DETERMINATION OF THE ELEMENTARY-EMITTER SPECTRUM LATENT IN AN INHOMOGENEOUSLY BROADENED SPECTRAL LINE

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It is shown that the radiation spectrum of an arbitrarily excited ensemble of atoms contains information regarding the energy structure of the initial (excited) state of the atoms; this structure cannot be resolved spectroscopically, owing to Doppler broadening. The information can be extracted by analyzing the radiation noise spectrum after preliminary coherent amplification. The 3.508-μ Xe line is investigated experimentally. The lifetime and Lande factor of the $5d[7/2]_3$ state are estimated.

1. INTRODUCTION

IN the present investigation we have demonstrated experimentally that the Doppler emission line contains information concerning the natural spectral width and the fine structure of elementary radiation packets (quanta) emitted by individual atoms from a source, irrespective of the degree to which the statistical broadening exceeds the spectral structure of the elementary processes. This information can be extracted with the aid of a special measuring procedure, which is the generalization of the beat technique.

Beats in the intensity of spontaneous and induced optical transitions were observed and intensively investigated in the past few years (see, for example, the review[1]). The beats accompany transitions from an initial state which is a superposition of states with a definite energy (up to the level width). As applied to the case of spontaneous emission, the beats can be particularly illustratively described as a result of interference between several optical harmonics emitted simultaneously by each atom, if this atom, in accordance with the sense of superposition, is simultaneously and partly at several energy sublevels. In order for beats to occur it is necessary to employ special measures to ensure phase matching of the states of the individual atoms. Among the measures employed are anisotropic excitation in conjunction with: a) harmonic modulation[2-6], b) a pulse procedure[7-8], c) modulation of the distance between the energy levels that take part in the superposition (parametric resonance)[10-14], and d) modulation of the spatial characteristics of the excitation.

The foregoing methods of excitation ensure a special preparation of the initial state with non-diagonal density-matrix elements. Otherwise the interference phenomena vanish. At the same time, when considering an individual atom, it can be stated that, with the exception of special cases, it always is excited in a superposition of a number of energy states, i.e., there are always beats in the elementary act of emission or absorption of such an atom[1)]. It has been heretofore assumed that disordered beats cannot be observed[1]. It is shown in the present paper that it is possible to observe the existence of such a hidden elementary coherence of states by analyzing an emission-intensity spectrum that has been first subjected to coherent amplification. By the same token, it becomes possible to observe the elementary spectral structure of the line under an excess of statistical broadening, without interfering with the radiating system.

2. BASIS OF THE METHOD

For simplicity we shall consider the simplest three-level scheme, Fig. 1a. Assume that starting with the instant t = 0 the atom is a superposition of closely lying upper levels 1 and 2, so that its state $\psi(t)$ is of the form

$$\psi(t) = C_1(t)|1\rangle + C_2(t)|2\rangle. \quad (1)$$

The emission intensity, with a certain polarization **e**, following spontaneous transition of the atom to the state $|0\rangle$, is proportional to the square of the modulus of the dipole moment of the transition $\langle 0|\hat{d}\mathbf{e}|\psi\rangle$, and is given by

$$I(t) \sim e^{-\Gamma t}[A + 2|B|\cos(\omega_{12}t + \varphi_{12})]. \quad (2)$$

We assume here that both levels have the same radiative width Γ, and we introduce the symbols

$$A = \sum |C_i|^2 |\langle 0|\hat{d}\mathbf{e}|i\rangle|^2, \quad B = C_1C_2^*\langle 0|\hat{d}\mathbf{e}|1\rangle\langle 2|\hat{d}\mathbf{e}|0\rangle,$$

C_i is the time-independent part of the amplitude of the state $|i\rangle$, $\omega_{12} = (E_1 - E_2)/\hbar$, $\varphi_{12} = \arg B$, and E_i is the energy of the state $|i\rangle$.

Expression (2) describes elementary beats. Figure 1b shows the time variation of the emission intensity for the characteristic case $|C_1| = |C_2|$ and $|\langle 0|\hat{d}\mathbf{e}|1\rangle|$

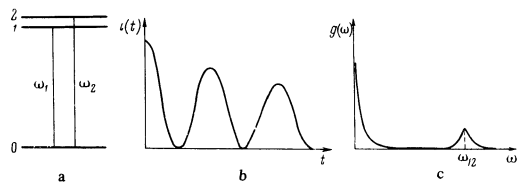

FIG. 1. a—Model level scheme, b—time variation of the spontaneous emission in the case of interference between the states 1 and 2, c—beat-power spectrum.

[1)]Under special conditions it is possible to excite an atom in an eigenstate, but even then one can speak of beats of harmonics within the limits of the width of the state. These "zero" beats, under anisotropic excitation, are responsible for the level-intersection effect.

$= |\langle 0|\hat{d}e|2\rangle|$. In all practical cases we have $\omega_{12} \ll \omega_{10} \approx \omega_{20} \approx \omega_0$, and consequently the beat frequency ω_{12} is not subject to the Doppler scatter.

By grouping correctly, in time and in phase, the momenta of type 1b from different atoms, we can obtain the collective intensity oscillations referred to in the introduction. We shall discuss below the possibility of revealing the "disordered coherence" of the states $|1\rangle$ and $|2\rangle$: we shall assume that the momenta of Fig. 1b are distributed randomly in time and have an arbitrary initial phase φ_{12}.

We call attention to the intensity spectrum of the elementary beat:

$$g(\omega) = \left| \int_{-\infty}^{\infty} I(t) e^{i\omega t} dt \right|^2 \sim g_0(\omega) + g(\omega, \varphi_{12}), \tag{3}$$

$$g_0(\omega) = (\Gamma^2 + \omega^2)^{-1} + {}^1/_4[\Gamma^2 + (\omega_{12} - \omega)^2]^{-1} + {}^1/_4[\Gamma^2 + (\omega_{12} + \omega)^2]^{-1}.$$

The second term $g(\omega, \varphi_{12})$ depends harmonically on the initial phase. As is well known from the theory of spectra, the spectrum of the sum of statistically independent processes is equal to the sum of their spectra. Therefore the power spectrum $G(\omega)$ of the random sequence of processes of type (2) is given by

$$G(\omega) = ng_0(\omega) + \sum_n g(\omega, \varphi_{12}^n), \tag{4}$$

here n—number of elementary events per unit time.

Averaging over the random initial phases φ_{12}^n leads to the vanishing of the second term in (4). The remaining part carries information concerning the spectrum of the elementary emitter, Fig. 1c.

In this reasoning, however, we took no account of the quantum character of the registration of radiation by the receiver. Indeed, the plot of Fig. 1b describes the intensity of emission from one atom. But one atom emits one energy quantum in the elementary act. During the course of registration, the probability of observing the photocurrent will follow (2) in time, but a single realization says nothing concerning the probability distribution. In the case of ideal registration, a quantum with spectrum of intensity (3) is exchanged for a current pulse from one photoelectron, the spectrum of the latter being close to a white spectrum, so that the entire information concerning the primary process (2) is lost[2)].

This obstacle can be overcome by introducing coherent amplification of light prior to the photoregistration (proposed by D. A. Varshalovich in 1966). Each of the primary quanta is multiplied in the amplifier, so that the output emission has a coarse-grained structure: it consists, as before, of a random superposition of packets of the type of Fig. 1b, but the energy of each of the packets will already amount to many quanta $\hbar\omega_0$. Registration of each packet will lead to the appearance of several time-correlated photoelectrons, the spectrum of the aggregate of which is the sum of the white spectrum and the spectrum $g_0(\omega)$ of the sought process.

In fact, the power spectrum of the process is obtained

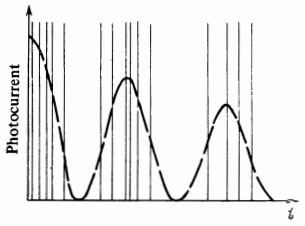

FIG. 2. Photocurrent of receiver under the influence of one amplified quantum.

by averaging the power spectrum of the random realization, an example of which is Fig. 2, which shows the time variation of the receiver photocurrent under the influence of a single amplified quantum is shown. The emission of the individual photoelectrons is represented by short identical current pulses. The power spectrum $h(\omega)$ of the process of Fig. 2 is given by the expression:

$$h(\omega) = \left| \int_{-\infty}^{\infty} e^{i\omega t} \sum_k f(t - t_k) dt \right|^2,$$

where f(t) is the impulse characteristic of the receiver. The summation is over all the pulses generated by a single amplified quantum. Integrating the series, we get

$$h(\omega) = kf(\omega) + f(\omega) \sum_{k,l} e^{i\omega(t_k - t_l)},$$

$$f(\omega) = \left| \int_{-\infty}^{\infty} f(t) e^{i\omega t} dt \right|^2.$$

The emission times t_k and t_l have a distribution given by (2), so that averaging over the realizations yields

$$\overline{h(\omega)} = kf(\omega) + k(k-1) f(\omega) g_0(\omega). \tag{5}$$

If the frequency characteristic $f(\omega)$ of the receiver is sufficiently broad, then the spectrum $\overline{h(\omega)}$, together with the summary spectrum $G(\omega)$, makes it possible to separate the characteristics ω_{12} and Γ of the sought process.

The foregoing conclusions are valid assuming there is no time overlap of the individual emission acts (2). In the general case, account must be taken of the interference between them, and the central optical frequency of each radiation pulse is a random quantity distributed over the Doppler contour. This leads to a broad (Doppler) spectrum of the beats, first observed in[16] (to be sure, under conditions of interference between two spectrally resolved lines). Against the smooth background made up of these beats together with the shot noise, it is possible to observe relatively narrow spectral maxima of the sought component of the spectrum $g_0(\omega)$.

3. EXPERIMENT

We chose xenon as the object of the experiment, since it is known that Xe in a gas discharge has the highest gain—up to 70 dB/m for the 3.508-μ line in thin tubes in the absence of saturation[17,18]. Xenon was attractive also because of its low working gas pressure, which ensured the absence of perturbation of the atoms by the collisions. Two experimental versions were employed. In the first, no distinction was made between the light source and the amplifier. One gas-discharge tube was used, in which the intrinsic spontaneous emission was amplified; in other words, we investigated the noise

[2)] It is assumed that the different atoms emit independently. Actually there is always a certain degree of coherence in the emission, making it possible to expect in principle that the analysis will be successful. An attempt to separate the beat spectrum against a noise background was made by one of the authors in 1962, but was unsuccessful.

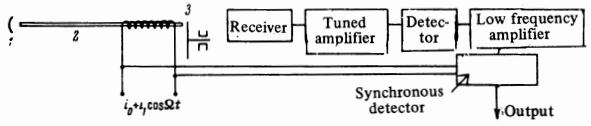

FIG. 3. Block diagram of experimental setup: 1 – mirror, 2 – discharge tube with Xe, 3 – analyzer.

spectrum of the so-called superradiance. A block diagram of the experimental setup is shown in Fig. 3.

We used a tube with inside diameter 6 mm, with discharge-channel length 2.2 m, and with oxide cathodes. The tube was fed with dc. The quartz windows of the tube were cemented to the ends at a small angle to the axis, so as to prevent back reflection and self-excitation of the system. In most experiments, an aluminum-coated mirror with radius of curvature 2 m was placed on one side of the tube. The radiation emerging from the opposite end was incident on a low-inertia receiver—an indium antimonide photodiode. The receiver was connected to a tuned amplifier with approximate bandwidth 100 kHz, tuned to different fixed frequencies. After amplification, the signal was detected and was fed to a synchronous detector. The reference voltage for the latter was the voltage modulating the magnetic audio-frequency field, superimposed together with the dc component on the discharge-tube section adjacent to the receiver. The magnetic field split the initial state of the emitting Xe atoms and determined the frequency of the sought beats. The emission intensity spectrum varied with varying magnetic field, and this should have led to a change in the voltage at the output detector with the magnetic-field scanning frequency. Since the frequency of the receiving channel was fixed, the form of the photodiode characteristic was immaterial. Thus, we registered in the described experimental setup only the high-frequency (magnetic-field dependent) part of the beat spectrum, Fig. 1c.

The main part of the radiation at the tube output is connected with the amplified spontaneous emission of the atoms located in that part of the tube which was closer to the receiver. It was therefore meaningless to extend the magnetic field over the entire length of the tube. In the experiment, the length of the section of the tube with the magnetic field ranged from 4 cm to 1 m. The greater part of the results were obtained when the length of this section was 85 cm. At the chosen direction of the magnetic field, parallel to the tube axis, we could observe beats connected with the interference of the $+\sigma$ and $-\sigma$ components of the radiation. To permit them to interfere, a linear analyzer was placed in front of the receiver.

In the second variant of the experiment, the source and the amplifier were separated. In this case the analyzer could be placed, with equal success, either past the source or past the amplifier. The source was a tube with a discharge length of approximately 1 m, and the amplifier was a tube employed in the first version. The main results obtained in these two versions agree. For concreteness, we shall henceforth refer to the first variant.

A preliminary study of the gas-amplifier conditions was made prior to the investigation of the photocurrent spectrum. It was established that a narrow radiation beam, which increased monotonically in power with decreasing Xe pressure, emerges from the output end of the tube. We chose an Xe pressure of 0.002 Torr, this being in equilibrium with the solid phase at liquid-nitrogen temperature[19], thereby fixing the discharge conditions in accordance with the gas pressure. Under these conditions, the optimum discharge current was 40–50 mA, and the order of magnitude of the radiation power was 10^{-4} W.

A number of observations have shown that the system was not excited and operated in the mode of amplification of the intrinsic noise, being saturated to a considerable degree. The gain was measured in the version in which the source and the amplifier were separated. To this end, a mechanical modulator was introduced between them, and the amplitudes of the alternating photocurrent was measured with the discharge in the amplifier turned on and off. The gain revealed a strong dependence on the input-signal intensity. At maximum intensity, the gain over the entire length (2.2 m) was approximately 200–300. This gives for the maximum gain an estimate on the order of 10^4–10^5 in the case of a double passage of the radiation in the version with the mirror. At such a gain, the calculated narrowing of the receiver band width does not exceed the three fold initial Doppler contour, with width of approximately 200 MHz at the 0.5 level. This gives an idea of the beat-frequency range that can be observed, since it is necessary that both harmonics of the elementary processes fall in the amplification band. Since the question of the amplification band is quite important, this question was approached also from a different direction. We plotted the dependence of the emission power on the magnetic field superimposed on the part of the amplifier-tube. It was found that the output power begins to decrease noticeably in fields exceeding 50 Oe. On the other hand, in experiments in which the intensity spectrum was investigated, the magnetic field did not exceed 12 Oe.

A preliminary rough check of the radiation noise has shown that in the frequency range from 25 MHz to practically zero the noise has approximately constant power (with accuracy up to several dozen per cent), and an equal illumination of the photoreceiver from the thermal source produced no observable noise under our conditions.

We now proceed to describe the main experiments. The resonant amplifier was tuned to the following frequencies: 5.1, 9.6, 18, and 26 MHz. The magnetic field varied from −12 to +12 Oe. The amplitude of its audio-frequency modulation was usually 0.08 Oe. For each fixed frequency of the amplifier, we plotted the readings of the synchronous detector as a function of the magnetic field. The obtained plots turned out to be symmetrical with respect to the origin. They are shown in Fig. 4 for one direction of the magnetic field. The majority of the results were obtained at a constant registration time of 10 seconds. As seen from Fig. 4, the observed signal shifted relative to the magnetic field with changing registration frequency, revealing a gradually developing structure.

A characteristic feature of the expected effect is the special role of the analyzer. Its presence is essential to reveal the effect, but the position of its plane of polarization is immaterial, in view of the randomness of the

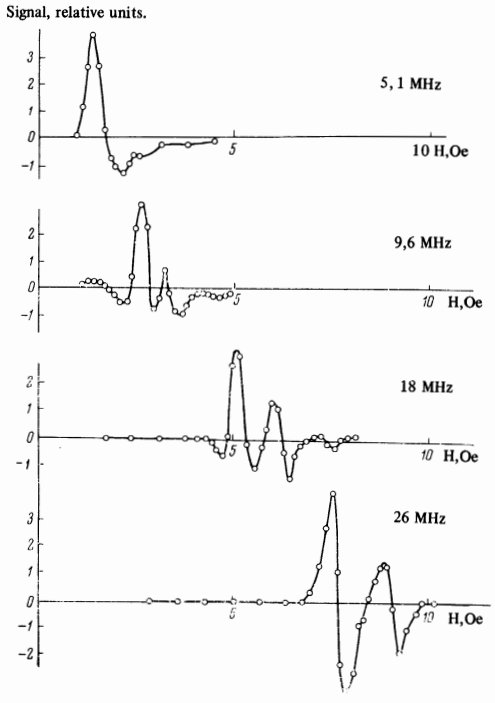

FIG. 4. Synchronous-detector signal as a function of the magnetic field intensity.

investigated beats. This circumstance was used for control. Removal of the analyzer led to an almost complete vanishing of the signal (the analyzing influence of the windows still remained), and rotation of the analyzed changed neither the amplitude nor the phase of the signal. The same test has made it possible to separate the parasitic signal, which occurs at any tuning of the receiver in the region of zero magnetic field, and which is connected, as we were able to establish, with the magnetic depolarization of the partly polarized radiation. Further measurements were made with a rotating analyzer. This was done to eliminate the Faraday effect, which occurs in the modulated magnetic field as a result of the residual polarization of the light. The Faraday effect has led to a flabby but in general nonmonotonic dependence of the readings of the synchronous detector on the magnetic field. Rotation of the analyzer at a frequency that was high compared with the time constant of the registration, averaged out the signal, although this signal distorted slightly the main resonant signal which is connected by us with the sought effect.

4. DISCUSSION OF RESULTS

Favoring our conclusion are the following additional observations. 1) The connection between the frequency of the registration channel and the position of the signal components with respect to the magnetic field is linear. The ratio of the frequency to the magnetic field lies in the region of typical values of the electronic g-factors. 2) The signals have a resonant character. The width of the component signal does not depend on the frequency band of the receiving channel or of the field-modulation frequency if the signals are decreased below the values indicated above. 3) The width of the signal components

Table I

Isotope	Nuclear angular momentum	Abundance, %	Total angular momentum of state $5d[7/2]_3$		g_F/g_J
128	0	1,9	J	3	1
129	1/2	26,2	F	5/2	8/7
				7/2	6/7
130	0	4	J	3	1
131	3/2	21,2	F	3/2	8/5
				5/2	34/35
				7/2	16/21
				9/2	2/3
132	0	26,9	J	3	1
134	0	10	J	3	1
136	0	8,9	J	3	1

does not depend on the registration frequency. 4) Displacement of the region of the superposition of the magnetic field over the tube from the output and towards the mirror (see Fig. 3) lowers the signal regularly. The signal is observed also without a mirror, when a magnetic field is imposed on the end of the tube opposite the receiver. Under all these changes, the form of the signal did not change noticeably.

It remains to discuss the structure of the signal. As seen from Fig. 3, at sufficiently high registration frequency, there are two strong lines of the dispersion type, and an additional less clearly pronounced structure (not always reproducible). The 3.508-μ emission in Xe is identified with the transition $5d[7/2]_3 - 6p[5/2]_2$. The beats are determined by the upper state.

The structure of the upper state in the magnetic field depends on the presence and on the magnitude of the nuclear moment. The natural isotope mixture contains noticeable amounts of seven isotopes, of which five are even. For the even isotopes, the upper state splits in the magnetic field into seven equidistant sublevels. Since beats of the $\pm\sigma$ components terminating at the common level are observed, there is a single beat frequency $\omega_{\pm} = 2\mu_B g_J H/\hbar$, where H—magnetic field intensity, μ_B—Bohr magneton, and g_J—Lande factor. The odd isotopes have several hyperfine levels with their own g_F factors. Information on the Xe isotopes are given in Table I, the lower line of which gives the calculated g_F/g_J ratios assuming that the magnetic splitting is small compared with the hyperfine splitting.

If the isotopic shift and the hyperfine splitting were to cause all the 17 structural lines of the listed isotopes to be separated in the vicinity of 3.508 μ beyond the limits of their contours, then in practice only the Xe^{132} line would be amplified, since the gain for this line is

Table II

Isotope	F of state $5d[7/2]_3$	F' of state $6d[5/2]_2$	Transition intensity
129	5/2	3/2	2/5
		5/2	1/35
	7/2	5/2	4/7
	3/2	1/2	1/10
		3/2	1/25
		5/2	1/350
131	5/2	3/2	4/25
		5/2	64/1225
		7/2	1/490
	7/2	5/2	12/49
		7/2	2/49
	9/2	7/2	5/14

approximately double that for the strongest of the remaining lines (Xe^{129}: F = 7/2, F′ = 5/2; the probabilities of the hyperfine transitions of Xe^{129} and Xe^{131} relative to the single line of the even isotope are given in Table II). It is therefore natural to ascribe the large signal to this even isotope and to determine g_J from its position. The only cause of the appearance of the smaller closely-lying signal may be superposition on the Xe^{132} spectral line of the hyperfine component of one of the odd isotopes, which is then amplified just as the main line. The position of the smaller component of the signal with respect to the magnetic field corresponds to a g_F-factor F = 7/2 of Xe^{129}. Unfortunately, this assumption cannot be verified in detail, since there are no data on the isotopic and hyperfine structures of the 3.508-μ line.

The results of the present work allow us to estimate the g_J-factor of the state $5d[7/2]_3$ of xenon and the width Γ of this state. For the larger signal, identified with Xe^{132}, we can get from the plots of Fig. 4 $g_J = 1.13 \pm 0.08$. Using this value, we find from the distance between the extrema of the large dispersion curve that $\Gamma = 3 \times 10^6$ sec^{-1}. This value is apparently close to the natural line width, since the experiment was performed at a rather low gas pressure. The already noted saturation of the gas amplifier offers evidence of the presence of a certain radiative broadening of the state, but no dependence of the signal width on the radiation power was observed by direct experiment.

5. CONCLUSION

Insofar as we know, this is the first analysis of the spectral structure of a Doppler line. Although this method can now be applied to a limited number of transitions in the infrared region, it is in essence universal and its prospects are determined by the progress of coherent-amplifier techniques. It should be noted also that although the light source and the present investigation was of the same nature as the amplifier, and was in a state with inverted population, this is not essential: the method makes it possible to analyze the structure of the radiation regardless of its source.

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