Quantum Electronics **48** (5) 464–467 (2018)

https://doi.org/10.1070/QEL16568

Precision measurement of the forbidden $2^1S_0-2^3S_1$ transition frequency in a helium atom

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Abstract. **We demonstrate the possibility of measuring the forbidden $2^1S_0-2^3S_1$ transition frequency (λ = 1557 nm) of a helium atom by the method of stimulated Raman scattering through the intermediate 2^3P_1 level. Singlet (2^1S_0) and triplet (2^3S_1) states have long lifetimes of 20 ms and 8000 s, respectively. The transition is important for the spectroscopy of the helium atom because it relates the singlet and triplet parts of the spectrum.**

Keywords: *lasers, spectroscopy, helium atom, singlet and triplet levels, forbidden transition, frequency standard.*

1. Introduction

The methods of high-resolution laser spectroscopy are a good tool for investigating the quantum mechanics of a helium atom. Its atomic structure is calculated with high accuracy [1]. Precision measurement of the transition frequencies of this atom, together with theoretical calculations, provides additional information for quantum electrodynamics, since a three-particle problem of the interaction of two electrons in the presence of a nucleus (radiative corrections, nuclear charge radius, etc.) is considered.

The measurement of the forbidden $2^1S_0-2^3S_1$ transition frequency is an important problem, since it experimentally relates the singlet and triplet parts of the spectrum of the helium atom. The line of this transition has a small radiation width (8 Hz), which is due to the two-photon decay of the 2^1S_0 state into the ground state. To measure the frequency of this transition, Baklanov et al. [2, 3] analysed the main methods of laser spectroscopy. However, specific calculations and estimates [4, 5] for a gas and an atomic beam have made it impossible to realise the experiment at room temperatures. The situation changed dramatically at the beginning of the 2000 s. A number of scientific groups [6–10] obtained and studied the Bose–Einstein condensation of cooled ^{4}He atoms in the 2^3S_1 state. The atoms were cooled in two stages. First, using laser methods, helium atoms in the 2^3S_1 state were cooled to temperatures of $\sim$1 mK and trapped in a magneto-optical trap, and then moved to a special cloverleaf magnetic trap, where they were further cooled by evaporation. As a result, about 10^9 helium atoms were obtained in the 2^3S_1 state at temperatures of $\sim$1 μK. This made it possible to perform a number of experiments, important both for the condensed state physics and for precision helium spectroscopy. For our investigations, of importance is paper [11], where the absolute frequency of the $2^1S_0-2^3S_1$ forbidden transition (λ = 1557 nm) was measured to within ~1 kHz, which made it possible to experimentally relate the singlet and triplet parts of the helium spectrum with the same accuracy.

Note that measuring the frequency of the $2^1S_0-2^3S_1$ forbidden (the Einstein coefficient of 10^{-7} s^{-1}) transition is quite a challenge. In this paper, we analyse the possibility of measuring the frequency of this transition by stimulated Raman scattering (SRS) (Fig. 1). It is known that for Λ-schemes in the form of a line of stimulated scattering, there is a resonance with a homogeneous width of the transition line between the lower levels, which has been well studied theoretically and experimentally (see monograph [12]). Formally, this resonance is present in any problem with the Λ-scheme, but the greatest interest in it is associated with precision spectroscopy

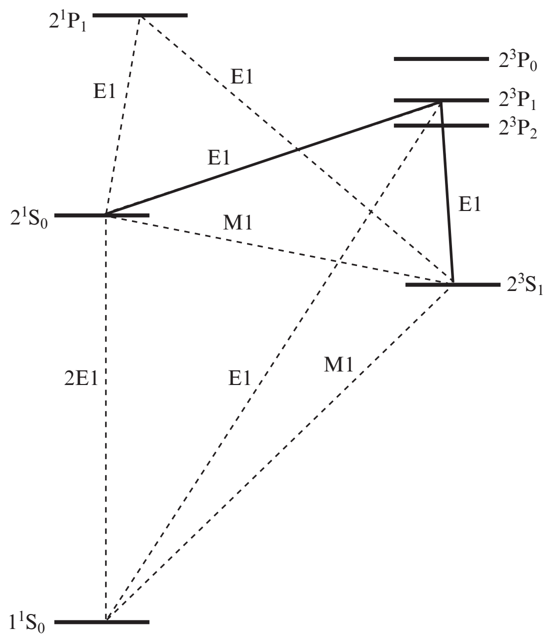

Figure 1. Diagram of low-lying levels of the helium atom and transitions between them: solid lines show the transitions involved in SRS; E1 are the electric dipole transitions; M1 are the magnetic dipole transitions; 2E1 is the two-photon electric dipole transition.

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Received 8 November 2017; revision received 12 January 2018
Kvantovaya Elektronika **48** (5) 464–467 (2018)
Translated by I.A. Ulitkin

and frequency standards. Under the name of 'a coherent population trapping resonance' (CPT resonance), it is used as a reference in compact commercial atomic clocks. The analysis of SRS resonance is usually based on the use of standard equations for the density matrix. At a temperature of cooled atoms on the order of 1 μK, we can neglect the motion of atoms and solve these equations for a stationary atom. In Section 2 of this paper, for the case of helium, we present a solution of these equations and a formula for the SRS resonance. The influence of Doppler broadening, the recoil effect, and the field shift is considered in Section 3. It was also shown there that by using the SRS method it is possible to measure the frequency of the $2^1S_0-2^3S_1$ transition in the helium atom with an accuracy of ~1 kHz.

2. Resonance in the form of the SRS line

We denote the levels 2^3P_1, 2^3S_1, 2^1S_0 by numbers 0,1,2 (Fig. 2), and the frequencies of the transitions $2^3P_1-2^3S_1$ ($\lambda = 1083$ nm) and $2^3P_1-2^1S_0$ ($\lambda' = 3561$ nm) by ω_{01} and ω_{02}, respectively. We consider SRS with the participation of laser fields at frequencies ω and ω':

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

at which the atom from level 1 passes to level 2 through intermediate level 0. A resonance approximation is used when the pump field frequency ω is close to the transition frequency ω_{01}, and the frequency ω' is close to the transition frequency ω_{02}. In our case, the line widths of the 0–1 and 0–2 transitions are $\Gamma = \gamma/2$, where $\gamma = 10^7$ s^{-1} is the probability of spontaneous emission at the 0–1 transition. For the density matrix elements, we have the equations:

$$\begin{aligned}
&\dot\rho_2 = -V(t)\rho_{20} - V^*(t)\rho_{02},\\
&\dot\rho_1 = \gamma\rho_0 - U(t)\rho_{10} - U^*(t)\rho_{01},\\
&\dot\rho_0 + \gamma\rho_0 = U(t)\rho_{10} + U^*(t)\rho_{01} + V(t)\rho_{20} + V^*(t)\rho_{02},\\
&\dot\rho_{01} + (\Gamma + i\omega_{01})\rho_{01} = U(t)(\rho_1 - \rho_0) + V(t)\rho_{21},\\
&\dot\rho_{02} + (\Gamma + i\omega_{02})\rho_{02} = V(t)(\rho_2 - \rho_0) + U(t)\rho_{12},\\
&\dot\rho_{21} + (\Gamma_{21} + i\omega_{21})\rho_{21} = -V^*(t)\rho_{01} - \rho_{20}U(t).
\end{aligned} \quad (2)$$

Here, $\rho_{ik} = \rho^*_{ki}$; $\rho_i = \rho_{ii}$ $(i,k = 0,1,2)$; Γ_{21} is the line width of the 2–1 forbidden transition;

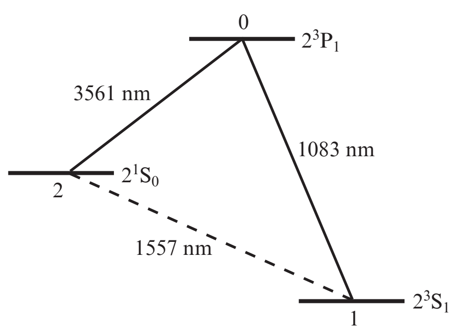

Figure 2. SRS scheme at the $2^1S_0-2^3S_1$ transition through the intermediate level 2^3P_1.

$$U(t) = U\exp(-i\omega t);\quad U = dE/(2i\hbar);$$

$$V(t) = V\exp(-i\omega' t);\quad V = d'E'/(2i\hbar);$$

d and d' are the projections of the dipole moment operators of the transitions to the field directions. We introduce new variables r_{01}, r_{02} and r_{21} in accordance with the equalities

$$\rho_{01} = r_{01}\exp(-i\omega t),\quad \rho_{02} = r_{02}\exp(-i\omega' t),$$

$$\rho_{21} = r_{21}\exp(-i\omega t + i\omega' t).$$

Taking into account the condition $\rho_0 + \rho_1 + \rho_2 = 1$ and assuming that $\rho_2 \ll \rho_0$, we obtain

$$\begin{aligned}
&\dot\rho_2 = -2\mathrm{Re}(r_{02}V^*),\\
&\dot\rho_0 + \gamma\rho_0 = 2\mathrm{Re}(r_{01}U^*) + 2\mathrm{Re}(r_{02}V^*),\\
&\dot r_{01} + (\Gamma - i\delta)r_{01} = U(1 - 2\rho_0) + Vr_{21},\\
&\dot r_{02} + (\Gamma - i\delta')r_{02} = -V\rho_0 + Ur^*_{21},\\
&\dot r_{21} + (\Gamma_{21} - i\Omega)r_{21} = -V^*r_{01} - Ur^*_{02}.
\end{aligned} \quad (3)$$

Here, $\delta = \omega - \omega_{01}$; $\delta' = \omega' - \omega_{02}$; and $\Omega = \omega - \omega' - \omega_{21}$. Obviously, the value of $\dot\rho_2$ is the probability of a transition from level 1 to level 2 under the action of two fields with frequencies ω and ω'. Denoting $\dot\rho_2$ by $W(1-2)$, we rewrite the first equation of system (3) in the form

$$W(1-2) = -2\mathrm{Re}(r_{02}V^*). \quad (4)$$

The remaining equations determine r_{02}. In their solution, the fields are considered weak, satisfying the conditions

$$|U|/\Gamma \ll 1,\quad |V|/\Gamma \ll 1. \quad (5)$$

In this case, the derivatives in (3) can be neglected, and in order to find r_{02}, we should solve the system of equations

$$\begin{aligned}
&\Gamma\rho_0 = \mathrm{Re}(r_{01}U^*) + \mathrm{Re}(r_{02}V^*),\\
&(\Gamma - i\delta)r_{01} = Vr_{21} - 2U\rho_0 + U,\\
&(\Gamma - i\delta')r_{02} = -V\rho_0 + Ur^*_{21},\\
&(\Gamma_{21} - i\Omega)r_{21} = -V^*r_{01} - Ur^*_{02}.
\end{aligned} \quad (6)$$

We substitute r_{02} from the third equation of this system into formula (4), and in the remaining equations we neglect the terms of the lowest order in the field. This gives

$$\begin{aligned}
&W(1-2) = \mathrm{Re}\left(\frac{2|V|^2\rho_0}{\Gamma - i\delta'}\right) - \mathrm{Re}\left(\frac{2UV^*r_{21}}{\Gamma - i\delta'}\right),\\
&r_{21} = -\frac{V^*U}{(\Gamma - i\delta)(\Gamma_{21} - i\Omega)},\\
&\rho_0 = \frac{q}{2\Gamma^2}\frac{\Gamma^2}{\Gamma^2 + \delta^2}.
\end{aligned} \quad (7)$$

Here,

$$q = 2|U|^2/\Gamma^2 \quad (8)$$

is a dimensionless saturation parameter for the 0–1 transition, which is considered to be much less than unity. As a result, for the transition probability of an atom from state 1 to state 2 under the influence of a two-frequency field, we have the expression

$$W(1\text{–}2) = A\frac{\Gamma^4}{(\Gamma^2+\delta'^2)(\Gamma^2+\delta^2)}$$

$$+ A\,\mathrm{Re}\left[\frac{\Gamma^3}{(\Gamma-\mathrm{i}\delta')(\Gamma-\mathrm{i}\delta)(\Gamma_{21}-\mathrm{i}\Omega)}\right], \qquad (9)$$

$$A = q\,|V|^2/\Gamma\,.$$

The probability of a two-photon transition (9) contains two terms, which have a different physical nature. The first one describes two independent transitions: absorption of a photon with the creation of a population at the upper level 0 and one-photon emission. The second term describes SRS (coherent absorption and emission of photons), the form of the line of which has a resonance with a uniform line width of the 2–1 forbidden transition.

When the conditions $|\delta| \ll \Gamma$ and $|\delta'| \ll \Gamma$ are fulfilled, we obtain the expression

$$W(1\text{–}2) = A\left(1+\frac{1+\Gamma_{21}\Gamma}{\Omega^2+\Gamma_{21}^2}\right).$$

In our case, $\Gamma \gg \Gamma_{20}$; therefore,

$$W(1\text{–}2) = W\frac{\Gamma_{21}^2}{(\omega-\omega'-\omega_{21})^2+\Gamma_{21}^2}, \qquad (10)$$

where

$$W = q\,|V|^2/\Gamma_{21}\,.$$

Thus, we have a resonance in the form of the SRS line, when $\omega - \omega' = \omega_{21}$.

3. Factors affecting the measurement accuracy

Doppler broadening. When measuring the transition frequency ω_{21}, cold atoms can be considered 'free'; therefore, the main factor of the line broadening is the Doppler broadening. When formula (10) is used for an atomic gas, we must take into account the Doppler shift of the frequencies ω and ω' for a moving atom and perform averaging over the velocities with a Maxwellian distribution function. If the waves with frequencies ω and ω' are unidirectional, then instead of (10) we obtain the expression

$$W(1\text{–}2) = W_{\mathrm{D}}\exp[(\omega-\omega'-\omega_{21})^2/\omega_{\mathrm{D}}^2], \qquad (11)$$

$$W_{\mathrm{D}} = \sqrt{\pi}\,q\,|V|^2/\omega_{\mathrm{D}},$$

where $\omega_{\mathrm{D}} = \omega_{21}v_{\mathrm{D}}/c$ is the Doppler width; and v_{D} is the thermal velocity of atoms. At a temperature of 1 μK, we have $\omega_{\mathrm{D}} = 2\pi\cdot 10^3$ s^{-1} (10 kHz). Tuning to the resonance centre with an accuracy of $0.1\omega_{\mathrm{D}}$ makes it possible to measure the transition frequency ω_{21} with an accuracy on the order of 1 kHz.

Intensity of fields. According to (8), the saturation parameter for the $2^3\mathrm{P}_1-2^3\mathrm{S}_0$ transition can be represented in the form $q = I/I_{\mathrm{sat}}$, where I is the radiation intensity at the frequency ω; and

$$I_{\mathrm{sat}} = 16\pi^2\hbar c\Gamma/\lambda^3 = 2\ \mathrm{W\ cm^{-2}}. \qquad (12)$$

We set $q = 10^{-2}$, then $I = 20\ \mu\mathrm{W\ cm^{-2}}$.

To estimate the radiation intensity at the $2^3\mathrm{P}_1-2^1\mathrm{S}_0$ transition, we write the expression for the number of atoms that, during the measurement time T, are in the $2^1\mathrm{S}_0$ state:

$$N = W_{\mathrm{D}}N_0T, \qquad (13)$$

where N_0 is the number of atoms in the initial state $2^3\mathrm{S}_1$. Since both fields are weak, then $N \ll N_0$. We set $N/N_0 = 0.1$. Then from expressions (11) and (13) we obtain the relation $|V|^2 = 10\omega_{\mathrm{D}}/T$. We express the field intensity at the frequency ω' in terms of $|V|^2$:

$$I' = 160\pi^2\hbar c\omega_{\mathrm{D}}/(\lambda'^3\gamma T), \qquad (14)$$

where $\lambda' = 3561$ nm and $\gamma = 2.7\times10^{-2}$ s^{-1} [1] is the wavelength of the $2^1\mathrm{S}_0-2^3\mathrm{P}_1$ transition and the spontaneous emission probability at this transition, respectively. In the experiment [11], the measurement time was several seconds. Assuming $T = 2$ s, we obtain $I' \approx 10\ \mu\mathrm{W\ cm^{-2}}$.

Field shift. To estimate the magnitude of the resonance shift $\Delta\Omega$ as a function of the intensity of the fields, we solved the system of equations (6) with an accuracy greater than that in obtaining (7), by using the smallness of the parameters $|U|/\Gamma$ and $|V|/\Gamma$. For $|\delta'| \ll \Gamma$, the shift is

$$\Delta\Omega = q\delta'/2. \qquad (15)$$

Previously, we set $q = 10^{-2}$. The frequency ω' can be easily tuned to the centre of the transition line with frequency ω_{02} with an accuracy of 10^{-2} from the line width. Setting $\delta' = 10^{-2}\Gamma = 0.5\times10^5$ s^{-1}, we obtain $\Delta\Omega/2\pi \approx 20$ Hz.

Recoil effect. The shift of the SRS resonance due to the recoil effect in the absorption and emission of photons with $\lambda = 1083$ nm and $\lambda' = 3561$ nm, respectively, is

$$\Delta\omega = \hbar(k'^2 - k^2)/(2M), \qquad (16)$$

where $k = 2\pi/\lambda$; $k' = 2\pi/\lambda'$; and M is the mass of the ^{4}He atom. The value of this shift $\Delta\omega/2\pi = 39.3$ kHz is calculated with an error of less than 1 kHz, and is taken into account when measuring ω_{21}.

Accuracy of recording the $2^1S_0-2^3S_1$ transition frequency. In experiment [11], more than 10^6 helium atoms in the $2^3\mathrm{S}_1$ state were obtained at a temperature of 1 μK. Therefore, for estimates we set $N_0 = 10^6$. We will register $N = 0.1N_0 = 10^5$ atoms in the $2^1\mathrm{S}_0$ state. We are guided by the method of paper [3], based on the detection of spontaneous VUV photons. Transitions involved in the measurement scheme are shown in Fig. 3. Using laser radiation at the $2^1\mathrm{S}_0-2^1\mathrm{P}_0$ transition of helium ($\lambda = 2058$ nm) [13], the atom from the $2^1\mathrm{S}_0$ state undergoes transition to the $2^1\mathrm{P}_0$ state, followed by spontaneous emission of a photon at $\lambda = 58$ nm upon transition to the ground state. We assume that $0.1N$ atoms participate in this process, i.e., using the vacuum ultraviolet technique, $N_{\mathrm{ph}} = 10^4$ photons are recorded. For a rough estimate of the detection accuracy, we can assume that the fluctuation of the number of photons is equal to $\sqrt{N_{\mathrm{f}}}$. In the absence of a background, the signal-to-noise ratio is $N_{\mathrm{ph}}/\sqrt{N_{\mathrm{f}}} = 10^2$. This allows us to register the shape of the resonance with an accuracy of 10^{-2}, which is enough to measure the $2^1\mathrm{S}_0-2^3\mathrm{S}_1$ transition frequency with an accuracy of 1 kHz. The presence of a

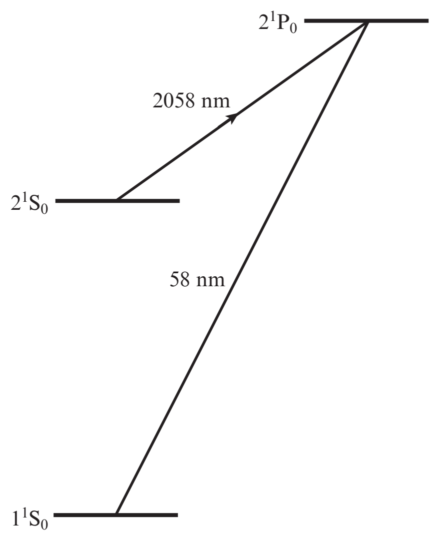

Figure 3. Transitions involved in the scheme for measuring the number of helium atoms at the 2^1S_0 level. Photons are recorded at $\lambda = 58$ nm.

background leads to a decrease in the signal-to-noise ratio. However, in real experiments the background can be strongly suppressed, since the resonance is recorded using a variety of techniques that allow this to be done [12]: the recording of the resonance with respect to the derivative, the use of frequency modulation, etc.

4. Conclusions

We have shown that the SRS method can be used to measure the frequency of the 2^1S_0–2^3S_1 transition of a helium atom with an accuracy of 1 kHz at a laser field intensity of $\sim$10 W cm^{-2}. Let us note the features of this transition, which make it promising for various investigations:

1. A small radiation width (8 Hz) allows this transition to be used to create a frequency standard.

2. Precision measurement of the 2^1S_0–2^3S_1 transition frequency provides additional information for testing quantum electrodynamics, since a three-particle system of two electrons and a nucleus is considered. It is possible to measure the difference of the radiative corrections for the levels 2^1S_0 and 2^3S_1 and compare it with the theoretical value.

3. Methods of Doppler-free laser spectroscopy can be used, since the SRS line exhibits a resonance with a uniform line width in the gas [12, 14, 15].

Acknowledgements. The authors thank D.V. Brazhnikov for discussion of the work and valuable remarks.

The work was supported by the Russian Foundation for Basic Research (Grant No. 17-02-00292).

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