Changes in the absorption spectrum and of dispersion of a two-level system in a rotating monochromatic radiation field

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An investigation was made of the changes induced by a high-power radiation field in the profiles of the absorption and dispersion lines of an optically oriented Cd^{113} vapor near the Zeeman splitting frequency of the ground state. A weak probing signal was amplified without population inversion when the high-power field frequency coincided with the atomic transition frequency or when there was a slight detuning between these two frequencies. When the high-power field frequency was much higher than the atomic transition frequency, the latter decreased to zero proportionally to the square of the high-power field amplitude and then it rose again. An amplification of the weak probing signal due to an inversion of the level populations was observed after the transition frequency passed through zero. The experimental results indicated that two-level systems subjected to high-power radiation fields could be used, without population inversion, as tunable laser amplifiers and oscillators.

1. INTRODUCTION

The action of high-power electromagnetic radiation on a quantum system may alter considerably the profiles of the spontaneous emission[1-3] and absorption lines. The latter effect can be detected using an independent weak probing beam.[4-8] An analysis of the changes in the spectra of atoms in the case when many levels are excited simultaneously is complicated by the possibility of many-photon parametric and stimulated cascade process[9-11] and by many-photon ionization of atoms.[12] It would therefore be interesting to study the changes in the profiles of absorption lines and dispersion curves under conditions permitting the application of the two-level approximation with respect to the applied strong field and to use the spontaneous emission or absorption of a weak probing beam (involving other levels) simply for the detection of changes in the spectrum.

It is shown theoretically[1-4] that a sufficiently strong resonance radiation field not only alters the width and displaces a fluorescence line of a two-level system but also splits it into three components (Fig. 1a). This phenomenon can be interpreted clearly using the concept of energy quasi-levels into which each state of a two-level system is split by the applied periodic electric field (Fig. 1b).[4,13-15]

It is much more difficult to interpret the theoretically predicted changes in the profiles of an absorption line of a probing beam by a two-level system in a strong quasi-resonance radiation field. Some time ago, Rautian and Sobel'man[1] predicted a far from trivial reversal of the sign of the absorption coefficient of a probing beam (i.e., amplification of such a beam) by a two-level system subjected to a quasiresonance monochromatic radiation in the absence of population inversion. Subsequently, the theory of this effect was developed and generalized in[4-6] and Klyshko, Konstantinov, and Tumanov[16] were the first to demonstrate this effect experimentally.

Direct quantitative investigations of the changes in the absorption and dispersion spectra of two-level atomic systems subjected to high-power optical radiation are very difficult to carry out because of the lack of lasers with a tunable emission wavelength and with a variable spectrum and duration of the emission pulses. In view of this, we used radio-frequency (rf) radiation and the transitions between the Zeeman energy levels of the ground state of cadmium atoms. Since the Zeeman splitting frequency was many orders of magnitude lower than the frequencies of transitions to the nearest levels, cadmium atoms could be regarded as a pure two-level system. The application of the optical orientation method made it possible to observe changes in the spectrum using radiation in the optical range and to employ very low cadmium vapor pressures. This allowed us to ignore the interatomic interactions and to study the changes induced by a strong radiation field in the absorption and dispersion spectra of a two-level system in the purest form, which permitted us to make quantitative comparisons between the experimental results and theory.

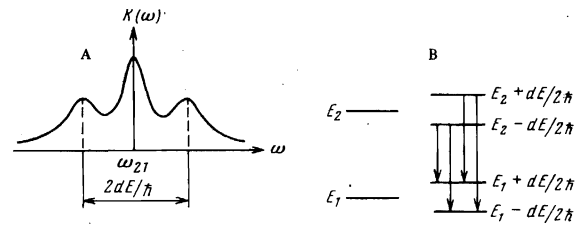

FIG. 1. a) Frequency dependence of the probability of spontaneous emission from a two-level system in a strong resonance radiation field. b) Scheme showing spontaneous transitions between levels in a two-level system subjected to a strong resonance radiation field.

2. EXPERIMENTAL METHOD

We investigated an atomic Cd^{113} vapor which only had the nuclear moment $I = \frac{1}{2}$ in the ground state. The application of a static magnetic field split this state into two levels corresponding to the orientations of the nuclear moment parallel and antiparallel to the magnetic field (the field intensity was $H_Z = 5.25$ Oe and the splitting of the levels corresponded to a frequency of 4.96 kHz). Under thermodynamic equilibrium conditions, the relative difference between the populations of the Zeeman sublevels was of the order of 10^{-10}. Therefore, when the concentration of cadmium atoms was 10^{10}–10^{11} cm^{-3}, it was impossible to observe the absorption of the rf field quanta. A considerable differ-

ence between the populations of the Zeeman levels ($\sim 10^{-2}$) was established by using the well-known optical orientation method, based on the difference between the probabilities of excitation of atoms from different Zeeman sublevels by circularly polarized resonance radiation.[17-19] Even this population inversion was insufficient for the direct observation of the rf absorption at the atomic concentrations employed but an indirect study was possible in which we used changes in the optical properties of the cadmium vapor revealed by the orienting resonance radiation. In fact, the absorption of the rf quanta, which altered the difference between the populations of the Zeeman sublevels, changed the absorption of the orienting resonance radiation. In the limit, one transition under the influence of the rf field altered the light flux transmitted by the cadmium vapor by one photon. In view of the high sensitivity of optical radiation detectors, this method enabled us to observe extremely small changes in the rf absorption spectra.

A. Changes in the Line Profile in Resonance and Quasiresonance Fields

Changes in the absorption line profile in resonance and quasiresonance fields were observed using one of the methods for the optical orientation of cadmium atoms in the presence of a buffer gas, discussed by Aleksandrov and Sokolov[18,19] (Fig. 2). One of the Zeeman levels was populated preferentially by illumination of the Cd^{113} vapor with a circularly polarized resonance line (3216 Å) produced by a Cd^{114} lamp subjected to a local axial magnetic field of ~0.5 kOe (the total power of this line was about 1 W). A cell with the Cd^{113} vapor and a buffer gas (xenon at 100 Torr) was heated to 220°C and placed at the center of a system of three pairs of Helmholtz coils, which produced mutually perpendicular magnetic fields, one of which was static $H_Z = 5.25$ Oe with an inhomogeneity $\Delta H = 10^{-3}$ Oe inside the cell, and the other two were rf fields: a strong field $H_X(t) = H_0 \cos \omega t$ ($H_0 = 10^{-2}$–10^{-1} Oe) and a weak field $H_Y(t) = H_1 \cos \omega t$ ($H_1 = 0.5 \times 10^{-3}$ Oe), whose frequency ω was varied near the transition frequency ω_{21}. An orienting beam was directed in the YOZ plane at an angle 45° with respect to the Y and Z axes and the same beam was used for recording purposes.

After passing through the cell, the light beam was modulated at the frequencies of motion of the transverse component of the angular momentum produced by the alternating field.[17] An F-4 photocell, a U2-6 amplifier, an SD synchronous detector, an ÉPP-09 automatic plotter, and a reference voltage of frequency ω were used in the measurement of the component of the modulation of light at the weak-field frequency ω. Under these conditions, the signal/noise ratio exceeded 10^3. When ω_0 was very close to ω, an alternating stray signal of frequency $|\omega_0 - \omega|$ was observed and this signal could not be removed completely by the RC filter in the synchronous detector (time constant ~1 sec)—see Fig. 3. This stray signal was eliminated by a subtraction circuit placed in front of the synchronous detector in the case when the weak field of frequency ω was not applied to the cell with the Cd^{113} vapor.

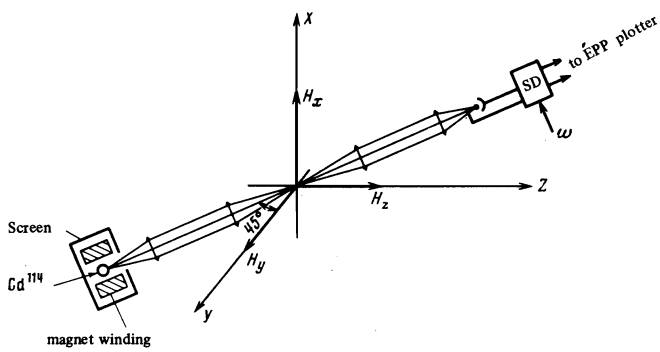

FIG. 2. Schematic diagram of the apparatus.

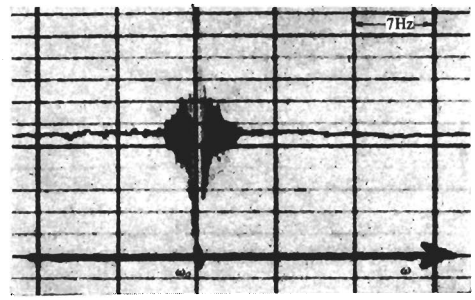

FIG. 3. Stray alternating signal near the strong field frequency.

Depending on the phase shift between the reference and recorded signals, we could measure either the real or imaginary part of the susceptibility of the cadmium vapor at the frequency ω: $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$, and these parts were proportional to the refractive index and the extinction coefficient of the vapor at the weak-field frequency. Figures 4 and 5 show the signals proportional to the extinction index (Fig. 4) and the refractive index (Fig. 5) in the case when the strong-field frequency ω_0 was in exact resonance with the Zeeman splitting frequency ω_{21} (ω_0

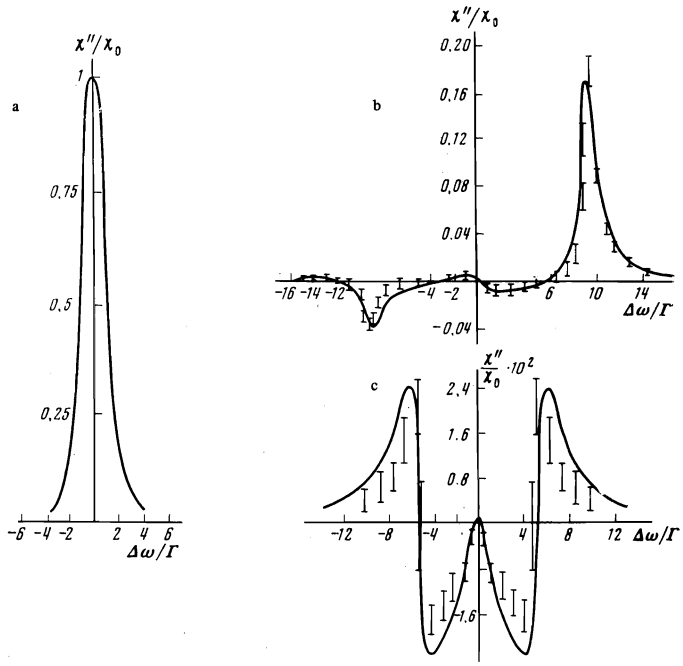

FIG. 4. Frequency dependences of the imaginary part of the susceptibility $\chi''(\omega)$: a) in $H_0 = 0$; b) for $\omega_{21} - \omega_0 = 3\Gamma$ and $|V_0|^2 = 20\Gamma^2$; c) for $\omega_0 = \omega_{21}$ and $|V_0|^2 = 6.8\Gamma^2$. The continuous curves in Fig. 4a are the experimental results and in Figs. 4b and 4c are the theoretical values; the vertical segments in Figs. 4b and 4c represent the scatter of the experimental values obtained in five independent measurements of these dependences.

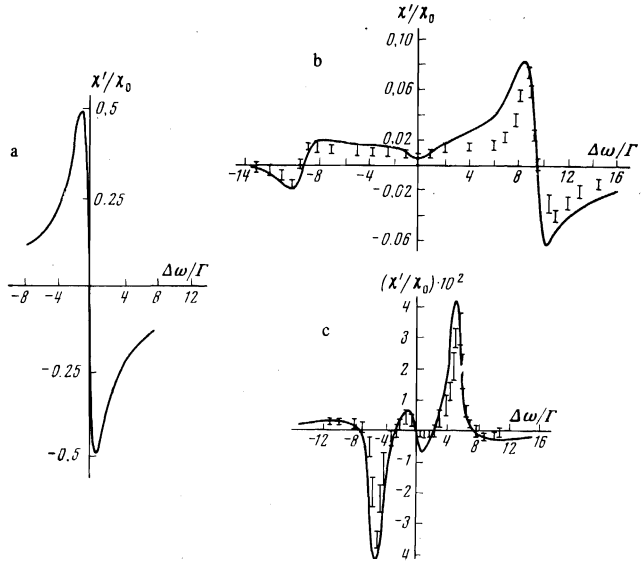

FIG. 5. Frequency dependences of the real part of the susceptibility $\chi'(\omega)$ for the same parameters as in Fig. 4.

$= \omega_{21}$) and in the case of some deviation from this resonance. The strong resonance field caused splitting of the absorption line, which increased linearly with the amplitude of the strong field H_0 (Fig. 6) and it reversed the sign of the weak-field absorption coefficient at the center of the split line.

The profiles of the recorded extinction and refractive indices were very sensitive to the detuning $\omega_{21} - \omega_0$ of the strong field: they varied considerably when ω_0 deviated from ω_{21} by an amount of the order of the line width even when the amplitude of the field H_0 was so large that the corresponding line splitting was considerably greater than the line width. In the off-resonance case, when $|\omega_0 - \omega_{21}|$ exceeded the line width Γ, the absorption line was split into two components of comparable amplitude and the sign of one of these components was reversed. In strong fields H_0, the splitting was proportional to H_0 and approximately equal to the splitting in the resonance field. The width of the gain band in a strong nonresonance field was approximately equal to the original line width and the maximum gain decreased with increasing detuning in proportion to $\Gamma/|\omega_0 - \omega_{21}|$. The amplification was observed on that side of the line which corresponded to the strong field H_0.

The absolute values of the amplitudes of the rf fields H_0 and H_1 were found by calibrating the apparatus. It was reported in[20] that the application of an rf field to a two-level system produced transient effects: the population M_Z and the transverse magnetization $M_\perp = (M_X + iM_Y)$ pulsated at a frequency which depended on the rf field amplitude. When this field was resonant, so that $T_1 = T_2 = T$ and $\gamma H_1 \gg T^{-1}$, the formula describing the transient oscillations of the projection of the transverse magnetization $M_\perp$ onto the OY axis simplified to

$$M_y = M_y(0)\, e^{-t/T} \sin 2\pi\gamma H_1 t \sin \omega_0 t. \qquad (1)$$

It is clear from the above formula that the component M_y of the transverse magnetization should oscillate rapidly at the field frequency ω_0 and its amplitude should be modulated at a frequency γH_1 and should decay exponentially with a time constant T. An electrical signal picked off the photocell load following application of the field was displayed on the screen of an oscillograph (Fig. 7). The pulsation period deduced from the oscillograms was used to find the field amplitude $H_1 = \pi/2\gamma\Delta t$, where $\gamma = 942$ Hz/G and Δt is the duration of a pulsation.

B. Changes in the Line Profile Far From Resonance

When the strong-field frequency was far from resonance ($\omega_0 \gg \omega_{21}$), the optical orientation signal was studied in two ways in order to eliminate secondary effects. In one case, the experimental geometry was the same as that shown in Fig. 2 except that the strong field of frequency ~2 kHz and the weak field were rotated (Fig. 8a). In the second case (Fig. 8b), the field H_Z was parallel to the pump beam and, at the same time, perpendicular to the probing beam (the optical orientation was observed along the transverse beam). The experimental results were independent of the observation method but, in the first case, the experimental conditions ensured a higher signal/noise ratio.

We observed a shift of the resonance frequency which was a quadratic function of the field H_0 (Fig. 9). In the region where the resonance frequency decreased, an increase in the amplitude of the rf field H_0 resulted in σ^+ transitions in the rotating weak field. Conversely, in the region where the resonance frequency rose with H_0, the weak field rotating in the opposite sense caused the σ^- transitions. This change in the direction of rotation of the weak field caused an inversion of the sign of

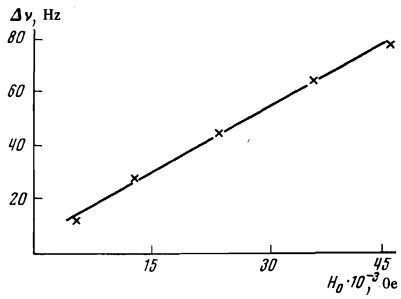

FIG. 6. Dependence of the splitting of an absorption line (separation between absorption peaks) on the amplitude of a strong resonance field H_0.

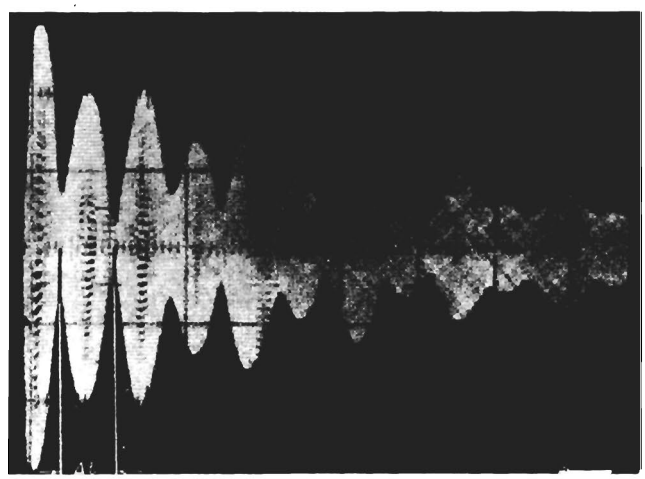

FIG. 7. Transient transverse magnetization signal used to measure rf field amplitude.

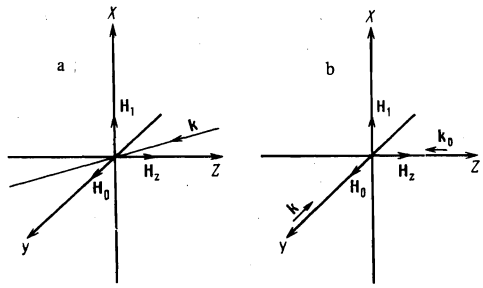

FIG. 8. Orientations of fields in an experimental study of the influence of a strong nonresonance field: a) observations at an angle of 45° with respect to the direction of H_Z; b) observations along a transverse beam.

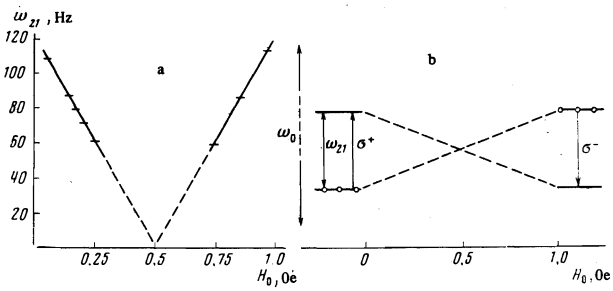

FIG. 9. Dependence of the Zeeman resonance frequency ω_{21} on the rf field amplitude H_0. b) Dependence of the relative positions of the levels on the field amplitude H_0.

the absorption coefficient of this field, i.e., the weak field was amplified. The amplitude of the signal in the strongest field H_0 decreased by a factor not exceeding 2 for a practically constant resonance line width. The reduction in the optical orientation signal in the strong nonresonance field could be attributed to a deterioration of the pumping efficiency in the practically transverse rotating magnetic field ($H_0 \gg H_Z$).

3. DISCUSSION OF RESULTS

The theoretical results most convenient for comparison with our experiments were those reported by Apanasevich.[5] His paper and similar theoretical investigations[1-4] are concerned with homogeneously broadened two-level systems subjected to monochromatic rotating fields. Since the quantity determined in the present investigation is proportional to the susceptibility of the system, we shall give the formulas for the imaginary and real parts of the susceptibility transformed to our case $T_1 = T_2 = T(2\pi\Gamma)^{-1}$:[1)]

$$\chi'(\omega) = -4\hbar\pi^2\gamma^2 q_{cr}\,\mathrm{Im}\left\{\frac{(\Gamma-i\mathscr{E})(\Gamma-i\mathscr{E}_0)(\Gamma-i\mathscr{E}_0-i\mathscr{E})+2i\mathscr{E}|V_0|^2}{(\Gamma-i\mathscr{E}_0)L(-i\mathscr{E})}\right\}, \quad (2)$$

$$\chi''(\omega) = 4\hbar\pi^2\gamma^2 q_{cr}\,\mathrm{Re}\left\{\frac{(\Gamma-i\mathscr{E})(\Gamma-i\mathscr{E}_0)(\Gamma-i\mathscr{E}_0-i\mathscr{E})+2i\mathscr{E}|V_0|^2}{(\Gamma-i\mathscr{E}_0)L(-i\mathscr{E})}\right\}, \quad (3)$$

$$q_{cr} = \frac{q_0}{1+4|V_0|^2(\mathscr{E}_0^2+\Gamma^2)^{-1}},$$

$$L(x) = x^3+3\Gamma x^2+(\mathscr{E}_0^2+3\Gamma^2+4|V_0|^2)x+\Gamma(\mathscr{E}_0^2+\Gamma^2)+4\Gamma|V_0|^2.$$

Here $V_0 = i\pi\gamma H_0$, $\mathscr{E} = \omega - \omega_0$, $\mathscr{E}_0 = \omega_{21} - \omega_0$, $q = \rho_{11} - \rho_{22}$, and q_0 is the initial difference between the populations of the lower and upper levels.

The continuous curves in Figs. 4 and 5 are the results of calculations based on Eqs. (2) and (3), on which the experimental results are superimposed. The quantities $\mathscr{E}_0$, V_0, and Γ were measured and controlled during the recording of the curves. The discrepancies in the case of the strong resonance field $\omega_0 = \omega_{21}$ could be partly due to the instability of the magnetic field H_Z due to fluctuations in the laboratory magnetic field, which caused the resonance frequency ω_{21} to fluctuate within ~1 Hz during the recording process. However, this failed to explain why the widths of the experimentally obtained resonance curves were half the theoretically predicted widths. This discrepancy could not be due to the contribution of the inhomogeneity of the magnetic field, which was negligible, because the line width of 2 Hz agreed with the measured relaxation times T_1 and T_2, which were both equal to $(80 \pm 10) \times 10^{-3}$ sec.

The longitudinal T_1 and transverse T_2 relaxation times were measured by a pulse method, described in detail by Abragam.[20] The transverse relaxation time T_2 was determined from the decay of the optical orientation signal after the application of a $\pi/2$ rf field pulse. The decay of the transverse component of the magnetic moment of the atoms $M_\perp$ in a homogeneous static magnetic field was characterized by the time constant T_2 (Fig. 10a). The results of measurements of the longitudinal relaxation time T_2 are shown in Fig. 10b. After the first π pulse, the longitudinal magnetization M_Z, proportional to the difference between the populations, assumed the value $-M_Z$ and relaxed from $-M_Z$ to $+M_Z$, in accordance with the law $M_Z = M_0(1 - 2e^{-1/T_1})$. If at a moment t_1, when $M_Z = 0$, a second π pulse was applied to the system, the response to this pulse was zero. We measured t_1 using the oscillograms and then calculated the time constant $T_1 = t_1/\ln 2$.

When the strong field was considerably off resonance so that $|\omega_0 - \omega_{21}| \gg \omega_{21}$ and $|V_0| \gg \Gamma$, the influence of this field on the atomic system depended strongly on the nature of its polarization. For the fields used in our experiments, which were circularly polarized, the expressions for $\chi'(\omega)$ and $\chi''(\omega)$ were

$$\chi'(\omega) = -4\hbar\pi^2\gamma^2 q_0\,\mathrm{Im}\left\{\frac{\omega_{21}-\omega-2|V_0|^2/\omega_0}{\Gamma^2+(\omega_{21}-\omega-2|V_0|^2/\omega_0)^2} - \frac{\omega_{21}+\omega-2|V_0|^2/\omega_0}{\Gamma^2+(\omega_{21}+\omega-2|V_0|^2/\omega_0)^2}\right\}, \quad (4)$$

$$\chi''(\omega) = 4\hbar\pi^2\gamma^2 q_0\,\mathrm{Re}\left\{\frac{\Gamma}{\Gamma^2+(\omega_{21}-\omega-2|V_0|^2/\omega_0)^2} - \frac{\Gamma}{\Gamma^2+(\omega_{21}+\omega-2|V_0|^2/\omega_0)^2}\right\}. \quad (5)$$

It is clear from the formulas (4) and (5) that an increase in amplitude of the strong field shifted the resonance frequency proportionally to H_0^2 in the direction of lower frequencies $\omega = \omega_{21} - 2|V_0|^2/\omega_0$. For $2|V_0|^2/\omega_0 \geq \omega_{21}$, the resonance of the circularly polarized right-handed component of the weak field [represented by the first terms in Eqs. (4) and (5)] disappeared and the resonance of the left-handed circularly polarized component (represented by the second terms in the same equations) was observed. The new resonance represented the amplification of the weak field and not its absorption. In this case, the strong field did not equalize the populations but shifted the levels without broadening and a population inversion resulted from the crossing of the levels (Fig. 9b). All these conclusions, deduced from the formulas (4) and (5), were confirmed by the experimental results (see Sec. 2).

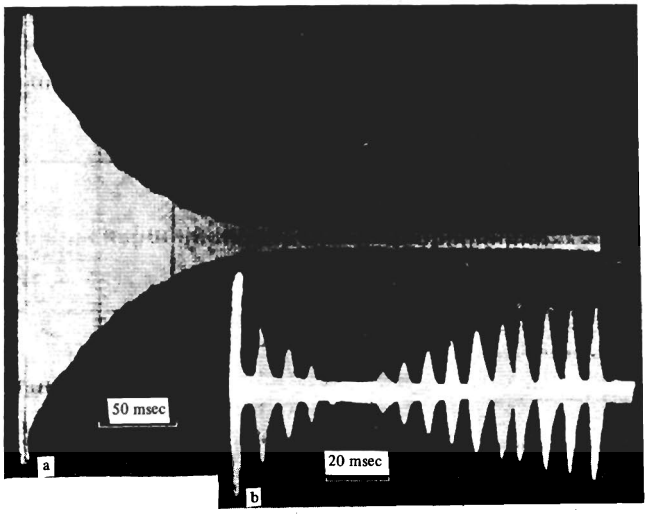

FIG. 10. a) Oscillogram of a signal produced by a $\pi/2$ rf field pulse in the measurement of the transverse relaxation time T_2. b) Oscillogram of a signal produced by two π rf field pulses in the measurement of the longitudinal relaxation time T_1.

4. CONCLUSIONS

It seems to us that amplification without population inversion provides a new interesting operation principle for laser amplifiers and oscillators. In the optical range, the most promising are atomic vapors with narrow absorption lines and large oscillator strengths. For example, in the case of saturated alkali metal vapors, the main absorption doublets have oscillator strengths of ~1 and the maximum absorption coefficients are of the order of 10^4 cm^{-1} for a line width of ~0.03 cm^{-1}. Estimates show that lasers capable of producing a power density of 10^8 W/cm^2 should ensure a gain of the order of 10^2 cm^{-1} in a tuning range of the order of 100 Å near the atomic lines and the width of the gain profile should be of the order of 10^{-2} Å.

An important point in the construction of such amplifiers would be the degree of monochromaticity of the exciting pump radiation. The characteristic features of changes in the absorption spectra of a two-level system in strong nonmonochromatic fields are being investigated by us at present.

The authors are deeply grateful to E. B. Aleksandrov for his help in the development of the experimental method and in building the apparatus used in the optical orientation of cadmium atoms. They are also indebted to A. L. Natadze and V. G. Fayans for writing a computer program.

[1)]We used linear rf fields to simplify the apparatus. The perturbation due to the second (circularly polarized left-handed) component of the field shifted the atomic transition frequency by $\Delta = \gamma^2 H_0^2/2\nu_0 = 10^{-2}$ Hz, which was much less than the line width $\Gamma = 2$ Hz. Therefore, we assumed that the effects of linear and rotating fields were identical.

[1] S. G. Rautian and I. I. Sobel'man, Zh. Eksp. Teor. Fiz. 41, 456 (1961) [Sov. Phys.-JETP 14, 328 (1962)]; 44, 934 (1963) [Sov. Phys.-JETP 17, 635 (1963)].
[2] P. A. Apanasevich, Opt. Spektrosk. 14, 612 (1963); 16, 709 (1964).
[3] T. I. Kuznetsova and S. G. Rautian, Zh. Eksp. Teor. Fiz. 49, 1605 (1965) [Sov. Phys.-JETP 22, 1098 (1966)].
[4] S. G. Rautian, in: Nelineĭnaya optika (Nonlinear Optics), Nauka, M., 1968, p. 3.
[5] P. A. Apanasevich, Dokl. Akad. Nauk B. SSR 12, 868 (1968).
[6] T. I. Kuznetsova, in: Nelineĭnya optika (Nonlinear Optics), Nauka, M., 1968, p. 116.
[7] N. N. Kostin, M. P. Sokolova, V. A. Khodovoĭ, and V. V. Khromov, Zh. Eksp. Teor. Fiz. 62, 475 (1972) [Sov. Phys.-JETP 35, 253 (1972)].
[8] A. M. Bonch-Bruevich, S. G. Przhibel'skiĭ, and V. A. Khodovoĭ, V. sb. trudov "Konferentsii po vzaimodeĭstviyu elektronov s sil'nym élektromagnitnym polem," Balatonfyured, 11-16 sentyabrya 1972 g. (Proc. Conf. on Interaction of Electrons with Strong Electromagnetic Fields, Balatonfüred, September 11-16, 1972), Budapest, 1973, p. 255.
[9] Yu. M. Kirin, S. G. Rautian, V. P. Safonov, and B. M. Chernobrod, Zh. Eksp. Teor. Fiz. 62, 466 (1972) [Sov. Phys.-JETP 35, 248 (1972)].
[10] A. M. Bonch-Bruevich, N. N. Kostin, S. G. Przhibel'skiĭ, V. A. Khodovoĭ, V. V. Khromov, and N. A. Chigir', in: Nelineĭnye protsessy v optike (Nonlinear Processes in Optics), No. 2, Novosibirsk, 1972, p. 75.
[11] A. M. Bonch-Bruevich, V. A. Khodovoĭ, and V. V. Khromov, ZhETF Pis'ma Red. 14, 487 (1971) [JETP Lett. 14, 333 (1971)].
[12] G. A. Delone, in: Sb. trudov "Konferentsii po vzaimodeĭstviyu élektronov s sil'nym elektromagnitnym polem," Balatonfyured, 11-16 sentyabrya 1972 g. (Proc. Conf. on Interaction of Electrons with Strong Electromagnetic Fields, Balatonfüred, September 11-16, 1972), Budapest, 1973, p. 77.
[13] A. M. Bonch-Bruevich and V. A. Khodovoĭ, Usp. Fiz. Nauk 93, 71 (1967) [Sov. Phys.-Usp. 10, 637 (1968)].
[14] Ya. B. Zel'dovich, Usp. Fiz. Nauk 110, 139 (1973) [Sov. Phys.-Usp. 16, 427 (1973)].
[15] V. G. Kolomiets, Zh. Prikl. Spektrosk. 19, 1020 (1973).
[16] D. N. Klyshko, Yu. S. Konstantinov, and V. S. Tumanov, Izv. Vyssh. Uchebn. Zaved. Radiofiz. 8, 513 (1965).
[17] L. N. Novikov, V. G. Pokazan'ev, and G. V. Skrotskiĭ, Usp. Fiz. Nauk 101, 273 (1970) [Sov. Phys.-Usp. 13, 384 (1970)].
[18] E. B. Aleksandrov and A. P. Sokolov, ZhETF Pis'ma Red. 3, 419 (1966) [JETP Lett. 3, 273 (1966)].
[19] E. B. Aleksandrov and A. P. Sokolov, Opt. Spektrosk. 27, 896 (1969).
[20] A. Abragam, The Principles of Nuclear Magnetism, Clarendon Press, Oxford, 1961 (Russ. Transl., IIL, M., 1963).

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