CHANGES IN THE ATOMIC ABSORPTION SPECTRUM IN THE FIELD OF A LIGHT WAVE. I.

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Submitted July 1, 1968

Zh. Eksp. Teor. Fiz. 56, 144–150 (January, 1969)

The shift and splitting of the absorption D line of the main doublet of atomic potassium ($4S_{1/2} - 4P_{1/2}$ transition of the D_1 line and $4S_{1/2} - 4P_{3/2}$ of the D_2 line) in the intense ruby laser emission field are investigated and interpreted theoretically. The observed change in the potassium absorption spectrum depends on the presence of resonance between the $6S_{1/2} - 4P_{3/2,1/2}$ transition wavelength and the laser emission wavelength. The latter was varied in the 6936-6943 Å range by varying the ruby rod temperature. The nonresonant field of the laser emission gave rise to an absorption D line shift proportional to the laser emission intensity and to new lines corresponding to two-photon transitions into the $6S_{1/2}$ state. Splitting of the D_2 line was observed when the laser wavelength coincided with $\lambda_0 = 6939$ Å of the $6S_{1/2} - 4P_{3/2}$ transition. The D line shift constants measured in the nonresonant laser field are compared with those predicted by the theory.

1. INTRODUCTION

D-line shift and splitting in the main doublet of a potassium atom in an intense ruby laser emission field were observed in[1,2]. The signal recording methods used in these experiments are applicable only to a qualitative study of changes in the atomic spectra occurring in intense fields of optical emission. Better results are obtained from photographic recording of the atomic absorption spectrum in the laser emission field[3]. The present paper reports on the results of photographic recording of the main doublet D-line shift in potassium due to a ruby laser emission field and gives a theoretical interpretation of the data.

2. THEORY

An exact theory of the changes in the atomic absorption spectrum due to a strong radiation field is very complex and a general computation of such changes is not practicable at this time[3,4]. The problem is much easier in the case of potassium atoms because the ruby laser emission wavelength (6943 Å) is close to the wavelengths of the $6S_{1/2} - 4P_{3/2}$ ($\lambda = 6939$ Å) and $6S_{1/2} - 4P_{1/2}$ ($\lambda = 6911$ Å) transitions. Therefore the computation of the spectral characteristics of the D_1 and D_2 lines of the main doublet (Fig. 1) can be limited to the perturbation of the $4P_{3/2,1/2}$ terms by stimulated transitions into the $6S_{1/2}$ state. If the ruby laser emission is linearly polarized, the selection rule $\Delta m_J = 0$ (where m_J is the projection of the electron shell momentum on the electric field direction of the laser emission) provides that levels $4P \pm 1/2$ of term $4P_{1/2}$ and $4P \pm 1/2$ of term $4P_{3/2}$ are perturbed, while levels $4P \pm 3/2$ of term $4P_{3/2}$ are not perturbed. Furthermore, if the emission polarization of a test source is parallel to that of the laser emission, the same selection rule ($\Delta m_J = 0$) prohibits the absorption of this emission with a transition into unperturbed states $4P \pm 3/2$. Consequently under these conditions the theoretical model of the potassium atom in which absorption is observed in each of the D lines reduces to a three-level system: level 1—$4S \pm 1/2$, level 3—$6S \pm 1/2$, and level 2—$4P \pm 1/2$ (different for the D_1 and D_2 lines) (Fig. 1b)[1)].

It can be shown that, in the approximation of a weak emission field with a broad spectrum in channel 1–2 allowing us to neglect the population of level 1, the stationary absorption regime in channel 1–2 is determined by the absorption coefficient

$$K(\omega) = K_0 \operatorname{Im} \frac{\gamma_{21}(\omega_{32} + \omega_{21} - \omega_2 - \omega - i\gamma_{31}^*)}{(\omega - \omega_1^*)(\omega - \omega_2^*)}. \quad (1)$$

Here, K_0 is the maximum absorption coefficient for $\omega = \omega_{21}$ in the absence of laser emission, ω_2 is the central laser emission frequency, γ_{21} is the reciprocal of the time constant of transverse relaxation in channel 1–2, $\gamma_{31}^* = \gamma_{31} + \Delta\omega$, where γ_{31} is the reciprocal of the time constant of transverse relaxation in channel 1–3, and $2\Delta\omega$ is the spectral width of laser emission

$$\omega_1^* = \omega_{21} + \frac{1}{2}\left\{\omega_{32} - \omega_2 - i(\gamma_{31}^* + \gamma_{21}) + \left[\frac{d_{32}^2 E_2^2}{\hbar^2} - [\gamma_{31}^* - \gamma_{21} + i(\omega_{32} - \omega_2)]^2\right]^{1/2}\right\},$$

$$\omega_2^* = \omega_{21} + \frac{1}{2}\left\{\omega_{32} - \omega_2 - i(\gamma_{31}^* + \gamma_{21}) - \left[\frac{d_{32}^2 E_2^2}{\hbar^2} - [\gamma_{31}^* - \gamma_{21} + i(\omega_{32} - \omega_2)]^2\right]^{1/2}\right\}, \quad (2)$$

where d_{32} is the matrix element of the dipole moment of transition 3-2 and $E_2 = (8\pi I_0/c)^{1/2}$, I_0 is laser emission power density, c is the velocity of light, $\hbar = h/2\pi$, and h is Planck's constant. The rate of establishment of the stationary absorption regime is found to be maximum from the time constants $T_{21} = 1/\gamma_{21}$ and $T_{31}^* = 1/(\gamma_{31} + \Delta\omega)$. We show below that the approximations of a weak field in channel 1-2 and of a stationary absorption regime hold well in our experiments. In the general case the analysis of (1) is quite complicated and therefore we consider only some

[1)] For the sake of simplicity we consider henceforth that levels 1, 2, and 3 are non-degenerate because in a linearly polarized laser emission field the levels $\pm m_J$ are perturbed equally and the consideration of degeneracy does not, in principle, contribute any new results.

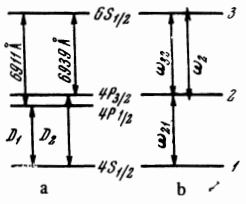

FIG. 1. Term diagram of a potassium atom. a – diagram of real terms; b – three-level theoretical model.

limiting cases relevant to the experiment described below.

a. Laser emission field is absent, $E_2 = 0$. In this case

$$K(\omega) = K_0 \frac{\gamma_{21}^2}{(\omega - \omega_{21})^2 + \gamma_{21}^2}. \quad (3)$$

The absorption line in channel 1-2 consists of a single component and has a Lorentz shape with a width of $2\gamma_{21}$ (at half altitude); the absorption coefficient at the maximum is equal to K_0.

b. Quasi-monochromatic nonresonant field, "weak", $\gamma_{31}^* \ll \gamma_{21}$, $d_{32}^2 E_2^2/\hbar \ll (\omega_{32} - \omega_2)^2 - \gamma_{21}^2$. Here

$$K(\omega) = K_0 \operatorname{Im} \frac{\gamma_{21}(\omega_{32} - \omega_2 + \omega_{21} - \omega - i\gamma_{31}^*)}{[\omega - \omega_{21} + \Delta_1 + i(\gamma_{21} - \gamma_1)][\omega - \omega_{21} - \omega_{32} + \omega_2 - \Delta_1 + i\gamma_1]} \quad (4)$$

where

$$\Delta_1 = \frac{1}{4} \frac{(\omega_{32} - \omega_2)}{(\omega_{32} - \omega_2)^2 + \gamma_{21}^2} \frac{d_{32}^2 E_2^2}{\hbar^2}, \quad (5)$$

$$\gamma_1 = \frac{1}{4} \frac{\gamma_{21}}{(\omega_{32} - \omega_2)^2 + \gamma_{21}^2} \frac{d_{32}^2 E_2^2}{\hbar^2} \quad (6)$$

(in the derivation of these equations it was assumed that $\gamma_{31}^* \ll \gamma_1$). The absorption line in channel 1-2 is split into two: a "one-photon" line with a frequency $\omega = \omega_{21} - \Delta_1$ and a "two-photon" line with a frequency $\omega = \omega_{21} + \omega_{32} - \omega_2 + \Delta_1$ (for the basis of this interpretation see[4]). The frequencies of these lines are shifted in opposite directions relative to their initial positions with $E_2 \to 0$ prorptional to E_2^2 (quadratic Stark effect). The shift sign of each line depends on the sign of the difference $\omega_{32} - \omega_2$; levels 2 and 3 are attracted when $\omega_2 > \omega_{32}$ and repelled when $\omega_2 < \omega_{32}$[4]. The width of the "one-photon" absorption line is in this case close to the initial width $2\gamma_{21}$ and the absorption coefficient in the maximum is close to K_0. The width of a "two-photon" line is less than that of the "one-photon" line and is proportional to E_2^2 and the absorption coefficient at the maximum equals K_0.

c. Quasi-monochromatic resonant field, "strong", $\omega_{32} = \omega_{21} \gamma_{31}^* \ll \gamma_{21}$, $d_{32} E_2/\hbar \gg \gamma_{21}$. In this case

$$K(\omega) = K_0 \operatorname{Im} \frac{\gamma_{21}(\omega_{21} - \omega - i\gamma_{31}^*)}{[\omega - \omega_{21} - d_{32}E_2/2\hbar + i\gamma_{21}/2][\omega - \omega_{21} + d_{32}E_2/2\hbar + i\gamma_{21}/2]} \quad (7)$$

The absorption line in channel 1-2 is split into two components that are shifted symmetrically with respect to ω_{21} by a distance proportional to E_2 (linear Stark effect). The line width and the absorption coefficient in the maximum of each component equal γ_{21} and K_0 respectively.

3. THE EXPERIMENTAL SETUP

The field power densities of the ruby laser emission that are high enough for the experimental detection of the potassium D line shift are currently available only in giant pulses 10^{-8}–10^{-7} sec long. Photographic recording of the changes in the potassium vapor absorption spectrum in the ruby laser field requires a pulse radiation test source with a spectrum overlapping the absorption D line region (~770 nm) and synchronized with the laser pulse. The brightness of ordinary (nonlaser) radiation sources is several orders lower than that required for the photographic recording of the spectrum in a single laser pulse. Additional difficulties involved in the use of ordinary radiation sources are due to the need to synchronize them with the laser pulse with an accuracy of $\sim 10^{-8}$ sec, posing a serious experimental problem.

In this work we used the stimulated fluorescence (SF) of dye solutions[5] as the test source of synchronous radiation. The experimental diagram is given in Fig. 2. A large portion of the emission of ruby laser 1 with a passive shutter of KS-19 glass ($\Delta t = 3 \times 10^{-8}$ sec, divergence angle $\sim 10^{-2}$ rad), separated by a stack of two glass plates (or a mirror with a reflection coefficient $R_2 = 40\%$), was directed by lens 2 (with a focal length $f_2 = 10$ cm) into cell 3 with potassium vapor. A dielectric spherical mirror with a radius of curvature of 1 m and $R_1 = 90\%$ was used as one of the laser resonator mirrors in order to improve the homogeneity of the spatial distribution of the laser emission field in the cell. A smaller portion of the laser emission emerging through the spherical mirror was focused by cylindrical lens 4 (having a focal length

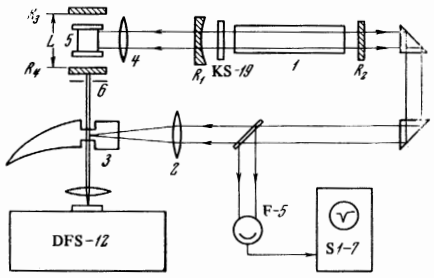

FIG. 2. The experimental setup. 1 – ruby laser; 2, 4 – lenses; 3 – potassium vapor cell; 5 – dye solution cell; 6 – diaphragm.

$f_4 = 10$ cm) into a cell with dye solution 5 ($l = 2$ cm) placed in the resonator ($L = 16$ cm) consisting of mirrors with reflection coefficients $R_3 = 99\%$ and $R_4 = 40\%$. The dye solutions used in the experiment were: kryptocyanine, 1,1′-diethyl-2,2′-dicarbocyanine iodide and 1,1′-diethyl-γ-nitro-4,4′-dicarbocyanine tetrafluoroborate in glycerine. The discrete structure of the SF spectrum[6] was eliminated by an appropriate solution concentration, the use of flat windows of equal thickness of 1.5 cm in the solution cell, and, if necessary, by the replacement of the flat mirror ($R_3 = 99\%$) with a spherical one having the same reflectivity and a curvature radius of 40 cm.

Such a system made it possible to reach an SF emission power of the order of 1 MW with a beam divergence of $\sim 10^{-2}$ rad for a ruby laser pump pulse power of ~10MW. The SF emission was linearly polarized in the direction of polarization of the laser emis-

sion and its spectrum represented a bandwidth from 50 Å (with a small excess over generation threshold) to 500 Å (with a multiple excess over generation threshold). The center of the band was in the region of ~770 nm and its intensity distribution near the D_1 (λ = 7665 Å) and D_2 (λ = 7699 Å absorption lines of the main potassium doublet was approximately uniform. The length of the SF pulse at the time instant corresponding to its intensity maximum substantially depended on the pumping conditions and was different for different spectral regions of the SF band. The general nature of these dependencies is similar to that described in[7]. In the spectral region of interest of 765–770 nm the SF pulse maximum approximately coincided with the maximum of the laser pulse and its length was equal to or somewhat less than (~1.5 times) the laser pulse length. Diaphragm 6 (7 × 0.3 mm) was placed in the path of the SF beam in the region of beam intersection to shape the SF beam. Buth beams were introduced into the cell containing potassium vapor (Wood horn of molybdenum glass) through flat sapphire windows. This construction of the cell preserved the beam configuration and the constant transparency of the windows (the molybdenum glass darkens rapidly at the temperatures of ~300°C due to the absorption of potassium).

The absorption spectrum of potassium vapor was recorded photographically by a spectrograph based on the DFS-12 spectrometer. The latter featured special diffraction gratings (600 lines/mm) having a considerable reflection coefficient in the region of 770 nm in the third order. The dispersion of the spectrograph was 1.85 Å/mm in the region of 770 nm. We used the I-810 film whose sensitivity in the 770 nm region ensured a measurable darkening for an emission energy density of ~1 erg/cm^2. A spectral power density of the SF beam of the order of 10^3 w/cm is necessary to measure the absorption spectrum in a single pulse for an input spectrograph slit of 0.05 nm (instrument width of 0.2 cm^{-1}).

4. EXPERIMENTAL RESULTS AND DISCUSSION

The appropriate potassium vapor pressure for the measurement of absorption spectrum significantly depends on the emission power density of the test source necessary for the photographic recording of the absorption spectrum in a single laser pulse. In our experiment, using the test source power density $I(\omega) = 10^3$ w/cm, saturation of absorption in channel 1-2 was absent at potassium vapor cell temperature of ~ 300°C; the Lorentz absorption line width was determined here by potassium atom collision and amounted to $2\gamma_{21} = 0.3$ cm^{-1}. The theoretical evaluation[8] of the transition rates in D_1 and D_2 lines shows that such values of $2\gamma_{21}$ and $I(\omega)$ do justify the weak field approximation in the test source channel permitting us to neglect the change in the ground level population. Preliminary experiments verified the fact that the test source emission does not cause broadening or shifting of the absorption D lines. Finally, the time constants T_{31}^{*}, $T_{21} < 0.3$ nsec for a laser pulse length of 30 nsec and spectral width (depending on the resonator parameters) of 0.02 – 0.1 cm^{-1}; i.e., the approximation of a stationary absorption regime is satisfied.

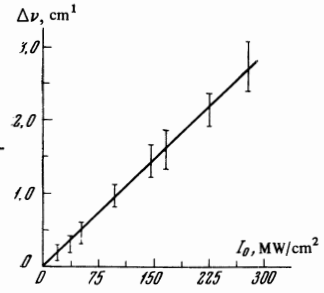

FIG. 3. Shift of the absorption D_2 line of a potassium atom as a function of ruby laser emission intensity.

We measured the absorption D_2 line shift as a function of laser emission intensity (Fig. 3) for a nonresonant ruby laser emission field (λ = 6942.5 Å). As reference we used the emission D_2 line of a potassium resonance lamp whose wavelength coincides with the center of the absorption D_2 line of potassium vapor in the absence of laser emission. According to the theory, the magnitude of the shift depends linearly on the laser emission intensity. Allowing for possible systematic errors due mainly to the measurement of the absolute value of emission density in the potassium vapor cell, the shift constant $\beta = (1 \pm 0.5) \times 10^{-8}$ cm/W.

The theoretical value of β can be computed from (5) using data available in the literature on the oscillator strength of the $6S_{1/2} \rightarrow 4P_{3/2}$ transition. According to[9-13] its value lies within the limits 0.033–0.044 which yields the value of $\beta = (1.1-1.5) \times 10^{-8}$ cm/W. In[14] the oscillator strength of this transition is estimated at 0.2 so that[2)] $\beta = 6.7 \times 10^{-8}$ cm/W. Thus the value of β measured by us is in good agreement with the data in[9-13], whereas the value of β computed from[14] lies consideraboy beyond the error of our measurements.

The shift constant of the D_1 line measured under the same conditions was found to be approximately 7 times lower which corresponds to the expected computed value if we use the data in[9-13] and allow for the stimulated transitions $4S_{1/2} - 4P_{1/2}$ in the laser emission field.

The width of the D_2 line in the laser emission field exceeded that of the initial line (in the absence of the laser pulse) by approximately the magnitude of the shift. The causes of this broadening are not yet clear although we assume that it is most probably due to the space-time inhomogeneity of the laser emission field distribution in the potassium vapor cell.

As we know the ruby laser wavelength depends on the temperature of the ruby rod. When the latter varies within the range from −200°C to +20°C the laser emission wavelength λ_2 varies from 6935 to 6943 Å. Under these conditions we observed the change in the nature of the absorption spectrum near the D_2 line (Fig. 4). According to the theory, two lines are observed when $\lambda_2 \neq 6939$ Å (λ_{32}): a one-photon line near λ = 7699 Å, and a two-photon line shifted with respect to the former by $\Delta\omega = \omega_2 - \omega_{32}$. The sign of the shift of the two-photon line (Stokes or anti-Stokes region) change with

[2)]The computation based on the data in [9,10,13] was performed without allowing for the spin-orbital interaction.

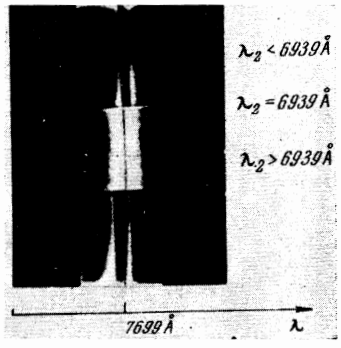

FIG. 4. Changes in the potassium absorption spectrum near the D_2 line with varying ruby laser emission wavelength λ_2.

the change of sign of $\Delta\omega$. Figure 4 shows three absorption spectra for three values of $\Delta\lambda$: positive, zero, and negative. The bright areas in the photograph correspond to absorption. The black narrow line passing through all three spectra is the reference line marking the center of gravity of the absorption D_2 line of the potassium resonance lamp. As predicted by theory the one-photon line is shifted by the laser emission, the sign of the shift changing in passing the resonance value of the laser emission wavelength, λ_{32} = 6939 Å. At the exact point of resonance we observed splitting of the absorption D_2 line; this is reflected in Fig. 4 by the absence of a shift in the absorption D_2 line and the considerable broadening of this line (~5 times).

Thus the results of the performed experiments are in agreement with theoretical considerations.

[1] E. B. Aleksandrov, A. M. Bonch-Bruevich, N. N. Kostin, and V. A. Khodovoĭ, ZhETF Pis. Red. 3, 85 (1966) [JETP Lett. 3, 53 (1966)].

[2] A. M. Bonch-Bruevich, N. N. Kostin, and V. A. Khodovoĭ, ZhETF Pis. Red. 3, 425 (1966) [JETP Lett. 3, 279 (1966)].

[3] V. A. Khodovoĭ. Investigation of the Stark Effect in Optical Frequency Fields. Author's abstract of a thesis for the degree of a Candidate of Physico-Mathematical Sciences, 1968.

[4] A. M. Bonch-Bruevich and V. A. Khodovoĭ, Usp. Fiz. Nauk 93, 71 (1967) [Sov. Phys. Usp. 10, 637 (1968)].

[5] B. I. Stepanov and A. N. Rubinov, ibid. 95, 45 (1968) [11, 304 (1968)].

[6] A. M. Bonch-Bruevich, N. N. Kostin, and V. A. Khodovoĭ, Opt. Spektrosk. 25, 2 (1968).

[7] M. Bass and J. I. Steinfeld, IEEE, J. Quant. Electron. 4, 53 (1968).

[8] V. G. Abramov, O. V. Konstantinov, N. N. Kostin, and V. A. Khodovoĭ, Zh. Eksp. Teor. Fiz. 53, 822 (1967) [Sov. Phys. JETP 26, 503 (1968)].

[9] E. M. Anderson and V. A. Zilitis, Opt. Spektrosk. 16, 177 (1964).

[10] D. S.Villars, J. Opt. Soc. Amer. 42, 552 (1952).

[11] E. F. M. Van der Held and J. H. Heierman, Physica 2, 71 (1935).

[12] E. F. M. Van der Held and J. H. Heierman, Physica 3, 31 (1936).

[13] L. S. Ornstein and J. Key, Physica 1, 945 (1934).

[14] H. Corliss and W. R. Bozman, Natl. Bur. Std. (US), Monograph, 53, 1962.

Translated by S. Kassel
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