

Superhigh-resolution spectroscopy based on interference of states

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We describe the current methods of superhigh-resolution spectroscopy based on interference of atomic states (quantum beats and level crossing) and compare them with the methods of Doppler-free laser spectroscopy. We pay major attention to the quantum-beat method, which has been developed in recent years in connection with progress in the technique of tunable lasers, and also to the phenomenon of alignment of atoms in a gas-discharge plasma. We treat examples of application of these methods for solving problems of superhigh-resolution spectroscopy.

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The ideas and methods of interference of states were intensively developed and implanted in atomic spectroscopy in the sixties. They allowed people to study the structure of energy levels at a resolution not limited by the Doppler broadening of spectral lines. The flourishing of studies on interference of states coincided in time with the rise of a new type of spectroscopy: superhigh-resolution nonlinear laser spectroscopy. Recent years have seen intensive technical progress in the field of tunable lasers; these promise to convert laser spectroscopy into a universal, all-encompassing method. In this regard the question arises of the competitive status of the former methods and of their rational field of application. This review will briefly summarize the fundamental information on interference methods of spectroscopy and their development in the laser period; we shall correlate the discussed methods with the problems and advances of laser spectroscopy.

1. INTERFERENCE OF ATOMIC STATES

Interference phenomena are based on the quantum-mechanical principle of addition of the probability amplitudes for transition of a system from an initial to a final state via a set of intermediate states when the specific transition pathway is not fixed by experiment.¹

Let an atom in the initial state Ψ_0 undergo a perturbation that can take it into either of two (for the sake of simplicity) intermediate states Ψ_1 and Ψ_2 . In

this case we have the superposition state Ψ , which can be described by the summation: $\Psi = C_1\Psi_1 e^{-i\omega_1 t} + C_2\Psi_2 e^{-i\omega_2 t}$. Here we have written out the temporal phase factors, which correspond to the energies E_1 and E_2 of the states Ψ_1 and Ψ_2 , and differ in the general case. A transition from the state Ψ to the final state Ψ_3 under the actions of the perturbation V leads to interference of the probability amplitudes. That is, the probability of transition to the state Ψ_3 , which is proportional to $|\langle\Psi_3|V|\Psi\rangle|^2$, contains three terms:

$$P(t) = |C_1|^2 |\langle\Psi_3|\hat{V}|\Psi_1\rangle|^2 + |C_2|^2 |\langle\Psi_3|\hat{V}|\Psi_2\rangle|^2 + 2 \operatorname{Re} C_1^* C_2 \langle\Psi_3|\hat{V}|\Psi_1\rangle \langle\Psi_3|\hat{V}|\Psi_2\rangle^* e^{-i(\omega_1 - \omega_2)t} \quad (1)$$

The first two of these correspond to independent transitions from the states Ψ_1 and Ψ_2 to Ψ_3 , while the third reflects the interference between the states Ψ_1 and Ψ_2 .

Let us rewrite the expression (1) for the time-dependence of the probability of transition from the superposition Ψ to the state Ψ_3 in a compact form, while in addition allowing for the decay of the states Ψ_1 and Ψ_2 . Here we assume the decay constant Γ to be the same for both states:

$$P(t) = e^{-\Gamma t} [A + B \cos(\omega_1 t + \varphi)], \quad \omega_{12} = \omega_1 - \omega_2 = \frac{E_1 - E_2}{\hbar} \quad (2)$$

The transition probability beats in time with the frequency of transition between the interfering states. The appearance of beats in the spontaneous decay of a superposition state is treated classically as resulting from interference of the harmonics of the field at the frequencies ω_1 and ω_2 emitted by a single atom.

The interference component is remarkable in its dependence on the energy difference between the interfering states, which in principle enables us to measure it. This is precisely the approach used in the various modifications of the quantum-beat method. A second approach is based on time-integral observations of the intensity of absorption or luminescence involving a superposition state. In an integral observation, the interference component of the transition probability averages to zero with the single exception of the case of equal energies of the interfering states. That is, they become degenerate, and then this component is determined by the phase difference ϕ , which depends on the polarization of the light. Interference of degenerate states is the basis of the method of level crossing.²⁹

2. PHENOMENA AND METHODS OF QUANTUM BEATS

Equation (2) describes the beats of the probability of the elementary process. In order to transform to observable quantities (intensity of emission or absorption), we must average over a multitude of emitting or absorbing particles. Without adopting special measures, this averaging leads to disappearance of the beats, and to preserve them we must synchronize the particles in phase.²

The frequency of beating is the same for an ensemble of identical atoms. Since it is determined by the energy difference between the sublevels of the given frequency, the simultaneous shift of the frequencies ω_1 and ω_2 by virtue of the Doppler effect hardly alters their difference ω_{12} , so that the frequency of beating is practically unaffected by Doppler broadening. The initial phase ϕ of the beats depends on the nature of the excitation and the conditions of observation. If the interfering states differ in angular characteristics (angular/momentum and its projection), then a necessary condition for the initial phase of the beats to be determined is that the excitation and observation should be anisotropic. Finally one attains phase synchronization by overcoming the phase scatter involving the scatter of angular momenta of excitation. Beats arise in the most obvious way in pulsed excitation in a time shorter than the period of beating. This is the simplest manifestation of beats, which has often been employed recently for spectroscopic purposes.

The existence of beats in spontaneous emission upon pulsed excitation was first demonstrated in Ref. 3. In that case the beats arose from interference of sublevels of the magnetic splitting of the 3P_1 state of cadmium and mercury. Demonstration of beats involved overcoming considerable difficulties in ensuring a sufficiently short and powerful pulse of optical excitation.

Free beats are of considerable interest as a means of studying the structure and lifetime of excited states. A very essential point is that the studied state is subjected to no perturbation—the beats are observed after the excitation pulse has ceased, while no restriction is imposed on the power of the latter. However, the

technical difficulties of exciting free beats in the “pre-laser” period restricted the application of this method to spectroscopic studies of two types. The first is the study of beats upon pulsed excitation of atoms by electron impact. After the first demonstration studies⁴⁻⁶ that showed the possibility of coherent electron excitation, pulsed excitation by electrons was used for spectroscopic purposes in Ref. 7, which estimated the possibility of exact measurement of Landé factors with the example of excited states of helium. Figure 1 shows an example of beats in emission from helium in the transition $2^1P_1-4^1D_2$ ($\lambda = 492.2$ nm). Excitation was performed with an electron beam directed perpendicular to a magnetic field. The time distribution of the spontaneously emitted photons was obtained by the technique of photon counting with transformation of the delay time of the photoelectronic pulse into an amplitude, with subsequent multichannel analysis.

A second type of studies of free beats without using lasers has become widespread. It involves experiments on charge-transfer of fast ion beams in thin carbon foils (the “beam-foil” technique).⁸ An ion beam of velocity $\sim 10^8$ cm/s penetrates a thin carbon film of thickness of the order of 10^{-5} cm. Charge transfer by the foil produces a beam of atoms excited into an extensive set of states. Here one realizes superpositions of states belonging to the components of the hyperfine (HF) and fine structure. For hydrogen and hydrogen-like ions, superposition of states separated by the Lamb shift is also realized. The fast motion of the particles after an extremely brief excitation creates a natural time scan that allows a resolution exceeding 10^{-10} sec. By this method people have measured both the splitting caused by intraatomic interactions (fine and hyperfine splitting) and that caused by external magnetic and electric fields.

The free-beat method has become intensively applied in spectroscopy in recent years in combination with laser excitation. This involves the progress in the field of tunable lasers. Most often people use dye lasers for this purpose pumped by a pulsed ultraviolet nitrogen laser. Thus one can easily produce high-power pulses of duration of the order of a nanosecond. Laser excitation of beats was first demonstrated in Ref. 9, where they observed beats in the magnetic structure of the $6s6p^3P_1$ state of ytterbium. The same sort of

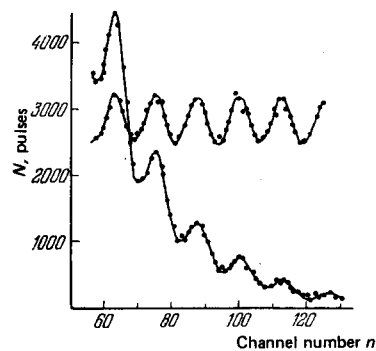


FIG. 1. Beats in the emission of helium in the transition $2^1P_1-4^1D_2$ ($\lambda = 492.2$ nm).⁷

beats has been observed in the 6^1P_1 state of barium and calcium.⁴⁶ Analogously, beats have been produced in the hyperfine structure of $7^2P_{3/2}$ of cesium by direct laser excitation. Spontaneous emission in the 4555-Å line with transition to the ground state $6^2S_{1/2}$ revealed an entire spectrum of beats at frequencies from 66.5 MHz to 149.4 MHz corresponding to interference of hyperfine sublevels differing in total angular momentum by 1 and 2.¹⁰

Beats have been observed^{11, 12} with two-step pulsed laser excitation. They are caused by interference of fine-structure components of the series $nD_{3/2, 5/2}$ of sodium for n ranging from 9 to 16. The first laser pulse excited the $3P$ state, from which the second pulse excited a superposition of the $nD_{3/2}$ and $nD_{5/2}$ levels. The beats were observed in spontaneous emission to the $3P$ level. Figure 2a shows the form of the beat signals that arise as recorded by the method of accumulation over a large number of excitation pulses. Figure 2b shows the Fourier spectra of some of these signals, which enable one to determine the beat frequency accurately. In these experiments the time resolution was restricted by the speed of action of the photomultiplier and corresponded to a limiting frequency of 150 MHz. Potentially the technique of recording beats in absorption demonstrated in Ref. 13 allowed much greater resolution. Here they observed interference of the hyperfine sublevels of the $3P_{1/2}$ state of sodium. The superposition of states was created by a laser exciting pulse, while the beats were detected by a second laser pulse that was arbitrarily shifted in time with respect to the first one, and which excited the atoms from the $3P_{1/2}$ to the $2^2S_{1/2}$ state. The production of the latter was detected by the ionization of the atoms in an external electric field. The probability of excitation of the atoms

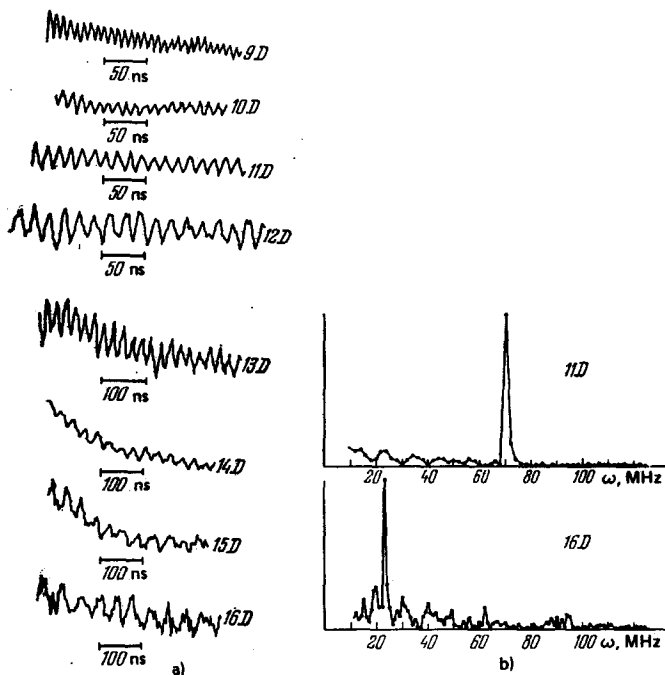


FIG. 2. a) Beats in the emission from the $nD_{3/2, 5/2} - 3P$ transitions of sodium¹²; b) Fourier spectrum of signals from a).

by the second pulse was subject to beating at the frequency of the hyperfine splitting at 192 MHz. This was detected by varying the delay of the second pulse. Here the time resolution was governed by the duration of the laser pulses.

Quantum beats have been observed¹⁴ in the light-echo and nutation signals upon laser excitation of molecules. These types of beats can no longer be termed free, since here the individual emitters exert a mutual influence, just as in Ref. 15, where beats were observed in cesium vapor in superluminescence. However, the merit of observing beats in a coherent emission processes is the high sensitivity of emission, which is methodologically essential in the long/wavelength region.

For the sake of completeness we shall point out a laser modification of beat spectroscopy in beams of atoms and ions.¹⁶ In these experiments, a fast beam was excited by laser radiation, rather than by charge transfer. The high beam velocity allows one to tune the effective frequency of the laser over a rather broad range through the Doppler effect by varying the angle of encounter of the particle beam and the light beam. Just as in the beam-foil technique, the beats are analyzed in terms of the emission from the particle beam as a function of the distance from the point of excitation. Figure 3 shows a diagram of the beats and the Fourier spectrum which is taken from Ref. 8, and which pertains to the laser excitation of a beam of barium ions in the transition $6^2S_{1/2} - 6^2P_{3/2}$.

In pulsed excitation, beats arise as a transition characteristic of the system. Other situations can occur in which beats are manifested in the form of stationary processes, to produce the so-called resonance beats. The most obvious one is amplitude resonance arising upon harmonic modulation of the excitation intensity. The phenomenon consists in a resonance increase in the depth of modulation of the spontaneous emission as the modulation frequency of

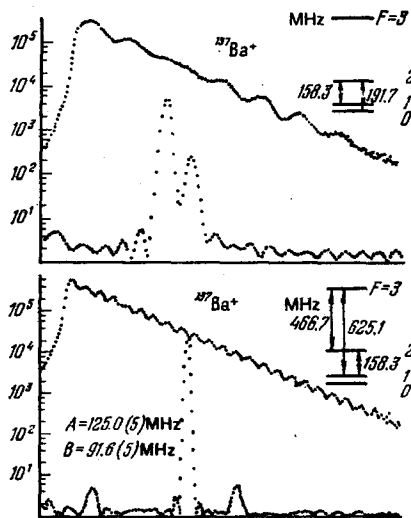


FIG. 3. Beats and Fourier spectrum of the emission of barium ions in the transition $6^2S_{1/2} - 6^2P_{3/2}$.⁸

the excitation approaches the free-beat frequency. The width of the resonance is determined by the width of the interfering levels. The phenomenon is not trivial in nature: the resonance frequency can exceed by an arbitrarily great factor the reciprocal of the lifetime of spontaneous emission, which is completely demodulated outside resonance owing to its time lag. We can understand the onset of resonance by examining the time grouping of the elementary emission events with account for the varying excitation intensity. Yet it is useful to point out another, spectral approach to explaining the formation of a resonance. The atom having a split upper level is modeled by a paired oscillator with a fixed frequency difference. When it is acted on by broad-spectrum radiation, both oscillators are excited with random relative phases, and summation over their ensemble does not lead to collective effects. In amplitude-modulated light, each harmonic of the spectrum acquires a pair coherent with it, although in sum the radiation remains spectrally "white". If the interval between the coherent harmonics coincides with the frequency splitting of the paired oscillator, both of its components are excited coherently, i.e., with the very same phase difference for all oscillators. Thus one can pass through resonance by varying the frequency of modulation of the light. One can say that the white light after harmonic modulation has acquired coherent properties. The degree of this coherence is determined by the near-zero width of the spectrum of the radiofrequency generator controlling the modulation of the light. It is precisely the width of this spectrum that determines the apparatus width of the method of amplitude beat resonance, which has practically unlimited resolution.

Amplitude resonance was first detected in luminescence in Refs. 17 and 18. Subsequently this method has been repeatedly applied in spectroscopic studies as a substitute for double radiooptical resonance. Beat resonance has an important advantage over the latter: the absence of the extra perturbation introduced into the studied system by the radiofrequency field, which gives rise to magnetic resonance. In addition to the substantial complication in deciphering the results in the double-resonance method, difficulties arise in connection with the need to provide enough power in the extra field, especially when one needs to employ a considerable splitting of levels having great breadth. Moreover, states can interfere to yield beats when the transitions between them approximation are generally forbidden in the one-photon dipole.

The invention of lasers has expanded the potentialities of amplitude beat resonance by easing the fundamental methodological difficulty of the method: the need for high-frequency modulation of the excitation. The most natural way to realize high-frequency modulation of laser radiation is to employ intermode beats. Beat resonance using a synchronized-mode laser has been observed in the $3s_2-2p_4$ transition of neon.²⁰ Narrow laser beams are easily modulated with electrooptic and electroacoustic modulators, and also by specific methods. An example of a "classical" application of beat resonance combined with laser ex-

citation is a study¹⁹ on determination of the g -factors of the terms of molecular selenium.

Employment of lasers enables an important modification of the amplitude-resonance method, which is called the mode-crossing method.^{21, 22} Its difference from beat resonance consists only in the fact that one doesn't record the modulation of the spontaneous emission from the interfering levels, but the gross absorption of the exciting line. An account for the nonlinear interaction of the intense laser radiation with the medium shows that the gross absorption by the medium changes when the frequency of beating of the modes coincides with the frequency of the sought splitting.

Other stationary manifestations of beats (parametric and phase resonance of beats^{23, 24} and the recently discovered relaxation resonance^{25, 26}) have not become widespread in spectroscopic practice. Parametric beat resonance has found a special application in a technique for measuring magnetic fields.^{27, 28}

The astrophysical literature has discussed the possibility of observing quantum beats in cosmic media with the example of employing the excitation of forbidden lines of oxygen ions in the filaments of the Crab Nebula by the radiation of the pulsar NP 0531.⁴⁵

3. LEVEL CROSSING

Interference of states having the same energy is an important special case of beats of zero frequency that are not averaged out in a time-integral observation. This type of interference phenomena is manifested as a singularity in the intensity of emission or absorption of light by a system subjected to an external field while any of its energy levels is near degeneracy. In the simplest case involving alignment it has the form

$$I = I_0 + \frac{I_0}{1 + (\omega_{12}^2/\Gamma^2)}. \quad (3)$$

This is obtained by integrating Eq. (2) over the time with $\phi = 0$. The region of formation of a singularity is determined by the width Γ of the levels. Of course, not every level crossing leads to interference—this requires that the levels must be excited coherently from a common initial state.¹⁾

An extensive literature has been devoted to the effect of level crossing (see, e.g., the monograph²⁹ and the reviews^{30, 31}). Let us take up briefly only a comparison of the methods of beats and of level crossing and the most recent modifications of experiments on level crossing.

The beat methods enable study of the energy structure of a studied system in the absence of external perturbations. Conversely, level crossing is brought about by an external, regulated perturbation—most

¹⁾In order to allow interference, the excitation of the atomic states need not be simultaneous. Reference 50 treats hypothetical experiments having time-separated excitation of the different states of an atom.

often a magnetic field, sometimes an electric field, or both together. Hence the interpretation of level-crossing signals requires existence of prior information on the system, on the basis of which one must calculate the evolution of energy and the transition probabilities in external fields. Comparison of the calculated with the experimental data allows one to extract information on the sought parameters of the unperturbed system—usually the constants of the fine and hyperfine interaction are in question, or the constants of the Stark and Zeeman splittings and the widths of levels. Thus level crossing is fundamentally less informative than beats. Yet it is methodically substantially simpler, since the level-crossing method requires neither the technique of pulsed excitation nor velocity methods of measuring the emission. As for the need for *a priori* information (angular momenta, type of bonding, etc.), this exists in the overwhelming majority of cases. For example, for the very simple case of degeneracy of magnetic sublevels in zero magnetic field (the Hanle effect, which is a special case of level crossing), the experimental signal enables one to determine the product of the *g*-factor by the lifetime of the state, which is usually the subject of study, while the *g*-factor is determined by independent experiments that don't require ultrahigh spectral resolution.

As arbitrarily chosen examples of level-crossing signals in the more general case, Fig. 4 shows the signals of crossing of the sublevels of the $6^2P_{3/2}$ state of rubidium-87 in a magnetic field; the dotted line shows the evolution of the signal upon applying an extra electric field. This allows one to determine the Stark constant.³⁰

In individual cases a study by the level-crossing method does not pursue the deciphering of their structure, but the solution of the inverse problem of measuring the intensity of the external perturbation, e.g., in measuring ultraweak magnetic fields.^{27, 28} A sensitivity of the order of 10^{-10} Oe has been realized by this method.²⁸

The overwhelming fraction of the studies by the level-crossing method in the period of the sixties and seventies has been performed with the resonance lines of atomic spectra. Attempts to extend the method of interference of states to energy levels not connected with the ground or metastable states by optical transitions face difficulties in exciting them. Some studies have employed resonance irradiation to populate an energy level above the one being studied, with transfer of coherence to the latter in the spontaneous decay of the initially excited state.⁵³ Excitation by an electron

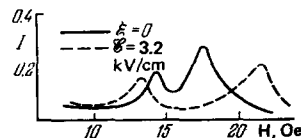


FIG. 4. Signals of crossing of the sublevels of $6^2P_{3/2}$ of ^{85}Rb in a magnetic field (solid curve) and the deformation of the signal by an imposed electric field $\mathcal{E} = 3.2$ kV/cm (dotted curve).³⁰

beam can easily yield occupation and coherence of the high-energy states. However, the dependence of the parameters of the exciting beam on the magnetic field and the coherent emission from a large number of states restrict the application of this method.

A significant forward step in extending the level-crossing method to highly excited states was the discovery of coherent population of the magnetic sublevels of excited atomic states directly in a gas discharge. The point in question is the so-called alignment and latent alignment of atoms in the discharge.³² As was found, the anisotropy of excitation necessary to create coherence occurs in any gas-discharge vessel of finite volume, and it mainly involves the anisotropy of the light fluxes within the light source. Upon isolating individual regions of the vessel for observation, people have been able upon applying a magnetic field, to detect the Hanle signals in the positive column of a discharge in all the inert gases,^{51, 52} in a dc discharge in mercury and cadmium vapor,⁴⁹ and in a hollow-cathode discharge in neon and helium.⁴⁸

The Hanle effect, which arises upon capture of radiation, opens up new potentialities of measuring magnetic fields in the Sun, and in recent years in astrophysics alignment has been intensively studied. In the solar crust and in protuberances the conditions of illumination are anisotropic, and this aligns the absorbing atoms. On the one hand, the magnetic field diminishes the degree of polarization of the radiation, and on the other hand, it changes its direction. Measurement of these two quantities is the source of information on the magnitude and direction of the magnetic field.⁴⁷

In addition to macroscopic alignment of atoms in isolated regions of a gas discharge, there is another process for onset of angular ordering of atomic states that doesn't involve directional fluxes of radiation. In particular, it occurs even in strictly isotropic irradiation. This process arises when the optical thickness of the medium is not too great, i.e., when the spectral lines of the discharge are not too greatly broadened in comparison with the Doppler width. Qualitatively, one can understand this process easily by transforming to a system of coordinates of an atom in random thermal motion. The moving atom, which is illuminated on all sides by the radiation from the other atoms, responds to it in various ways owing to the Doppler frequency shift. The radiation propagating to face the atom is blue-shifted in the system of coordinates of the latter. Therefore the probability that the atom will be excited by it is depressed in comparison with radiation illuminating the atom in a plane normal to its motion, since the latter radiation is in resonance with the atomic frequency. In precisely the same way, the efficiency of excitation by light overtaking the moving atom from the rear is depressed. Thus the atom is excited anisotropically with an axis of symmetry in the direction of motion. This causes the ensemble of moving atoms, while remaining isotropic on the average, to acquire a latent anisotropy with respect to the instantaneous velocity vector of

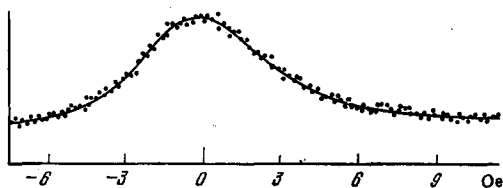


FIG. 5. Latent-alignment signal (dependence of intensity on the magnetic field) of the 3P_1 state of neon observed in the 607.4-nm line.

each atom. One can show upon accounting for capture of radiation that this anisotropy causes the population of the state and the associated emission intensity of the ensemble to depend on the intensity of the external field.²⁾³² As compared with ordinary Hanle signals, latent-alignment signals have a more complicated form:

$$I = I_0 + I' \left[\frac{1}{1 + (\omega_{\text{Lar}}^2 / \Gamma^2)} + \frac{1}{1 + (4\omega_{\text{Lar}}^2 / \Gamma^2)} \right]. \quad (4)$$

Here ω_{Lar} is the Larmor frequency. This relationship allows one to determine the width of states when the gyromagnetic ratios are known.⁵¹ Figure 5 shows a latent-alignment signal in the 607.4-nm line of neon, the initial state of which is spherically symmetric ($\gamma = 0$).

Application of lasers as excitation sources has also allowed people substantially to extend the set of objects accessible to the level-crossing method. Here one can apply stepwise excitation, which proves rather efficient at high laser intensities, or excitation of the studied state from a level that is being populated in a gas discharge. The Hanle effect was first observed in laser transitions in neon.³⁵ A substantial contribution to extension of the application of lasers was a proposal to employ pulsed laser excitation with time-integral observation.³⁶ This technique has been used to study a series of states of cadmium.³⁷ The perspectives of studies of this type are very broad in view of the large and ever-growing list of laser lines that can be generated in a pulsed regime. However, the application of lasers has led to complications involving perturbations of the system by the high-power light field and saturation of the studied transitions. In particular, crossing signals have acquired an additional structure involving the introduction into the signal of parameters of the initial state along with those of the studied state³⁸ (Fig. 6). On the other hand, the Hanle effect in laser levels is of interest as a means of studying an inverted medium and it is employed as one of the ways of controlling the types of oscillations of lasers.³⁹

In closing this section we point out that level-crossing signals often amount to some percent or tenths of a percent of the measured intensity, which *per se* may be slight. For efficient analysis of weak signals,

²⁾The change in population of a state having latent alignment in a discharge entails a change in the population of other levels also. Hence, a state-alignment signal arises in the emission of certain spectral lines that don't directly participate in this dipole transition.

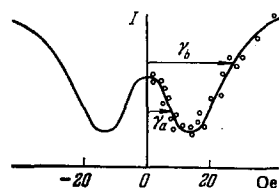


FIG. 6. Distortion of a level-crossing signal by nonlinear interactions with the laser radiation.³⁸

modern experiments employ balanced photodetection circuits, synchronized detection, and transformation from analog to digital form of the signal, which enables numerical methods of accumulation without time limit. The received signals are processed by computer in order to select parameters that allow optimal correspondence of the experimental data with the calculated curves.

4. COMPARISON OF THE METHODS OF INTERFERENCE OF STATES AND OF NONLINEAR LASER SPECTROSCOPY

The modern methods of nonlinear laser spectroscopy⁴⁰ enable one to measure the frequencies of optimal transitions without restrictions imposed by Doppler broadening of the spectral lines. If one knows the structure and width of a transition, then with additional information one can get the structure of an individual state and its width, e.g., as in the case of transitions to a studied excited state from the ground state, whose width one can neglect. Thus the methods of laser spectroscopy are universal. Under favorable conditions they allow one to solve both problems accessible to interference methods and broader problems—absolute measurement of frequencies of independent quantum systems (e.g., analysis of isotopic composition). However, in the overlap region of the problems of interference and laser methods, i.e., in superhigh-resolution studies of the structure and widths of states, the interference methods may prove preferable.

a) The methods of interference spectroscopy yield direct information pertaining to the given state. In order to decipher the structure of a state from data of laser spectroscopy, one generally³¹ needs information on the structure of the second state entering into the transition.

b) The nonlinear methods of laser spectroscopy require an additional account for the perturbation introduced by the laser into the studied system, which can be considerable. The interference methods are fundamentally linear. Their laser variants can become complicated by light perturbation, but at least the pulsed beat method is completely free of introduced perturbations. We note that the study of light perturbation can be of independent interest. Interference methods can also be utilized for such studies.

In addition to the complications involving decipherment of the results, the nonlinearity of the methods of

³¹⁾In certain special experimental designs in which interference effects again play the essential role, one can obtain signals that stem from the characteristics of a single state.

laser spectroscopy can be a source of errors in analyzing a weak transition in the vicinity of a strong one. In this situation the interference methods are preferable. This problem has been discussed in Ref. 14, where beats have been used to determine the frequency of the hyperfine splitting of an excited state of the molecule CH_3D , which had not been detected previously by the saturated-absorption method. The matrix elements d_{i0} of the transition from the split state $|i\rangle$ to the ground state $|0\rangle$ differ by a factor of about five, while the ratio of the saturation parameters of the two transitions, which is proportional to $(d_{i0}/d_{i'0})^4$, amounted to about 500 in the studied example. Therefore, at the power needed to saturate the weak line, the broadening of the strong line would be so great that it would conceal the sought structure. At the same time, the beat signal is proportional to the product $d_{i0} \cdot d_{i'0}$, so that the strong transition "aids" the detection of the weak one.

c) Methodologically the interference measurements are usually much simpler. Here it is natural to compare the experiments that are closest in methodology of experiment: laser spectroscopy and laser-excited beats. As was mentioned in Refs. 11 and 12, the fine splitting of cesium levels has been measured by beat methods in the series $nD_{3/2,5/2}$ for $n=9$ to 16. An analogous study has been performed by two-photon spectroscopy for $n=3$ to 8.^{41,42} The two methods impose quite different requirements on the spectrum and emission stability of the lasers. For two-photon spectroscopy, the width of the laser spectrum determines the resolution of the method. In Refs. 41 and 42 this was brought down to 10 MHz. In combination with the requirement on stability and possible tuning of the frequency over several GHz, this is a complicated technical problem. For laser excitation of beats, the only requirement is overlap of the spectrum of the laser pulse with the absorption line of cesium. The laser spectrum had a width of about a gigahertz, while frequency scanning was not needed. The lack of a need for high monochromaticity and stability of spectrum allows one to employ the entire stock of pulsed lasers, and primarily the very versatile dye lasers excited by a nitrogen laser. These lasers team very well with the pulsed beat method, which requires high-power short-period excitation.

d) The limiting spectral resolution attainable in laser spectroscopy is limited by the width of the laser spectrum, which plays the role of the apparatus width of the method. In some laser spectrographs, based on a gas laser tunable in frequency within the narrow limits of the width of the working line, one can reduce the width of the spectrum down to tens of hertz.⁵⁷ Yet it is hard in practice to make the spectrum narrower than 1 MHz for the dye lasers tunable over a broad range that constitute the modern basis for laser spectroscopy.

Upon turning to the resolution of the interference methods, we should speak separately of the interference methods for degenerate and nondegenerate states. For the former, i.e., the methods of crossing and anti-

crossing of levels, the only restriction on the resolution is imposed by the system for creating and controlling the external fields. For example, inhomogeneity of the field broadens the signal. This type of restriction depends on the field intensities, and thus on the energy interval to be measured. It is hard in practice to ensure a relative homogeneity of the field in the working volume better than 10^{-4} – 10^{-5} (more difficult in regions of high intensity). This gives an estimate of the relative resolving power.

For the beat methods, the restrictions on the resolving power are established by the Doppler broadening of the frequency of the interval to be measured, which has the relative value of \bar{v}/c , where \bar{v} is the relative velocity of the thermal motion of the particles, i.e., a value of the order of 10^{-6} times the frequency to be measured.⁴¹

Thus, in contrast to the methods of laser spectroscopy, the resolution of the interference methods depends on the size of the energy interval to be measured. The resolution proves to be very great for small intervals. Thus, in the above-cited measurements of supersmall magnetic fields, the splitting of the levels of the ground state of rubidium was fixed with an accuracy of the order of 10^{-4} Hz.

The above-said pertains to the comparison of the limiting attainable accuracies. If one doesn't pose the problem of realizing them (e.g., in studying rather broad excited states), then a comparison of the accuracies attainable by the different methods (see Refs. 43,44) is not especially indicative. This is because these accuracies primarily depend on the investments in experimental technique. In this regard we can compare the above-cited Refs. 11, 12, and 41, 42. At the compared general level of technique of experimentation, the accuracy of the measurements by the beat method was ten times as great as by the method of two-photon spectroscopy.

Thus there is every ground for asserting that it is preferable to study problems of measuring the structure of states by the methods of interference of states. These methods are especially appropriate in the region of small splittings (0 – 10^8 Hz), which doesn't impose especially severe requirements on the speed of action of the apparatus (in the beat method) or on the values and homogeneity of the external fields (in the level-crossing). There are also specific problems solvable only by interference methods. These are problems of measuring the constants of collisional relaxation of orientation, alignment, etc., of states possessing angular momentum. At the same time, as we have pointed out, there is a broad class of problems that can be solved only by methods of nonlinear laser spectroscopy.

As we see, the two fields of superhigh-resolution spectroscopy (nonlinear laser and interference) do not

⁴¹The restrictions of accuracy of measurement of radiofrequencies for commercial apparatus lie in a range of much smaller values, $\sim 10^{-6}$, and hence can be neglected.

so much compete with as supplement one another. We also point out that interference effects have been playing an ever greater role in recent years in nonlinear spectroscopy,⁵⁴⁻⁵⁶ while people have had to take account of the role of nonlinear processes ever more often in the methods involving interference of states in connection with applications of lasers.^{35,38}

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