V. S. Letokhov. Nonlinear Narrow Molecular Resonances Induced by Laser Radiation, and Their Application in Spectroscopy and Quantum Electronics

One of the characteristic marks of the development of physics in the last two decades is, perhaps the production of narrow, frequency-stable resonances of the interaction of an electromagnetic radiation with quantum transitions. The techniques for obtaining narrow resonances in the microwave region with the aid of atomic and molecular beams were developed in the fifties. Such narrow radio-frequency resonances were the basis of quantum standards of frequency and of the atomic time scale now adopted the world over. Extremely narrow resonances in the higher frequency spectral region in nuclear transitions were discovered by Mössbauer in 1958. The narrow resonances of recoilless nuclear transitions in a crystal now ensure the highest sensitivity of physical experiment, on the order of 10^{-15}—10^{-16}. In the intermediate optical region of the spectrum the resonance width was until recently usually not less than 10^{-6}. We consider in the paper how we can with the aid of laser radiation produce narrow resonances inside a Doppler absorption line of relative width 10^{-8}—10^{-9} of the molecules in a gas (see[1-3]). The values of the relative width of the narrowest quantum-transition resonances in the microwave, optical, and γ-ray regions of the spectrum are shown in Fig. 1.

Doppler broadening is nonuniform—the center ω of the spectral absorption line of each particle depends on the velocity $\mathbf{v}$ of the particle ($\omega = \omega_0 + \mathbf{k}\cdot\mathbf{v}$, where ω_0 is the center of the line for the stationary particles and $\mathbf{k}$ is the wave vector of the light wave). The Doppler width $\Delta\nu_D$ may considerably exceed the homogeneous width 2Γ determined by radiation damping and collisions. A plane coherent light wave of frequency ν and intensity sufficient for absorption saturation excites only the particles whose velocity satisfies the resonance condition

$$|\omega_0 + kv - \nu| \leqslant \Gamma. \tag{1}$$

As a result, a "hole" of width $\Delta\nu = 2\Gamma(\sqrt{1+G^2})$, where $G^2 = a\,\xi^2$ is the degree of absorption saturation determined by the field intensity, is burnt out at the frequency ν in the Doppler contour. The saturation-absorption coefficient is determined by the expression $\kappa = \kappa_0(1 + G^2)^{-1/2}$, where κ_0 is the absorption coefficient for a weak field. Thus, if the light wave is: 1) monochromatic, 2) plane, and 3) intense, then in the wide Doppler contour is formed a "hole" containing information about the narrower homogeneous width.

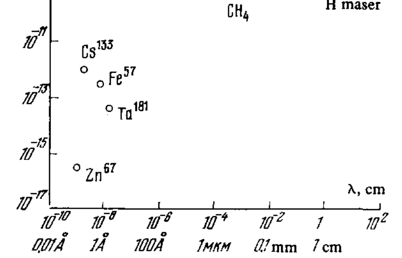

FIG. 1. Relative width of the narrowest resonances in the spectra of atoms (Cs, H) in the radio-frequency region, of molecules (CH_4, I_2, SF_6) in the optical region, and of nuclei (Cs^{133}, Zn^{67}, Fe^{57}, Ta^{181}) in the α-ray region.

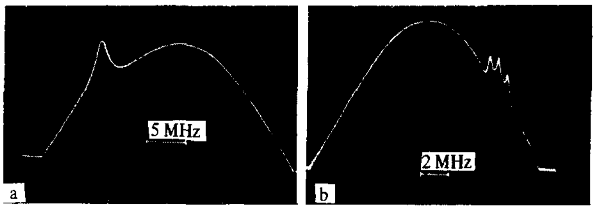

FIG. 2. Narrow resonances observable during absorption saturation in rotational-vibrational transition lines of the ν_3 band of the SF_6 molecule under the action of the radiation of a CO_2 laser tuned on: a) the P(18) and b) the P(16) lines of the 10.6-μ wave band (the width of the Doppler line of SF_6 is equal to 30 MHz).

To produce a narrow resonance at the center of a Doppler line, it is sufficient to send, in exact opposition to a strong wave, a weak plane test wave of the same frequency as the strong wave. The test wave interacts with the molecules whose velocity satisfies another resonance condition:

$$|\omega_0 - kv - \nu| \leqslant \Gamma, \tag{2}$$

i.e., with particles having the same velocity component in the direction opposite to the direction of propagation of the strong wave. It follows from the conditions (1) and (2) that if the frequency ν of the waves does not coincide with the frequency ω_0 of the quantum transition, then both waves interact with entirely different groups of molecules, i.e., the test wave does not feel the strong wave. If, however, the frequency of the waves coincides with the center of the line ($\nu = \omega_0$), then both waves interact with the same group of molecules having a null velocity component in the direction of the waves. Thus, the absorption of the test wave decreases resonantly at the center of the Doppler line, since it interacts with molecules the absorption of which has been saturated by the strong field[4]. This is the narrow molecular resonance induced by laser radiation.

The width of the narrow resonance is determined by the homogeneous width and, at a low pressure of the gas ($\sim 10^{-3}$ Torr), can be limited only by a finite time of flight of the molecules through the light beam, i.e., is $\sim 10^5$ Hz[1]. Figure 2a shows the shape of the narrow resonance observable at the center of the Doppler line of one rotational-vibrational transition of the ν_3 band of the SF_6 molecule with the aid of the radiation of a CO_2 laser tuned to the P(18) line of the 10.6-μ band[5]. If an absorption Doppler line consists of several lines masked by the Doppler broadening, then a narrow resonance is produced at the center of each line, i.e., this method allows the realization of laser spectroscopy inside the Doppler line. An example of such an observation with the molecule and the P(16) radiation line of a CO_2 laser is shown in Fig. 2b[6].

A high-precision stabilization of a laser frequency can be realized on a narrow molecular resonance belonging to a symmetric molecule without a dipole moment, i.e., to a molecule not subjected to an appreciable influence of external electric and magnetic fields[1-4]. We consider in the report ways for increasing the stability and reproducibility of a laser frequency to within 10^{-13}—10^{-14} with the aid of narrow molecular resonances, and also consider the use of lasers for measuring small changes in optical lengths with a sensitivity $\Delta l/l$ of up to 10^{-15}.

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[1] V. S. Letokhov, ZhETF Pis. Red. 6, 597 (1967) [JETP Lett. 6, 101 (1967)].

[2] V. N. Lisitsyn and V. P. Chebotaev, Zh. Eksp. Teor. Fiz. 54, 419 (1968) [Sov. Phys.-JETP 27, 227 (1968)].

[3] R. L. Barger and J. L. Hall, Phys. Rev. Lett. 22, 4 (1969).

[4] V. S. Letokhov and V. P. Chebotaev, ZhETF Pis. Red. 9, 364 (1969) [JETP Lett. 9, 215 (1969)].

[5] N. G. Basov, O. N. Kompanets, V. S. Letokhov, and V. V. Nikitin, Zh. Eksp. Teor. Fiz. 59, 394 (1970) [Sov. Phys.-JETP 32, 214 (1971)].

[6] O. N. Lompanets and V. S. Letokhov, Zh. Eksp. Teor. Fiz. 62, 1302 (1972).

V. A. Zverev and E. F. Orlov. Optical Methods of Information Processing in Radiophysics and Medicine.

Optical methods of information processing possess a number of advantages over electronic methods when performing multichannel integral transformations (measurement of spectra, correlation functions, and probability distributions). These operations can be carried out simultaneously in many parallel channels with the aid of optical systems. In connection with the appearance of lasers and the advances made in the field of holography, considerable attention is being given to the development of the optical methods of information processing based on the properties of coherent light. However, the requirement of coherence of light, which is the basis of the calculating machine, is difficult to fulfil when the signal under investigation is introduced into the processing scheme. Optical calculators are known which allow the use of noncoherent light, but the comparative ease with which information can be fed into them reduces considerably the accuracy of the computations. The decrease in the computational accuracy occurs owing to the fact that the result of the computations is observed in a strong interfering background, which is absent when coherent light is used.

We describe in the report a new principle of construction of optical calculators which allows us to simply feed the information in question into the machines and at the same time maintain a high computational accuracy. The principal idea of the proposed method consists in the application of a matched spatial and time modulation of the luminous flux. One type of modulation is used for computations: either spatial or time modulation, and the other type is used for raising the computational accuracy by means of spatial or time filtering of the useful signal. Experimental investigations of the principles considered showed that it is possible to construct the corresponding apparatus for optical information processing (the OSA system)[1], which will not be inferior in respect of computational accuracy to the apparatus using coherent light. At the same time the OSA systems utilize better the region of low spatial frequencies and, therefore, less accurate optical elements can be used in them[1].

The application of the OSA methods for solving concrete problems led to the development of an effective method for preliminary information processing—the method of generalized two-dimensional holograms[2]. A generalized two-dimensional hologram is obtained by representing the initial information $f(t)$ in the form $f(t)f(t-\tau)$, i.e., as functions of two coordinates t and τ. By inverting the generalized hologram with respect to τ and averaging over t, we can measure the spectral parameters of the process $f(t)$ with a high resolution and with a simultaneous simplification of its structure, which is necessary for the solution of a number of problems. The method has been successfully applied for the measurement of the technical width of the line of the quartz generator[3], for the measurement of spatial correlation functions of wave fields with a long averaging time[4], to some problems of stereophonic audio reproduction[2], and in operational interference spectroscopy[5].

The application of the method for the solution of a number of problems of medical diagnosis proved to be fruitful[6,7]. The recording of a phonocardiogram signal in the form of a generalized hologram in the frequency-time plane allows us to efficiently use the surplus information in the signal and to get rid of it when it is fed into the calculating machine; in a number of cases such a representation makes it possible to establish simple diagnostic symptons[6]:

A generalized hologram of a ballistic cardiogram permits an easy measurement of the absolute levels of the forces acting on the cardiovascular system[8]. The measurement of the relative levels of certain harmonic components of these forces allows objective estimates of the force of a heat muscle, the relative activity of operation of the right and left halves of the heat, and a number of other parameters of physiological and diagnostic importance[7].

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[1] V. A. Zverev and E. F. Orlov, Opticheskie analizatory (Optical Analyzers), Sov. Radio, M., 1971.

[2] V. A. Zverev, Radiotekhnika 26, No. 6, 2 (1971).

[3] V. A. Zverev, E. F. Orlov et al., Izv. VUZov (Radiofizika) 12, 1169 (1969).

[4] V. A. Zverev and I. K. Spiridonova, Akust, Zh. 7, 428 (1961) [Sov. Phys.-Acoust. 7, 346 (1962)].

[5] S. M. Gorskiĭ and V. A. Zverev, Izv. VUZ. (Radiofizika) 11, 1205 (1968) [Radiophysics and Quantum Electronics 11, 687 (1968)].

[6] V. A. Zverev et al., Pediatriya No. 8, 21 (1970).

[7] V. A. Zverev et al., Med. Tekh. No. 6, 28 (1971).

[8] K. V. Zvereva, V. A. Zverev, and I. K. Spiridonova, Vrachebnoe Delo No. 1, 10 (1970).

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