

CROSSING AND ANTICROSSING OF ATOMIC LEVELS AND THEIR USE IN ATOMIC SPECTROSCOPY

V. G. POKOZAN'EV and G. V. SKROTSKIĬ

Ural Polytechnic Institute, Sverdlovsk; Moscow Physico-technical Institute

Usp. Fiz. Nauk 107, 623-656 (August, 1972)

The phenomena discussed can arise in sufficiently rarefied vapors of atoms at definite values of the external magnetic field. They are accompanied by interference of the corresponding atomic states, and this affects noticeably the character of the polarization and the angular distribution of the resonant emission of the atoms. The study of the microstructure of the resonant emission of atoms situated in an external constant field and a radiofrequency field can yield valuable information on the structure of the atomic states, on the character of the interatomic interactions, and on the phenomena associated with them. New methods greatly extend the capabilities of the interference technique and have an exceedingly high resolving power (which can amount to a fraction of a hertz for certain gas systems). The phenomena in question have found various scientific and technical applications, and serve in particular as a basis for the operation of high-sensitivity magnetometers, which make it possible to perform absolute measurements of ultraweak magnetic fields (to 10^{-11} Oe). The physical principles of the phenomena are considered, a theory is developed for them, and certain possible scientific and technical applications are indicated. The bibliography covers all the principal papers published before 1972.

CONTENTS

1. Introduction	452
2. Interference of States in Crossing of Energy Levels	453
3. Crossing of Excited-State Atomic Levels	455
4. Anticrossings of Atomic Levels	464
5. Crossing of Levels in the Ground State	467
6. Bibliography	469

1. INTRODUCTION

THE positions of the Zeeman sublevels of the excited state of an atom situated in external magnetic fields depend on the intensity of this field. At definite values of the field, crossing of the sublevels can take place and be accompanied by interference of the corresponding states. This circumstance affects noticeably the character of the polarization and the angular distribution of the resonant emission of the atoms. The influence of a magnetic field on the polarization of resonant emission of atoms was observed by Hanle back in 1923^[1]. It was soon explained^[2] on the basis of the Lorentz classical theory. A more rigorous theory was developed by Breit^[3b]. Hanle established already in the first experiments that the polarization of resonant emission is exceedingly sensitive to the influence of weak magnetic fields, weaker than that of the earth. This phenomenon, which is a particular case of level crossing in a zero field, is called the Hanle effect. For a long time it attracted the attention of many workers in the field of optical spectroscopy, and was extensively used to determine the lifetimes of excited states. It is the subject of an extensive literature (see, for example, ^[4]).

In 1959, Colegrove et al.^[5] again renewed interest in this phenomenon, using the level-crossing effect in a nonzero constant magnetic field to investigate the fine structure of the 2^3P excited state of helium, and proposing by the same token a new spectroscopic

method for the study of excited atomic states and for the measurement of the corresponding atomic and nuclear constants. Its limiting resolution was determined by the natural width of the emitting term and, what is particularly important, did not depend on the thermal motion of the atoms that leads to Doppler broadening of resonance fluorescence.

Investigations of atomic systems by the level crossing method have made it possible not only to obtain new very valuable information on the structure of atomic states, but also to observe many new phenomena, as well as to extend greatly the capabilities of the interference technique. Thus, methods of "anticrossing" of levels^[6,7,8a,8c] and methods of crossing of magnetic sublevels of atoms in the ground state^[9,10] came into being. An important role in the theoretical and experimental development of these methods was played by the work of Kastler^[11] and Podgoretskiĭ^[8a,b,c].

The crossing and anticrossing energy levels arise not only in atomic systems, but can also appear in the study of molecular spectra and of the properties of atomic nuclei and elementary particles, as first pointed out by Podgoretskiĭ^[8a,b,c,12]. Their classical analogs have been known for a long time and are widely used in the theory of oscillations^[13], and are also realized in energy spectra of solids of various kinds.

Investigations of the crossing of atomic sublevels of the ground state have by now become particularly timely, owing to the unusually large resolving power of this method, determined by the lifetime of the ground

state. For certain gas systems it can amount to a fraction of a hertz. Therefore the crossing of sublevels of the ground state (particularly in zero magnetic fields) uncovers ways not only of a thorough study of the character of intraatomic interactions and the phenomena associated with them, but also for the solution of a number of important applied problems. In particular, it has served as a basis for the development of high-sensitivity magnetometric apparatus that makes it possible to measure ultraweak magnetic fields of the order of 10^{-9} Oe^[14].

It is of interest to compare the possibilities of the level crossing method with another presently well-known method based on a phenomenon in which features of both optical and radio-frequency (RF) spectroscopy are manifest, namely the method of double radio-optical resonance^[7, 15a]. It yields information on the level structure in the case when the Zeeman or the hyperfine splitting is larger, comparable with, or smaller than the natural width of the investigated state. Unfortunately, the RF field in which the system is placed shifts the atomic sublevels by an amount proportional to the square of the amplitude of the field, and broadens these levels. For the practice of spectroscopy, this phenomenon is highly undesirable, especially if it is recognized that an analytic solution of the double-resonance problem is known only for a limited number of simplest situations. The existing theory of radiooptical resonance makes it possible to estimate quantitatively the effect produced by the RF field, and to eliminate it. In addition, the amplitude of the field is connected in definite manner with the lifetime of the state: the shorter the lifetime, the larger should be the amplitude of the RF field. The latter circumstance, in particular, makes it difficult to use the method of radio-optical resonance for investigations of the short-lived atomic states. This, however, does not pertain to the crossing method.

The simplicity and reliability of the experimental devices used to observe this phenomenon, the high accuracy, which is not to the accuracy of the radiooptical resonance, make the crossing method quite valuable in atomic spectroscopy. Since in many cases the fields of application of both methods do not overlap, their conjunction can greatly increase the capability of each of them individually.

In the present review, principal attention is paid to the physical picture of the phenomena that accompany the crossing and anticrossing of the levels. These phenomena serve as a basis for the corresponding spectroscopic methods. The authors did not undertake to gather and systematize all the experimental material available on this question. If experimental results are being cited, it is only to illustrate the possibilities of the method in question, or else to confirm a definite physical concept.

2. INTERFERENCE OF STATES IN CROSSING OF ENERGY LEVELS

It was shown theoretically and confirmed experimentally that the crossing of atomic magnetic sublevels is accompanied by a change in angular distribution of the radiation. This change is due to the interference of

the corresponding atomic states^[16]. The explanation of the interference phenomenon is based on one of the fundamental premises of quantum mechanics, namely the principle of superposition of states. As is well known, this principle leads in a quite natural fashion to the concept of interference of atomic states, and also to the choice of physical conditions for its experimental observation. It is therefore advantageous to dwell briefly on the main consequences of this principle as applied to a circle of the problems touched upon here.

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

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

where the expansion coefficients $a_m(t)$, generally complex numbers, are characterized by moduli and phase shifts. We shall henceforth choose the energy operator for this purpose. The mean value of the operator \hat{g} in the state $|\psi(t)\rangle$ is defined by the expression

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

where $g_{mn} = \langle \varphi_m | \hat{g} | \varphi_n \rangle$ is a matrix element of the operator \hat{g} . If this operator commutes with the energy operator, the mean value of \hat{g} is determined only by the first term of (2), and the second interference term, which depends on the ratio of the phases of the coefficients, vanishes. On the other hand, if the operators \hat{E} and \hat{g} do not commute, then the expression for the mean value includes besides the first term, also an interference term whose value depends on the phase difference between the coefficients a_m and a_n in the states $|\varphi_m\rangle$ and $|\varphi_n\rangle$, i.e., in the general case on $\varphi = (E_m - E_n)t/\hbar$, accurate to within an initial phase shift. In this case, the mean value \bar{g} is a periodic function of the time and varies at the frequency of the transitions between the interfering states. To calculate an experimentally measured mean value of a physical quantity $\langle \bar{g} \rangle$, relation (2) must be averaged over a statistical ensemble, which we assume to consist of N independent systems. Then

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

where the subscript i numbers the atoms and $\hat{\rho}$ is the density matrix for the ensemble of the atoms. In accordance with the definition, its elements are given by

$$\rho_{mn} = N^{-1} \sum_{i=1}^N \rho_{mn}^i, \quad (4)$$

where $\rho_{mn}^i = a_m^i a_n^{i*}$ are the elements of the density matrix of the i -th atom.

The presence of an interference term in (2), which is equivalent to stating that $\rho_{mn} \neq 0$ when $n \neq m$, still does not mean that this term is contained in expression (3). Its appearance in (3) depends also on the results of averaging the density matrix (4) over the statistical ensemble. In this case one can encounter two physically real cases. The first is realized when the ensemble of the atoms is prepared in such a way that the phase shifts φ_i of the matrix elements ρ_{mn}^i which enter in (4) are equally probable. For a suffic-

iently large N , this leads to the vanishing of the resultant off-diagonal matrix element ρ_{mn} . The density matrix is then diagonal, and one can observe only these system properties which are possessed by the atoms in pure states. If the phase shifts φ_i are distributed over the atoms non-uniformly, then the elements ρ_{mn} differ from zero and the observed physical quantity experiences beats with a frequency determined by the energy difference between the interfering states. One says in this case that coherence exists between the states $|\varphi_m\rangle$ and $|\varphi_n\rangle$. This coherence leads to a macroscopically observable interference effect, and the expansion (1) is a coherent superposition of the atomic states.

Thus, to observe experimentally the interference of atomic states in pure form, the system must be constructed in such a way that the phase difference between the interfering states is the same for all the atoms of the ensemble, or in other words, that each atom be in a coherent superposition of states.

There are many known methods of producing coherence. All involve the use of some external perturbation. These methods are well known in the fields of magnetic and radio-optical resonance, optical orientation of atoms, etc. The most common is the use of an RF field whose frequency is close to the frequency of the Zeeman splitting of the atoms oriented by an external constant magnetic field H_0 (magnetic resonance). The interaction of the atoms with the RF field under magnetic-resonance conditions leads to stimulated oscillations of the atomic states having the phase of field, thereby excluding their uniform distribution. The interference of the states becomes manifest in this case in the appearance of a macroscopic magnetization component perpendicular to H_0 and varying with the frequency of the RF field. It can be shown that this result follows from (3) if the basis vectors in the latter are taken to be the eigenvectors of the operator of the z-component of the total angular momentum J of the atomic system, and \hat{g} is taken to be the operator of a transverse component of the angular momentum J_x or J_y , since $[J_x, J_y] \neq 0$.

This example leads to the following interesting conclusion: In the case under consideration, there is a complete one-to-one correspondence between the appearance of a magnetization component that is transverse with respect to the constancy of H_0 , on the one hand, and the coherence ρ_{mn} on the other. One can speak of a magnetization that is transverse with the magnetic field and take it to mean a coherent superposition of states, and vice versa. If we consider other examples of coherence production, we can arrive at the more general conclusion that introduction of coherence into an atomic system is accompanied by the onset of atomic orientation, i.e., polarization or alignment. In the latter case no magnetization is produced. If we use this circumstance, it is easy to present a qualitative description of the interference effect that results from the interaction of an atomic system with a "phasing" perturbation, and, in particular, with optical excitation that makes it possible to observe the crossing effect. In such a formulation, the phenomenon considered here has a direct bearing on a phenomenon already known in physics, namely optical orientation of atoms, the gist

of which is that when radiation interacts with atoms the latter can acquire a fraction of the angular momentum of the radiation field (within the limits of the angular-momentum conservation law), and this changes the total angular momentum J of the atomic system. The foregoing can be formulated in terms of magnetization, using the well-known relation $\mathbf{M} = \gamma\mathbf{J}$.

Knowing what changes occur in the angular state of the atoms when they interact with light, we can find the conditions under which a coherent superposition of atomic states is produced.

Let us consider an atomic system in whose excited state two magnetic sublevels m and m' ($m \neq m'$) are characterized by an energy difference $\Delta E = E_m - E_{m'}$, determined by a constant magnetic field H_0 . We assume that, depending on the field, ΔE can vary from zero (level crossing) to a value exceeding the natural width of the magnetic sublevels. To excite atoms from a definite sublevel μ of the ground state into a superposition state, it is necessary that the incident light be represented by a superposition of components with σ^+ , σ^- , and π polarizations. Such an excitation is referred to customarily as coherent or possessing coherent polarization. If the atoms are excited in pure states when they absorb light, such an excitation is called incoherent. Obviously, the concept of coherent or incoherent excitation is relative and is meaningful only with respect to the chosen direction of the quantization axis. For example, circularly polarized light propagating in the direction H_0 constitutes an incoherent excitation if the H_0 direction is chosen to be the quantization direction. In this case the atoms are excited only in pure states. The same radiation, when propagating in a direction perpendicular to H_0 , excites atoms in a superposition state, and the magnetization quantum numbers of the ground and excited states differ then by 0 and ± 1 .

The direction of the constant magnetic field, the direction of the oscillations of the magnetic vector of a linearly polarized wave, and the direction in which the atomic ensemble is irradiated in the case of circular polarization, are all physically singled-out quantization directions. Preference to any one of them is dictated, of course, only by considerations of convenience in solving the particular problem. We assume in the example that follows that the quantization axis is in the direction of propagation of circularly polarized light (the Ox axis in Fig. 1a), and that it is perpendicular to the vector $H_0 = H_z$. The magnetic sublevels M and M' , defined relative to the Ox axis, are incoherently excited by the incident light. Since $M \neq M'$ and the probabilities of the optical transitions to these sublevels are not the same, a definite difference arises between the populations of the latter, and leads to the appear-

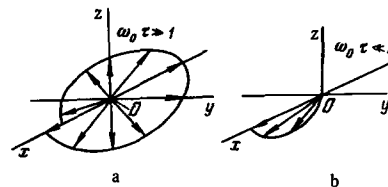


FIG. 1. Magnitude and position of the magnetization vector during one Larmor period.

ance of macroscopic magnetization of the excited atoms in the irradiation direction (optical orientation of the atoms in the Ox direction). Since the system is in a magnetic field, the magnetization produced in the Ox direction begins to precess with frequency $\omega_0 = \gamma H_0 = \Delta E/\hbar$ about the direction of H_0 and by the instant t it negotiates an angle $\omega_0 t$. By virtue of the radiative spontaneous decay of the excited state, this magnetization decreases with time and has at the instant of time t the value $\exp(-t/\tau)$, where τ is the lifetime of the excited atom and is connected with the natural width Γ by the relation $\tau\Gamma = 1$. If the magnetization goes through a large number of cycles during the lifetime τ , i.e., $\omega_0\tau \gg 1$, then its mean value taken over this time interval is equal to zero. In other words, if the distance between the levels $\Delta E/\hbar$ is much larger than the natural width of the levels, no transverse magnetization relative to the magnetic field takes place. Consequently, under these conditions optical excitation does not introduce coherence into the atomic system.

When the precession frequency ω_0 is small, so that the condition $\omega_0\tau \ll 1$ is satisfied, the magnetization produced by the light decays without hardly beginning to precess about the direction of H_0 (Fig. 1b). In the case of continuous optical excitation this produces in the Ox direction a macroscopic magnetization (polarization of the atoms in the Ox direction), meaning that the atoms are excited into a coherent superposition of states $|\varphi_m\rangle$ and $|\varphi_{m'}\rangle$.

Thus, optical excitation can introduce coherence into an atomic system if (a) the distance between the energy levels is comparable with or smaller than their natural width, and (b) the optical excitation is coherent. These are precisely the conditions under which experiment can reveal the crossing of atomic magnetic sublevels.

Besides crossing, it is possible to produce in atomic systems conditions under which the levels approach each other to within a distance shorter than or of the order of their width, but without crossing. In atomic spectroscopy, this interesting phenomenon was revealed quite recently by a change in the angular distribution of light in the "anticrossing" region. The conditions for the occurrence and observation of this phenomenon are considered in detail in Chap. 4 below.

4. CROSSING OF ATOMIC LEVELS OF A DEGENERATE STATE

a) **Crossing signal and conditions for its observation.** The magnitude and form of the expected signal of the crossing of magnetic hyperfine sublevels of an excited state of atoms can be estimated with the aid of the expression^[15a, 17]:

$$S_F = S_0 \sum_{FmF'm'} G_{mm'}^{FF'} \rho_{FmF'm'} \quad (5)$$

where

$$G_{mm'}^{FF'} = \sum_{f\mu} \langle Fm | eD | f\mu \rangle^* \langle F'm' | eD | f\mu \rangle,$$

which represents the intensity of the light spontaneously emitted by the atoms as they return from the excited state to the ground state, $G_{mm}^{FF'}$ (in the standard notation) is the matrix of radiation characterized

by a single polarization vector e , and D is the electric dipole moment. The state vector $|Fm\rangle$ describes the hyperfine Zeeman sublevel of the excited state, and the vector $|f\mu\rangle$ describes the hyperfine sublevel of the ground state, while F and f are the values of the total angular momentum in the corresponding states and m and μ are the magnetic quantum numbers determining the projections of F and f on the direction of the constant magnetic field H_0 which is parallel to the z axis.

The density matrix $\rho_{FmF'm'}$ describes the evolution of the excited state of the atoms and depends mainly on the method of their excitation. If the atoms go over into an excited state as a result of absorption of resonant light characterized by a single polarization vector e^0 , then the matrix $\rho_{FmF'm'}$ can be obtained from the equation^{[51a, 17] 1)}

$$\begin{aligned} (d\rho_{FmF'm'}(t)/dt) + (\Gamma + i\omega_{FmF'm'}) \rho_{FmF'm'}(t) \\ = T_r^{-1} \sum_{f\mu f'\mu'} \langle Fm | e^0 D | f\mu \rangle \langle F'm' | e^0 D | f'\mu' \rangle^* \rho_{f\mu f'\mu'}(t), \end{aligned} \quad (6)$$

which is well known in the theory of optical orientation of atoms^[17]; here, T_r is the relaxation time, equal to the average lifetime of the atoms in the ground state and is determined by the intensity of the optical excitation, $\omega_{FmF'm'}$ is the energy interval between the excited magnetic sublevels, Γ is the decay constant (here and throughout we used a system of units in which $\hbar = c = 1$), and $\rho_{f\mu f'\mu'}(t)$ are the density-matrix elements that describe the populations ($f = f'$, $\mu = \mu'$) and the coherence (off-diagonal elements) of the ensemble of atoms in the ground state. In deriving the equation for $\rho_{FmF'm'}$ it was assumed that the damping of the populations and of the coherence of the excited atoms is determined only by spontaneous-decay processes. It will be shown subsequently that these assumptions are quite reasonable.

For the light intensities customarily used in experiments on level crossing, the probability of the induced atomic transitions is much shorter than the probabilities of the spontaneous ones. Taking this circumstance into account, Eq. (6) can be easily integrated:

$$\begin{aligned} \rho_{FmF'm'}(t) = T_r^{-1} \sum_{f\mu f'\mu'} \langle Fm | e^0 D | f\mu \rangle \langle F'm' | e^0 D | f'\mu' \rangle^* \\ \times \{\Gamma + i[\omega_{FmF'm'} - \omega_{f\mu f'\mu'}]\}^{-1} \rho_{f\mu f'\mu'}(t). \end{aligned} \quad (7)$$

The obtained expression represents the coherence produced in the excited state by the resonant light, which is determined in many respects by the behavior of the atoms in the ground state. In particular, the coherence between the sublevels m and m' is not the same for different distributions of the magnetic sublevels of atoms of the ground state. On the other hand, the presence of coherence in the ground state leads to the interesting conclusion that even if there is no coherence in the excited state at the initial instant of time, coherence can be introduced into it subsequently from the ground state. The phenomenon of coherence transfer does not occur if $|\omega_{FmF'm'} - \omega_{f\mu f'\mu'}| \gg \Gamma$, as follows from Eq. (7).

Let us assume that there is no coherence in the ground state and that the populations are equal. Then

¹⁾ Atoms can be excited into a coherent superposition of states by using a beam of slow electrons or ions. As a rule, this method is used if it is impossible for some reason to excite the atoms optically. For details see the review [7] and the papers in [18].

the expression for the radiation intensity (5), when account is taken of (7), takes the form

$$S = S'_0 \sum_{F'F''m'm'} \langle Fm | eD | f\mu \rangle \langle F'm' | eD | f\mu \rangle (1 + i\tau\omega_{FmF'm'})^{-1} \times \langle F'm' | eD | f'\mu' \rangle \langle Fm | eD | f'\mu' \rangle, \quad (8)$$

in which it is easy to recognize the well-known Breit expression^[3a, 5D] for the crossing signal of sublevels of excited atoms. An interference signal is observed experimentally only when certain conditions are satisfied with respect to the magnitude of the constant magnetic field and the properties of the exciting and detected light.

The requirements imposed on the magnetic field H_0 are determined by the factor in the round brackets of (8). If the magnetic sublevels $|Fm\rangle$ and $|F'm'\rangle$ are completely resolved, i.e., $\tau\omega_{FmF'm'} \gg 1$, then only the terms with $F = F'$ and $m = m'$ remain in the expression for the radiation intensity. The terms with $F \neq F'$ and $m \neq m'$, which describe the interference effect, drop out. The appearance of interference effects is expected only when two or more excited states are sufficiently close to one another, so that $\tau\omega_{FmF'm'} \ll 1$. We are already acquainted with this condition.

The properties of the resonant radiation, exciting the atoms into a coherent superposition of states $|Fm\rangle$ and $|F'm'\rangle$ are determined by the excitation matrix $A_{mm'}^{FF'}$, which enters in (8)

$$A_{mm'}^{FF'} = \sum_{f'\mu'} \langle F'm' | eD | f'\mu' \rangle \langle Fm | eD | f'\mu' \rangle = \sum_{f'\mu'q} \epsilon_{qq'}^0 \langle F'm' | D_q | f'\mu' \rangle \langle Fm | D_q | f'\mu' \rangle, \quad (9)$$

where D_q are the spherical components of the vector D and are simultaneously the components of the irreducible tensor of the operator D_q^1 , while $\epsilon_{qq'}^0 = e_q^0 e_{q'}^0$ is the polarization matrix, with

$$e_q^0 = -(q/\sqrt{2})(e_x^0 + iq e_y^0) \quad (q = \pm 1), \quad e_0^0 = e_z^0.$$

The indices $q = 1$ and $q = -1$ correspond to right-hand and left-hand polarization of the light, respectively.

If we use the Wigner-Eckart theorem^[16]

$$\langle Fm | D_q^1 | f\mu \rangle = \langle F || D || f \rangle (f1\mu q | Fm),$$

where $\langle F || D || f \rangle$ is the reduced matrix element of the atomic transition and $(f1\mu q | Fm)$ is the Clebsch-Gordan coefficient, then we can reduce (9) to the form

$$A_{mm'}^{FF'} = \sum_{f'\mu'q} \langle F' || D || f' \rangle \langle F || D || f \rangle (f'1\mu'q' | F'm') (f'1\mu'q | Fm) \epsilon_{qq'}^0.$$

from the properties of the Clebsch-Gordan coefficients it follows that $m - m' = q' - q$, where $q = 0, \pm 1$, with zero characterizing the transitions with absorption of light with π polarization, and ± 1 transitions with absorption of right- and left-polarized light. It follows therefore that when atoms are irradiated with, say, circularly polarized light ($q = 1$) in the direction of the magnetic field we have $q = q'$ and $m - m' = 0$. In other words, the atoms are excited in pure states. To the contrary, if circularly polarized light propagates in a direction perpendicular to H_0 , all the sublevels for which $\Delta m = 0, 1$, and 2 are excited. Thus, to excite atoms into a superposition state the light should have coherent polarization.

The properties of the re-radiated light are determined by the radiation matrix

$$G_{mm'}^{FF'} = \sum_{f\mu\eta} \langle F || D || f \rangle \langle F' || D || f \rangle (f1\mu\eta | Fm) (f1\mu\eta' | F'm') \epsilon_{\eta\eta'},$$

where $\epsilon_{\eta\eta'}$ is the polarization matrix. It follows from this expression that to observe level crossing with $m \neq m'$ we must have η different from η' . In other words, the detected radiation must be represented by a mixture of components with different polarizations.

Thus, the crossing signal, which is proportional to the reradiated light intensity and is described by the expression

$$S = S'_0 \sum_{\substack{FmF'm' \\ f'1\mu'q'q\eta\eta'}} D(FF'; ff') (f'1\mu'q' | F'm') (f'1\mu'q | Fm) \times \epsilon_{qq'}^0 \epsilon_{\eta\eta'} (1 + i\tau\omega_{FmF'm'})^{-1} (f1\mu\eta | Fm) (f1\mu\eta' | F'm'), \quad (10)$$

where

$$D(FF'; ff') = \langle F' || D || f \rangle \langle F || D || f \rangle \langle F || D || f \rangle^* \langle F' || D || f \rangle,$$

is different from zero if coherent excitation and detection are realized, and the magnetic field satisfies the condition $\tau\omega_{FmF'm'} \lesssim 1$.

To produce the correct experimental geometry for the study of crossing of excited atomic sublevels, it is necessary to know the exact dependence of the probability of the optical transitions on the propagation direction and polarization of the light; this dependence is contained in $\epsilon_{qq'}$. It is convenient to express the polarization in terms of the polar and azimuthal angles θ and φ , which define the direction of the unit vector r^0 , which is also the direction of radiation propagation (Fig. 2). The polarization unit vector e , specified relative to the direction r^0 in the system $(\theta^0, \varphi^0, r^0)$ makes an angle α with the direction of the unit vector θ^0 connected with the angle θ . In the case of arbitrary polarization of the exciting light, the vector e in the system $(\theta^0, \varphi^0, r^0)$ can be represented in the form

$$e = \theta^0 \cos \alpha + \varphi^0 \sin \alpha e^{i\beta}. \quad (11)$$

If $\beta = 0$, then expression (11) describes a linear-polarization vector making an angle α with the θ^0 axis, and describes at $\beta = \pm \pi/2$ and $\alpha = \pi/4$ a vector representing right- and left-polarized radiation, respectively. Transforming from the system $(\theta^0, \varphi^0, r^0)$ to (x, y, z) and introducing then the complex unit vectors u_0 and u_{\pm} in accordance with the formulas

$$u_{\pm} = \mp (1 \pm i)/\sqrt{2}, \quad u_0 = k,$$

we can represent the light polarization vector in the form

$$e = \sum_{q=-1}^1 (-1)^q e_{-q} u_q,$$

where

$$e_q = -(q/\sqrt{2})(\cos \theta \cos \alpha + iq \sin \alpha e^{i\beta}) e^{i\varphi} \quad (q = \pm 1), \quad e_0 = -\sin \theta \cos \alpha.$$

Thus, the polarization matrix is given by

$$\epsilon_{qq'} = \begin{pmatrix} \epsilon_{11} & \epsilon_{10} & \epsilon_{1,-1} \\ \epsilon_{01} & \epsilon_{00} & \epsilon_{0,-1} \\ \epsilon_{-1,1} & \epsilon_{-1,0} & \epsilon_{-1,-1} \end{pmatrix}, \quad (12)$$

where

$$\begin{aligned} \epsilon_{\pm 1, \pm 1} &= (1/2)(1 - \sin^2 \theta \cos^2 \alpha \mp \cos \theta \sin 2\alpha \sin \beta), \\ \epsilon_{0, -1} &= \epsilon_{-1, 0} = (1/\sqrt{2}) \sin \theta \cos \alpha (\cos \theta \cos \alpha + i \sin \alpha e^{-i\beta}) e^{i\varphi}, \\ \epsilon_{10} &= \epsilon_{01} = (1/\sqrt{2}) \sin \theta \cos \alpha (\cos \theta \cos \alpha + i \sin \alpha e^{i\beta}) e^{i\varphi}, \\ \epsilon_{1, -1} &= \epsilon_{-1, 1} = -(1/2)(\cos^2 \theta \cos^2 \alpha - \sin^2 \alpha + i \cos \theta \sin 2\alpha \cos \beta) e^{2i\varphi}, \\ \epsilon_{00} &= \sin^2 \theta \cos^2 \alpha. \end{aligned}$$

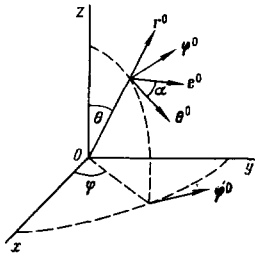


FIG. 2. Coordinate system for the description of the light polarization.

Expression (12) enables us to calculate the crossing signal for any experimental geometry and for an arbitrary polarization of the exciting and re-radiated light.

b) **The Hanle effect.** The first experiments in which they observed coherence of states produced by optical excitation were the experiments of Hanle on magnetic depolarization of light^[1,2,3,4] We shall examine the gist of these experiments using the even isotopes of mercury as an example.

Mercury vapor is irradiated in the direction of the Ox axis by resonant light of the 2537 Å line, polarized in the Oy direction. The following is then observed. In the absence of a magnetic field, the re-radiated light is strongly polarized (~90%), and the electric vector of the radiation is parallel to the unit vector of polarization of the exciting light. The angular distribution of the fluorescence is determined by the directivity pattern of the radiation of the electric dipole, the axis of which coincides with the direction of the electric vector of the exciting light. Superposition of a magnetic field parallel to e^0 does not change the character of the radiation (incoherent excitation). However, if $H_0 \perp e^0$ (coherent excitation), the picture changes, and as the field increases from zero the degree of polarization of the light emitted in the direction of H_0 decreases and vanishes when the Zeeman splitting of the excited radiation greatly exceeds the natural width Γ .

A quantitative description of these results can be obtained in the basis of expression (10), in which F should be equated to F' , and $\omega_F m F' m'$ must be replaced by $\omega_F(m - m')$, inasmuch as the atomic magnetic sublevels belonging to a definite F are equidistant in a weak field:

$$S_F = S_0' \sum_{\substack{\mu\mu' m m' \\ q q' \eta \eta'}} D(F F'; f f) (f 1 \mu' q' | F m') (f 1 \mu q | F m) \\ \times e_{q q'}^0 e_{\eta \eta'} [1 + i \tau \omega_F (m - m')]^{-1} (f 1 \mu' \eta | F m) (f 1 \mu \eta' | F m').$$

The ground 6^1S_0 state of even isotopes of mercury is a singlet (nuclear spin equal to zero), and the excited 6^3P_1 state in a magnetic field is a system of three equidistant magnetic sublevels with $m = 0$ and ± 1 . The conditions for the excitation of these sublevels depends on the polarization and on the propagation direction of the radiation. In particular, if the atoms are eliminated in the Oy direction with light polarized in the Ox direction ($H_0 \parallel Oz$), only the sublevels $|1\rangle$ and $|-1\rangle$ are excited, as follows from the form of the matrix $\epsilon_{qq'}^0$, in which only the components with $q = q' = \pm 1$ and $q = -q'$ differ from zero, and from the properties of the Clebsch-Gordan coefficients. The difference Δm between the magnetic quantum numbers is equal to zero and ± 2 . The case $\Delta m = 0$ corresponds to excitation of the atom in the pure state, and $\Delta m = \pm 2$ corre-

sponds to excitation in a superposition of states $|1\rangle$ and $|-1\rangle$ upon absorption of a linearly-polarized photon. This case corresponds to alignment in the atomic system. Therefore the expression for the intensity of $S_F(\theta', \varphi'; \alpha')$ of light re-radiated in a direction defined by the angles θ' and φ' and polarized at an angle α' to the θ^0 axis can be represented in the form of a sum of two terms, $S_F^{(0)}$ and $S_F^{(1)}$, with $S_F^{(0)}$ characterizing the intensity of that part of the radiation for which $\Delta m = 0$, and $S_F^{(1)}$ connected with coherent excitation ($\Delta m = 2$). This part depends on the value of the magnetic field H_0 . The expressions for $S_F^{(0)}$ and $S_F^{(1)}$ are

$$S_F^{(0)} = (1/2) S_0' (1 - \sin^2 \theta' \cos^2 \alpha'),$$

$$S_F^{(1)} = (1/2) S_0' (1 + 4\tau^2 \omega_F^2)^{-1} [(\cos^2 \theta' \cos^2 \alpha' - \sin^2 \alpha' - \cos \theta' \sin 2\alpha' \sin 2\varphi') + 2\tau \omega_F (\cos^2 \theta' \cos^2 \alpha' \sin 2\varphi' - \cos \theta' \sin 2\alpha' \cos 2\varphi')].$$

In particular, the intensity of light emission in the direction of the z axis ($\theta' = \varphi' = 0$) of the laboratory frame (x, y, z), polarized along Ox ($\alpha' = 0$) and Oy ($\alpha' = \pi/2$), is described by the well-known relation

$$S_F(x) = (1/2) S_0' [1 + (1 + 4\tau^2 \omega_F^2)^{-1}], \quad (13a)$$

$$S_F(y) = (1/2) S_0' [1 - (1 + 4\tau^2 \omega_F^2)^{-1}]. \quad (13b)$$

The loss of coherence is directly connected with the quantity $S_F(y)$. It becomes complete when $S_F(y) = S_0'/2$, that is, when the splitting of the Zeeman sublevels of the excited state exceeds the natural line width ($4\omega_F^2 \tau^2 \gg 1$). In this case the magnetic field no longer influences the character of the polarization of the radiation, and the radiation intensity is described by the expression for $S_F^{(0)}$, as if the atoms were to be excited in pure states.

One can present a qualitative albeit insufficiently consistent semiclassical interpretation of these results^[20]. If the linearly polarized excitation is regarded as a superposition of radiations with left- and right-hand polarizations, then the latter produce σ^+ and σ^- transitions with equal probability. Obviously, in the general case, the employed method of excitation transforms the atoms not into pure states $|1\rangle$ and $|-1\rangle$, but into a state representing the coherent combination $|\psi\rangle = 2^{-1/2}(|1\rangle - |-1\rangle)$ for all the atoms. In this state, the electric moment of the atom lies in the direction of the Ox axis, and the spatial-distribution diagram of the radiation of the atoms in the state $|\psi\rangle$ is not simply a sum of the radiation diagrams of the state $|1\rangle$ and $|-1\rangle$. If now the atoms are placed in a magnetic field, then the electric dipoles begin to precess about the direction of this field. The angle rotation of the moments in the (x, y) plane is proportional to the magnitude of the magnetic field, and therefore, being excited at different instants of time, they will turn through different angles relative to the Ox axis, forming a sort of fan in the (x, y) plane. If the period of the Larmor frequency is much larger than the decay time of the excited state, then the angle through which the moments have time to turn as a result of the precession will be negligible (partially open fan). In this case, partial depolarization of the light is observed with simultaneous rotation of the plane of polarization relative to the direction of the polarization vector of the exciting light. On the other hand, if the dipoles

execute many revolutions about the direction of the magnetic field H_0 during the lifetime, then the resultant radiation will be fully depolarized.

A comparison of expressions (13) with the experimental results shows that in addition to providing a perfectly satisfactory description of the general character of the observed phenomenon, they also make it possible to calculate the line shape of the level crossing signal in a weak magnetic field.

c) **Line width of crossing signal.** Expression (10) for the crossing signal was obtained under the assumption that the damping of the coherence ρ_{nm} in the excited state is due to spontaneous decay of the atoms. In this approximation the level-crossing signal width is determined by the decay constant Γ of the investigated state. The experimental data indicate, however, that this approximation is not always valid and in some cases the relaxation time of the coherence of the crossing levels may greatly differ from $1/\Gamma$. In the experiments considered here, this is caused mainly by two phenomena, resonant collisions and coherence diffusion. When the atoms are excited by an electron or laser beam, by electric discharge, or by some other method, other causes of crossing-line broadening occur, but their analysis is beyond the scope of the present review^[8b, 18, 21, 22].

Let us formulate briefly the conditions under which the broadening mechanisms considered here are significant. A detailed analysis of these conditions can be found in^[7].

1) **Resonant collisions.** When an excited atom in the resonant state collides with an identical atom in the ground state, the electrostatic dipole-dipole interaction gives rise to appreciable probability of excitation transfer from one atom to the other (the **Holtmark effect**, see^[23]). Interactions of this type are the cause of additional relaxation of the excited atoms, increasing the width Γ of the emission line by an amount^[24] $g = \alpha\Gamma N/k_0^3$, where α is a numerical coefficient that depends on the experimental conditions, k_0 is the frequency of the resonant transition of the atom, and N is the number of atoms per unit volume. The broadening due to the collisions depends significantly on the vapor pressure of the investigated substance and on the frequency of the atomic transition. Thus, for $N \sim 10^{15}$ at/cm³ we have $g \sim \Gamma$ near the ultraviolet radiation region. Figure 3 shows the width of the crossing signal of the levels of the 5^3P_1 state of cadmium in a vanishingly weak magnetic field as a function of the concentration^[25]. An analysis of the experimental result shows that the effect of the collisions on the line width can be neglected if N does not exceed $10^{13} - 10^{14}$ at/cm³. Thus, the collisions have practically no effect at low concentration of the atoms.

a) **Coherence diffusion.** This interesting phenome-

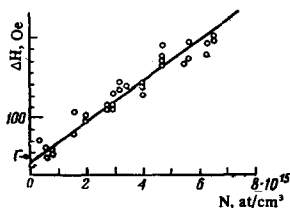


FIG. 3. Holtmark broadening of the crossing signal.

non, which consists in a narrowing of the resonant-signal line because of coherent multiple scattering of the light quantum in the resonance cell, was observed in an investigation of the 6^3P_1 state of mercury. Since that time it has been the subject of many experimental^[26a, b, 27] and theoretical studies^[26b, c, 28]. Coherent narrowing can be illustrated on the basis of the following analysis. Let, for example, mercury atoms in a magnetic field H_0 be excited to a definite magnetic sublevel of the 6^3P_1 state. Starting with the instant of excitation, the orientation of the atom in this state, owing to precession in the magnetic field, varies with time. In the decay of the state, the emitted quantum carried definite information on the orientation of the atom and the phase of the state, which can be transferred to another atom that absorbs this photon. This phase memory is preserved as a result of multiple scattering acts, which lengthen the damping time $T_c = 1/\Gamma_c$ of the off-diagonal components ρ_{nm} that determine the width Γ_c of the contracted line. For T_c , theory yields the value

$$T_c = \tau/(1 - \beta p), \quad (14)$$

where $\beta < 1$ is a function of the quantum numbers of the ground and excited states, and p is the probability of multiple scattering. According to (14), the width of the observed crossing signal decreases. Figure 4 shows the results of an experimental study^[26b, c] of the coherent narrowing of the signal in the Hanle effect for the 3^3P_1 state of Hg²⁰². Owing to multiple diffusion of the radiation, as follows from the given relation, one can obtain a line with one-third the natural width. However, transitions of atoms to other magnetic sublevels, following absorption of the re-radiated photons, as well as disorienting collisions, interrupt the coherent process and by the same token broaden the line. Therefore, to avoid disorienting collisions when the pressure is increased and causes a growth of T_c as a result of resonant narrowing (although T_c is decreased by depolarization), it is necessary that the photon absorption cross section be large enough. Transitions to other sublevels following absorption of the emitted photon can be eliminated by making the Zeeman splitting larger than the Doppler width of the optical excitation. In this case it is possible to obtain an even larger coherent narrowing^[7, 27] of the optical emission line. Unfortunately, this process is accompanied by depolarization of the light, and thus makes observation of the crossing difficult.

The foregoing analysis leads to two important practical conclusions: (1) the width of the crossing signal is determined with great accuracy by the decay constant of the excited state, if the concentration of the atoms in the resonance cell is low enough to prevent

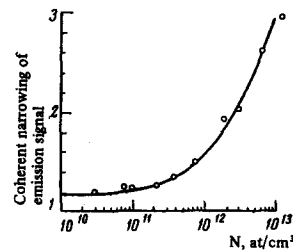


FIG. 4. Coherent narrowing of emission signal vs the concentration N .

the Holtsmark effect and coherent narrowing from setting in. For mercury atoms (see Fig. 4), at concentrations 10^{10} – 10^{11} cm^{-3} , the observed line width practically coincides with the natural width. (2) The crossing signal can be used to investigate relaxation processes in excited atoms, the mechanisms of interaction between them, collision cross sections^[23,29], and coherent-narrowing phenomena^[30].

The level crossing method makes it possible in some cases to separate the contributions made to the crossing line width by spontaneous transitions from the broadening due to modulation of the point of the level crossing by the fluctuating electric and magnetic fields in which the atoms (or nuclei) in the medium are situated^[12].

By way of example, we consider the case when the crossing signal $S_F(x)$ is given by (13a) in the absence of fluctuating fields. If the atoms are acted upon by a field h that is parallel to $H_0 = \omega_F/\gamma$ and changes the splitting by a certain amount $\Delta\omega = \gamma h$, then the position of the maximum of $S_F(x)$ as a function of ω_F will occur no longer at $H_0 = 0$, but at $H_0 = -h$. The crossing line contour then shifts without changing shape, and the area under the curve

$$\mathcal{F}(\tau) = \int_{-\omega_m}^{\omega_m} [S_F(x) - (1/2)S_0] d\omega_F = \pi S_0' / 4\tau$$

is a function of τ and (an important factor at sufficiently large values of $\omega_m > \Delta\omega$ and $1/\gamma\tau, \omega_m \gg 1$) is no longer dependent on h .

Thus, if h is the intensity of the random magnetic field, then the crossing-line contour becomes inhomogeneously broadened and is made up of the envelope of lines (13a) with randomly shifted maxima, while the area \mathcal{F} remains the same as at $h = 0$. This circumstance enables us to determine the line width $1/\tau$ of the spontaneous transition against the background of its inhomogeneous broadening. The coefficient can be determined experimentally by measuring the emission signal in strong magnetic fields ($\omega_F\tau \gg 1$).

d) Determination of the lifetimes of excited atoms with the aid of the Hanle effect. As already mentioned, a characteristic feature of the crossing signal (if the conditions in Sec. (c) are satisfied) is the absence of line-broadening sources, so that the experimentally observed line width practically coincides with the natural width. It is precisely this circumstance which makes it possible to use experiments on level crossing, particularly in a zero field, for a very accurate determination of the lifetime of an excited state.

The experimental geometry used to determine τ does not differ from that considered in Sec. (c): linearly polarized excitation propagates along Oy, the polarization vector is parallel to the Ox axis, and $H_0 \parallel Oz$. The crossing signal, which satisfies (13) in the case of mercury atoms, is observed in the direction of the field H_0 . One determines in the experiment the degree of polarization of the emitted light

$$P(H_0) = [S_F(x) - S_F(y)] / [S_F(x) + S_F(y)]$$

at two values of the magnetic field—one zero the other nonzero. If we denote by $P(0)$ the degree of polarization of $H_0 = 0$, then we obtain with the aid of (13) the following relation for $P(H_0)$:

$$P(H_0) = P(0) / (1 + 4\omega_F^2\tau^2) = P(0) [1 + (eH/m)g_F\tau]^2)^{-1}, \quad (15)$$

from which, knowing the atomic g_F -factor, we can estimate the lifetime of the excited state. It can be shown that although (15) was derived with mercury-atom sublevel crossing as an example, it is valid for any atomic system.

It should be noted that the crossing signal in a zero field can be influenced by level crossing near the investigated range of fields. Therefore, for an exact measurement of τ , it is necessary to know the contribution of other crossings to the investigated signal. Nonetheless, even without taking this factor into account, the experimentally observed values of τ are perfectly satisfactory and are not inferior in their accuracy to those obtained by other methods. According to^[31], for example, the lifetime of Na^{23} atoms in the state $3^2P_{3/2}$, determined by the technique of level crossing in a zero field, is equal to $\tau = (1.63 \pm 0.05) \times 10^{-8}$ sec, and the lifetime of this state measured with the method of double radiooptical resonance^[32,33] is $\tau = (1.63 \pm 0.04) \times 10^{-8}$ sec.

The Hanle effect was used to measure the lifetimes of many states of different elements. In addition to such classical substances as $\text{Hg}^{[33,34]}$ and alkali metals^[35,36,37], at the present time the experimental techniques make it possible to investigate excited states of heavy elements: Mn, Eu, Sm, and $\text{Tm}^{[38]}$, Sn and $\text{Pb}^{[39]}$, $\text{Pd}^{[40]}$, $\text{Co}^{[41]}$, $\text{Ba}^{[42]}$, and others.

e) Crossing of atomic levels in arbitrary constant magnetic fields. A study of the phenomena connected with the level crossing in nonzero magnetic fields yields new information on the atomic structure^[43]. The most important are experiments on the determination of the constants of the fine and hyperfine structure and on the quadrupole interaction. The method can also be used to investigate Stark splitting.

The crossing signal in nonzero fields has a much more complicated structure than in the case of the Hanle effect. For its interpretation it is therefore necessary to produce in each experiment conditions that make it possible to simplify the comparison of the experimental results with the theory. We consider below several highly simplified experimental setups customarily used in practice. The character of these setups is determined by the distinguishing features of the observed phenomena and by the characteristics of the light emitted by the atoms under the crossing conditions.

It was shown above that in the electric dipole approximation it is possible to observe crossing of atomic sublevels whose magnetic quantum numbers m and m' differ by ± 1 and ± 2 . The case $\Delta m = 0$ is not considered, since levels with equal magnetic quantum numbers do not cross according to Neumann's theory^[44]. Therefore, the fluorescence light contains information on crossings both with $\Delta m = \pm 1$ and with $\Delta m = \pm 2$, crossings that can take place at any given value of the magnetic field H_0 , including the zero value. Through a definite choice of the geometry of the beams of the excited and re-radiated lights, their polarization, the modulation technique, etc., it is possible to separate experimentally the crossing signals with $\Delta m = \pm 1$ and $\Delta m = \pm 2$.

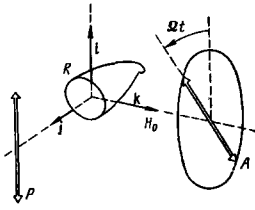


FIG. 5. Scheme providing coherent excitation and detection of crossing signals with $|\Delta m| = 2$.

1) Crossing signal with $|\Delta m| = 2$ and the conditions for its observation. Figure 5 shows one of the possible experimental setups for the observation of the crossing signal of levels with $\Delta m = 2$. The exciting light propagates along the Oy axis and its polarization vector e^0 is parallel to the Ox axis. The investigated substance is placed in the resonance cell R and the light emitted by its atoms is registered in the direction of the magnetic field $H_0 \parallel Oz$. After passing through analyzer A, which rotates with frequency Ω in the (x, y) plane, the light proceeds to a photomultiplier, the ac signal from which is fed to a synchronous detector. A rotating analyzer is used to improve the signal/noise ratio and to eliminate the influence of the incoherent part of the spontaneous emission on the signal.

At a given experimental geometry, only crossings of levels with $\Delta m = 2$ will be observed. To predict this result, it suffices to consider the form of the polarization matrix $\epsilon_{qq'}$, without going into details of the calculations connected with (10). In this case

$$\epsilon_{qq'} = \frac{1}{2} \begin{pmatrix} 1 & 0 & -1 \\ 0 & 0 & 0 \\ -1 & 0 & 1 \end{pmatrix}.$$

Since $m - m' = q' - q$, the states that can be coherently excited are those for which $m - m' = \pm 2$, and the interference effect will be due only to these states.

Recognizing that the polarization matrix of the light re-radiated in the direction of H_0 is given by

$$\epsilon_{\eta\eta'} = \frac{1}{2} \begin{pmatrix} 1 & 0 & -\cos 2\alpha' - i \sin 2\alpha' \\ 0 & 0 & 0 \\ -\cos 2\alpha' + i \sin 2\alpha' & 0 & 1 \end{pmatrix},$$

we obtain with the aid of (10), for the coherent part of the fluorescence from two arbitrary sublevels $|F_m\rangle$ and $|F'm'\rangle$ ($|m - m'| = 2$), the expression

$$S_F^{(\Delta m=2)} \sim \Gamma^2 (\Gamma^2 + \omega_{F'mF'm'}^2)^{-1} \cos 2\Omega t + \Gamma \omega_{F'mF'm'} (\Gamma^2 + \omega_{F'mF'm'}^2)^{-1} \sin 2\Omega t, \quad (16)$$

where account is taken of the fact that $\alpha' = \Omega t$. In the general case, the modulation part of the fluorescence is determined from the sum of different crossing signals with $\Delta m = 2$, of the type (16). Depending on the choice of the phase of the synchronous detector, we can separate experimentally either the first or the second term of (16).

2) Crossing signal with $|\Delta m| = 1$. The experimental setup usually employed to observe the crossing signal of state with $\Delta m = 1$ is shown in Fig. 6. The exciting light passes in the direction of the Oy axis through a polarizer P rotating with frequency Ω , and the fluorescence is observed in the direction of the Ox axis perpendicular to the magnetic field H_0 . The analyzer A makes an angle α' with the direction of the magnetic

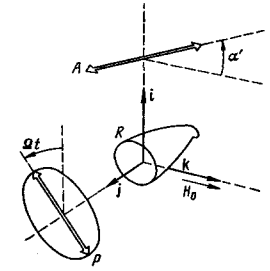


FIG. 6. Scheme for the observation of crossing signals with $|\Delta m| = 1$.

field. For the excitation employed in this case, the polarization matrix $\epsilon_{qq'}^0$ is

$$\epsilon_{qq'}^0 = \frac{1}{2} \begin{pmatrix} \sin^2 \alpha' & -(1/\sqrt{2}) \sin 2\alpha' & -\sin^2 \alpha' \\ -(1/\sqrt{2}) \sin 2\alpha' & 2 \cos^2 \alpha' & (1/\sqrt{2}) \sin 2\alpha' \\ -\sin^2 \alpha' & (1/\sqrt{2}) \sin 2\alpha' & \sin^2 \alpha' \end{pmatrix}, \quad \alpha' = \Omega t. \quad (17)$$

It is easy to note that in this case the coherently excited levels are those with $\Delta m = \pm 1$ and ± 2 . At no experimental geometry is it possible to excite atoms into a coherent superposition of states with $\Delta m = 1$, in contrast to the case of sublevels with $\Delta m = 2$. The re-radiated light now constitutes an aggregate of crossing modes, and the problem of detection is to separate the sought signal from the overall signal. This problem can be easily solved by using the time dependence of the spatial orientation of the plane of polarization and the experimental geometry. Indeed, let the fluorescence be received in the direction of the unit vector i , and let the polarization vector e make an angle with the direction of the field H_0 . The polarization matrix then takes the form

$$\epsilon_{\eta\eta'} = \frac{1}{2} \begin{pmatrix} \sin^2 \alpha' & (1/\sqrt{2}) i \sin 2\alpha' & \sin^2 \alpha' \\ -(1/\sqrt{2}) \sin 2\alpha' & 2 \cos^2 \alpha' & -(1/\sqrt{2}) i \sin 2\alpha' \\ \sin^2 \alpha' & (1/\sqrt{2}) i \sin 2\alpha' & \sin^2 \alpha' \end{pmatrix}. \quad (18)$$

Substitution of (17) and (18) in (10) shows that in this case the crossing signals of sublevels with $|\Delta m| = 1$ and with $|\Delta m| = 1$ and with $|\Delta m| = 2$ can be observed independently. Indeed, the crossing signal of sublevels for which $|\Delta m| = 1$ depends on the time via $\sin 2\Omega t$, i.e.,

$$S_F^{(\Delta m=1)} \sim \Gamma \omega_{F'mF'm'} (\Gamma^2 + \omega_{F'mF'm'}^2)^{-1} \sin 2\alpha' \sin 2\Omega t, \quad (19)$$

and the coherent-radiation component due to the crossing with $|\Delta m| = 2$ depends on t via $\cos 2\Omega t$, i.e.

$$S_F^{(\Delta m=2)} \sim \Gamma^2 (\Gamma^2 + \omega_{F'mF'm'}^2)^{-1} \sin^2 \alpha' (1 - \cos 2\Omega t).$$

The time variation of the incoherently scattered radiation is similar. By synchronous detection and by a definite choice of the phase it is possible to obtain a signal of crossing with only $|\Delta m| = 1$, proportional to $\sin 2\Omega t$, eliminating the influence of the crossings with $|\Delta m| = 2$ and of the incoherently scattered light.

Expressions (16) and (19), generally speaking, describe more readily the qualitative aspect of the phenomenon and not the quantitative one (with the exception of the case when the individual crossings have good resolution). For a comparison with the experimental results it is necessary to use somewhat modified expression, for example (8) with allowance for the

spectral composition of the exciting light. It is known that the relative intensity I_f of the light absorbed by the atoms of the f -th hyperfine level of the ground state has different values for different f . Therefore the crossing signal depends on the ratio r of their intensities. Taking this circumstance into account, the excitation matrix should be taken in the form

$$A_{mm'}^{F'F} = \sum \langle F'm' | e^0 D | f'\mu' \rangle^* I_{f'} \langle f'\mu' | e^0 D | Fm \rangle. \quad (20)$$

f) Use of the crossing method in atomic spectroscopy. Since its discovery, the crossing method was used to study different problems in atomic spectroscopy. However, the principal among them are the fine and hyperfine structures of atomic states, lifetimes of the excited state, Stark shifts^[45] and atomic g -factors, nuclear and electronic moments^[46,47,48], etc. To illustrate the crossing method, we stop to discuss only the study of the fine and hyperfine interactions. Data on the remaining questions can be found in the cited papers.

1) Fine structure. High-accuracy measurements of the fine structure are of great importance for light atoms that are well described by modern theory. Inasmuch as in all these cases the splitting is small in comparison with the Doppler width of the corresponding spectral lines, ordinary spectroscopy is not accurate enough. The crossing method however, has been used successfully to investigate the states of hydrogen and helium with $n = 2$ and the state of lithium with $n = 2, 3$, and 4 ^[49]. The accuracy of the results is higher by several orders of magnitude than that of optical spectroscopy.

Let us consider by way of example the study of the fine structure of the 2^3P state of He^4 atoms, first undertaken in^[5], where this method was initiated. Figure 7 shows the scheme of the Zeeman sublevels of the 2^3P state. The circles mark the crossings with $\Delta m = 2$. One more crossing of this type, $(0, 0) - (2, 2)$, is observed in the strong field 8200 Oe; it is not shown in the figure. The squares mark the crossings with $\Delta m = 1$. As shown in^[5], all the crossings with $\Delta m = 2$ are well resolved, and on the basis of (13) we estimate the fine structure of the splitting, i.e., the frequency of the $2^3P_1 - 2^3P_2$ transition, which turned out to be 2291.200(22) MHz in these measurements. It is interesting to compare this value with the recently published 2291.195(7) MHz, obtained by the atomic-beam method.^[50]

Experiment on the determination of the fine structure of atoms turned out to be relatively simple and easy to interpret. Much greater difficulties are en-

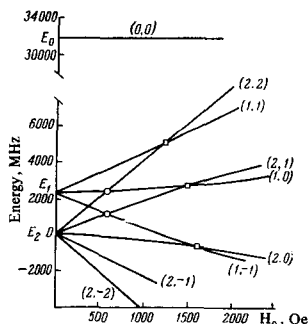


FIG. 7. Zeeman splitting of 2^3p state of He^4 in a magnetic field.

countered in the study of the crossings of hyperfine levels, to which we now turn.

2) Determination of atomic and nuclear constants from the crossings of hyperfine sublevels. From the experimentally obtained crossing curve it is possible to determine the atomic characteristics only if one knows the character of the dependence of the energy of the crossing sublevels on the magnetic field, i.e., $\omega_{FmF'm'}(H_0)$. For excited states, this dependence as a rule has a more complicated form, particularly in those cases when the atomic nuclei have a spin $I > 1/2$. As is well known, this makes it necessary to include in the Hamiltonian \mathcal{H} of the system not only the magnetic hyperfine interaction but also the quadrupole interaction

$$\mathcal{H} = A\mathbf{I}\mathbf{J} + B[2I(2I-1)(2J-1)J]^{-1}[3(\mathbf{I}\mathbf{J})^2 + (3/2)(\mathbf{I}\mathbf{J}) - I^2J^2] + \mu_B g_J H_0 J_0 \quad (21)$$

where A and B are the hyperfine and quadrupole interaction constants, respectively, \mathbf{J} is the total angular momentum of the electron shell, μ_B is the Bohr magneton, and g_J is the electronic g -factor. The interaction of the nuclear spin with the magnetic field is very small compared with the corresponding interaction of the electron shell, and is therefore omitted from (21).

In the general case it is impossible to diagonalize the matrix of the energy operator \mathcal{H} for arbitrary values of I , J , and H_0 , and it is consequently impossible to determine the sublevel energies as functions of H_0 . An exact solution of the problem is possible only in the region of very weak or very strong magnetic fields, which are of no particular interest from the point of view of the level-crossing technique, since crossings are observed mainly in intermediate fields for which the Zeeman splitting of the levels is comparable with the hyperfine splitting. If computer techniques are used, however, it is possible to obtain the sublevel energies also for this region, and it is possible to reproduce with definite accuracy the energy spectrum of any excited state, depending on the ΔH_0 intervals used. It should be noted that whereas until recently the experimental investigations were carried out without computers, this was due only to the simplicity of the investigated spectrum. The study of crossings of heavy elements without computers is quite unthinkable. A clear example of this is^[38], in which the spectrum of the $3d^5 4s 4p^2 {}^6P_{7/2}$ state of Mn^{55} , represented by hyperfine magnetic sublevels^[48b], was calculated.

As an example illustrating the possibilities of the crossing method for the determination of different atomic constants, let us consider the crossings of the magnetic sublevels of the $3^2P_{3/2}$ state of sodium atoms, which have been sufficiently well studied. We consider first crossings with $\Delta m = 2$.

The energy spectrum of the $2^3P_{3/2}$ state, calculated in^[51] on the basis of the Hamiltonian (21), is shown in Fig. 8 ($A = 18.8$ MHz, $B = 2.9$ MHz, $g_J = 1.3344$; a-d correspond to crossings with $\Delta m = 2$, and e and f to crossings with $\Delta m = 1$). The circles mark the crossings of the levels with $\Delta m = 2$ in a nonzero magnetic field, and the squares show crossings with $\Delta m = 1$. In addition to these crossings, there are nine crossings of the same time in a zero magnetic field (the Hanle effect). Therefore the experimentally obtained interference-signal curve is a superposition of

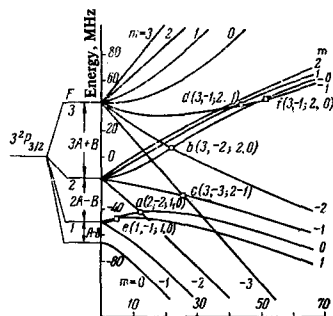


FIG. 8. Hyperfine splitting of $3^2P_{3/2}$ state of Na^{23} as the function of the magnetic field.

$F, m; F'm'$	Position of the crossing point, Oe	Signal amplitude, rel. un.
3, 3; 3, 1	0.0	0.075
3, 2; 3, 0	0.0	0.150
3, 1; 3, -1	0.0	0.180
3, 0; 3, -2	0.0	0.150
3, -1; 3, -3	0.0	0.075
2, 2; 2, 0	0.0	0.000
2, 1; 2, -1	0.0	0.000
2, 0; 2, -2	0.0	0.000
1, 1; 1, -1	0.0	0.262
2, -2; 1, 0	12.5	0.319
3, -2; 2, 0	22.5	0.135
3, -3; 2, -1	26.0	0.137
3, -1; 2, +1	44.5	0.0096

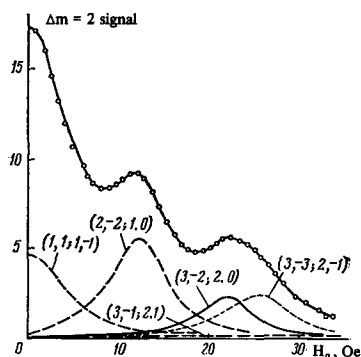


FIG. 9. Signal of crossings with $|\Delta m| = 2$.

signals of all these crossings. Figure 9 shows the dependence of the signal of crossings with $\Delta m = 2$ on the magnetic field. The circles on the upper curve show the experimental results. As seen from the plot of the obtained signal, the individual crossings are not resolved in the experiment, and only one group of crossings is slightly separated from the other. Now knowing the contributions of the individual crossings to this common signal, measurements do not make it possible to extract from this signal the expected information on the structure of the investigated spectrum. In such cases one therefore proceeds in the following manner^[38, 51]. Assuming that the expression for the interference signal (10) with allowance for (20) is true, one calculates the curves of the signals for each pair of crossing levels at definite values of the parameters A, B, τ , and r (in this case r represents the ratio of the light intensity absorbed by the atoms by the hyperfine sublevels $f = 1$ and 2 of the ground state). These signals are shown in Fig. 9. They are then added to form the theoretically calculated crossing-signal plot. By varying the parameters A, B, τ , and r , one obtains their optimal values at which the calculated resultant curve (upper curve of Fig. 9) agrees best with the experimental one. According to^[51], these parameters are $A = 18.80 \pm 0.15$ MHz, $B = 2.9 \pm 0.3$ MHz, $\tau = (1.59 \pm 0.04) \times 10^{-8}$ sec, and $r = 1.44$. The relative signal amplitude of each of the crossings and the values of the magnetic field at which crossings with $\Delta m = 2$ take place, calculated with the aid of these parameters, are listed in the table. We note for comparison that the constants A, B , and τ obtained by the method of double radio-optical resonance coincide within the limits of measurement error with those given above^[52, 53]. Experiments on crossing make it possible

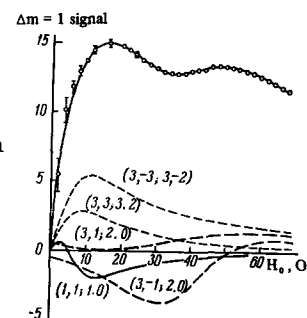


FIG. 10. Signal of crossings with $|\Delta m| = 1$.

to determine also the value of the quadrupole moment. Its value for Na^{23} was found to be^[36 a]

$$Q = (0.109 \pm 0.03) \cdot 10^{-24} \text{ cm}^2.$$

With the aid of the crossing signal for levels with $\Delta m = 1$ we can obtain the same information concerning the spectrum of the state as in the case of crossings with $\Delta m = 2$. Figure 10 shows the calculated (solid) and experimental curves obtained in the already cited paper^[51] and represent the signal of the crossing of the hyperfine sublevels of the $3^2P_{3/2}$ state of sodium. The resultant signal sums the $\Delta m = 1$ crossing signals (the notation is the same in Fig. 9) in a zero magnetic field and two signals $(1, 1; 1, 0)$ and $(3, -1; 2, 0)$ in approximate fields 5 and 52 Oe. For the calculated curve they used the same optimal parameters as for the calculation of the crossing curve with $\Delta m = 2$. For best agreement with the experimental curve, however, the value of r was taken here to be 1.28. The difference in the values of r for the considered cases is due to the difference in the experimental geometry.

The example considered demonstrates quite well the possibilities of the level-crossing procedure for the study of the spectrum of excited atomic states. To be sure, to use it successfully it is necessary to have information calculated or obtained by other methods. Thus, for example, to obtain the atomic constants given above, the value of the atomic g -factor was taken from experiments on double radio-optical resonance^[52]. Consequently, it is always desirable to combine these methods, which ensure practically the same accuracy of the experimental results. By now there are data on more than 50 excited states, obtained by these methods for systems such as $In^{[54]}$, $Yb^{[55]}$, $C^{[56]}$, $Ca^{[57]}$, $Ba^{[42]}$, $Cd^{[48]}$, $Pb^{[58]}$, $K^{[59]}$, $Cs^{[60]}$, $Hg^{[46, 61]}$, $Xe^{[62]}$ and others.

g) Methods of increasing the resolution of the crossing curves. At present we know three methods that

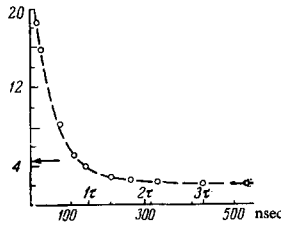


FIG. 11. Line with b as a function of the time, obtained with the aid of the gamma-ray absorber $K_4Fe(CN)_6$.

make it possible to separate, at least in part, crossing signals that are completely unresolved under the ordinary experimental conditions considered in the preceding section.

The first method was introduced into optics from nuclear physics, where it was employed for a long time to investigate the structures of nuclear states. It was developed most fully in experiments on the Mössbauer effect, in which the hyperfine structure of nuclear states was determined by resonant gamma-fluorescence^[63]. These studies have confirmed that when measuring the spectrum of photons emitted a short time θ after the formation of the excited state of the nuclei (short in comparison with the average lifetime τ), the emitted line is broader than in the normal Mössbauer experiments (on the order of Γ). The line becomes narrower, however, even narrower than the natural width, if the delay time θ is increased, i.e., if the detector registers only those quanta which were emitted after an appreciable time interval following the formation of the excited state. Physically this means that only photons emitted by nuclei with lifetimes larger than the mean value τ in this state are detected.

Figure 11 shows the dependence of the gamma-ray emission line width of Co^{57} on the decay time^[64]. We see from it that at a delay exceeding the lines become narrower than the usually measured value (the arrow shows the width of the normally measured line).

A similar phenomenon is observed for the crossing signal in resonant fluorescence of atoms. Let the probability for photon emission by two crossing levels at the instant after excitation, at a definite excitation geometry and observation geometry, be equal to^{[65] 2)}

$$P(t, \omega_0) = (1 - \cos \omega_0 t) e^{-t/\tau},$$

where ω_0 is the distance between the sublevels. If all the photons do not reach the detector by the instant θ after the excitation of the atoms (the photomultiplier is shut off), then the probability of observing the photon from this instant of time is constant:

$$P(\omega_0, \theta) = \int_0^\infty P(t, \omega_0) dt = \tau e^{-\theta/\tau} [1 - (\cos \omega_0 \theta - \sin \omega_0 \theta) (1 + \omega_0^2 \tau^2)^{-1}].$$

A plot of this function is shown in Fig. 12. It follows from the plot that starting with $\theta = \tau$ the frequency distribution of the probability has a wavy character, and the central peak becomes narrower with increasing time.

The delayed-observation principle was used to study the crossings of the levels of the $^3P_{3/2}$ state of sodium^[65]. A cell with sodium vapor was irradiated with pulses (of approximate duration 20 nsec) of reso-

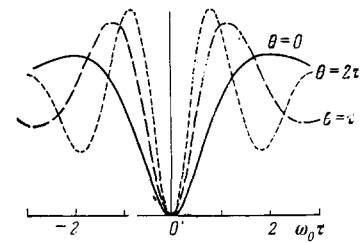


FIG. 12. The function $P(\omega_0, \theta)$ at different values of θ .

FIG. 13. Signal of crossings with $\Delta m = 2$ of the levels of the $^3P_{3/2}$ states of Na^{23} for different delay times.

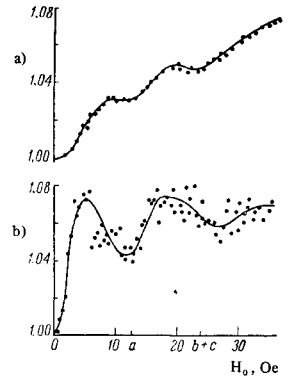
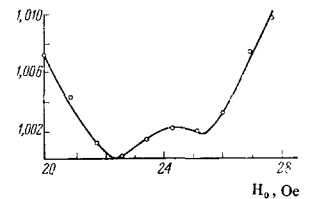


FIG. 14. Partial resolution of the crossings b and c in Na^{23} .



nant light. The fluorescence light produced at a time θ after the excitation was registered by delayed-coincidence technique. Figure 13a shows a plot of the signal of the crossings with $\Delta m = 2$ for $\theta = 0$. It shows how the crossing signal in the zero field becomes smoothed out and distorts the curves from the crossings a and b + c, making it impossible to determine the positions of the latter. The results of the experiment for $\theta = 2.5\tau$ are shown in Fig. 13b. One observes a strong narrowing of the line, corresponding to crossings in a zero magnetic field, even at a poor statistical accuracy. The experimental setup has made it possible to investigate the regions of magnetic fields near the crossings with better statistical accuracy, in comparison with the results shown in Fig. 13b. This has resulted in a partial resolution of the crossings b and c (Fig. 14), and a more accurate separation of the signal of the crossing b, the position of which corresponds to a field $H_0 = 124 \pm 0.2$ Oe, which agrees with the value calculated in^[51]. The results obtained by this method are very good. However, in view of the unusual complexity of the experimental setup, work in this direction is developing very slowly.

To understand the gist of the second method, let us turn to expression (10) for the intensity of the crossing signal. It was obtained assuming equal populations of the magnetic sublevels of the ground state, an assumption quite valid for room temperatures in the assumed excitation method. Because of this, the rate of excitation of the atoms to the crossing levels from different

2) $|\exp(i\omega_1 t) - \exp(i\omega_2 t)|^2 \exp(-t/\tau) = 2(1 - \cos \omega_0 t) \exp(-t/\tau)$.

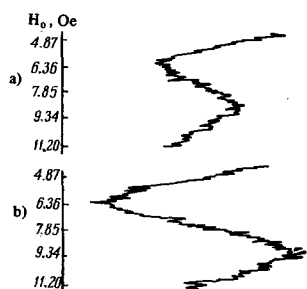


FIG. 15. Signal of the crossings b and c of the $3^2P_{3/2}P_{3/2}$ state of K^{39} .

sublevels of the ground state depends only on the probability of the optical transitions and does not depend on the populations. These conditions determine the form of the experimental crossing signal, which agrees well with the theoretical calculations, as showed earlier. Naturally, the character of this signal changes if the populations of the Zeeman sublevels of the ground state are significantly different. In this case the rates of populations of different sublevels of the excited state change, and consequently also the values of the relative signals of the individual crossings. This question was already discussed in Sec. (a) of the present chapter. The most effective methods of producing nonequilibrium distribution of the ground-state atoms over the sublevels is the optical-orientation method. The described method was first used to study the crossings of K^{39} ^[66]. Its spectrum is the same as for sodium, and crossings with $\Delta m = 2$ are observed in fields 3.02, 6.75, 8.21, and 13.21 Oe (a, b, c, and d, respectively).

From the selection rules and the conditions for coherent excitation of the atoms in the "crossing," it follows that the contribution made to the resonant fluorescence from the level crossing is determined in final analysis by the absorption of light by the atoms at definite sublevels of the ground state. In the case of alkali metals, the crossings a and b are connected with absorption of light on the levels ($f = 2, \mu = -1$) and ($1, -1$) of the ground state, while the crossing is connected with the level ($2, -2$). A selective increase of the populations of the magnetic sublevels with quantum numbers $\mu = -1$ and -2 leads to an increase of the relative signal of the aforementioned crossings. The experimental geometry is indeed chosen to take this circumstance into account. In addition to the linearly polarized light that propagates in the H_0 direction and excites the atoms into a coherent superposition state, circularly polarized radiation, also in the H_0 direction, is applied to the system to produce optical orientation of the atoms.

Figure 15a shows the signal of the crossings b and c without pumping (optical-orientation) light, and Fig. 15b shows the same in the presence of an orienting signal beam. In the latter case, the crossing signal was increased 2.5 times in comparison with the former. At the same time, its position changed and shifted into the region of stronger magnetic fields. This is due to the increased contribution of the crossing c to the common signal $b + c$.

By the same token, this demonstrates one more possibility of resolving crossing signals (to be sure, without additional quantitative estimates), which is worthy of certain attention because of its obvious simplicity.

It is possible that under certain physical conditions, discussed for example in^[67], single crossings can also be separated by this method. But no such investigations have been reported so far.

Finally, one more method was proposed quite recently, based on the change of the relative intensities of the hyperfine components of the exciting light^[68]. It makes it possible to amplify one group of crossings and simultaneously attenuate others, if their occurrence is due to absorption of different hyperfine components of the resonant radiation. Applications of this method can be found in^[36d].

4. ANTICROSSINGS OF ATOMIC LEVELS³⁾

Interference effects arise in resonant fluorescence not only when the atomic levels cross, but also when the distance between them becomes smaller than or of the order of their natural width in the presence of a static perturbation V that couples these levels. In atomic spectroscopy, this phenomenon was predicted by Podgoretskii^[60] and was observed relatively recently^[6, 69]. In an investigation of crossings of the 2^3P levels of lithium, the authors of^[6, 69] observed an appreciable asymmetry of the expected crossing signal, something that could not be explained within the framework of the crossing theory. Further investigations have shown that this asymmetry could be traced in the presence of an initially unnoticed interference signal due to anticrossing of the atomic sublevels.

Anticrossings of energy levels have been known in atomic physics a long time ago. Their theory as an independent problem has been developed most completely in^[19, 69], and we shall therefore consider only some general ideas and simple calculations for a qualitative interpretation of the results obtained with the aid of a fluorescence signal. Let $\mathcal{H} = \mathcal{H}_0 + V$ be the Hamiltonian of the system, where V is the operator of a time-independent perturbation coupling the states $|a\rangle$ and $|b\rangle$ of the unperturbed Hamiltonian \mathcal{H}_0 . As the initial zeroth approximation we can replace $|a\rangle$ and $|b\rangle$ by linear combinations of the type

$$|\psi\rangle = a|a\rangle + b|b\rangle.$$

Substituting this expression in the "perturbed" Schrödinger equation

$$\mathcal{H}|\psi\rangle = E|\psi\rangle,$$

we obtain after simple transformations the sought eigenvalues of the energy in the first approximation in the perturbation

$$E = \langle \frac{1}{2} (\mathcal{H}_{aa} + \mathcal{H}_{bb}) \pm \langle \frac{1}{4} (\mathcal{H}_{aa} - \mathcal{H}_{bb})^2 + |V_{ab}|^2 \rangle^{1/2}. \quad (22)$$

If the energy values of both terms become different (the levels cross), this means that both values of E determined by (22) coincide. To this end it is necessary that the radicand in (22) vanish, i.e.,

$$\begin{aligned} \mathcal{H}_{aa} - \mathcal{H}_{bb} &= E_a - E_b + V_{aa} \\ -V_{bb} &= 0, \quad V_{ab} = 0. \end{aligned}$$

These equations are satisfied simultaneously if $V = 0$. We see therefore that if there are transitions between

³⁾An extensive bibliography on the questions considered here is given in the review [7].

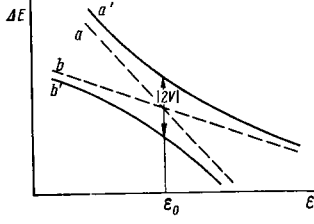


FIG. 16. Position of the perturbed states $|a'\rangle$ and $|b'\rangle$ as functions of $\mathcal{H}_{aa} - \mathcal{H}_{bb} = \epsilon$ the shortest distance between levels is $\epsilon = \epsilon_0$.

the two states ($V_{ab} \neq 0$), when the level crossing that could take place in the absence of the perturbation vanishes, and the distance between them becomes equal to

$$\Delta E = [(E_a - V_{aa} - E_b + V_{bb})^2 + 4 |V_{ab}|^2]^{1/2}.$$

Figure 16 shows a diagram of two anticrossing levels. The symbols a and b pertain to levels and wave functions in the absence of coupling, and a' and b' to levels and states when $V \neq 0$. The levels a and b (dashed lines) cross at $E_a = E_b$, whereas a' and b' do not cross. For this case, the wave function of each state is given by

$$\begin{aligned} |a'\rangle &= \cos(\beta/2) |a\rangle + \sin(\beta/2) |b\rangle, \\ |b'\rangle &= -\sin(\beta/2) |a\rangle + \cos(\beta/2) |b\rangle, \\ \text{tg } \beta &= 2V_{ab}/\epsilon, \quad \epsilon = E_a - E_b - V_{aa} + V_{bb}. \end{aligned} \quad (23)$$

For $\beta = \pi/2$ it is a 50% mixture of the wave functions of uncoupled states. It follows therefore that in the presence of a perturbation coupling the states $|a\rangle$ and $|b\rangle$, even incoherent excitation of these pure states leads to the occurrence of superposition states (23) in the anticrossing region. This is indeed the main reason why anticrossing can be observed for levels whose coherent excitation is forbidden by the selection rules and for which it is impossible to use the crossing method in principle. Whereas in experiments on level crossing the "coupling" between the corresponding magnetic sublevels is part and parcel of coherent optical excitation, in experiments on anticrossing it results from a certain static perturbation (in the double radio-optical resonance this coupling is effected by a radio-frequency field of suitable frequency)⁴.

These qualitative results with respect to the character of the resonant forces in the anticrossing state of atomic levels are confirmed by the theory^[96]. Let $|a\rangle$ and $|b\rangle$ be the eigenstates of the operator \mathcal{H}_0 with energies ω_a and ω_b , and let V , as before, be the binding operator of these states. Then the change of the excited state of the atom $|t\rangle$ with time is determined by the equation

$$i \frac{\partial}{\partial t} |t\rangle = (\mathcal{H}_0 + \mathcal{H}_D + V) |t\rangle, \quad (24)$$

where the damping of the states $|a\rangle$ and $|b\rangle$ is taken into account by introducing an operator \mathcal{H}_D , having eigenvalues $-i\Gamma_a/2$ and $-i\Gamma_b/2$ in the \mathcal{H}_0 representation. The solution of (24) is sought in the form

⁴It can be shown that from the point of view of double radio-optical resonance the transition in experiments on anticrossing are induced by a transverse magnetic field, which can be represented in the form of two components that rotate in opposite directions with zero frequency^[70]. In other words, double resonance in a rotating coordinate system can be represented as anticrossing of levels.

$$|t\rangle = a(t) e^{-i\omega_a t} + b(t) e^{-i\omega_b t}$$

under the condition that the atom was excited at the instant $t = 0$. We then obtain in place of (24)

$$i\dot{a} = -i(\Gamma_a/2)a + 2\pi V e^{i\omega t} b, \quad i\dot{b} = -i(\Gamma_b/2)b + 2\pi V^* e^{-i\omega t} a, \quad (25)$$

with

$$\omega = \omega_a - \omega_b, \quad V = \langle a | V | b \rangle / 2\pi.$$

Substitution of the obvious solution

$$a = \sum_{n=1}^2 A_n e^{(\alpha_n + i\omega_a)t}, \quad b = \sum_{n=1}^2 B_n e^{(\alpha_n + i\omega_b)t}$$

in (25) yields

$$\begin{aligned} \alpha_{1,2} &= -[(1/4)(\Gamma_a + \Gamma_b) + (1/2)i(\omega_a + \omega_b)] \\ &\quad \pm \{[(1/4)(\Gamma_a - \Gamma_b) + (1/2)i\omega]^2 - |2\pi V|^2\}^{1/2}, \\ B_n &= (A_n/2\pi V) i(\alpha_n + i\omega_a + (1/2)\Gamma_a), \\ A_1 &= -[(\alpha_2 + i\omega_2 + (1/2)\Gamma_a) f_{a\mu} + 2\pi i V f_{b\mu}] / (\alpha_1 - \alpha_2), \\ A_2 &= [(\alpha_1 + i\omega_1 + (1/2)\Gamma_a) f_{a\mu} + 2\pi i V f_{b\mu}] / (\alpha_1 - \alpha_2), \end{aligned} \quad (26)$$

where it was assumed that at the instant of time $t = 0$ we have $a(0) = f_{a\mu}$, $b(0) = f_{b\mu}$, where f is, apart from an inessential constant, the polarization vector of the exciting light and $|\mu\rangle$ is a magnetic sublevel of the ground state.

The fluorescence intensity is proportional to the square of the matrix element $\langle \mu' | \mathbf{g} \cdot \mathbf{r} | t \rangle$, where \mathbf{g} is the polarization vector of the emitted light. To find the stationary emission signal from atoms excited by the instant $t = 0$ with equal rate, it is necessary to replace t by $t - t_0$ and integrate $|\langle \mu' | \mathbf{g} \cdot \mathbf{r} | t - t_0 \rangle|^2$ with respect to t_0 from $-\infty$ to t . Taking (26) into account, this yields

$$\begin{aligned} S = & \Gamma_a^{-1} \sum |f_a|^2 |g_a|^2 + \Gamma_b^{-1} \sum |f_b|^2 |g_b|^2 \\ & + (\Gamma_a \Gamma_b / \gamma D) \sum (f_a f_b^* g_a g_b^* + \text{compl. conj.}) \\ & - i \Gamma_a \Gamma_b \Delta / (\gamma D) \sum (f_a f_b^* g_a g_b^* + \text{compl. conj.}) \\ & - (2\Gamma_a \Gamma_b / \gamma D) |V|^2 \sum [fg] + (2/\gamma^2 D) \sum (V^* f_a f_b^* + V f_a^* f_b) (V^* g_a g_b^* + V g_a^* g_b) \\ & + (\Delta \Gamma_a \Gamma_b / \gamma^2 D) \sum [f(V g_a g_b^* + V g_a^* g_b) + g(V^* f_a f_b^* + V f_a^* f_b)] \\ & + i(\Gamma_a \Gamma_b / \gamma D) \sum [f(V g_a g_b^* - V^* g_a^* g_b) + g(V^* f_a f_b^* - V f_a^* f_b)], \end{aligned} \quad (27)$$

where

$$D = \Gamma_a \Gamma_b + 2 |V|^2 + |\Gamma_a \Gamma_b \Delta|^2 / \gamma^2,$$

$$f = (|f_a|^2 / \Gamma_a) - (|f_b|^2 / \Gamma_b), \quad g = (|g_a|^2 / \Gamma_a) - (|g_b|^2 / \Gamma_b),$$

$$\gamma = (\Gamma_a + \Gamma_b) / 2.$$

The summation is carried out here over the magnetic quantum numbers of the ground state.

The first two terms in this expression represent the previously considered incoherent radiation, constituting the background. The third and fourth describe the already known crossing signal corrected with allowance for the perturbation V , the observation of which calls for coherent excitation and detection. This known criterion of the crossing signal means that the optical excitation should transform the atoms into the states $|a\rangle$ and $|b\rangle$ from one and the same sublevel or from several sublevels of the ground state, and the detector should "observe" the transitions from $|a\rangle$ and $|b\rangle$ to one and the same or to several sublevels of the final state.

The fifth term in (27) is the only pure anticrossing signal. Its observation requires neither coherent exci-

tation nor coherent detection. Moreover, it becomes maximal when only one of the states $|a\rangle$ or $|b\rangle$ is excited, and one detects radiation from only one of them. If $\Gamma_a = \Gamma_b$ and the excited sublevels are equally populated, then $\sum |f_a|^2 = \sum |f_b|^2$ and the anti-precession signal vanishes. It vanishes also in the case when it is impossible to separate the radiation from the sublevels $|a\rangle$ and $|b\rangle$ during the detection, since $\sum |g_a|^2 = \sum |g_b|^2$, and consequently also $g = 0$.

The last three terms of (27) describe radiation constituting a mixture of crossing and anticrossing signals, which require for their observation both $V \neq 0$ and coherence on one of the sections of the resonance-fluorescence processes, either in the excitation channel or in the detection channel. Recently such signals were observed experimentally^[71].

When there is no perturbation, the last four terms of (27) drop out and there remains the crossing signal i.e., in the zeroth approximation in V . If $\Gamma_a = 0$ or $\Gamma_b = 0$, i.e., one of the sublevels is nonradiative, then all but the first two terms of (27) vanish. These are the general characteristics of the fluorescence light, which contains information on the crossings and anticrossings in the presence of a static perturbation coupling the states under consideration.

To explain the distinguishing features of the anticrossing signal and to assess the feasibility of using this effect for the spectroscopy of atomic states, let us consider certain experimental results obtained for lithium atoms. In the 2P state of lithium there are two crossings of the fine-structure levels: ($J = 3/2$, $m_J = -3/2$) - ($J = 1/2$, $m_J = 1/2$) in a 3200-Oe field and ($3/2$, $-3/2$) - ($1/2$, $-1/2$) in a 4800-Oe field (Fig. 17). The last crossing pertains to the region of strong magnetic fields. However, since the spin of the lithium nucleus differs from zero, $I(\text{Li}^6) = 1$, $I(\text{Li}^7) = 3/2$, the hyperfine interaction causes each m_J level of the fine structure to split into $2I + 1$ magnetic sublevels. Figure 18 shows the energy spectrum of the Li^7 atoms in the crossing region of the levels with $m_J = -1/2$ and $m_J = -3/2$, with allowance for the hyperfine interac-

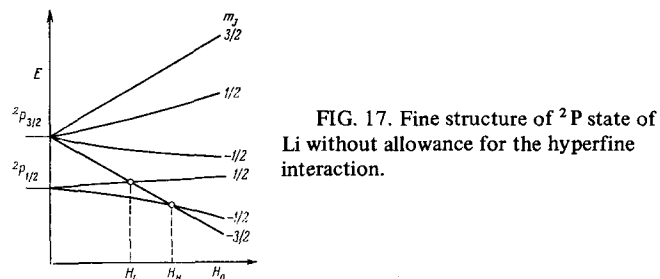


FIG. 17. Fine structure of 2P state of Li without allowance for the hyperfine interaction.

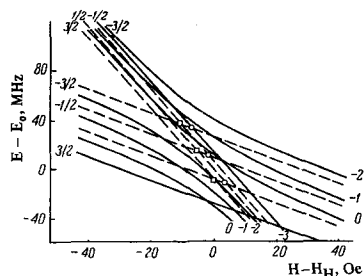


FIG. 18. Hyperfine structure of Li^7 atoms in 2P state.

tion. The levels obtained neglecting the coupling perturbation are designated on the left in the figure by the quantum numbers m_J , while the real levels are characterized by the numbers $m_F = m_I = m_J$ and are shown by solid lines. The coupling leads to a unique repulsion of these levels whose quantum numbers m_F are identical but the m_J are different. From this condition one finds the three anticrossing positions marked by squares in the figure. The circles mark the crossing positions that would be observed in the absence of this coupling. Although the crossing signal is nevertheless observed (in accord with (27)), it is distorted by the shift of the crossing positions and also by the fact that the wave functions of six out of eight states are strongly altered in the crossing region. This means that it is very difficult to extract information concerning the fine and hyperfine structures from the crossing signal in a strong magnetic field. Fortunately, the anticrossing signal is not distorted and can be observed in pure form. Since a pair of levels of each of the three crossings has different m_J , they cannot be either excited from one sublevel of the ground state, nor decay to it, for in strong magnetic fields this is forbidden by the selection rules for the electric dipole transitions. Taking this fact and the condition $\Gamma_a = \Gamma_b = \Gamma$ into account, the expression for the anticrossing signal becomes simpler for the interpretation of the experimental results:

$$S_a = -[2|V|^2/\Gamma(\Gamma^2 + \Delta^2 + 2|V|^2)] \left[\sum_{\mu} |f_{a\mu}|^2 - \sum_{\mu} |f_{b\mu}|^2 \right] \left[\sum_{\mu} |g_{a\mu}|^2 - \sum_{\mu} |g_{b\mu}|^2 \right], \quad (28)$$

where the sum over μ and μ' do not overlap.

The general anticrossing signal is a superposition of three such signals (with strong overlap) for Li^7 and two such signals for Li^6 . It is interesting to note that according to (28) the signal has a Lorentz shape for any geometry of the experiment; the mixing of the Lorentz and the dispersion curves, which takes place in experiments on level crossing, does not occur here. The dependence of the anticrossing signal on the polarization of the exciting and detected light can be determined as a result of an estimate of the matrix elements f_a , as was done in Sec. (a) of Chap. 3. For the case when the propagation vectors of the exciting and re-radiated light are perpendicular to each and to the direction of the constant magnetic field H_0 we have

$$S_a \sim (3\cos^2\theta - 1)(3\cos^2\theta' - 1), \quad (29)$$

where θ and θ' are the angles between H_0 and the planes of polarization of the incident and detected light. To be sure, there is a certain discrepancy between this dependence and that obtained in experiment. We leave out here the discussion of this question, which is considered in detail in^[69].

On the basis of (27), we can estimate the dependence of the crossing signals for the same experimental geometry in the absence of a perturbation:

$$S_F \sim \sin 2\theta \cdot \sin 2\theta'. \quad (30)$$

The perturbation distorts the signal line shape, but does not change the character of the dependence of S_F on θ and θ' . Equations (29) and (30) lead to a very important conclusion: a pure anticrossing signal can

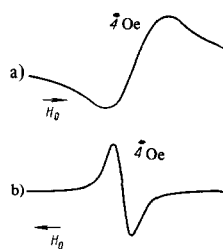


FIG. 19. Plots of anticrossing signal in the 2P stages of the atoms Li^7 (a) and Li^6 (b).

be obtained for θ or $\theta' = \theta$ or $\pi/2$, while a pure crossing signal is obtained at θ or $\theta' = \cos^{-1}(\sqrt{3/3})$. These conditions determine the choice of the experimental geometry in the study of anticrossing of levels in pure form.

Figure 19 shows plots of pure anticrossing signals for Li^7 and Li^6 . The width of the curve for Li^6 was found to be 18.22 ± 0.20 MHz, and that of Li^7 was 58.71 ± 0.67 MHz. If the level width Γ is known, then V can be determined from the width of the observed signal. For Li^6 and Li^7 , the interaction is equal to

$$V(\text{Li}^6) = 7.33 \pm 0.08 \text{ MHz},$$

$$V(\text{Li}^7) = 25.25 \pm 0.30 \text{ MHz}.$$

Theoretical estimates lead to the same results.

Thus, by using the anticrossing technique we can successfully determine the structure of the excited atomic states when the crossing technique is not applicable or gives unsatisfactory results. Its applications can be quite varied. With the aid of the anticrossing signal one can investigate the first and second orders of the Stark interaction in atoms and the Lamb shift^[72], the character of the interaction between states with different orbital angular momenta^[73], and also a steady excited state which are not connected with the fundamental electric dipole transitions. For alkali metals, such a state is the B-state, which is populated upon decay of the S state. The ordinary crossing experiment becomes impossible to perform because of the almost complete loss of the angular correlations, due to the additional step in the process of V-state formation. Although the foregoing analysis pertains to free atoms, one must not think that the anticrossing method cannot be used for condensed media. For example, it is shown in^[74] that the anticrossing signal can be used to study crystalline fields in solids by determining the perturbation of the narrow energy levels of substituted rare-earth ions.

One must not forget that the light itself can be regarded as a perturbation that couples magnetic sublevels, and consequently can lead to a lifting of the crossing. Thus, in the experiment of Dupont-Roc et al.^[75] they succeeded in observing a coherent superposition of the states of Hg^{199} in a zero magnetic field with lifting of the degeneracy over the atomic sublevels of the ground state by an additional optical nonresonant excitation. This question is described in detail in the review^[17].

It should be noted in conclusion that, in principle, crossings are never realized in atomic systems, since all the states of the atom are coupled with one another in a definite order in the magnitude of the perturbation, determined by the random electric and magnetic fields. If a crossing signal is observed, this is due to the

smallness of V in comparison with Γ_a and Γ_b . In this case the signal apparently differs from the signal at $V = 0$.

5. CROSSING OF LEVELS IN THE GROUND STATE

If interference of states is one of the general laws of nature, would it not be possible to produce coherence in an atomic system also by crossing of magnetic sublevels of the ground state, and if it were possible, under what conditions and how could it be observed^[22]? Experiments^[11] and theory^[15a] provide an affirmative answer to this question; moreover, the conditions for its production are analogous to those considered for the case of an excited state: the distance between the crossing sublevels should be smaller than or of the order of their width, and the optical excitation must be coherent.

On the other hand, in experience of optical orientation of atoms these conditions determine the possibility of realizing the so-called transverse pumping, that is, orientation of the atoms in a direction perpendicular to the direction of the constant magnetic field H_0 ^[15b]. Using the formalism of the theory of optical orientation^[15a,76], we can obtain an equation describing the change of the coherence $\rho_{\mu\mu'}$ of the ground state of atoms, determined by the field H_0 and the properties of the optical excitation, and show that the Hanle effect in the ground state and the transverse optical pumping are similar phenomena.

We confine ourselves here to the case of crossing in a zero field, which is customarily called the "Hanle effect in the ground state." This case admits of a very simple and intuitive interpretation of the results on the basis of a semiclassical model of the phenomenon of optical orientation of atoms^[16,77].

a) Transverse "pumping." We assume for simplicity that the investigated atoms can be located at two Zeeman sublevels both in the ground and in the excited states. Let the resonance cell be illuminated in a zero magnetic field by circularly (σ^+) polarized light propagating in the direction of the Ox axis. Since the only physically preferred direction is the light-beam propagation direction, we take Ox to be the quantization axis. Figure 20 shows the energy levels of the atoms in a zero field (the magnetic quantum numbers m of the excited states and μ of the ground state pertain to the Ox axis). Under thermal equilibrium conditions and in the absence of optical transitions, the two sublevels with $\mu = \pm 1/2$ are equally populated and the total magnetization of the vapor \mathbf{M} in the ground state is equal to zero.

Owing to the σ^+ polarization of the exciting light

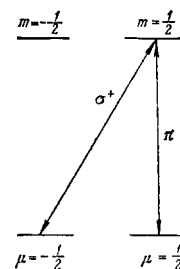


FIG. 20. Scheme of Zeeman sublevels of the ground and excited states.

(incoherent excitation), the atoms situated at the sublevel with $\mu = -1/2$ absorb photons and go over to an excited sublevel with $m = 1/2$. Then, as the result of the decay of the excited state, they return to the ground state, partly to the sublevel with $\mu = -1/2$ and partly to the sublevel with $\mu = 1/2$. The net result of this cycle is a "pumping over" of atoms from the sublevel with $\mu = -1/2$ to the sublevel with $\mu = 1/2$, whose populations thus become unequal, which leads in turn to the appearance of magnetization along the Ox axis. In other words, some of the angular momentum of the radiation field is transferred to the atoms. If we neglect all the relaxation processes, then after a definite number of "pumping" cycles all the atoms will go over to the sublevel with $\mu = 1/2$ and the magnetization in the Ox direction reaches the saturation value M_0 . If we denote by $d^{(1)}M/dt$ the rate of change of M under the influence of the pump light only, we easily obtain the equation

$$d^{(1)}M/dt = T_R^{-1}(M_0 - M), \quad (31)$$

i.e., M tends to M_0 with a characteristic constant T_R , called the pumping time and dependent on the radiation intensity. We note that the intensity S_A of the absorbed light is proportional to the number of atoms capable of absorbing the σ^+ light, in other words, to the number of atoms at the sublevel with $\mu = -1/2$. It is equal to zero when $M = M_0$, and is maximum when $M = -M_0$. It is easy to verify that the intensity of the absorbed light S_A is proportional to the quantity $(M_0 - M_x T_R)$, or apart from a constant simply to M_x .

During the course of the thermal relaxation, M tends to an equilibrium value, that is, to zero, with a time constant T . If we denote the rate of change of M as the result of a thermal relaxation by $dM/dt = -M/T$, then the equation for the magnetization, with allowance for (31) can now be written in the form

$$dM/dt = \tau^{-1}(M'_0 - M), \quad (32)$$

where $\tau^{-1} = T^{-1} + T_R^{-1}$ is the overall time constant, describing the combined effect of optical and thermal relaxation. $M'_0 = M_0 T / T_R$ is the overall magnetization obtained in the stationary regime. It becomes equal to M_0 if $1/T_R > 1/T$, i.e., if the optical pumping precedes the thermal relaxation.

If the spin system is in a magnetic field H_0 , then (32) is replaced by

$$dM/dt = \tau^{-1}(M'_0 - M) + \gamma[MH_0]. \quad (33)^*$$

This equation has the same structure as Bloch's equation. However, the nature of the occurrence of M'_0 is different in the present case, namely, in Bloch's equations the magnetization in thermal equilibrium, determined by the field H_0 , is parallel to the latter, whereas in the present case M'_0 is not parallel to H_0 . If we choose $H_0 \parallel Oz$, then $M'_0 \perp H_0$, and we speak in this case of transverse pumping.

The last equation can be derived by a rigorous quantum-mechanical analysis. We have used here a semiclassical analysis, since in quantum theory the quantities τ and M'_0 are regarded as constants characterizing the physical system under definite conditions.

* $[MH_0] \equiv M \times H_0$.

b) **Hanle effect in the ground state.** Let us consider transverse pumping in a magnetic field H_0 that varies slowly about a zero value. Circularly polarized light propagates in the Ox direction, and we detect the absorbed-light intensity S_A , which is proportional to M_x . It is necessary to determine the change in the absorption signal with changing field H_0 .

To this end we find the stationary solution (33). Making the substitution $\omega_0 = \gamma H_0$, $M_{\pm} = M_x \pm iM_y$ in (33), we obtain

$$dM_{\pm}/dt = (M'_0/\tau) - (M_{\pm}/\tau) \mp i\omega_0 M_{\pm}, \quad dM_z/dt = -M_z/\tau.$$

If $\dot{M}_{\pm} = \dot{M}_z = 0$, then we obtain directly from the preceding equations

$$M_z = 0, \quad M_{\pm} = M'_0/(1 + i\omega_0\tau),$$

or

$$M_x = M'_0/(1 + \omega_0^2\tau^2), \quad M_y = -M'_0\omega_0\tau/(1 + \omega_0^2\tau^2), \quad M_z = 0.$$

The component M_z is always equal to zero, while M_x and M_y represent the absorption and dispersion curves as function of $\omega_0\tau$ with center at zero field and with line width $\Delta = 2$ or $\Delta\omega = 2/\tau$. Thus, the quantity

$$\Delta H_0 = 2/\gamma\tau, \quad (34)$$

which is directly proportional to the width of the magnetic sublevels, determines the signal line width in Oe. In a zero field, M_y and M_z are equal to zero, and $M_x = M_0$, i.e., to the magnetization in the direction of Ox, as we have already seen earlier. In a strong field ($\omega_0\tau \gg 1$), all three components of M are equal to zero, and the optical excitation does not produce a preferred orientation of the atoms. These results can be understood on the basis of the following analysis. Pumping with a light beam produces a system of atomic spins oriented in the Ox direction. After they are oriented, they begin to precess in the (x, y) plane with a frequency ω_0 about the direction of the field H_0 . However, their orientation becomes perturbed in time (with a time constant τ), owing to the optical and thermal relaxation mechanisms. If the field is such that within the lifetime $\sim\tau$ the spins execute many revolutions in the (x, y) plane ($\omega_0\tau \gg 1$), then the resultant magnetization, averaged over this time interval, is equal to zero. On the other hand, if $\omega_0\tau \ll 1$, that is, the external field is very weak in comparison with the half-width $1/\gamma\tau$ of the crossing signal determined by (34), and consequently also by the level width, then all the vectors are practically parallel to one another. As a result we obtain maximum magnetization in the direction of the Ox axis. Notice should be taken here of the importance of the transverse pump direction with respect to H_0 : It is precisely in this case that M_x and M_y change near $H_0 = 0$. If the pump is longitudinal, ($H_0 \parallel Ox$), then the spins are oriented along the Ox axis and remain immobile regardless of the value of the magnetic field.

We shall now attempt to connect the obtained results with coherence of the ground state. With respect to the Ox axis, the optical excitation is incoherent. However, with respect to the magnetic field, it is coherent and produces in the optical-pumping cycle a coherent superposition of states $|\xi\rangle$ and $|\xi'\rangle$, if $\omega_0\tau \lesssim 1$ (ξ and ξ' characterize the magnetic sublevels with respect to

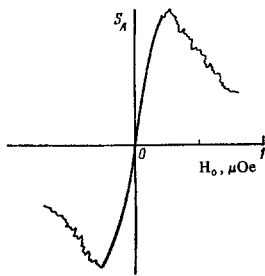


FIG. 21. Crossing signal of magnetic sublevels of the ground state of Rb^{87} .

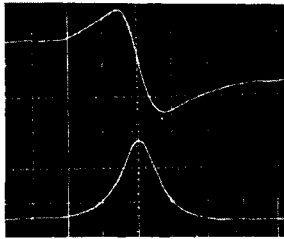


FIG. 22. Crossing signals in the ground state of Cd, revealed by fluorescence light.

the field H_0). Indeed, the mean value of the magnetization, determined with the aid of the density matrix $\rho_{\xi\xi'}$, is precisely $M_x = M'_0$, which differs from zero if the presented inequality is valid. Consequently, under the same conditions, the off-diagonal matrix elements $\rho_{\xi\xi'}$, which indicate the existence of coherence in the atomic system, differ from zero.

Thus, the Hanle effect in the ground state is observed in coherent optical excitation, provided $\omega_0\tau \lesssim 1$ in this case.

In contrast to the Hanle effect in the excited state, the crossing signal of the levels in a zero field of the ground state is characterized by an exceedingly narrow line. Let us examine by way of example the atoms Hg and Cd. For these, the paramagnetism in the ground state is purely nuclear: $\gamma/2\pi \sim 1$ kHz/Oe. If we assume $\tau \sim 1$ sec, which is of the order of the usual relaxation time for cells with paraffin coating, then, according to (34), ΔH_0 is of the order of several millioersted. For alkali metals, paramagnetism has an electronic nature, γ is about a thousand times larger, and ΔH_0 is of the order of microersted. It is easy to see that this yields a very sensitive method for the study of the Zeeman structure of atoms and nuclei in weak magnetic fields^[14,79].

The crossing of magnetic sublevels of the ground state of Rb^{87} atoms was recently observed experimentally^[9,14]. A plot of the signal is shown in Fig. 21. We see from it that the line width is very small, $\Delta H_0 \sim 0.5 \mu\text{Oe}$. This enables us to measure magnetic fields accurate to 10^{-9} Oe^[14].

Finally, we wish to note an interesting feature observed in experiments on the Hanle effect in the ground state. The production of coherence in crossing of ground-state levels leads to a dependence of the density matrix $\rho_{mm'}$ of the excited state on the off-diagonal elements $\rho_{\xi\xi'}$ of the ground state (formula (7)). This means that the crossing signal in the ground state can be detected by the fluorescence light. The phenomenon of coherence transfer from the ground state to an excited state has been known for a long time in the region of the double radio-optical resonance and was used many times to study the influence of the ground state

on the behavior of excited atoms. The feasibility of using this phenomenon in experiments on the Hanle effect was recently confirmed in^[79]. Figure 22 shows dispersion and absorption curves of the magnetic-sublevel crossing signals for the ground state of Cd, obtained with the aid of resonance fluorescence. The results of this study offer evidence that optical excitations can introduce coherence into the excited state in two ways: (a) if it is coherent and the levels in the excited states cross, (b) if coherence exists in the ground state.

It should be noted that transfer of coherence from the ground state to the excited state can take place both for coherent and incoherent irradiation. If the coherence in the ground state is induced with light, then the excitation should be coherent, as in the case considered above.

¹W. Hanle, *Naturwiss.* 11, 691 (1923); *Zs. Phys.* 30, 93 (1924); 35, 346 (1926); 41, 164 (1927); 85, 300 (1933); W. Hanle, R. Pepperl, *Phys. Bl.* 27, 19 (1971).

²P. Soleillet, *C. R. Ac. Sci.* 185, 198 (1927); 187, 212 (1928); *J. Phys. Rad.* 7, 478 (1936).

³G. Breit, a) *Rev. Mod. Phys.* 5, 91 (1933); b) *Phys. Rev.* 46, 590 (1934).

⁴A. C. G. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms*, Macmillan, 1934, Chap. 5; P. P. Feofilov, *Polyarizovannaya lyuminescentsiya atomov, molekul i kristallov* (Polarized Luminescence of Atoms, Molecules, and Crystals) Fizmatgiz, 1959, Chap. 2.

⁵a) R. Sands et al., *The Ann Arbor Conference on Optical Pumping* (Ann Arbor, Michigan, June 15–18, 1959), Ann. Arbor, Univ. of Michigan, p. 184;

b) P. A. Franken, *Phys. Rev.* 121, 508 (1961).

⁶T. Eck et al., *Phys. Rev. Lett.* 10, 239 (1963).

⁷G. Putlitz, *Atomic Physics* (Proc. of the 1st Intern. Conference on Atomic Physics (New York, 1968)), ed. by B. Bederson et al., N. Y., Plenum Press, 1969, p. 227.

⁸a) M. I. Podgoretskiĭ, *JINR Communication* (Soobshchenie) R-491, Dubna, 1960; O. A. Khrustalev, *JINR Preprint* R-574, Dubna, 1960; M. I. Podgoretskiĭ and O. A. Khrustalev, *Usp. Fiz. Nauk* 81, 217 (1963) [*Sov. Phys.-Usp.* 6, 682 (1964)]; L. G. Zastavenko and M. I. Podgoretskiĭ, *Zh. Eksp. Teor. Fiz.* 45, 706 (1968) [*Sov. Phys.-JETP* 18, 485 (1969)]; *JINR Communication* (Soobshchenie) R-1222, Dubna, 1963; c) L. G. Zastavenko and O. A. Khrustalev, *JINR Commun.* R-629, Dubna, 1960; d) L. G. Zastavenko and O. A. Khrustalev, *Opt. i spektr.* 11, 441 (1961); V. A. Onishchuk and M. I. Podgoretskiĭ, *JINR Commun.* R-2897, Dubna, 1966; *Opt. i spektr.* 24, 170 (1968).

⁹J. Dupont-Roc et al., *Phys. Lett.* A26, 638 (1969).

¹⁰E. Geneux et al., *Adv. Electron. and Electron. Phys.* 27, 19 (1969).

¹¹A. Kastler, *C. R. Ac. Sci.* 252, 2396 (1961).

¹²V. G. Baryshevskiĭ and M. I. Podgoretskiĭ, *JINR Communications* (Soobshchenie) R4-5786, Dubna, 1971.

¹³L. I. Mandel'shtam, *Collected Works* (in Russian), Vol. 4, AN SSSR, 1955, *Lecture on Oscillations* No. 23, p. 234.

¹⁴Colloque Intern. (Champ Magnetiques Faibles

- d'Interet Geophysique et Spatial), *Rev. Phys. Appl.* 5 (1), 95, 102 (1970).
- ¹⁵C. Cohen-Tannoudji, a) Thèse (Paris, 1962), b) *C. R. Ac. Sci.* 258, 4463 (1964).
- ¹⁶F. Bertin, *Principles of Quantum Electronics* (Russ. transl.) Mir, 1971, Chaps. 4-5.
- ¹⁷L. N. Novikov, V. G. Pokozan'ev, and G. V. Skrotskiĭ, *Usp. Fiz. Nauk* 101, 273 (1970) [*Sov. Phys.-Usp.* 13, 384 (1970)].
- ¹⁸J. C. Pebay-Peyroula, Thèse, *J. Phys. Rad. L'Univ. de Paris, ser. A, Nr. 3296 et 4168* (1959); *J. de Phys.* 20, 669 (1959).
- ¹⁹L. D. Landau and E. M. Lifshitz, *Kvantovaya mekhanika* (Quantum Mechanics), 2nd ed., Fizmatgiz, 1963, Chap. XIV [Addison-Wesley, 1965]; cf. also^[16], p. 607.
- ²⁰J. Brosnel, *Advances in Quantum Electronics*, J. R. Singer, ed., 1961, p. 95.
- ²¹O. Nédélec et al., *C. R. Ac. Sci.* 257, 3130 (1963); *J. P. Descoubes*, *ibid.* 259, 3733 (1964).
- ²²M. Dumont and B. Decomps, *J. de Phys.* 29, 181 (1968).
- ²³J. Meunier et al., *C. R. Ac. Sci.* 261, 5033 (1965).
- ²⁴W. Furssov and A. Wlassow, *Phys. Zs. Sowjetunion* 10, 378 (1936).
- ²⁵M. Grance and A. Omont, *C. R. Ac. Sci.* B269, 1230 (1969).
- ²⁶a) J. Barrat, J. Brosnel, *C. R. Ac. Sci.* 246, 2744 (1958); E. Otten, A. Winnacker, *Phys. Lett.* 23, 462 (1966); b) J. Barrat, *C. R. Ac. Sci.* 244, 2785 (1957); c) *J. de Phys. Rad.* 20, 541, 633, 657 (1959).
- ²⁷E. Otten, *Naturwiss.* 7, 157 (1964).
- ²⁸A. Omont, *J. de Phys. Rad.* 26, 2785 (1965).
- ²⁹C. Cohen-Tannoudji and S. Haroche, *J. de Phys.* 30, 125 (1969).
- ³⁰J. Meunier and A. Omont, *C. R. Ac. Sci.* 262, 260 (1966).
- ³¹G. V. Markova and M. P. Chaĭka, *Opt. i spektr.* 17, 319 (1964).
- ³²B. Kibble et al., *Phys. Rev.* 153, 9 (1967).
- ³³H. Hirsch and G. Stager, *J. Opt. Soc. Am.* 50, 1052 (1960).
- ³⁴C. Cohen-Tannoudji and A. Kastler, *Progr. Opt.* 5, 3 (1966).
- ³⁵G. Markova et al., *Opt. i spektr.* 23, 835 (1963); E. Al'tman and M. Chaĭka, *ibid.* 19, 968 (1965); E. Al'tman and S. Kazantsev, *ibid.* 28, 805 (1970); E. Al'tman, *ibid.*, p. 1029; H. Bucka et al., *Zs. Phys.* 194, 193 (1966); J. Feichtner et al., *Phys. Rev.* 164, 44 (1967).
- ³⁶A. Mashinskiĭ, *Opt. i spektr.* 28, a) 3, b) 201 (1970).
- ³⁷V. Meyer-Bekkhaut, *Zs. Phys.* 141, 185 (1955).
- ³⁸F. Handrish et al., *Proc. of the Intern. Conference on Optical Pumping and Atomic Line Shape (OPALS)* (Warsaw, June 25-28, 1968), ed. by T. Skalinski, Warszawa, PWN, 1969, p. 417.
- ³⁹R. De Zafra and A. Marshall, *Phys. Rev.* 170, 28 (1968).
- ⁴⁰B. Budick, *Phys. Rev.* 168, 89 (1968).
- ⁴¹M. Hartrott, *Zs. Naturforsch.* 24a, 1202 (1969).
- ⁴²M. Chenevier et al., *Phys. Lett.* A26, 291 (1968).
- ⁴³A. Khadjavi et al., *Phys. Lett.* 17, 463 (1965).
- ⁴⁴M. A. Élyashevich, *Atomnaya i molekulyarnaya spektroskopiya* (Atomic and Molecular Spectroscopy), Fizmatgiz, 1963, Chap. 14, Sec. 5.
- ⁴⁵B. Budick et al., *Phys. Rev.* A140, 1041 (1965).
- ⁴⁶G. Oppen, *Zs. Phys.* 213, 254 (1968).
- ⁴⁷M. Chantepic and J. Margeril, *C. R. Ac. Sci.* B265, 302 (1967).
- ⁴⁸M. Chantepic, *C. R. Ac. Sci.* a) B269, 522 (1969); b) B270, 596 (1970).
- ⁴⁹R. Isler et al., *Bull. Am. Phys. Soc.* 10, 1096 (1965); B. Budick et al., *Phys. Rev.* 147, 1 (1966); K. Brog et al., *ibid.* 153, 91 (1967).
- ⁵⁰F. Pichanick et al., *Phys. Rev.* 169, 55 (1968).
- ⁵¹M. Baumann, *Zs. Naturforsch.* 24a, 1049 (1969).
- ⁵²J. Dodd and R. Kinnear, *Proc. Phys. Soc.* 75, 51 (1960).
- ⁵³H. Ackermann, *Zs. Phys.* 194, 253 (1966); M. Baumann et al., *ibid.* 194, 270 (1966).
- ⁵⁴M. Briger et al., *Zs. Naturforsch.* 24a, 903 (1969).
- ⁵⁵M. Baumann et al., *Zs. Phys.* 221, 245 (1969).
- ⁵⁶S. Stromberg and S. Rydberg, *Zs. Phys.* 227, 216 (1969).
- ⁵⁷H. Kluge et al., *J. Phys.* 30, 15 (1970).
- ⁵⁸E. Solomon and W. Happer, *Phys. Rev.* 144, 7 (1966).
- ⁵⁹J. Ney, *Zs. Phys.* 223, 126 (1969).
- ⁶⁰F. Ackermann et al., see^[38], p. 545.
- ⁶¹D. Lecler, *J. Phys.* 30, 10 (1969).
- ⁶²J. Levine et al., *Phys. Rev. Lett.* 22, 267 (1969).
- ⁶³F. Lynch et al., *Phys. Rev.* 120, 513 (1960); C. Wu et al., *Phys. Rev. Lett.* 5, 432 (1960); W. Newirth, *Zs. Phys.* 197, 473 (1966).
- ⁶⁴K. Albrecht et al., *Hyperfine Structure and Nuclear Radiation* (Proc. of the Conference), Amsterdam, North-Holland, 1968, p. 357.
- ⁶⁵B. Kibble et al., see^[38], p. 445.
- ⁶⁶M. Krainska-Miszczak, see^[38], p. 441.
- ⁶⁷B. Baglis, *Phys. Lett.* A26, 414 (1968).
- ⁶⁸E. Al'tman et al., *Opt. i spektr.* 26, 127 (1969).
- ⁶⁹H. Wieder, and T. Eck, *Phys. Rev.* 153, 103 (1967).
- ⁷⁰G. W. Series, *Phys. Rev. Lett.* 11, 13 (1963).
- ⁷¹B. Budock and J. Snir, *Phys. Rev. Lett.* 20, 177 (1968).
- ⁷²R. Riscol, *Phys. Rev.* A138, 22 (1965).
- ⁷³M. Leventhal, *Phys. Lett.* 20, 625 (1966).
- ⁷⁴J. Van der Ziel and N. Bloembergen, *Phys. Rev.* A138, 1287 (1965).
- ⁷⁵J. Dupont-Roc et al., *Phys. Lett.* A25, 87 (1967); *C. R. Ac. Sci.* B264, 1811 (1967).
- ⁷⁶J. Barrat and C. Cohen-Tannoudji, *J. de Phys. Rad.* 22, 329 (1961).
- ⁷⁷W. Bell and A. Bloom, *Phys. Rev.* 107, 1539 (1957); G. V. Skrotskiĭ and T. G. Izyumova, *Usp. Fiz. Nauk* 73, 423 (1961) [*Sov. Phys.-Usp.* 4, 177 (1961)]; T. R. Carver, *Science* 141, 599 (1963).
- ⁷⁸E. B. Aleksandrov et al., *Opt. i Spektr.* 23, 282 (1967).
- ⁷⁹J. Lehman, Thèse, *Ann. de Paris* 2, 345 (1967).

Translated by J. G. Adashko