levels $\pm 1/2$. Thus, in this case the coherence of the states is due not to pulsed excitation but to pulsed lifting of the degeneracy.

In fact, in [16a] the pulsed splitting of the state was effected not by a magnetic field, but by an electromagnetic field of a second more powerful light beam directed at a right angle to the first orienting beam, and polarized in suitable manner. As shown by a number of preliminary investigations (a review can be found in [5]), a light beam not at full resonance with the atomic transition shifts the levels that take part in the transition; the magnitude of the shift depends in the general case on the quantum numbers of the level. In the example considered, an auxiliary beam was used with two close spectral components, and the line of the $6^1S_0 - 6^3P_1$, $F = 1/2$ atomic transition of Hg^{199} fell in the middle between these components. This led to a shift of the sublevels with $m = \pm 1/2$ of the state 6^1S_0 in opposite directions, i.e., to a lifting of the degeneracy and to formation of a superposition (8) of states with definite energy and definite projections of the angular momentum on the direction of the second powerful light beam. Since we are dealing with a superposition of the sublevels of the ground state, this superposition is optically manifest by absorption of light of definite polarization[4]. Concretely, after a pulse of the second light beam was turned on in [16a], damped beats were observed in the intensity of the first orienting beam after its passage through the mercury vapor. An oscillogram of the registered beats is shown in Fig. 6. In concluding this chapter we indicate that one can treat as optical pulsed beats also all other variants of excitation of the free precession of oriented atoms in the ground state, registered by means of optical absorption. Such a precession was observed in the early 60's in most investigations on optical orientation of atoms, and did not attract special attention.

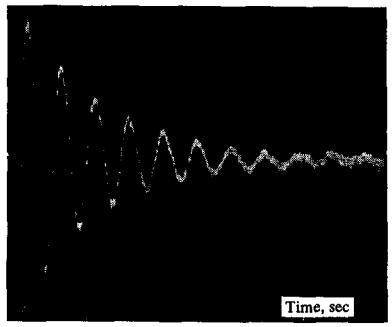

FIG. 6. Oscillogram of beats produced by pulsed lifting of the degeneracy [16a].

4. RESONANCE OF BEATS

In this and in the following two chapters we consider experiments in which the interference of states becomes manifest under stationary conditions. By resonance of beats we mean the appearance of modulation in the luminescence or in the absorption coefficient when the system is periodically excited at a frequency close to the frequency of the free beats.

This phenomenon was first observed in [7b], where atoms were optically oriented by modulated light directed across a magnetic field. Under these conditions, a superposition of magnetic states is produced, in analogy with the description given in the preceding chapter as applied to [16a]. The phenomenon was treated in a particular manner, in terms of precession of the magnetic moments[5]. The resonance of beats was demonstrated in a much clearer form in [20b,22a], in the excited 5^3P_1 state of cadmium. As already mentioned, if magnetic splitting of this state is used and the polarization of the exciting radiation is chosen perpendicular to the magnetic field, then it is possible to produce a coherent superposition of the sublevels with $m = \pm 1$, i.e., to realize the simplest three-level model considered in Chap. 2. Let the exciting light be modulated in accordance with the law

$$\mu(t_0) = \mu_0 (1 + \varepsilon \cos \Omega t_0).$$

Substituting this expression in (5) and using (4) we have the following expression for the intensity I(t) of the luminescence registered perpendicular to the magnetic field:

[4] As indicated in Chap. 1, the beats are observed on going from the superposition state to the eigenstate under the influence of some perturbation. So far, only spontaneous emission was considered, and the perturbations were the fluctuations of vacuum. In this case the perturbation is an external light beam that causes transitions from a lower state to a higher one.

[5] These investigations are discussed in somewhat greater detail later on in Chap. 7.

$$I(t) = C[\Gamma^{-1} + \varepsilon(\Gamma\cos\Omega t + \Omega\sin\Omega t)(\Gamma^2+\Omega^2)^{-1} + (\Gamma\cos\varphi + \omega_{12}\sin\varphi)(\Gamma^2+\omega_{12}^2)^{-1} + 0{,}5\varepsilon\{[\Gamma\cos(\Omega t-\varphi) + (\Omega-\omega_{12})\sin(\Omega t-\varphi)][\Gamma^2+(\omega_{12}-\Omega)^2]^{-1} + [\Gamma\cos(\Omega t+\varphi) + (\Omega+\omega_{12})\sin(\Omega t+\varphi)][\Gamma^2+(\omega_{12}+\Omega)^2]^{-1}\}; \quad (9)$$

Here φ is double the angle between the direction of the electric vector of the exciting light and the plane normal to the observation direction, and C is a proportionality coefficient. This expression takes into account the fact that the probabilities of transitions to the lower state are the same for the two interfering sublevels.

The first term in the formula corresponds to the constant component of the population of the excited state. The second term, which decreases rapidly with increasing excitation frequency, reflects the modulation of the population of the excited state. The remaining terms are connected with interference of the states. The third term describes the interference contribution to the constant component of the intensity of the scattered light, which is significant only in the region where the levels overlap noticeably. This is the well-known and already mentioned effect of level crossing (see, for example,[32]). The fourth term describes the alternating component of the luminescence. We see that the depth of the modulation increases resonantly in the vicinity in the region where the excitation modulation frequency becomes equal to the frequency of the transition between the interfering sublevels ($\Omega = \omega_{12}$). The left term is analogous but "antiresonant," and goes over into the resonant term when the magnetic field is reversed. Under conditions of considerable level splitting, $\omega_{12} \gg \Gamma$, an important role is played only by the resonance term. We note that its amplitude (maximum) value does not depend on the frequency; the beat resonance can occur at any frequency, regardless of the inertia of the spontaneous emission.

Beat resonance in luminescence was first observed in[22a]. A quantitative interpretation of this phenomenon was given in[19]. Similar investigations were performed in[20]. We present the results of an investigation of the modulation of the spontaneous 3261 Å emission line of cadmium[25]. In that study, the excitation was produced by modulated light polarized at an angle of 45° to the direction of the magnetic field, so that a superposition of all the excited levels with m = ±1 and 0 was produced. Since these levels are equidistant, the system has two resonant beat frequencies, one of the transition between the outer sublevels ±1, and the other between the central and the outer ones. In the experiment, the light modulation frequency was fixed at 1000 kHz. The magnetic field was varied, so that two resonant values of the magnetic field were expected. Registration was by a synchronous detection technique. Figure 7 shows the dependence of the obtained signal on the magnetic field, together with the theoretically calculated curve. The resonances occurred in the vicinity of ±0.24 and ±0.48 Oe, and the luminescence was modulated in these regions with a phase shift of 90°.

Beat resonance in mercury vapor was observed under analogous conditions in[33,34]. Experiments on beat resonance excited by an electron beam are described in[22b,35a,36a]. A modification of this type is of interest because it becomes possible to excite states that are not connected with the ground state by an optical transition[32,33]. Problems of the theory of coherent

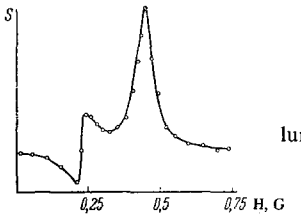

FIG. 7. Signal of beat resonance in luminescence of cadmium [25].

excitation by electron impact are discussed in[35c,36a]. The advantages of using an electron beam are demonstrated particularly clearly in[36a], where a modulated electron beam was used for coherent population of the mercury metastable state 6^3P_2. If the beam is perpendicular to the magnetic field, then a superposition of states with m = ±1 is obtained[6]. This superposition was registered with a light linearly polarized beam directed along the magnetic field was used from an additional lamp, with wavelength 5461 Å ($6^3P_2 - 7^3S_1$). On passing through the mercury vapor, this beam exhibited absorption modulation when the resonance conditions were satisfied.

The qualitative picture of the onset of beat resonance, described in Chap. 2, makes use of the concept of in-phase addition of the oscillatory processes of decay of various atoms. It is clear from this picture that what we have here is free beats for each individual atom taking part in the resonance. At ordinary intensities, each atom is rarely excited, the intervals being much longer than the lifetime of the excited atom. Therefore modulation of the excitation does not affect in any way the character of the emission of each individual atom, and only leads to a phasing of the beats of the individual atoms. This explains why the width of the beat resonance is determined only by the damping parameter Γ of the atom. In other words, the radiospectroscopic methods are characterized by a saturation that is connected with the perturbing action of the alternating field.

A kinetic treatment of the onset of the beat resonance is convenient because it is universal and is equally well applicable to any type of excitation[7]. When optical excitation is used, it is possible to use with success a spectral treatment of the resonance[19], in which the beat resonance is regarded as the result of an increase in the effectiveness of the scatter of correlated harmonics of the modulated-light intensity when the splitting of the spectral components of the scattering atoms coincides with the frequency of the light modulation. This treatment is convenient for the understanding of the onset of resonance also when modulated laser radiation is used for the excitation, i.e., in a situation that does not fit the concept of pulsed excitation. The resonance picture turns out to be the same in this case. The variant with laser excitation was investigated in[39]. The laser radiation was modulated by a clever device, using synchronized beats of longitudinal modes of a neon-helium laser (6328 Å). A theoretical description of the

[6] Generally speaking, a superposition of states with m = ±2 also takes place, but the existence of such coherence cannot be observed in single-photon electric dipole transitions, owing to the selection rules.

[7] This treatment was applied to nuclear physics in [37,38].

beat resonance using quantum-electrodynamic methods, which leads to the same results, is given in [40a].

In all the experiments, the beat resonance was realized at the magnetic-splitting level, with the exception of [25], where the Stark structure of the 5^3P_1 level of cadmium was investigated. A theoretical investigation of beat resonance in a system of hyperfine-splitting levels was carried out in [41]. In [40b], quantum electrodynamics methods were used to investigate theoretically beat resonance in a system of levels with different lifetimes. In this case there are differences in the result of the excitation of the system by a broad spectrum and by a laser. Questions of interference of states with different lifetimes were treated also by semiclassical methods in [1c].

5. PARAMETRIC RESONANCE

Parametric resonance is an interference effect connected with modulation of the energy interval between the interfering states. It is manifest in the appearance of modulation in the spontaneous emission (or in the absorptivity) of a system of atoms when the interval between the sublevels of the excited states is modulated at a frequency equal to the level splitting frequency or is smaller by an integer factor. Modulation and radiation occurs at frequencies close to the modulation frequency of the splitting. Like all interference effects, parametric resonance can be registered also in absorption.

The influence of modulation of the position of energy levels on the polarization [42] and the spectrum [43] of atom emission was discussed long ago and many times. However, the idea of interference resonance in the case of such modulation was first advanced in [9], where there are also theoretical hints, which were formulated in somewhat more extended form in [44]. Further development of these ideas, carried out in [18], led to a thorough change in the predictions concerning the experimental manifestations of the process. In that reference, this phenomenon was called parametric resonance.

Parametric resonance can be easily described on the basis of the general analysis of Chap. 2. Confining ourselves to the simplest case of two interfereing sublevels (corresponding to the conditions of the first experiments), we present an expression for the intensity of spontaneous emission with a given polarization

$$I(t) = C \operatorname{Re} \sum_{k=-\infty}^{\infty} B_k e^{ik\Omega t},$$

where C is a proportionality coefficient, and the harmonic amplitudes B_k are given by the expression

$$B_k = i^k \sum_{n=-\infty}^{\infty} J_{k+n}(\omega_1/\Omega) J_n(\omega_1/\Omega) [\Gamma + i(\bar{\omega}_{12} - n\Omega)]^{-1};$$

Here $J_n(\omega_1/\Omega)$ is a Bessel function of the first kind. It is assumed that the modulation of the interval ω_{12} is given by $\bar{\omega}_{12}(t) = \bar{\omega}_{12} + \omega_1 \cos \Omega t$. Thus, parametric resonance is characterized by an infinite set of harmonics in the radiation intensity, and for each harmonic there is an infinite set of resonances. This phenomenon can be qualitatively explained in various ways. Probably the clearest of them is the spectral approach [18]. Assume that we are dealing with parametric resonance in an excited state following excitation by light. Since the light is not modulated in these experiments, it can be represented in the form of a set of independent harmonics of the field. An atom with two levels in the excited state can be described by an aggregate of two oscillators with different frequencies. In experiments on modulated excitation, the modulation of the light has led to the appearance of two correlated satellites for each optical harmonic. When the frequency difference between the two oscillators coincided with the modulation frequency, both oscillators could be excited by two coherent harmonics of the light, and consequently could interfere coherently (with respect to all other oscillators in the system). What is transformed in parametric resonance is not the spectrum of the exciting light, but the absorption spectrum of the oscillators. As is well known, frequency modulation produces in the oscillators an infinite set of discrete absorption lines separated from the fundamental frequency by multiples of the modulation frequency. When the average distance between the sublevels of ω_{12} and the modulation frequency Ω are equal or are exact multiples, the same harmonic of the exciting light is capable of exciting one of the oscillators at the fundamental frequency, and another at one of the sideband frequencies coinciding with it. Thus, one optical harmonic excites two correlated sets of oscillations, which are able to produce various beats between each other, including also a beat at zero frequency. The latter, in the case of interference of magnetic sublevels, corresponds to a change in the average directivity pattern of the emission of the system of atoms. We note that, unlike in beat resonance, no modulation of the populations takes place in parametric resonance, and the effect has a pure interference character. This is a radical difference between parametric resonance and the methodologically similar double resonance, which is characterized by induced transitions between the sublevels. In the case of magnetic levels, both experiments differ only in the orientation of the alternating magnetic field. In experiments on double resonance, the alternating field is perpendicular to the constant field and leads to magnetic resonance. In experiments on parametric resonance, both fields are parallel—the alternating field modulates the splitting without causing any transitions. This difference gives rise to a difference in the forms of the signals. The widths of the parametric-resonance signals do not change with increasing modulation of the energy gap.

There are relatively few experimental studies of parametric resonance. The effect was first observed in the luminescence of cadmium [18] in a magnetic field. Shortly thereafter, a similar experiment was performed [45]. Parametric resonance in the ground state was observed almost simultaneously in [15b,c], the objects of the investigation being optically oriented cesium vapor and He^{199} vapor, respectively. In all these experiments, the level splitting was modulated by a magnetic field. The only exception was [25], in which parametric resonance of the Stark sublevels was observed when the electric field was modulated. More detailed information, with a demonstration of the experimental results, can be found in a recent review [13]. In [46], parametric resonance is treated theoretically as a multiphoton process.

6. PHASE RESONANCE OF BEATS

Phase resonance is an interference effect that becomes manifest also in the appearance of modulation (sometimes in accordance with a complicated law) of absorption or emission of a system of atoms, and is connected with the action of the interfering states on the initial phase difference. In practice, in the case of interference in a magnetic level structure, the initial phase difference depends on the direction of the unisotropic exciting action relative to the quantization axis, so that we are dealing with modulation of the polarization or of the direction of the exciting light, or else of the direction of the flux of exciting fast particles. Phase resonance takes on significantly different forms, depending on the time variation of the initial phase. In the simplest form, the phase varies with time linearly (see Chap. 2). Such an experiment could be realized by exciting luminescence with light along a magnetic field with a rotating plane of polarization. Another variant, proposed by Series [1b], consists of exciting an atomic vapor with two beams of light orthogonal to each other and to the magnetic field, and modulated in intensity with a phase shift of 90°. In both cases, the resonance would be given by a formula corresponding to the first and third terms of (9). It is difficult to devise a method for varying the phase linearly. Therefore the first study of phase resonance was made with harmonic modulation of the phase. If the variation of the initial phase is given by $\varphi_0(t) = \varphi + \varphi_1 \cos \Omega t$, we can easily obtain an expression for the intensity of the scattered light

$$I(t) = C \operatorname{Re} e^{i\varphi} \sum_{k=-\infty}^{\infty} i^k J_k(\varphi_1) e^{ik\Omega t} [\Gamma + i(\omega_{12} + k\Omega)]^{-1}; \qquad (10)$$

Here again $J_k(\varphi_1)$ is a Bessel function of the first kind. It follows from the last expression that at a fixed frequency Ω of the phase modulation, the beats occur at multiple frequencies $k\Omega$, and the amplitude of the k-th harmonic has a maximum value when the distance between the interfering sublevels ω_{12} is k times larger than the modulation frequency. In the presence of a spectrum of harmonics, a similarity appears between the phenomenon and parametric resonance. The difference between the phenomena lies in the different number of resonances. In phase modulation, each harmonic has one resonance and one "antiresonance" under the condition $\omega_{12} = \pm k\Omega$, whereas in parametric resonance the harmonic with number k has an infinite number of resonances $\omega_{12} = n\Omega$, where $n = 0, \pm 1, \pm 2, \ldots$ etc. At a small depth of modulation, that is, when $\varphi_1 \ll 1$, we can confine ourselves in the preceding formula to terms with $k = \pm 1$ and $k = 0$. Then the expression for the luminescence intensity coincides with the expression for the beat resonance (9), in which it is necessary to discard the second term and replace ϵ by $\varphi_1/2$.

An experiment with harmonic modulation of the phase was realized in [22b]. A traditional object, cadmium and the transition $5^3P_1 - 5^3S_0$ was used. The vapor was excited along the magnetic field with linearly polarized light passing through a Faraday cell, so that it was possible to swing the plane of polarization at high frequency (the resonant Faraday effect was used in the vapor of the same cadmium with magnetic splitting of the absorption line in the modulated magnetic field).

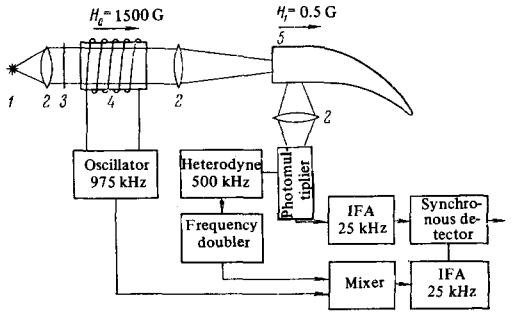

FIG. 8. Block diagram of setup for the observation of phase resonance of beats [22d].

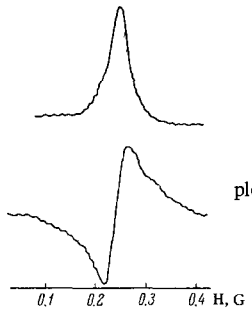

FIG. 9. Two limiting forms of experimental plot of phase-resonance signal [22d].

The experimental setup and plots of the resonance signal are shown in Figs. 8 and 9 (in Fig. 8: 1—cadmium lamp, 2—lens, 3—polarizer, 4—cell with cadmium vapor to modulate the position of the plane of polarization of the light, 5—resonance vessel, IFA—intermediate frequency amplifier). The waveform of the signal depends on the average position of the plane of polarization of the exciting light, which determines the phase φ in expression (10). Depending on the choice of this phase, the signal after synchronous detection is one superposition or another of a Lorentz contour and a dispersion contour, as can be seen from (10). Figure 9 shows plots of the signal in these two limiting forms.

Phase resonance of cadmium nuclei in the ground state, under conditions approaching a linear phase variation, was realized in [47]. It should be noted that mixed situations arise sometimes, corresponding to a superposition of beat resonance and phase resonance: in theory this corresponds to simultaneous modulation of the modulus and phase of the first part of expression (6). This case corresponds to a resonant experiment on optical orientation of mercury vapor with light perpendicular to the magnetic field, using a rotating phase plate to ensure, together with the polarizer, the modulation of the type of polarization of the light beam [16b].

7. NONLINEAR MANIFESTATIONS OF INTERFERENCE OF ATOMIC STATES

The interference phenomena considered in the preceding chapters were linear in the intensity of the action that produces the excited superposition state, or in the intensity of the controlling light beam (in experiments with absorption). In these experiments, the formation of the superposition state and its observation were independent. There is also a group of experiments in which the same action (optical) produces the interference state and reveals its occurrence. The effects ob-

served thereby are nonlinear and require the use of higher orders of perturbation theory for their description. In fact, nonlinear manifestations of the interference of states were already mentioned in connection with experiments on beats in the absorption in transitions from the ground state. In these experiments, the superposition state was prepared with the aid of optical orientation, which is essentially a nonlinear process[5,31]. However, if the intensity of the orienting light beam is fixed, and an auxiliary weak light beam is used to register the beats, then the observed phenomena do not differ in any way from their analogs observed in the interference of excited states. The situation is different when the registration is carried out in an orienting beam. We consider below briefly three types of experiments on nonlinear manifestations of the interference of states. The first of them[7b], historically the earliest, is nonlinear resonance of beats and can be easily treated with the aid of the phenomenological picture of optical orientation. The second, which is of great practical significance and called the mode crossing phenomenon[48a], requires a rigorous description, but can be understood in analogy with the first. Finally, the third type of nonlinear experiments, in which the so-called incoherent beats are observed[49], will be described with the aid of perturbation theory.

In[7b] the atoms were optically oriented with the aid of circularly polarized resonant radiation perpendicular to the magnetic field. The orientation process can be treated as transfer of the angular momentum of the photons of the polarized light to the atoms, which become aligned in this case parallel or antiparallel to the light beam. This is accompanied by a change (most frequently, a decrease) of the absorption of light by the oriented atoms. Were there no magnetic field, then only the relaxation processes would prevent complete orientation of the atoms. The absorption produced thereby is evidently connected in nonlinear fashion with the intensity of the oriented light, which competes with the relaxation orientation-loss processes. The presence of a magnetic field perpendicular to the beam alters the picture decisively. With respect to the magnetic field, the oriented atom falls into a superposition state that is classically described by its precession about the field vector. Since the phase distribution of such a precession is uniform for different atoms, the ensemble of atoms turns out to be not oriented in the mean. However, if the orienting beam is modulated in intensity at the precession frequency of the atoms, then a group of atoms that precesses in phase is produced and now interacts with the light as a system of oriented atoms, since the periodically turned-on light always illuminates the atoms at the same precession phase, namely, then they are aligned along the direction of the light. This results in an integral change in the light absorption by the atoms in comparison with the average absorption in the absence of orientation. In essence, the described experiment is a typical beat resonance, but is revealed by the change of the integral absorption[8]. This indeed is a main feature of the nonlinear variant. The absorption increment attained in this experiment decreases with decreasing light intensity. At low intensity, this connection is quadratic. The small parameter is the ratio of the average number of excitations of the atom per unit time to the average relaxation time. The latter can take on values in the range 10^{-3}–10^{3} sec for atoms in the ground state, so that the optical nonlinearity comes into play already at rather moderate intensities. The development of lasers with their high spectral power has greatly extended the region of nonlinear interactions with atoms, since laser radiation is capable of overcoming much faster relaxation processes, including spontaneous emission. Lasers have made it possible to use nonlinear resonance of beats to investigate excited atoms, by a procedure given the new designation mode crossing. It reduces to the following. Consider a system (atom, molecule) having two close levels 1 and 2 in the excited state, coupled by a radiative transition with a common level 0, which can be either higher or lower than the levels 1 and 2. If radiation of a multimode laser in which the spacing between neighboring modes is equal to the spacing between levels 1 and 2, or is smaller by an integer factor, is made to pass through a medium of particles with such an energy scheme, then the coefficient of absorption (amplification) of the laser radiation in such a system is minimal. For a radiation power $P_{1,2}$ absorbed or emitted (by stimulation) by the medium under the influence of laser radiation, an expression $P_{12} \sim I_1 I_2 (N_{12} - N_0) \times [\Gamma_{12}^2 + (\omega_{12} - \Delta)^2]^{-1}$ was obtained in[48a] in third order of perturbation theory; here N_{12} is some of the populations of sublevels 1 and 2, N_0 is the population of level 0, I_1 and I_2 are the intensities of the two modes of the laser, Γ_{12} is the arithmetic mean of the widths of levels 1 and 2, Δ is the frequency difference between the modes, and ω_{12} is the distance between the levels 1 and 2. We call attention to the nontrivial character of the phenomenon: as a rule, in experiments of this kind the spacing ω_{12} is much smaller than the Doppler broadening of the spectral absorption (amplification) line of the medium. Therefore the absorption coefficient for each of the modes taken separately does not depend on the value of the splitting Δ. Such a dependence comes into play only when the interference of states is taken into account.

From the principal point of view, there is complete analogy between the mode crossing phenomenon and nonlinear resonance of beats in optical orientation by means of modulated light, since the excitation in the case of mode crossing is also carried out by light of variable intensity, owing to the beats between the components of the mode structure of the radiation. There

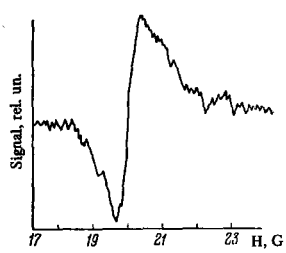

FIG. 10. Mode crossing signal in xenon discharge [52].

[8] Complete identity with beat resonance is obtained by using an auxiliary unmodulated light beam, which is also circularly polarized and lies in a plane perpendicular to the magnetic field. Such a beam, by interacting with the system of precessing atoms, becomes intensity-modulated.

are, however, differences in the details. In the case of optical orientation with modulated light, the total population of the ground state remains unchanged, and only a definite distribution of the interference phase shifts of the magnetic sublevels is established. In the laser variant, the total population of the aggregate of sublevels 1 and 2 is altered by the laser emission[9)], but additional interference of states 1 and 2, coherent over the ensemble of the particles, arises under the mode crossing condition $\omega_{12} = \Delta$ and leads to an additional change of the absorption (or amplification) of the laser emission with the interfering modes.

We present an example of the use of the mode crossing effect for an analysis of the magnetic structure of the excited state of xenon $5d[5/2]^0_2$. Radiation from a multimode xenon laser at a wavelength 3.37 μ (transition $5d[5/2]^0_2 - 5p[3/2]_1$) is made to pass through a gas discharge in xenon. When the intensity of the magnetic field that splits the $5d[5/2]^0_1$ level is varied, resonance is observed in intensity of the transmitted light. An experimental plot is shown in Fig. 10. The resonance curve has a dispersion character, since a magnetic-field scanning technique was used with subsequent synchronous detection of the photoreceiver signal. The gyromagnetic ratio and the homogeneous width of the state were determined from the experimental results.

Let us discuss, finally, recent experiments[49] in which incoherent beats likewise pertaining to nonlinear interference phenomena were observed. In all the previously considered manifestations of interference of states, the necessary condition was that the elementary oscillations in the emission of the individual atoms be in phase. However, the absence of such phase equality is not a principal obstacle to the observation of beats. Indeed, if the number of excited atoms is large enough, then, when the phase distribution of the elementary beats is random, it is possible to choose a large group of atoms with beat phases that are identical (within specified limits). If the entire ensemble of the radiating atoms is broken up into such groups, it can be seen that the emission of the entire system is made up of a large set of light radiative processes of type (4) with a random distribution of the phases. It is easy to verify that the resultant radiation, while not having a regular modulation of the amplitude, contains in the spectrum of its fluctuations information concerning the characteristic parameters Γ and ω_{12} of the terms of processes of type (4), parameters characterizing the elementary radiative processes.

It follows from the foregoing that it is possible in principle to extract information concerning the atomic parameters by analyzing the spectrum of the intensity fluctuations of the radiated light. To establish quantitative criteria of the realizability of such a measuring procedure, it is necessary to take into account the shot noise in the photoregistration and the wave noise of the interference of radiation from different atoms. The corresponding estimates show that a useful signal in measurements of this type is very weak and calls for excessively large accumulation times to be reliably registered. Nonetheless, incoherent beats could be observed in a nonlinear variant of the experiment. The experiments were performed as follows: the investigated object was low-density xenon vapor excited in a thin tube by an electric discharge. As shown in a number of recent investigations (reference to which can be found in[53]), this causes appreciable population of the states forming the transitions $5d[7/2]_3 - 6p[5/2]_3$ (3.507 μ and $5d[7/2]_4 - 6p[5/2]_3$ (5.57 μ), and in a definite interval of xenon pressure and discharge currents, it is possible to observe in these transitions both appreciable absorption and amplification of the radiation, corresponding to an optical density on the order of 0.1 cm^{-1}. The cell was a short tube (about 10 cm long). Unpolarized resonant radiation with wavelengths 3.508 and 5.57 μ, generated by an external source, was passed through the discharge in the vapor. The source was a similar tube, but longer, with discharge conditions such that the intrinsic spontaneous radiation in these lines was amplified. The spectral width of these lines was of the order of 10^8 Hz, which exceeded all the splittings of the levels whose interference was investigated. The levels of the atoms in the investigated volume were split by an actual magnetic field. After passing through the tube, the radiation was fed through a linear analyzer and was registered with a low-inertia photoreceiver. Leaving out the description of the inessential details of the setup (Fig. 11), we mention only that the further reduction of the photoreceiver signal consist of an analysis of the power spectrum of the photocurrent fluctuations as a function of the magnetic field intensity (in Fig. 11, LA is a linear analyzer, while HF and LF stand for high and low frequency).

It was observed that the fluctuation power at a given frequency has narrow extrema when the magnetic field is varied. The widths of these extrema were of the order of the natural width of the states $5d[7/2]_{3,4}$ and their position with respect to the magnetic field corresponded to the chosen frequency interval of observation, in accord with the available data on the splitting of these states in the magnetic field. An example of the experimental curve is shown in Fig. 12, which shows a plot of the derivative of the spectral density of the radiation at 3.507 μ at the point 22 MHz ± 50 kHz as a function of the magnetic field intensity for Xe^{136}.

The sign of the extremum was determined by the conditions of the passage of radiation through the tube, namely, if the light was amplified in the tube, then a narrow dip appeared in the noise spectrum, and its position was determined by the magnetic field. If absorption took place in the tube, the dip gave way to a maximum.

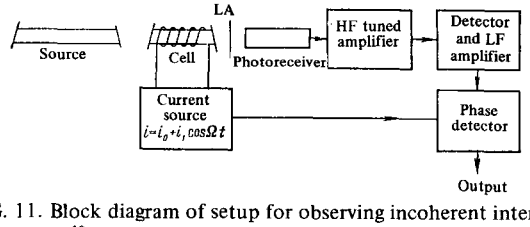

FIG. 11. Block diagram of setup for observing incoherent interference of states [49].

9) Since laser emission consists of a set of very narrow spectral lines, we are dealing here with a change in the populations of only those Doppler-ensemble atoms which have a suitable velocity projection in the direction of the light beam. The phenomenon of selective excitation of atoms by laser radiation is called the Bennet "hole burning" effect [51].

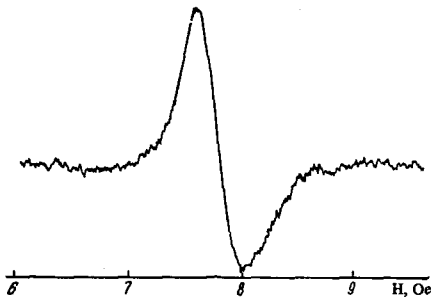

FIG. 12. Derivative of spectral density of xenon emission as a function of the magnetic field intensity (see the text).

These phenomena are explained as follows[49]. Assume that at a given polarization λ there is incident on the investigated object a radiation with intensity given by the random function of time $I_\lambda(t)$. After passing through the investigated volume, the intensity decreases or increases by an amount $W_\lambda(t)$. The output radiation has an intensity $J_\lambda(t) = I_\lambda(t) - W_\lambda(t)$. To find the power spectrum $S(\omega)$, we determined the correlation function $\langle J_\lambda(t)J_\lambda(t+\tau)\rangle = S(\tau)$, where the angle brackets denote averaging. If it is assumed that $W_\lambda(t) \ll I_\lambda(t)$, then we can see that

$$S(\tau) = \langle I_\lambda(t)\, I_\lambda(t+\tau)\rangle - \langle I_\lambda(t)\, W_\lambda(t+\tau)\rangle - \langle I_\lambda(t+\tau)\, W_\lambda(t)\rangle = S_0(\tau) - S_1(\tau),$$

where $S_0(\tau)$ is the correlation function of the incident radiation, and $S_1(\tau)$ is a correction connected with the interaction with the medium. This correction is thus directly connected with the energy $W_\lambda(t)$ released or absorbed in the medium.

For the quantity $W_\lambda(t)$, assuming that the emission line and the absorption line of the atom have a considerably broad spectrum, owing to the Doppler broadening, we can obtain in first order perturbation theory the following expression:

$$W_\lambda(t) = AI_\lambda(t)\left[\sum_{m,\mu,\mu'}\lambda_{m\mu}\lambda_{\mu m}\sigma_{\mu\mu'} - \sum_{m,m',\mu}\lambda_{\mu m}\lambda_{m'\mu}\sigma_{mm'}\right]; \quad (11)$$

here A is a proportionality coefficient, and the remaining notation is the same as in Chap. 2. The first sum corresponds to absorption of energy with a transition from a superposition of the lower levels $\sigma_{\mu\mu'}$ to all the upper states $|m\rangle$. The second sum describes an analogous contribution of the stimulated emission. For the density matrix components with $\sigma_{mm'}$ and $\sigma_{\mu\mu'}$, which are connected with the action of the light I(t), Eqs. (6) and (7) are valid.

As explained in Chaps. 2 and 3, the harmonic component of the intensity $I_\lambda(\omega)$ leads to the appearance of a corresponding harmonic of the density matrix $\sigma_{mm'}$ or $\sigma_{\mu\mu'}$, which increases resonantly when the frequency ω approaches the frequencies $\omega_{mm'}$ or $\omega_{\mu\mu'}$, namely,

$$\sigma_{mm'}(\omega) = -NkI_\lambda(\omega)\sum_{\mu'}\lambda_{m\mu}\lambda_{\mu' m'}/[\Gamma_m + i(\omega_{mm'} - \omega)],$$
$$\sigma_{\mu\mu'}(\omega) = NkI_\lambda(\omega)\sum_{m'}\lambda_{\mu m}\lambda_{m'\mu'}/[\Gamma_\mu + i(\omega_{\mu\mu'} - \omega)]; \quad (12)$$

here k is a proportionality coefficient, N is the population difference between the aggregate of the upper levels m and the lower levels μ, and Γ_m and Γ_μ are the widths of the upper and lower levels.

When (11) and (12) is taken into account, it can be seen that the increment of $W_\lambda(t)$, together with the correlation function $S_1(\tau)$, contains the Fourier components $s(\omega)$, which increase linearly in absolute magnitude in the vicinity of the frequencies of the interference between the upper and lower systems of levels, so that the power spectrum $S(\omega)$ of the transmitted light, together with the spectrum $S_0(\omega)$ of the initial emission, contains a term $s(\omega)$ that carried information concerning the interference of the sublevels of the object:

$$s(\omega) \approx NS_0(\omega)\,\mathrm{Re}\sum_{\substack{m,m',\\ \mu,\mu'}}\{\lambda_{m\mu}\lambda_{\mu' m'}\lambda_{\mu m'}\lambda_{\mu m}[\Gamma_m + i(\omega_{mm'} - \omega)]^{-1} + \lambda_{m\mu}\lambda_{\mu' m'}\lambda_{m'\mu}\lambda_{\mu m}[\Gamma_\mu + i(\omega_{\mu\mu'} - \omega)]^{-1}\}. \quad (13)$$

The complete expression for the power spectrum of the transmitted light contains also other terms analogous to the terms of formula (9) for the simplest beat resonance.

The off-diagonal terms of the sum in (13) describe interference of nondegenerate states. The experiments revealed only resonance of the upper levels, since the lower levels which were much broader, remained in essence degenerate in the employed magnetic fields. The diagonal terms of the sum were responsible for the singularity of the power spectrum at frequencies in the vicinity of zero. The corresponding dips in the spectrum were also observed experimentally[9].

The described theoretical picture of the phenomenon reduces the matter to a certain beat resonance: the initial light is represented as consisting of independent intensity harmonics, each of which excites its own partial nonlinear beat resonance registered with the aid of the integral intensity of the induced transition. The aggregate of these independent resonances leads to a summary singularity in the spectrum of the fluctuations.

The close relationship between the phenomenon of coherent beats and the mode crossing effect is quite evident. In the former case the system of atoms is subjected to excitation with a broad spectrum of fluctuations and transforms it in a characteristic manner. In the latter case, there is realized harmonically modulated excitation with successive registration of the response of the system to different modulation frequencies.

8. METHODOLOGICAL SIGNIFICANCE OF INTERFERENCE OF NONDEGENERATE ATOMIC STATES

It follows from all the foregoing that all the variants of the beats can serve as sources of information on the mutual distances between interfering sublevels and on their homogeneous widths. Inhomogeneous broadening of the Doppler type does not limit the accuracy of such measurements. It is useful to compare the beat methods with the main competing method, that of double radio-optical resonance. The double resonance method[16,54] consists of inducing, by means of a radio-frequency field, transitions between the investigated states, which are revealed by the change of the optical properties of the system. The inducing alternating field perturbs the investigated system and causes a broadening and a shift of the levels. The degree and character of this perturbation can be calculated easily only in simplest cases.

In interference-beat methods, the investigated state does not experience any additional perturbation. From general points of view, this can be attributed to the fact that in these methods the observed quantity is connected not with an induced change in the populations, but with a natural time variation of the phase difference between the interfering states, and the rate of this variation is proportional to the splitting energy. This advantage of the beat method becomes particularly clearly pronounced in investigations of multi-level systems. If one deals with a magnetic equidistant structure, then the use of, say, beat resonance leads to the appearance of two nonsaturating resonances of simplest form, corresponding to interference of states that differ by one or by two in the projections of the angular momentum. The shape of the double-resonance signal under these conditions is predictable but very complicated[54]. To obtain information on the parameters of the investigated system it is necessary to measure in addition the absolute intensity of the alternating field, which is not always possible. If the system is non-equidistant, then the use of beat resonance leads to the occurrence of two series of resonances ($|\Delta m| = 1, 2$), whose positions yield directly the distances between the unperturbed sublevels, and whose widths are determined by the homogeneous broadening of the levels. When double resonance is used, there is produced a complicated system of resonances whose number, positions of extrema, and widths depend on the external field intensity. The double-resonance signals were obtained analytically, under certain favorable conditions, only for three levels. We present for comparison the analytic form of the signal of (a) beat resonance and (b) double resonance for a system of three non-equidistant levels with m = 0 and ±1. Such a situation arises when parallel magnetic and electric fields are simultaneously applied to a state with unity angular momentum.

a) Beat resonance: $\omega_{-1,0}, \omega_{0,+1}, \omega_{-1,+1} \gg \Gamma$, the signal I from the output of the synchronous detector, at a definite choice of phase, is given by[25]

$$I \sim \Gamma^2[\Gamma^2+(\Omega-\omega_{01})^2]^{-1} + \Gamma^2[\Gamma^2+(\Omega-\omega_{-1,0})^2]^{-1} - 0{,}5\Gamma(\Omega-\omega_{+1,-1})^2[\Gamma^2+(\Omega-\omega_{+1,-1})^2]^{-1}$$

here, as before, Ω is the excitation modulation frequency and $\omega_{ik} = \omega_i - \omega_k$, where the subscripts correspond to the projections of the angular momentum (Fig. 13).

b) Double resonance: the signal is proportional to the intensity of the transitions to the lower S state with change of angular-momentum projection by ±1, under the condition that only the level with m = 0 becomes populated upon excitation[55][10]:

$$I \sim A^2\{\Gamma^4+\Gamma^2(\varepsilon^2+5\omega^2+4A^2)+2[2\omega^2\varepsilon^2+(\omega^2+2A^2)(2\omega^2+A^2)]\} \times \{\Gamma^2[\Gamma^2+\varepsilon^2+3(2A^2+\omega^2)]^2+4[\omega^2(\omega^2-\varepsilon^2)+2A^2\omega^2(3\omega^2+5\varepsilon^2) + A^4(\varepsilon^2+12\omega^2)+8A^6]\}^{-1}, \quad (14)$$

where $A = \gamma H_1/\sqrt{2}$, γ is the gyromagnetic factor, H_1 is the intensity of the alternating magnetic field with frequency Ω; $\omega = \Omega - 0.5\omega_{+1,-1}$, $\epsilon = \omega_0 - 0.5(\omega_1 + \omega_{-1})$ (see Fig. 13). A comparison of these expressions shows that whereas the signal in the case of beats breaks up into three simplest resonance, the double resonance signal

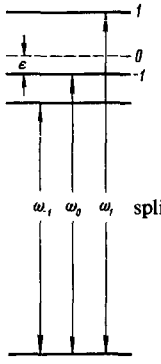

FIG. 13. Four-level scheme with non-equidistant splitting of the upper state.

is so complicated that it can hardly be analyzed. If we compare different modifications of the beats, then the most convenient from the methodological point of view is parametric resonance, which combines the methodological advantages of double resonance with the general advantages of the beat methods. Parametric resonance, however, is convenient only at low level splittings (on the order of 1 MHz and lower), for when the splitting is increased the requirements concerning the power of the alternating field and its homogeneity become more stringent. Parametric resonance was used successfully in experiments in which unprecidentedly weak magnetic fields were measured[56].

The most universal of all methods of interference of nondegenerate states is apparently the beat resonance. Its use entails modulation of the excitation at high frequencies, which frequently is a difficult technical problem. Nonetheless, this method has the advantage of experimental purity and maximum simplicity of the connection between the experimental results and the measured parameters. In addition to numerous investigations in which beat resonance was studied as an independent phenomenon, it was successfully used as an applied method in[35a] to measure the cross sections of the interaction of the 5^3P_1 state of cadmium with inert gases. A variant of beat resonance in accordance with the idea of Series[1c] was used in[36b] to determine the Lamb shift of a helium-ion excited state. The beat resonance, developed and first used in optics as a research method, was extended to nuclear spectroscopy. The possibility of using the method in nuclear physics was indicated in[37]. In[38], the beat resonance (called there the stroboscopic method of observing the nuclear Larmor precession) was used in practice to investigate the nuclear excited state of Ge^{69} and to investigate relaxation of oriented nuclei in solid germanium. Beat resonance turned out to be particularly convenient for the investigation of long-lived nuclear states.

Returning to optics, we note that the technical difficulties of realizing excitation modulation at high frequencies, as well as difficulty in the reception of high-frequency signals, are responsible for the tendency to reduce, where possible, a nondegenerate energy structure to a degenerate one. This makes it possible to use the level crossing method, which, being also an interference method, has the advantages of the beat method in conjunction with technical simplicity. Such a change in the measurement problem is not always possible, however, not all the levels can be made to cross in ex-

[10] Formula (14) of the journal version of [55] contains misprints.

ternal fields, in addition, the research problem itself can exclude such a formulation (for example, when the beat methods are used to measure a magnetic field).

By way of example, we can point to a special case when the method of beats is preferable, in spite of the fact that the same information concerning the level width can be obtained by the Hanle method—level crossing in a zero magnetic field. Such a situation arises in measurements of widths of very narrow levels, for which the crossing takes place at such weak fields, that the problem of exact cancellation of the laboratory fields comes into play. The transition to relatively large splittings eliminates this difficulty, since addition of a sufficiently strong controlled magnetic field causes a contribution to the total intensity to be made only by the coaxial component of the random laboratory field. This component can be easily taken into account by reversing the sign of the controlled field.

A method that promises to be particularly valuable is the nonlinear modification of the beat resonance, or the mode crossing method[48,52,58]. Methodologically it is much simpler: there is no need for high-speed modulation of the excitation, since the modulation is produced in the laser source automatically as a result of the intermode beats. In addition, the resonance is registered by using the integral change of the transparency of the investigated medium, so that there is no need for an inertialess photoreceiver. Different variants of the method already exist. In[48b], the investigated medium was simultaneously the working medium of the laser and was placed in a resonator. In[52], the laser played a role of only the excitation source. In[58], beats in the radiation of two tunable CO_2 single-mode lasers were used to investigate the dipole moment of the CH_3F molecule. It can be assumed that with further development in laser technology this spectroscopy method will be used more and more extensively.

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