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ATOMS, MOLECULES, OPTICS

Ionization of *nS*, *nP*, and *nD* Lithium, Potassium, and Cesium Rydberg Atoms by Blackbody Radiation

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Abstract—The results of theoretical calculations of the blackbody ionization rates of lithium, potassium, and cesium atoms residing in Rydberg states are presented. The calculations are performed for *nS*, *nP*, and *nD* states in a wide range of principal quantum numbers, $n = 8$–65, for blackbody radiation temperatures $T = 77$, 300, and 600 K. The calculations are performed using the known quasi-classical formulas for the photoionization cross sections and for the radial matrix elements of transitions in the discrete spectrum. The effect of the blackbody-radiation-induced population redistribution between Rydberg states on the blackbody ionization rates measured under laboratory conditions is quantitatively analyzed. Simple analytical formulas that approximate the numerical results and that can be used to estimate the blackbody ionization rates of Rydberg atoms are presented. For the *S* series of lithium, the rate of population of high-lying Rydberg levels by blackbody radiation is found to anomalously behave as a function of *n*. This anomaly is similar to the occurrence of the Cooper minimum in the discrete spectrum.

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1. INTRODUCTION

Equilibrium thermal blackbody radiation, or, simply, blackbody radiation, which is always present in space, can appreciably affect the dynamics of population of highly excited (Rydberg) states [1] of any atoms. Interactions of atoms in Rydberg states with blackbody radiation have been theoretically and experimentally studied in a number of works, in which the greatest attention was concentrated on transitions in the discrete spectrum and on the decrease in the lifetime of Rydberg states under the action of blackbody radiation [2–4]. Detailed theoretical investigations of the depopulation of Rydberg levels were performed in [5]. Also, cascade transitions in the discrete spectrum were theoretically and experimentally studied [6].

Recent growing interest in the interaction of Rydberg atoms with blackbody radiation was stimulated by investigations of ultracold plasma. In first experiments, conversion of a cloud of cold Rydberg atoms in a magnetooptical trap into ultracold plasma was observed [7, 8]. It was assumed that this occurs due to avalanche ionization of Rydberg atoms in the cloud, which is initiated by photoionization of Rydberg atoms in the blackbody radiation field.

To construct a model of formation of ultracold plasma, calculation data on the total rates of ionization of different Rydberg states by blackbody radiation are necessary. However, analysis of the literature available shows that only a few works were devoted to transitions to the discrete spectrum, which are responsible for photoionization of atoms. In [9], the temperature dependence of the blackbody photoionization rate of the 17*D* state of sodium was theoretically calculated and experimentally measured. A simple analytical formula for estimating the ionization rate proposed in that study is still in use [7], although it takes into account only the ionization potential of the Rydberg state, while the orbital momentum *L* is ignored. This makes this formula inapplicable in calculations of photoionization rates of non-hydrogen-like states with small *L*. The rates of direct photoionization of hydrogen and sodium atoms by blackbody radiation were numerically calculated in [10] to a high accuracy using the model potential method. Our recent results for sodium obtained in [11] agree well with these calculations. In [11], we also calculated the total ionization rates for rubidium atoms. Theoretical calculations of direct photoionization by blackbody radiation were recently calculated for helium atoms as well [12]. We also note that radial matrix elements for transitions between coupled and

free states of Rydberg atoms of alkali metals were systematically calculated in [13]; however, the process itself of the photoionization by blackbody radiation was not considered.

In our previous studies [14–16], the signal of blackbody ionization was used as the reference signal in measurements of the rate constants of associative ionization of sodium atoms. For this purpose, we measured the blackbody ionization rate constants of Rydberg *nS* and *nD* states (n = 8–20) in relation to the principal quantum number n. The theory agreed well with experiment for *nD* states and disagreed with it for *nS* states at $n > 15$. We also calculated blackbody ionization rates of Rydberg *nS*, *nP*, and *nD* sodium and rubidium atoms for a wide range of principal quantum numbers, n = 8–65, which are of interest in experiments on ultracold plasma [11].

In this study, we calculated for the first time the ionization rates of Rydberg *nS*, *nP*, and *nD* states of lithium, cesium, and potassium atoms in the range of principal quantum numbers n = 8–65 for three values of temperature, T = 77, 300, and 600 K. These temperatures correspond to the majority of experiments that are performed using heat shields cooled by liquid nitrogen, or at room temperature, or in heated optical cells. In numerical calculations, we used the known quasi-classical formulas for photoionization cross sections and for transition probabilities in the discrete spectrum [17]. Special attention is paid to the effect of blackbody-radiation-induced transitions in the discrete spectrum of a Rydberg atom on its ionization rate measured under laboratory conditions. In particular, blackbody-radiation-induced population of high-lying Rydberg states leads to their ionization by electric pulses (selective ionization by a pulsed electric field [18]) that are usually used in experiments for extracting ions from the interaction region of Rydberg atoms with blackbody radiation. It was shown that the ionization rate by electric pulses can be comparable with the rate of direct photoionization by blackbody radiation. Based on the quasi-classical formulas for photoionization cross sections [19], we obtained simple analytical expressions that approximate the numerical calculations and that can be used to estimate the rate of direct photoionization by blackbody radiation, as well as the rate of ionization by an electric field.

In addition, blackbody radiation efficiently populates neighboring Rydberg states, which it subsequently ionizes. The role played by this effect is especially large under such experimental conditions when the time interval between excitation of Rydberg states and their detection is comparable with the lifetime of the Rydberg atom [14].

As a rule, in studies of blackbody ionization of a Rydberg atom A initially excited to a state *nL*, the consideration is restricted only to the case of direct photoionization of the atom by a blackbody photon,

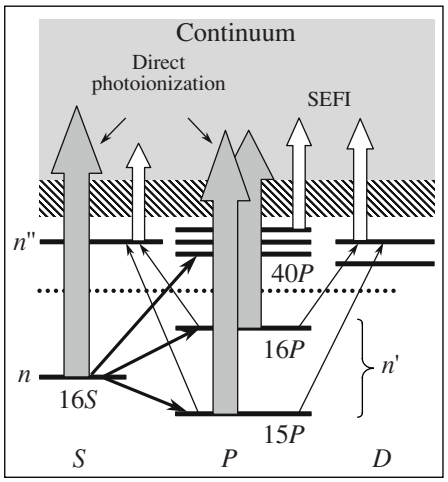

Fig. 1. Schematic diagram of ionization of the lithium atom in the 16*S* Rydberg state by blackbody radiation (SEFI is the selective electric-field ionization; high-lying Rydberg states are in the hatched region).

$$A(nL) + \hbar\omega_{BBR} \longrightarrow A^+ + e^-. \quad (1)$$

Here, $\hbar\omega_{BBR}$ is the energy of a blackbody photon; A^+ is an atomic ion; and e^- is a free electron, which is formed as a result of ionization.

In reality, blackbody ionization of a Rydberg atom is a more complicated process. Figure 1 schematically shows various ionization channels that were taken into account in this study for lithium atom initially excited to the state 16*S*. The total rate of blackbody ionization of Rydberg atoms can be represented as a sum [11]

$$W_{BBR}^{\text{tot}} = W_{BBR} + W_{SFI} + W_{BBR}^{\text{mix}} + W_{SFI}^{\text{mix}}. \quad (2)$$

Here, W_{BBR} is the rate of direct photoionization of the initially excited state *nL*; W_{SFI} is the rate of ionization by electric pulses upon population of high-lying states from the initially excited *nL* state; W_{BBR}^{mix} is the total rate of direct photoionization of neighboring Rydberg levels by the blackbody radiation; and W_{SFI}^{mix} is the rate of ionization by electric pulses of high-lying states populated as a result of two-step transitions induced by the blackbody radiation. In what follows, each of the terms of Eq. (2) will be considered separately.

2. DIRECT PHOTOIONIZATION BY BLACKBODY RADIATION

The rate of direct blackbody photoionization of Rydberg states (here and below the atomic units are used) is expressed as follows [9]:

$$W_{BBR} = c\int_{\omega_{nL}}^{\infty} \sigma_\omega \rho_\omega d\omega. \quad (3)$$

Scaling coefficients C_L in Eq. (4)

	C_S	C_P	C_D
Li	–	1	1
Na	0.15	1	1
K	0.12	0.15	0.4
Rb	0.2	0.2	0.4
Cs	0.1	0.6	0.3

Here, c is the speed of light; $\omega_{nL} = 1/2n_{\text{eff}}^2$ is the photoionization threshold frequency for the atom in the nL Rydberg state with the effective principal quantum number $n_{\text{eff}} = n - \delta_L$, where δ_L is the quantum defect (exact quantum defects of Rydberg states of lithium, potassium, and cesium were given in [20–22]); and σ_ω is the photoionization cross section at the frequency ω. The volume density ρ_ω of blackbody photons at a temperature T is given by the Planck distribution.

For isotropic and nonpolarized thermal radiation, the photoionization cross section σ_ω is expressed via radial matrix elements $R(nL \longrightarrow E, L \pm 1)$ of transition dipole moments between states nL of the discrete spectrum and states of the continuum spectrum with the orbital momentum $L \pm 1$ and photoelectron energy E.

The most complicated problem in determining the rate of photoionization by blackbody radiation is the calculation of radial matrix elements. For exact calculations of photoionization cross sections, the analytical quasi-classical formulas derived by Dyachkov and Pankratov [17] are convenient. In comparison with other quasi-classical methods [19, 23, 24], the Dyachkov–Pankratov method has a number of advantages. Thus, it uses orthogonal and normalized quasi-classical wave functions, and photoionization cross sections determined by this method agree well with those obtained from the most sophisticated and exact quantum-mechanical calculations by the model potential method [25]. Also, calculations based on the Dyachkov–Pankratov model agree well with the well-known calculations by the quantum-defect method [26–28].

Approximate analytical expressions for W_{BBR} are also useful in consideration of photoionization of Rydberg atoms by blackbody radiation, since they illustrate how the ionization rate depends on the principal quantum number n, orbital momentum L, and temperature T. Such expressions we previously obtained in [11] using the quasi-classical analytical formulas for radial matrix elements of dipole transitions reported in [19] (the Goreslavsky–Delone–Krainov (GDK) model),

$$W_{BBR}\ [\text{s}^{-1}] = C_L T \frac{14\,423}{n^{7/3}} \ln\left(\frac{1}{1 - \exp\left(-\frac{157\,890}{Tn^2}\right)}\right). \quad (4)$$

Expression (4) was obtained for hydrogen atoms. To estimate the photoionization rate of Rydberg atoms of alkali metals, it is necessary to replace n by the effective quantum number n_{eff}. The coefficient C_L depends on the orbital momentum L and takes into account the non-hydrogen-like character of the wave functions of nS, nP, and nD Rydberg states of alkali metals, whose quantum defects are large.

The values of C_L that best approximate the results of our exact numerical calculations previously published in [11] (sodium, rubidium) and performed in this study (lithium, potassium, cesium) are given in the table. Figure 2a shows the numerically (solid curves) and analytically (dashed curves) calculated rates of direct blackbody photoionization of nS Rydberg cesium atoms at $T = 77$, 300, and 600 K. Good agreement between the approximate analytical formula and exact numerical calculation is observed. For nP and nD Rydberg states, the ionization rates analytically calculated using the scaling coefficients C_L from the table also agree well with exact numerical data (not shown). For comparison, circles in Fig. 2a show the calculation results according to the frequently used analytical formula from [9]. For this formula, we also introduced scaling coefficients, since it describes only the relative dependence of the photoionization rate on the principal quantum number and radiation temperature. It is clearly seen from Fig. 2a that the dependence of the photoionization rate on the principal quantum number obtained with our expression (4) best coincides with the numerical calculations compared to the approximation from [9].

Figure 2b shows the calculation results on direct photoionization of potassium atoms in nP Rydberg states. Good agreement between numerical and analytical calculations is observed not only for nP states, but also for nS and nD states.

Figure 2c presents the rates of direct photoionization of nS lithium Rydberg atoms. In this case, the dependence on the principal quantum number analytically calculated by formula (4) considerably differs from the numerically calculated dependence. It also should be noted that the rates of direct blackbody photoionization of non-hydrogen-like nS lithium atoms are anomalously low; they are two orders of magnitude lower than the photoionization rates of hydrogen-like nP atoms (Fig. 2d).

It is known that nS states of the lithium atom show other anomalous properties. Thus, the photoionization cross section near the threshold does not increase with increasing n, as is the case with other alkali metals, but

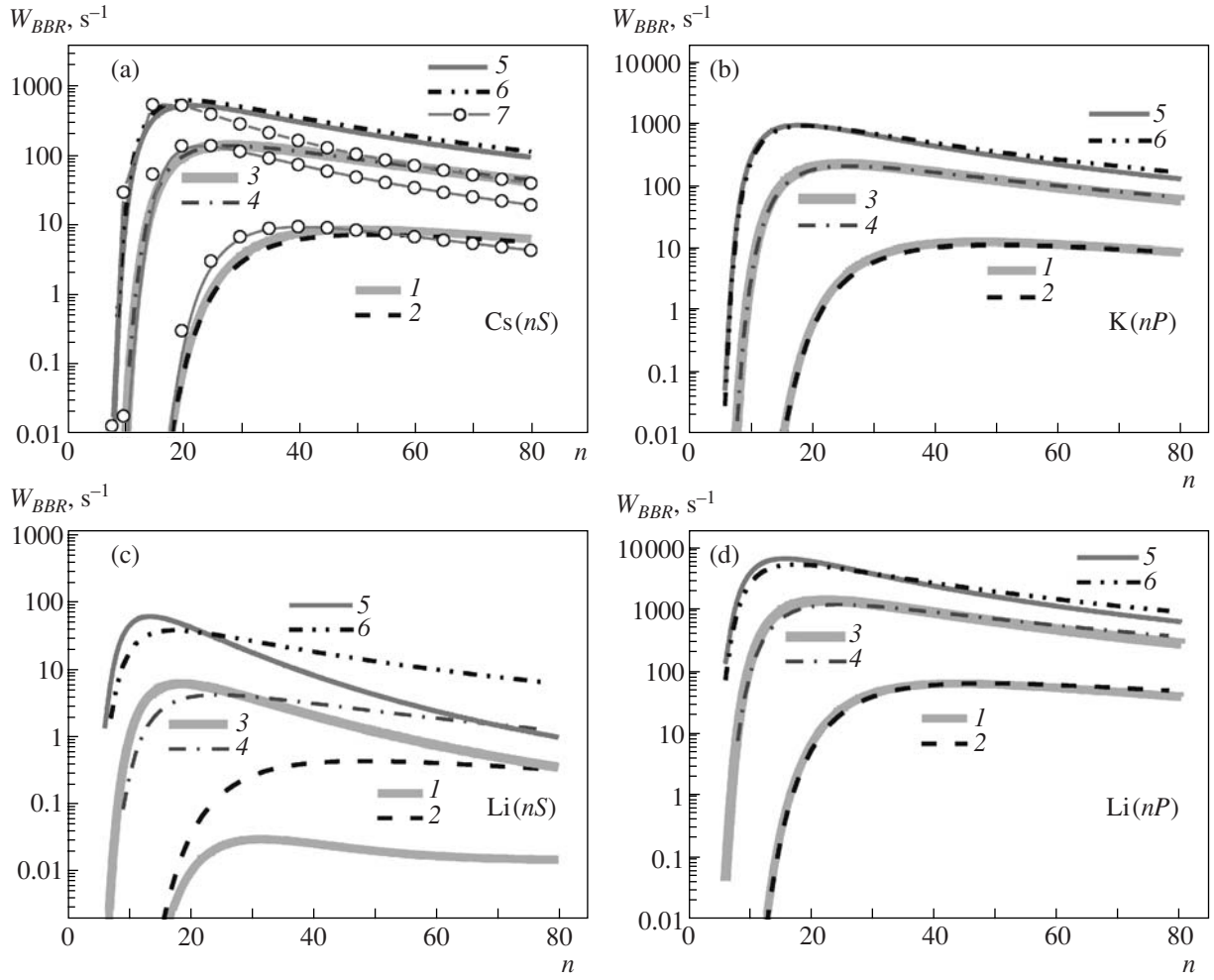

Fig. 2. Dependences of the direct blackbody photoionization rate W_{BBR} on the principal quantum number n calculated numerically (curves *1*, *3*, and *5*) and analytically (curves *2*, *4*, and *6*) at $T = 77$, 300, and 600 K for Rydberg atoms: (a) nS cesium, (b) nP potassium, (c) nS lithium, and (d) nP lithium. Curve *7* in (a) is the calculation by the analytical formula from [9] for these temperatures.

rather decreases [29]. It turned out that simple analytical formulas of the GDK model do not yield proper rates of blackbody ionization of nS lithium atoms, and to determine these quantities, exact numerical calculations are necessary. At the same time, for hydrogen-like nP states (Fig. 2d) and nD states (not shown), numerical and analytically calculations again agree well with each other.

3. IONIZATION OF HIGH-LYING STATES BY ELECTRIC FIELD

Electric field pulses, which are commonly used in ionization experiments to extract ions from the ionization zone, ionize states with a nearly unit probability if their effective quantum number obeys the condition $n_{\text{eff}} > n_c$, where n_c is a certain critical value. This critical value n_c depends on the amplitude of the applied electric field, which, in the classical approximation is described by the formula [18]

$$E_c\ [\text{V/cm}] \approx 3.2 \times 10^8 n_c^{-4}. \quad (5)$$

Consequently, if the blackbody radiation populates Rydberg states with $n' \geq n_c$, these states will be ionized and will contribute to the observed ionization signal. This contribution is comparable with the contribution from direct blackbody photoionization [9]. Since the probability of the field ionization of states with $n' \geq n_c$ is almost unity, to find the field ionization rate W_{SFI}, it suffices to calculate the rate of population of high-lying states caused by the blackbody radiation [11]. We calculated the values of the rate $W(nL \longrightarrow n'L')$ of dipole transitions to states $n'L'$ with $n' \geq n_c$ and $L' = L \pm 1$ using the Dyachkov–Pankratov quasi-classical formulas [17]. The values of W_{SFI} were obtained for electric field amplitudes of 5 and 10 V/cm, which are most frequently applied in experiments with high-lying Rydberg states.

We also compared our numerical results with the analytical data obtained by formulas based on the GDK

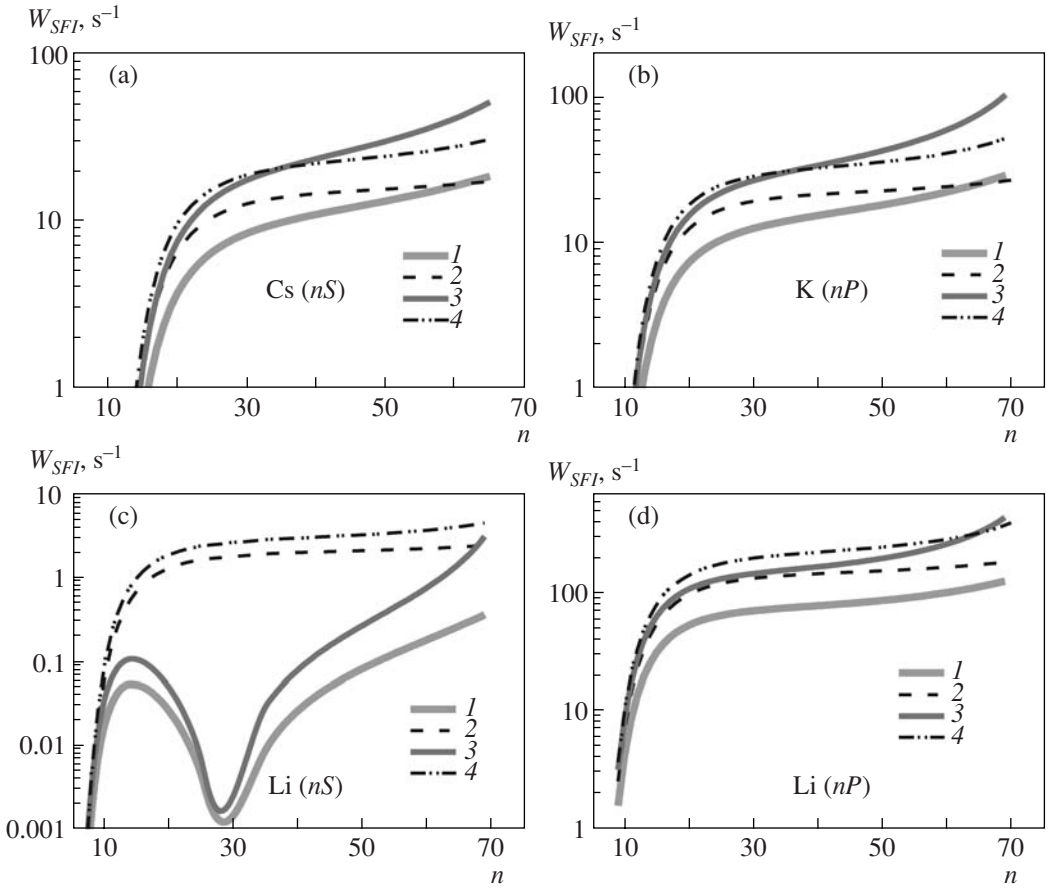

Fig. 3. Dependences of the photoionization rate W_{SFI} by electric pulses on the principal quantum number n calculated numerically (curves *1* and *3*) and analytically (curves *2* and *4*) at $T = 300$ K and electric field amplitudes of 5 and 10 V/cm for Rydberg atoms: (a) nS cesium, (b) nP potassium, (c) nS lithium, and (d) nP lithium. Ionization occurs as a result of population of high-lying Rydberg states induced by the blackbody radiation.

model, which are similar to Eq. (4) and where, instead of integration over the continuum spectrum, the transition probabilities to states with $n' \geq n_c$ were summed. Accordingly,

$$W_{SFI}\ [\mathrm{s}^{-1}] = C_L T \frac{14\,423}{n^{7/3}} \left(\ln \frac{1}{1 - \exp\left(\frac{157\,890}{T n_c^2} - \frac{157\,890}{T n^2} \right)} - \ln \frac{1}{1 - \exp\left(-\frac{157\,890}{T n^2} \right)} \right), \quad (6)$$

where T is expressed in kelvins, and the coefficients C_L are listed in the table.

The results of numerical and analytical calculations for nS states of cesium and nP states of potassium are presented in Figs. 3a and 3b, respectively. These figures show that Eq. (6) yields a correct estimate for the field ionization rate of high-lying Rydberg states. At the same time, exact rates can be obtained only by numerical calculation.

An interesting result is obtained for the rate of ionization W_{SFI} of Rydberg nS lithium atoms by electric pulses (Fig. 3c). The dependence $W_{SFI}(n)$ has an anomalous shape with a clearly pronounced minimum. Theoretical analysis shows that this anomaly is caused by the phenomenon, which is similar to the occurrence of the Cooper minimum in the discrete spectrum for transitions between nS and $n'P$ states of lithium [30]. For the hydrogen atom, radial matrix elements of transitions between Rydberg states monotonically decrease with increasing energy interval between the levels under consideration. The phase shift $\pi\delta_l$ in the radial wave functions of Rydberg states of alkali metals varies in different series. As a result, the overlap integral in the matrix elements of the dipole moment may turn to zero for some transition energies [31]. This leads to occurrence of minima for the probability of optical transition, which is proportional to the squared radial matrix element. As in the case of direct photoionization of

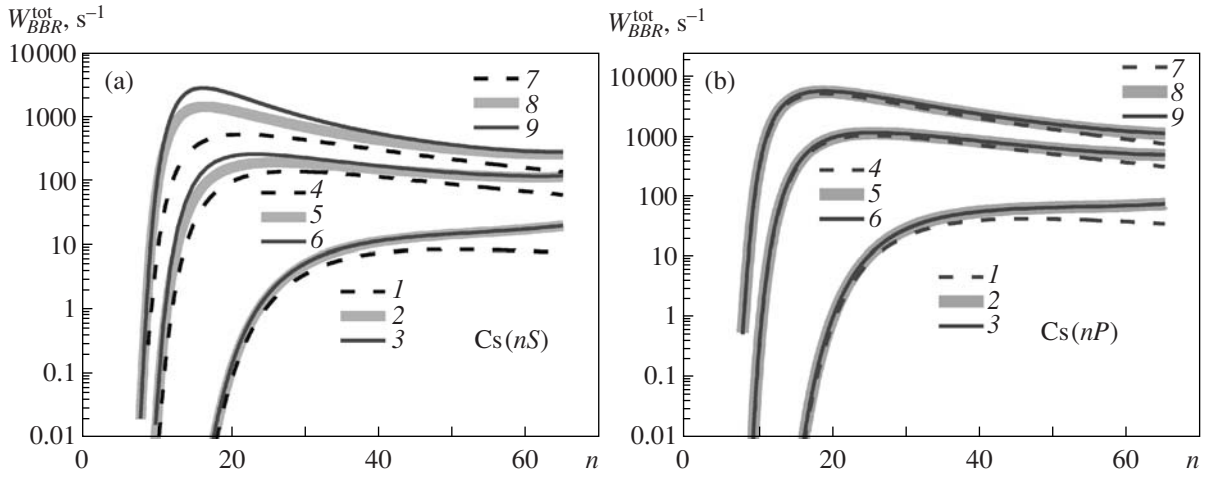

Fig. 4. Dependences of the total blackbody photoionization rate W_{BBR}^{tot} on the principal quantum number n for Rydberg atoms: (a) nS cesium and (b) nP cesium calculated at $T = 77$, 300, and 600 K; for an electric field amplitude of 10 V/cm; and at the ion accumulation interval $\Delta t = (2, 5, 8)$ 2 and $(3, 6, 9)$ 5 μs. For comparison, curves *1*, *4*, and *7* show the direct blackbody photoionization rates at $T = 77$, 300, and 600 K.

Rydberg nS lithium atoms, the hydrogen-like GDK model proves to be inadequate for the description of the data presented in Fig. 3c, since it ignores the presence of the quantum defect.

4. EFFECTIVE IONIZATION RATE OF NEIGHBORING STATES

Blackbody radiation populates Rydberg states that are neighboring to the initially excited state (see Fig. 1). In [11], we obtained the following expression for the effective ionization rate of neighboring states:

$$W_{BBR}^{mix}(nS) = \sum_{n' \geq n_c} \frac{[W(nS \longrightarrow n'P) + A(nS \longrightarrow n'P)]}{(\tau_{eff}^{n'P})^{-1} - (\tau_{eff}^{nS})^{-1}} \times W_{BBR}(n'P)\left(1 - \frac{t_{eff}^{nS}}{t_{eff}^{n'P}}\right). \quad (7)$$

Here, τ_{eff}^{nS} and τ_{eff}^{nP} are, respectively, the effective lifetimes of Rydberg nS and nP states at a given temperature;

$$t_{eff}^{nS} = \tau_{eff}^{nS}\left[\exp\left(-\frac{t_1}{\tau_{eff}^{nS}}\right) - \exp\left(-\frac{t_2}{\tau_{eff}^{nS}}\right)\right]$$

is the effective interaction time of Rydberg atoms with blackbody radiation provided that ions are accumulated in the interaction region during the time interval (t_1, t_2); and $W(nS \longrightarrow n'P)$ and $A(nS \longrightarrow n'P)$ are, respectively, the rates of blackbody-radiation-induced and spontaneous transitions between nS and $n'P$ states.

The term W_{SFI}^{mix} in Eq. (2) is described in a similar way. The expression for $W_{SFI}^{mix}(nS)$ is identical to sum (7) where $W_{BBR}(n'P)$ should be replaced by $W_{SFI}(n'P)$, and the summation over n' should be limited by n_c.

Figure 4a shows the total ionization rates of Rydberg nS cesium atoms at an electric field amplitude of 10 V/cm for ion accumulation intervals of 2 and 5 μs. Changes in these intervals lead to marked change in the total ionization rate of Rydberg nS atoms. For nP states (Fig. 4b) and nD states (not shown), the contributions from mixing of neighboring Rydberg states are significantly smaller.

It is seen that taking into account both the field ionization of high-lying states and the photoionization of neighboring Rydberg states by blackbody radiation considerably affects not only the absolute values of the ionization rate, but also the shape of the dependence on the principal quantum number.

Figure 5 presents the total blackbody ionization rates of Rydberg lithium (nS and nP) and potassium (nS and nD) atoms that were calculated for an ion accumulation interval of $\Delta t = 2$ μs ($t_1 = 0$, $t_2 = 2$ μs). Comparison with the direct photoionization rates (curves *1*, *4*, *7*) shows that, in calculations of the experimentally measured total ionization rate of Rydberg atoms, it is necessary to take into account the mixing of Rydberg states. This is especially important for nS states of lithium, for which, due to extremely low ionization rates of initially populated states (because of the occurrence of the Cooper minimum), the contribution of mixing processes to the total ionization rate proves to be dominating. In particular, the Cooper minimum in the discrete spectrum mentioned above ceases to be pronounced in the total ionization rate.

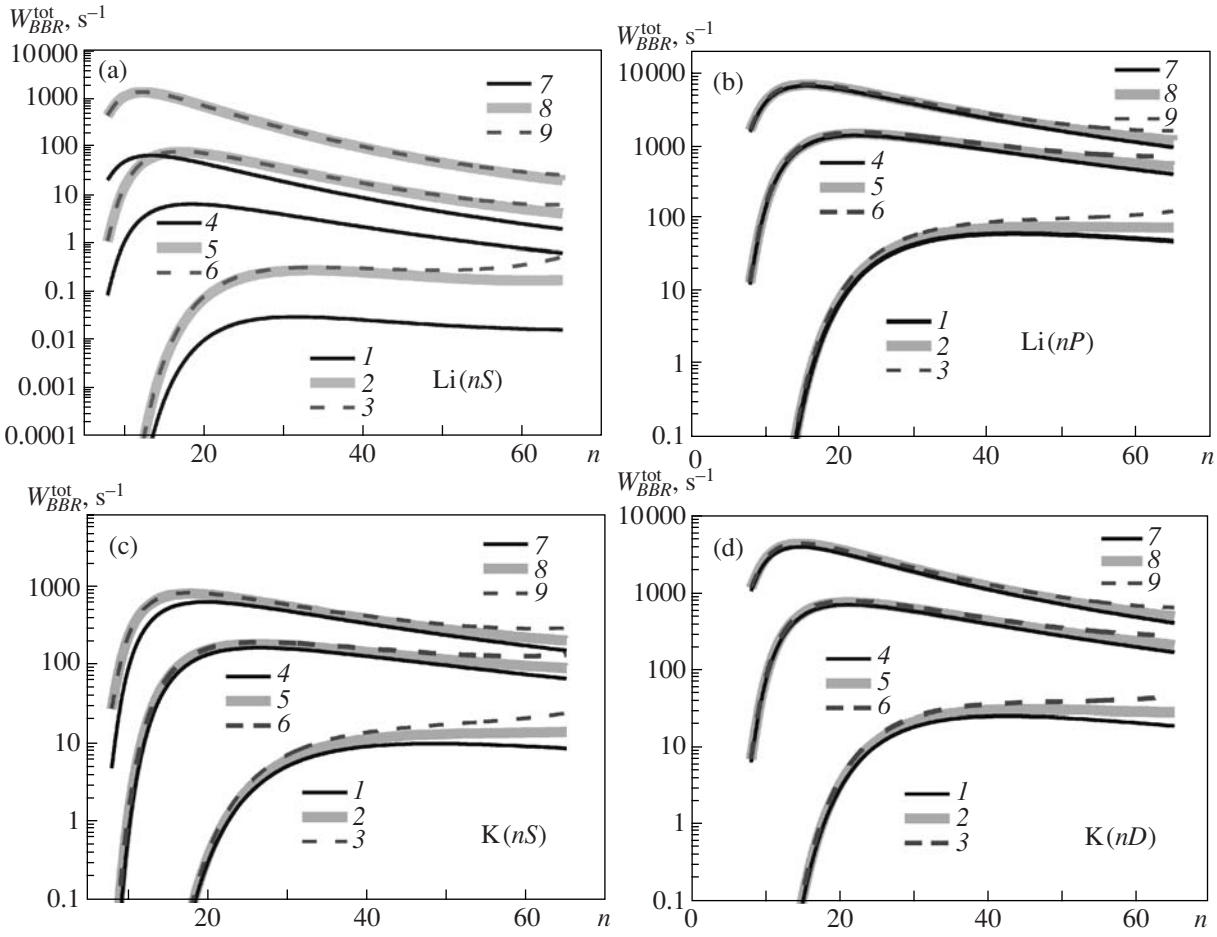

Fig. 5. Dependences of the total blackbody photoionization rate W^{tot}_{BBR} on the principal quantum number n for Rydberg atoms: (a, b) nS and nP lithium and (c, d) nS and nD potassium calculated at $T = 77$, 300, and 600 K; *2*, *5*, and *8* correspond to an electric field amplitude of 5 V/cm; *3*, *6*, and *9*, to 10 V/cm, and at an interval of accumulation of ions of $\Delta t = 2$ μs. For comparison, curves *1*, *4*, and *7* show the direct blackbody photoionization rates at $T = 77$, 300, and 600 K.

5. DISCUSSION OF RESULTS

The Dyachkov–Pankratov method used in this study for the calculation of dipole moment matrix elements has a high reliability, which is confirmed by good agreement with photoionization cross sections calculated by the model potential method [25, 29]. Direct photoionization rates that we calculated for sodium Rydberg atoms [11] agree well with calculation data of [10], which were also obtained using the model potential method. Our calculations of the depopulation rate for Rydberg states of lithium, sodium, potassium, and rubidium, which were necessary for calculating the lifetimes of Rydberg states and rates of population of neighboring states, coincide with the calculation results of [5], where the Coulomb approximation was applied.

Our calculations on the total ionization rate of nD sodium Rydberg atoms were experimentally confirmed in [14]. At the same time, discrepancies for nS states with $n > 15$ were found. For a more detailed comparison between theory and experiment, further experimental investigations of ionization processes of Rydberg atoms by blackbody radiation are necessary.

The analytical formulas for the rates of direct photoionization of Rydberg atoms obtained in [11] and the scaling coefficients C_L calculated in this study make it possible to estimate with a high accuracy the rate of photoionization of Rydberg atoms of alkali metals by blackbody radiation except for nS states of lithium, for which one has to use only numerical calculations because of an anomalous behavior of dipole matrix elements. The accuracy of the analytical formulas for the calculation of the ionization rates of high-lying Rydberg states by electric pulses is lower, and these formulas can be used only for approximate estimates.

6. CONCLUSIONS

The results that we presented in this study along with our previous results published in [11] are the first systematic investigation of ionization of Rydberg

atoms of alkali metals by blackbody radiation taking into account mixing of Rydberg states. Our results can be useful in analysis of experimental conditions under which ultracold plasma is formed [7, 8]. We showed that, to determine the rate of total ionization of Rydberg atoms in real experimental conditions, it is necessary to take into account the contribution to the detected ionization signal from the ionization of neighboring and high-lying states populated by blackbody radiation. We observed the appearance of the Cooper minimum in the discrete spectrum for transitions between nS and $n'P$ states of lithium, which is consistent with the results obtained in [30].

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