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About measuring the forbidden 1S-2S transition frequency of a hydrogen atom by stimulated Raman scattering

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Abstract. We demonstrate the possibility of measuring the forbidden 1S-2S transition frequency (121 nm) of hydrogen atom by the method of stimulated Raman scattering (SRS) when atom from the 1S level moves to the 2S level through the intermediate 3P level. The frequency of the pumping field (103 nm) is close to the frequency of transition 3P-1S, and the frequency of the stimulated scattering (656 nm) - to the frequency of transition 3P-2S.

INTRODUCTION

The hydrogen atom is the simplest quantum system, the levels of which are measured and calculated with high accuracy. The possibility of precision measurement of the 1S–2S transition frequency using the method of two-photon absorption without Doppler broadening was considered in [1,2], and the measurement of the transition frequency was performed by the same method with relative accuracy $\sim 10^{-15}$ [3,4]. The use of this method is associated with two important factors - the interaction with the atom of two opposite waves of the same frequency eliminates both the Doppler broadening and the recoil effect in the absorption of photons. The main drawback of the method is that high field intensities are required, since their frequencies are far from resonance. However, there are now prerequisites for addressing this shortcoming. When atoms are cooled in traps, temperatures of the order of nanokelvin are obtained, which makes it possible to consider atoms at rest, and with precision measurement of frequencies, Doppler broadening is considered as a small value. Therefore, at two-photon absorption, the wave frequencies can be chosen to be different and close to the frequencies of the allowed transitions. Essentially, we have a stimulated Raman scattering.

In this article the possibility of measuring the forbidden 1S-2S transition frequency of the hydrogen atom, when the atom from the 1S level moves to the level 2S through the intermediate level 3P. It is known that, for Λ-schemes, the stimulated scattering lineshape has a resonance with a homogeneous width of the transition between the lower levels (SRS-resonance), which has been well studied theoretically and experimentally [5]. Formally, this resonance takes place in any problem that involves the Λ-scheme, but the greatest interest in it relates to precision spectroscopy and frequency standards. This resonance is used as a reference in compact commercial atomic clocks under the name of coherent population trapping resonance (CPT- resonance). Standard equations for the density matrix are used to analyze this resonance. At a temperature of cooled atoms of the order of 1 µK, the motion of atoms can be neglected, and these equations can be solved as for an atom at rest. For the 1S-2S transition of the hydrogen atom in the work [4] the solution of these equations is given and the expression for the SRS-resonance is obtained . Here we give estimates of Doppler broadening, atomic collisions, field shift, and the recoil effect. It is shown that using the SRS method it is possible to measure the 1S-2S transition frequency of the hydrogen atom with an accuracy of 1 kHz. Note that the stimulated Raman scattering method for this transition is essentially an alternative to the Doppler-free two-photon one.
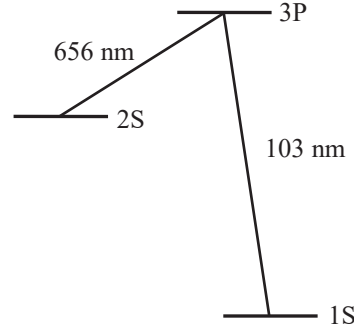
FIGURE 1. The hydrogen atom moves from the ground 1S state
to the metastable 2S state through the intermediate 3P state.

RESONANCE IN THE SRS LINESHAPE

The levels of a three-level scheme (1S, 2S, 3S) will be enumerated by indices 1, 2, and 3 (Fig. 1). We will consider SRS that is associated with the field

$$E(t) = E\,exp(-i\omega t) + E'exp(-i\omega' t) + c.c., \tag{1}$$

in which the atom passes from level 1 to level 2 via intermediate level 3. The resonance approximation is considered in which pump field frequency ω is close to transition frequency ω_{31}, while stimulated scattering frequency ω' is close to transition frequency ω_{32}. The expression for the probability of transition of an atom from state 1 to state 2 under the action of a field of two frequencies is given in the work [6] The probability of a two-photon transition consists of two terms, the physical natures of which are different. The first term consists of two stepwise transitions: (i) the absorption of a photon to create a population at upper level 3 and (ii) single-photon emission of radiation. The second term is SRS (coherent absorption and emission of photons), the lineshape of which has a resonance with a homogeneous width of the forbidden transition:

$$W(1 \to 2) = W \frac{\Gamma_{21}^2}{(\omega - \omega' - \omega_{21})^2 + \Gamma_{21}^2}, \tag{2}$$

$$W = 2a_1 a_2 \Gamma^2 / \Gamma_{21}.$$

Resonance occurs when the frequency difference $\omega - \omega'$ is equal to the frequency of the forbidden transition ω_{21}; Γ_{21} is the width of the forbidden transition $2 \to 1$, and $\Gamma = \gamma/2$, where γ is the radiation probability of the decay of level 3P; values a_1 and a_2 are small dimensionless saturation parameters at transitions 3→1 and 3→2, respectively:

$$a_1 = I/I_{31}, \quad a_2 = I'/I_{32},$$

where I and I' is the intensity of the fields E and E'; for the hydrogen atom $I_{31} = 10\,W/cm^2$, $I_{32} = 0.25\,W/cm^2$ [6].

MEASUREMENT ACCURACY

To determine the necessary measurement accuracy of the 1S-2S transition frequency, we will consider basic factors that affect resonance broadening and shift.

Doppler broadening
In measurements of the ω_{21} transition frequency, cold atoms can be free and therefore, Doppler broadening becomes the main line broadening factor. Therefore, it is necessary to perform averaging at speeds with the

Maxwell distribution function. If the waves propagate in the same direction, then, instead of (2), we obtain

$$W(1 \rightarrow 2) = W_D exp((\omega - \omega' - \omega_{21})^2/\omega_D^2) \tag{3}$$
$$W_D = 2\sqrt{\pi}a_1 a_2 \Gamma^2/\omega_D,$$

where $\omega_D = (\omega_{31} - \omega_{32})v_D/c$ is the Doppler width and v_D is the thermal velocity. At temperature $T = 1\mu K$, we have $\omega_D = 2\pi \cdot 10^6 \ s^{-1} (1 \ MHz)$. Tuning to the resonance center with an error of $10^{-3}\omega_D$ allows one to measure transition frequency ω_{21} with an error on the order of 1 kHz. In the future, we will focus on this accuracy.

Atomic collisions
Collisions of hydrogen atoms in a trap lead to a line shift. The value of this shift was measured in [7]. For the shift at a temperature of $T = 1\mu K$ to be about 1 kHz, the density of atoms should be rather low (on the order of $10^{10} \ cm^{-3}$). Atoms in the trap occupy a region on the order of $1 \ mm^3$; therefore, their number should be no larger than 10^7.

Field-induced shift
The value of the resonance shift from the field intensity is given in [6]. At $|\omega' - \omega_{21}| \ll \Gamma$, this shift is given by

$$\Delta\omega_f = a_2(\omega' - \omega_{21}) . \tag{4}$$

For intensity $I' = 2.5 \ mW/cm^2$, we have $a_2 = 10^{-2}$. At $\omega' - \omega_{21} = 10^{-2}\Gamma$, we obtain $\Delta\omega_f/2\pi = 160 \ Hz$.

The recoil effect
The shift of a SRS resonance due to the recoil effect upon absorption and emission of counterpropagating photons is given by

$$\Delta\omega = \hbar(k^2 - k'^2)/2M, \tag{5}$$

where $k = 2\pi/\lambda$, $k' = 2\pi/\lambda'$, and M is the mass of a hydrogen atom. The value of this shift is 17 MHz, and it is calculated with an error that is much smaller than 1 kHz.

Registration accuracy
Let N_0 be the number of atoms in the initial 1S state and N be the number in the 2S state. Because $N \ll N_0$, we can write

$$\partial N/\partial t = W_D N_0.$$

By substituting W_D, we have

$$\Gamma^{-1}\partial N/\partial t = 2\sqrt{\pi}a_2 a_1 N_0 \Gamma/\omega_D. \tag{6}$$

To measure the number of atoms in the 2S state, an electric field is usually applied to mix the 2S and 3P states. Then, fluorescence is recorded at the Lyman line at $\lambda = 121 \ nm$ (in [7], there were about 10^5 photons per second, which provided the resonance registration error of 1 kHz). If we assume that only one tenth of atoms are detected, then, for estimates, we can set $\partial N/\partial t = 10^6 s^{-1}$. Taking into account that $\Gamma = 10^8 s^{-1}$, $\Gamma/\omega_D = 16$, $a_2 = 10^{-2}$, $N_0 = 10^7$, we find from relation (6) that $a_1 = 2 \times 10^{-9}$. Since $I = a_1 I_{31}$, then

$$I = 0.2 \ \mu W/cm^2$$

The size of the region of atoms in the trap is usually a few millimeters, and, therefore, the necessary radiation power at a wavelength of 103 nm is on the order of $I \times 10^{-2} cm^2$; i.e., it is a few nanowatts.

CONCLUSIONS

This paper shows the possibility of measuring the frequency $1S - 2S$ transition of a hydrogen atom using the method of stimulated Raman scattering. We note several points that make the considered method promising for precision spectroscopy. Atoms at temperatures of $1\mu K$ have a Doppler width of the SRS resonance of the order of 1MHz, which ensures the accuracy of the transition frequency measurement ~1kHz. In this case, there is no second Doppler shift, which is a source of error in experiments with atomic beams, the field shift is excluded, the required field intensities are significantly reduced. In estimating the parameters, we focused on experimental work [7], where the hydrogen atoms in the trap were cooled to a temperature of ~$1\mu K$. At lower temperatures, you can expect better accuracy in measuring the frequency of this transition.

ACKNOWLEDGMENTS

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