Observation of two-photon dynamic Stark effect in a three-level Rydberg Na atom

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A two-photon dynamic Stark effect has been observed for the first time in the field of cw monochromatic radiation on transitions of Rydberg Na atoms. The total cross sections for the two-photon absorption have been measured.

The giant dipole moments corresponding to transitions between Rydberg states[1] should result in pronounced field effects at extremely low power densities of a microwave field. A Rydberg atom thus becomes a convenient tool for studying the system consisting of an atom and a strong resonant field. The observation of a dynamic Stark effect on single-photon transitions[2,3] makes it possible to carry out direct measurements of the microwave field. With a calibrated field, it becomes possible to measure transition dipole moments. The high probabilities for the two-photon transitions, which result from the specific values of the quantum defects of the P and S series of alkali metal atoms, make it possible to formulate an experiment to observe a dynamic two-photon Stark effect.

The Rydberg Na atoms in the $36P$ state are prepared by a three-stage excitation scheme, $3S_{1/2} \rightarrow 3P_{3/2} \rightarrow 4S_{1/2} \rightarrow nP_{1/2,3/2}$, as described in detail in our earlier papers.[4,5] In the present experiments, in contrast with Refs. 4 and 5, the regions in which the atomic beam interacted with the microwave radiation and in which the selective detection of Rydberg states was carried out by a field-ionization method were surrounded by a cooled ($T = 77$ K) copper shield. The shield suppressed the residual electric fields (5 mV/cm) and thermal-emission effects. The efficiency of the excitation of the $36P$ state was 10^{-4}, and at an atomic density of 5×10^6 at/cm^3 one Rydberg atom was present in the region of the interaction with the microwave radiation, on the average per excitation pulse. In the experiments we used the monochromatic radiation from two backward-wave-tube microwave sources, tunable over the range 53–78 GHz, with a linewidth ~ 1 MHz. (The linewidth reached ~ 3 MHz in the case of pulsed modulation.) One of the microwave sources was operated under conditions corresponding to a continuous, electrically controllable tuning of the frequency. This type of operation made it possible to carry out two-photon absorption spectroscopy.

We selected the $36P$–$37P$–$38P$ sequence of a three-level Rydberg atom with two two-photon transitions as the working sequence (Fig. 1a). The "strong" field at a fixed frequency was at resonance with the $36P_{3/2} \rightarrow 37P_{3/2}$ two-photon transition. During the measurement of the two-photon absorption on this transition, on the basis of the density of atoms in the $37P$ state, the fine structure of the two-photon absorption was resolved. This structure contains three components. The "weak" probing field was tuned near the $37P$–$38P$ two-photon transition; with detection of the atoms

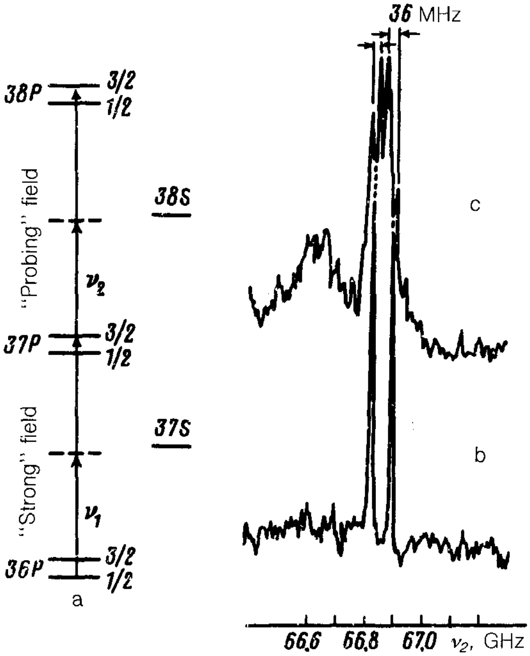

FIG. 1. Spectrum of two-photon absorption on the $37P$–$38P$ transition of Na in the presence of a field which is resonant with the $36P_{1/2}$–$37P_{3/2}$ transition. a—Scheme of working levels; b—two-photon absorption spectrum at a power density $I \approx 5 \times 10^{-7}$ W/cm^2 of the microwave radiation at the frequency ν_1; c—the same as in part b, but for $I \approx 2.8 \times 10^{-4}$ W/cm^2.

in the $38P$ state, this field produced two fine-structure components in the spectrum (Fig. 1b). As the intensity of the strong field was raised, we observed a field-induced broadening and then a splitting of the resonances, associated with a two-photon dynamic Stark effect (Fig. 1c). This probably constitutes the first direct observation of the effect in the field of coherent cw radiation. Figure 2 shows the experimental results on the splitting of the fine-structure component of the two-photon transition versus the field intensity. An absolute scale was established for the electric component of the microwave field from measurements of the single-photon dynamic Stark effect on the

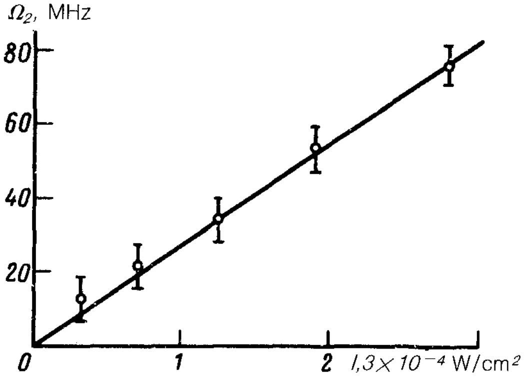

FIG. 2. Experimental results on the two-photon Rabi frequency versus the microwave power density.

38S–37P transition, which was also detected on the basis of the splitting of the line of the two-photon 36*P*–37*P* absorption.

The two-photon Rabi frequency, which determines the splitting of a level into two quasienergy levels, is given by[6]

$$\Omega_2 = \frac{1}{2}\sqrt{(\Delta_2)^2 + 4(V_{mn}^{(2)})^2} \quad . \tag{1}$$

Appearing in this expression is the frequency deviation from the two-photon resonance,

$$\Delta_2 = \omega_{mn} - 2\omega + V_{mm}^{(2)} - V_{nn}^{(2)} \quad , \tag{2}$$

and the nondiagonal two-photon matrix element $V_{nm}^{(2)}$, which are measure of the probability for two-photon absorption. A calculation of the magnitude of the matrix element $V_{nm}^{(2)}$ would generally require a summation over a large number of intermediate states. In the case at hand, the deviation of the virtual level from the real $37S_{1/2}$ level is small ($\delta \approx 2.48$ GHz). We can thus assume that a three-level approximation will be sufficient for a description of the two-photon absorption on *P*-*P* transitions. The two-photon matrix element then takes the form

$$V_{nm}^{(2)} = \frac{V_{nk}^{(1)} V_{km}^{(1)}}{4(\omega_{nk} - \omega)} = \frac{V_{nk}^{(1)} V_{km}^{(1)}}{4\delta} \quad , \tag{3}$$

where $V_{nk}^{(1)}$ and $V_{km}^{(1)}$ are the matrix elements of the dipole interaction of the atom with the field on single-photon transitions and $\omega_{nk} - \omega = \delta$ is the frequency deviation of the virtual level from the real level in this case ($37S_{1/2}$). At the exact resonance ($\Delta_2 = 0$), the two-photon Rabi frequency determines the distance between the split components, $\Omega_2 \approx V_{mn}^{(2)}$. We see that, in contrast with the Autler-Townes effect, the two-photon Rabi frequency is a linear function of the intensity, in agreement with our experimental results (Fig. 2). This result is further confirmation that the observed splitting is of a "two-photon" nature. This behavior opens up some new possibilities for studying the statistical properties of microwave radiation, in particular, in the two-photon micromaser which was recently discovered.[7] In the three-level approximation, the two-photon Rabi frequency can be written

$$\Omega_2 = \beta E^2 \, , \qquad \beta = \frac{d_{nk}\, d_{km}}{4\delta h^2} \quad . \tag{4}$$

The dipole-moment matrix elements calculated from the equations of Ref. 8 turn out to be $d_{36P-37} = 1591\ ea_0$ and $d_{37S-37P} = 1716\ ea_0$. For a frequency deviation $\delta = 2.48$ GHz we find $\beta_{\rm theo} = 447$ MHz/(V/cm)^2. The experimental value $\beta_{\rm expt} = 438 \pm 60$ MHz/(V/cm)^2 is evidence that the three-level approximation holds very accurately for the two-photon 36*P*–37*P* transition.

The cross section for two-photon absorption on the 36*P*–37*P* transition can be calculated from

$$\sigma = \frac{\pi^2 \omega_{mn}}{c^2 \hbar^3} |d_{mn}^{(2)}|^2 \frac{I}{\Delta\omega_L} , \tag{5}$$

where $d_{mn}^{(2)}$ is the two-photon matrix element found from the measured two-photon Rabi frequency, and $\Delta\omega_L$ is the homogeneous linewidth, determined in our case by transit-time effects. Using the measured microwave power density of 2.9×10^{-4} W/cm^2, we find from (5) the first reliable measurement of the cross section for two-photon absorption: $\sigma = 6\times10^{-13}$ cm^2. By way of comparison, the cross section for two-photon absorption corresponding to the strong $3s$–$4d$ transition of Na is, for the same level of the power density of optical radiation, $\sigma \approx 4\times10^{-24}$ cm^2, i.e., smaller by 11 orders of magnitude. The basic field properties of the observed spectrum are described by Eqs. (1)–(3), but it should be noted that a field resonance shifted ~200 MHz in the low-frequency direction appears. For it we do not have a satisfactory interpretation at this point.

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