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MEASUREMENT OF THE PERTURBATION OF ATOMIC LEVELS BY INTENSE LIGHT USING THE PROCESS OF RESONANT MULTIPHOTON IONIZATION

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A new method is proposed for measuring the Stark shift of high atomic levels in a strong optical field. The method is based on observing resonant multiphoton ionization of the atom. The multiphoton ionization of the 2^3S metastable state of the helium atom is investigated, and the values of the Stark shift are obtained for a number of levels.

The possibilities of investigating the perturbation of atomic levels in an optical field have been greatly expanded of late. In the experimental domain, this is due to the development of high-power lasers, which make it possible to observe the perturbations in a strong field [1]; theoretically, this is made possible by exact knowledge of the electron wave functions in a complex atom and by the development of various new computation methods, which make it possible to calculate the dynamic polarizability of atoms [2]. The fundamental circumstance in this case is that sufficiently high level can be excited in an optical field only as a result of multiphoton transitions, the probability of which is high enough only if the field intensity is high enough. Typical of such excited levels is the large probability of the induced transition to a high-energy state, when compared with the probability of spontaneous relaxation. Under these conditions no classical method, whether emission spectroscopy or absorption of auxiliary light, makes it possible to observe the perturbation of the levels. We have developed a new method of measuring the perturbation of such states; this method is based on observing resonant multiphoton ionization of the atom.

Within the framework of perturbation theory, the probability of multiphoton ionization of an atom is connected with the radiation intensity F by the relation [3]

$$W(F) = A(F)F^{k_0} = \text{const}\left|\sum_{l_1 \ldots i \ldots n} \frac{\langle f|r|l\rangle \ldots \langle n|r|0\rangle}{\ldots [E_i(F) - E_0(F) - k\hbar\omega - i\gamma_i(F)] \ldots}\right|^2 F^{k_0}, \quad (1)$$

where $k_0 = \langle (I/\hbar\omega) + 1\rangle$; I is the ionization potential of the atom; $k < k_0$; r is the electron coordinate; $E_i(F)$ and $\gamma_i(F)$ are the energy and width of the i-th level in the radiation field.

If the energy of a certain number $k < k_0$ of quanta coincides with the energy $E_i(F)$ of some atomic level, the dependence of the multiphoton-ionization probability on the light intensity has a typically resonant character and contains information on the shift and broadening of the resonant state in the light field [4]. We shall henceforth assume for simplicity that only one level is at resonance [5]. In this case, the corresponding energy denominator in expression (1) is much smaller than all the remaining ones, we can neglect all but the resonant term in the summation, and (1) goes over into

$$W(F) = \text{const}\frac{F^{k_0}}{[E_i(F) - E_0(F) - k\hbar\omega]^2 + \gamma_i^2(F)}. \quad (2)$$

We assume that the change of the level energy depends linearly on the light intensity:

$$E_i = E_{i0} + \Delta E_i(F) = E_{i0} + c_i F \tag{3}$$

and then (2) takes the form

$$W(F) = \text{const} \frac{F^{k_0}}{(\Delta\omega_i - c_{i0}F)^2 + \gamma_i^2(F)}, \tag{4}$$

where $\Delta\omega_i = E_{i0} - E_{00} - k\hbar\omega$ is the static detuning of the resonance (in the absence of a field), while $c_{i0} = c_i - c_0$ is the Stark constant and characterizes the change of the energy of the transition between the states 0 and i.

Assuming a Gaussian distribution of the optical-field intensity and a Lorentz contour of the laser line, the effective width of the resonance is $\gamma_{ieff}(F) = k\gamma_L + \gamma_i(F)$ [6], where γ_L is the laser line width. The width γ_0 of the initial level can be assumed equal to zero.

If the light is chosen to have a frequency ω_1, the resonance denominator in [5] becomes equal to zero at a definite light intensity $F_{ik}(\omega_1)$, and dynamic resonance sets in, i.e.,

$$E_{i0} - E_{00} - k\hbar\omega_1 - c_i(\omega_1)F_{iR}(\omega_1) = 0. \tag{5}$$

On going over to a radiation frequency

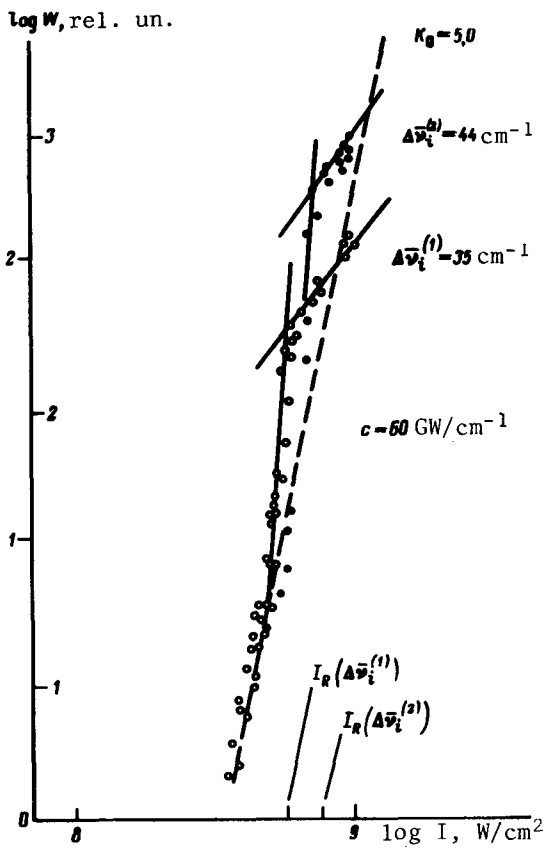

Fig. 1. Probability of five-photon ionization of triplet metastable helium atoms vs. the light intensity (F) in the case of resonance with the 13^3S state.

On going over to a radiation frequency ω_2, dynamic resonance sets in at another light-intensity value $F_{iR}(\omega_2)$. The plot of the resonant light intensity against the radiation frequency will therefore show also the dependence of the resonant-state energy on the intensity:

$$E(F_{iR}(\omega)) = k\hbar\omega + c_i(\omega)F_{iR} \tag{6}$$

if it is assumed that $E_0(F) = 0$.

In a small interval of variation of the light intensities near the intensity of the dynamic resonance, the slope of the log W(F, ω = const) plot changes very rapidly. But far from resonance the slope varies slowly, and we can therefore use a very simple approximate graphic method wherein the resonant light intensities $F_R(\omega)$ are determined as the points of in-ersection of the tangents of the log W(F, ω) curve just ahead and immediately past the resonance point.

By way of illustration, Fig. 1 shows two experimental curves obtained with a previously-described setup [7]. They show the probability of five-photon ionization of the triplet metastable 2^3S helium atoms as a function of the light intensity in the case of four-photon resonance with the 13^3S level; the parameter of the curves is the static detuning of the four-photon resonance.

It is possible to calculate the Stark shift constant from these two curves knowing only the static detunings, by determining the resonant light intensities from the intersections of the tangents. Assuming that the Stark constant remains unchanged in the investigated small range of frequency variation, we obtain for this constant the value $c_{i0} = 60\ \text{cm}^{-1}/\text{GW-sec}^{-2}$. The solid curve in Fig. 1 is drawn by using this constant for a Lorentz shape of the

resonant atomic line and for a rectangular distribution of the laser radiation.

By observing the resonant change of the multiphoton-ionization process in a sufficiently wide range of variation of the static deviation from resonance (the radiation frequency) and of the radiation intensity, we can measure the dependence of the resultant Stark shift on the radiation intensity. Figure 2 shows the result for the transition from the state 2^3S to the state 14^3S.

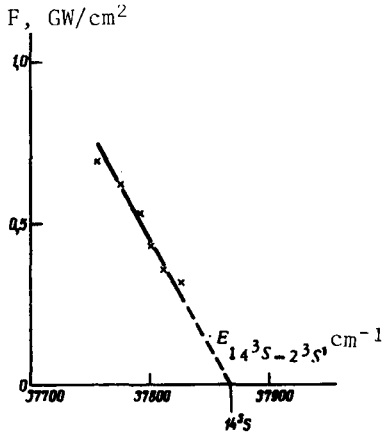

Fig. 2. Energy of transition between the states 2^3S and 14^3S in the field of a neodymium laser.

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EFFECTIVE MODULATION OF LIGHT WITH BARIUM-STRONTIUM NIOBATE SINGLE CRYSTALS

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$Ba_{0.25}Sr_{0.75}Nb_2O_6$ single crystals were used to modulate light from an He-Ne laser was modulated to a depty of 98% at a beam aperture 4×4 mm, a bias voltage ∿ 500 V/cm, and a half-wave voltage 50 V for a sample of unit dimensions.

Ferroelectric crystalline solid solutions of barium and strontiun niobates, $Ba_xSr_{1-x}Nb_2O_6$, have large transverse electro-optical effect coefficients, especially in the case of strontium-rich compositions [1]. Their use as electro-optical light modulators was limited until recently by the difficulty of obtaining homogeneous single crystals of these solid solutions. We report here effective modulation of a laser beam with crystals of composition $Ba_{0.25}Sr_{0.75}Nb_2O_6$.

The measurements were performed by a polarization-optical method [2] at a wavelength 0.63 μ. The voltage was applied to the sample either from an acoustic generator or from an infralow-frequency oscillator. The radiation source was an He-Ne laser and the indicator was a photomultiplier. The spontaneous birefringence of the crystals was calcelled out in the measurements with a quartz compensator.

The samples were plates cut along the c-axis of the crystal, with thickness 3 - 4 mm and cross section from 3×3 to 7×7 mm. The samples did not have the usual growth bands typical of barium-strongium niobate crystals [3, 4]. The refractive-index gradients did not exceed 1×10^{-5}/cm and 5×10^{-5}/cm for n_o and n_e, respectively. The transmission of the residual light flux, with the polarizers crossed at 45° to the crystal optical axis, was 2 - 4% at a beam aperture up to 4×4 mm. The faces of the plates were oriented perpendicular to the [100], [010], and [001] directions with accuracy ±2°, and were optically polished. The light beam was directed