The two-electron mechanism of Ba^{2+} ion formation in the ionization of Ba atoms by YAG-laser radiation

I. I. Bondar' and V. V. Suran

Uzhgorod State University, Ukraine

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We study Ba^{2+} ion formation in the ionization of Ba atoms by a YAG laser in the presence of tunable dye-laser radiation. We find that the Ba^{2+} ion yield is considerably lower when the dye-laser radiation excites the Ba atom than when this atom is not excited. The effect is a strong indication that Ba^{2+} ion formation at the YAG laser frequency is due to the two-electron mechanism.

1. INTRODUCTION

Since the discovery of the phenomenon of formation of doubly charged ions in multiphoton ionization of alkali-earth atoms[1] there have been many experimental and theoretical papers devoted to identifying the mechanism by which such ions form (see, e.g., the review by Delone and Fedorov[2]). From the very beginning two possible mechanisms have been discussed, cascade and two-electron. In the cascade mechanism, within one laser pulse, neutral atoms (we designate these by A) are assumed to be first transformed into singly charged ions A^+ ($A + k_1\hbar\omega \to A^+ + e$) and then, as a result of further ionization, doubly charged ions A^{2+} are formed ($A^+ + k_2\hbar\omega \to A^{2+} + e$). When the two-electron mechanism is realized, A^{2+} ions are formed as a result of simultaneous detachment of two electrons from the neutral atom ($A + k_3\hbar\omega \to A^2 + 2e$).

In recent years a number of papers[3–11] have appeared that unambiguously prove that the cascade mechanism of formation of doubly charged ions (A^{2+}) is valid. This occurs, for one thing, in the optical and ultraviolet ranges. As for the two-electron mechanism, up to now its realization has not been proved unambiguously, although studies have shown that in some spectral ranges (e.g., the infrared) the formation of A^{2+} ions cannot be explained by the cascade mechanism.[12,13] In view of this we conducted a study to see whether the two-electron mechanism of A^{2+} ion formation is valid in the infrared range.

2. THE EXPERIMENT

The idea behind the two-electron mechanism of A^{2+} ion formation is to introduce an additional perturbation into the neutral atom and thus charge the probability of atom ionization by laser radiation. We studied how doubly ionized barium atoms, Ba^{2+}, are formed when Ba atoms are ionized by radiation from a YAG laser with a lasing frequency $\omega = 9395\ \text{cm}^{-1}$. Absorption of the following numbers of photons is required for the formation of Ba^+ and Ba^{2+} ions: $k_1 = 5$ for the formation of Ba^+ ions, $k_2 = 9$ for the formation of Ba^{2+} ions from Ba^+ ions, and $k_3 = 14$ for the formation of Ba^{2+} ions from neutral Ba atoms. Note that the formation of Ba^{2+} ions at this frequency was studied earlier in Refs. 14 and 15, but the data does not lead to an unambiguous conclusion about the mechanism of Ba^{2+} ion formation.

For additional perturbation that in our experiments acted on Ba atoms we chose the excitation of these atoms. To this end we used the radiation of a tunable dye laser. Pumping of this laser was synchronized with the operation of the YAG laser. The frequency of the dye laser was varied between 17 860 and 18 160 cm^{-1}, and the lasing linewidth was $\Delta\omega \approx 2$–$3\ \text{cm}^{-1}$. According to the data of Ref. 16, the following processes are possible in this range: one-photon excitation of state $6s6p\,^1P_1^0$ with an excitation frequency $\omega = 18\,060\ \text{cm}^{-1}$, two-photon excitation of states $6s7d\,^3D_2$ ($\omega = 17\,881\ \text{cm}^{-}$) and $5d\,6d\,^3D_2$ ($\omega = 18\,100\ \text{cm}^{-1}$), and one-photon transition from state $6s6p\,^1P_1^0$ to state $5d\,6d\,^3D_2$ ($\omega = 18\,140\ \text{cm}^{-1}$).

The radiations from the YAG and dye lasers were linearly polarized, and the pulse length was practically the same, 40 ns. The beams from the two lasers were combined in space and focused on a beam of neutral Ba atoms. The YAG radiation intensity was selected in such a way that ionization of Ba atoms was sure to be saturated by the radiation, and in our experiment it was set at $F = 6.5\times10^{10}\ \text{W cm}^{-2}$. Both Ba^+ and Ba^{2+} ions were observed, and the yield of the Ba^+ ions was found to be higher by a factor of 40. The dye-laser radiation intensity was $F = 3.4\times10^{6}\ \text{W cm}^{-2}$, and only Ba^+ ions were observed as the product of ionizing Ba atoms.

The quantities measured in the experiments were the yields of Ba^+ and Ba^{2+} ions in the three following cases: only the beam from the YAG laser acts on the beam of Ba atoms, only the beam from the dye laser acts on the beam of Ba atoms, and both laser beams act on the beam of Ba atoms.

3. EXPERIMENTAL RESULTS

The results are depicted in Fig. 1. The dashed curves correspond to the ionization process when both laser beams act on the beam of Ba atoms, the dot-dash curves when only the dye-laser radiation acts on the Ba atoms, and the solid horizontal lines when only the YAG-laser radiation acts. The vertical dot-dash lines designated by the letters *A*, *B*, *C*, and *D* on the frequency axis indicate the frequencies of the dye-laser radiation corresponding to the following transitions in the spectrum of a neutral Ba atom:

$$A - 6s^2\,{}^1S_0 + 2\hbar\omega \to 6s7d^3D_2 \quad (17881\ \text{cm}^{-1}),$$

$$B - 6s^2\,{}^1S_0 + \hbar\omega \to 6s6p^1P_1^0 \quad (18060\ \text{cm}^{-1}),$$

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$$C - 6s^2\,{}^1S_0 + 2\hbar\omega \to 5d6d^3D_2 \quad (18100\,\mathrm{cm}^{-1}),$$

$$D - 6s6p^1P_1^0 + \hbar\omega \to 5d6d^3D_2 \quad (18140\,\mathrm{cm}^{-1}).$$

The data depicted in Figs. 1a and 1b was obtained with the following YAG-laser and dye-laser radiation intensities: $F_{YAG} = 3.5\times10^{29}$ $s^{-1}cm^{-2}$ and $F_{dye} = 9.4\times10^{24}$ $s^{-1}cm^{-2}$, and the data in Fig. 1c with $F_{YAG} = 1.5\times10^{29}$ $s^{-1}cm^{-2}$ and $F_{dye} = 2.0\times10^{24}$ $s^{-1}cm^{-2}$.

Let us first consider Fig. 1a, which shows the yield of Ba^{2+} ions. The reader can see that when the combined radiation of the YAG and dye lasers acts on the atomic target the Ba^{2+} ion yield is generally lower than when only the YAG-laser acts. Also, the dips in the Ba^{2+} ion yield occur at dye-laser frequencies corresponding to the excitation of neutral Ba atoms. The lowest dip occurs at a dye-laser frequency ω equal to 18 060 cm^{-1}, which corresponds to the one-photon excitation of the state $6s6p^1P_1^0$.

Let us now turn to Fig. 1b, which depicts the data on the Ba^+ ion yield obtained in the same conditions as the data on the Ba^{2+} ion yield. Clearly, the Ba^+ ion yield is higher when the combined radiation of the YAG and dye lasers acts on the target than when only the YAG-laser radiation acts on the target. In other words, adding the dye-laser radiation to the YAG-laser radiation raises the probability of Ba^+ ion formation. The peaks in the Ba^+ ion yield in this case correspond in frequency to the dips in the Ba^{2+} ion yield.

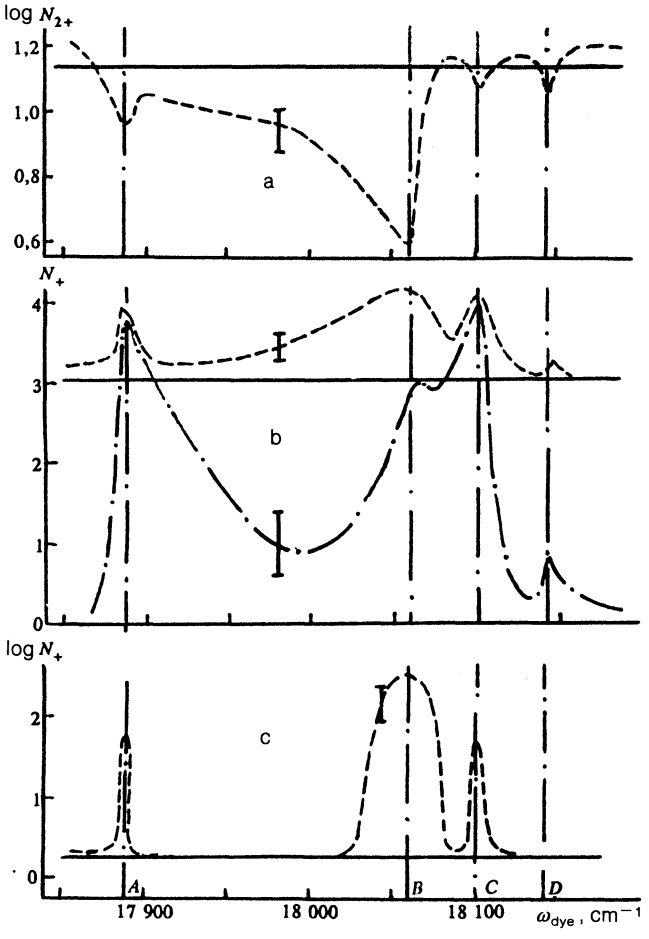

FIG. 1. Results of measuring the yields of Ba^+ and Ba^{2+} ions as functions of the dye-laser radiation frequency (for notation see the text).

Recall that the data shown in Figs. 1a and 1b was obtained when the process of ionizing Ba atoms by the YAG-laser radiation was saturated. The increase in the Ba^+ ion yield and the formation of peaks when the dye-laser radiation is added are caused by the fact that the increase in the probability of Ba^+ ion formation leads to an increase in the effective volumes of the interaction of laser radiation with atoms. Because of the saturation effect, these peaks have approximately the same height, which makes it impossible to estimate the magnitudes of the probability of Ba^+ ion formation at these peaks. For this reason we measured the Ba^+ ion yields at lower values of YAG-laser and dye-laser radiation intensities. The relevant data is depicted in Fig. 1c, which shows that when the combined radiation of the YAG and dye lasers acts on the target the highest probability of Ba^+ ion formation occurs if the dye-laser radiation excites the $6s6p^1P_1^0$ state.

The sharp increase in the probability of Ba^+ ion formation at $\omega = 18\,060$ cm^{-1} occurs because adding radiation with such a frequency transfers approximately 50 percent of the Ba atoms from the ground state to the excited state $6s6p^1P_1^0$, and it is from this state that ionization by YAG-laser radiation takes place. In view of this, Ba^+ ion formation transform from a five-photon process (when barium is ionized by YAG-laser radiation from the ground state $6s^2S_0^1$) into a three-photon (ionization of barium by the same radiation from state $6s6p^1P_1^0$), which in turn leads to a sharp increase in the probability of Ba^+ ion formation. As for the other peaks, here the increase in the probability of Ba^+ ion formation is moderate because exciting the states from which ionization occurs requires the absorption of two photons from the dye-laser radiation, and the probability of a two-photon process is, naturally, lower than that of a one-photon.

Thus, the results of the experiment show that the additional excitation of Ba atoms by a dye-laser in the ionization by YAG-laser radiation increases the probability of Ba^+ ion formation and reduces the Ba^{2+} ion yield. And the greater the increase in the probability of Ba^+ formation, the greater the decrease in the Ba^{2+} ion yield.

Such an effect cannot be explained by the cascade mechanism. For instance, as noted earlier, with this mechanism the A^+ ions serve as the target for A^{2+} ion formation. Hence, obviously, every additional excitation of neutral atoms that leads to an increase in the probability of A^+ ion formation must also lead to an increase in the A^{2+} ion yield. This is certainly true if the process of atom ionization is not saturated. But if saturation is present, additional excitation of atoms may not lead to an increase in the A^{2+} ion yield (the yield will remain constant). Indeed, experimental studies[5] have shown that additional excitation of neutral barium atoms within the cascade mechanism of formation of barium ions leads to a situation in which the yield of these ions as a function of frequency either increases or remains constant. But the effect we observed, where because of additional excitation of neutral atoms the probability of A^+ ion formation increased and at the same time the A^{2+} ion yield decreased, cannot be realized by the cascade mechanism. At the same time, such an effect can be realized through the two-electron mechanism. Let us explain this in greater detail.

4. CALCULATIONS

We assume that the ionization process is saturated since it was in these conditions that our experiment took place. Saturation in this case means that the concentration of neutral atoms, n, decreases within a single laser pulse, that is, is a function of time, $n = n(t)$. Since neutral atoms in the model of the two-electron mechanism are a target for the formation of A^+ and A^{2+} ions, the ion yields per unit volume (n_+ and n_{2+}, respectively) are

$$n_+ = \int n(t) W_+(t) dt, \tag{1}$$

$$n_{2+} = \int n(t) W_{2+}(t) dt, \tag{2}$$

where $W_+(t)$ and $W_{2+}(t)$ are the probabilities of formation of A^+ and A^{2+} ions, respectively. Here and in what follows we assume that the distribution of laser radiation depends only on time and not on spatial coordinates. The integration in Eqs. (1) and (2) is over the duration of a single laser pulse.

Note that generally saturation means that $n_+ + n_{2+} = n_0$, where n_0 is the initial concentration of neutral atoms. In a real situation, however, the process of atom ionization is determined chiefly by the formation of A^+ ions, since the probability of A^{2+} ion formation is always considerably lower than that of A^+ ion formation, that is, the saturation condition transforms in this case into $n_+ \approx n_0$. The time dependence of $n(t)$ can then be determined by integrating the equation

$$\frac{dn(t)}{dt} = -n(t) W_+(t).$$

This yields $n(t) = n_0 \exp[-\int W_+(t) dt]$. Equations (1) and (2) can then be written as

$$n_+ = n_0, \tag{1'}$$

$$n_{2+} = n_0 \int \exp[-\int W_+(t) dt] W_{2+}(t) dt. \tag{2'}$$

We now assume that an additional excitation of atoms has been introduced into the ionization process. This increases the probability of formation of both A^+ and A^{2+} ions. Such an increase results in the ionization process becoming saturated in less time than without additional excitation. Naturally, the A^+ ion yield does not change, that is,

$$n_+' \approx n_+ \approx n_0. \tag{1''}$$

But what happens to the A^{2+} ions? Their number is

$$n_{2+}' = n_0 \int \exp[-\int W_+'(t) dt] W_{2+}'(t) dt, \tag{2''}$$

where $W'_+(t)$ and $W'_{2+}(t)$ are the probabilities of formation of both A^+ and A^{2+} ions under additional excitation. Since additional excitation changes the functions $W_+(t)$ and $W_{2+}(t)$, the value of the integral in Eq. (2'') may be larger than, smaller than, or equal to, the value of the integral in Eq. (2'), depending on the behavior and values of the functions $W_+(t)$, $W_{2+}(t)$, $W'_+(t)$, and $W'_{2+}(t)$.

Thus, introducing additional excitation when the process of atom ionization is saturated leads to a situation in which the A^{2+} ion yield may not only increase but also remain constant or even decrease. The reader will recall that this is accompanied by an increase in probabilities of formation of both A^+ and A^{2+} ions.

To illustrate the possibility of the above cases involving the A^{2+} ion yield being realized in the two-electron mechanism, we calculated the time dependences (within a single laser pulse) of the concentrations of neutral atoms and of singly and doubly charged ions. As Eqs. (2') and (2'') imply, the realization of the above-noted cases involving the A^{2+} ion yield in real experiments depends in a complicated manner on the interplay between the functions $W_+(t)$, $W_{2+}(t)$, $W'_+(t)$, and $W'_{2+}(t)$.

We used a simplified model in our calculations. We assumed, for one thing, that the time dependence of the probability of A^{2+} ion formation is the same as that of A^+ ion formation, irrespective of whether additional excitation is present. Bearing in mind that the probability of A^{2+} ion formation is much lower than that of A^+ ion formation, we expressed the criteria for realizing the above cases of A^{2+} ion yield in such conditions by fairly simple formulas. For instance, if

$$\frac{W_{2+}'}{W_{2+}} \gg \frac{W_+'}{W_+}, \tag{3}$$

when additional excitation is introduced, the A^{2+} ion yield grows. But if

$$\frac{W_{2+}'}{W_{2+}} \approx \frac{W_+'}{W_+}, \tag{4}$$

this yield remains constant, and if

$$\frac{W_{2+}'}{W_{2+}} \ll \frac{W_+'}{W_+} \tag{5}$$

it decreases. Here W_+, W'_+, W_{2+}, and W'_{2+} denote the peak values (within a single laser pulse) of the probabilities of A^+ and A^{2+} ion formation. Note that the values of some parameters used in the calculations are close to those realized in our experiment. For one thing, we allowed for the fact that the laser beam that ionizes the atoms has a Gaussian intensity distribution in time with a duration $\tau = 40$ ns at half-height and a maximum intensity of $F = 6.5 \times 10^{10}$ W cm^{-2}. We also took it into account that as a result of the transition from five-photon to three-photon ionization the probability of A^+ ion formation increases.

The values of five- and three-photon ionization probabilities needed in the calculations were determined from the known cross section of five-photon ionization of Ba atoms by the YAG laser and from the typical values of the cross sections of three-photon ionization of alkali-earth atoms.[17] As for the probability of A^{2+} ion formation, it was assumed to be a factor of 100 smaller in magnitude than that of A^+ ion formation, both with and without additional excitation, and the specific value of this probability was selected in such a way as to satisfy conditions (3)–(5).

The results of calculations are depicted in Figs. 2(a)–2(c). Figure 2d depicts the Gaussian envelope of the temporal distribution of the laser radiation, which was used in the calculations:

$$F = e^{-1,73 \cdot 10^{15} (\tau - 5 \cdot 10^{8})^2}. \tag{6}$$

The solid curves correspond to ionization without additional

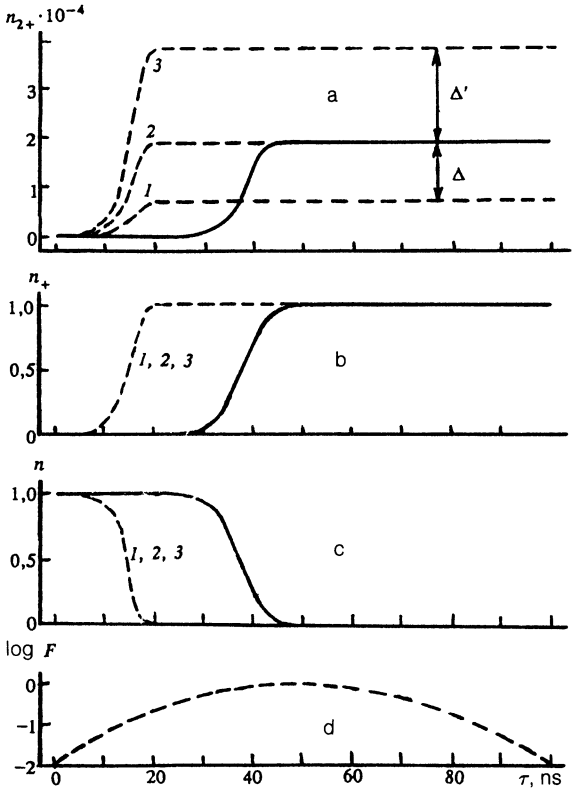

FIG. 2. (a)–(c) Results of calculating the time dependence of the concentrations of doubly charged ions (a), singly charged ions (b), and neutral atoms (c) in the duration of a single laser pulse for different values of W'_{2+}: curve *1*, 10^7 s^{-1}, curve *2*, 2.8×10^7 s^{-1}, and curve *3*, 6×10^7 s^{-1}. (d) The Gaussian envelope of the temporal distribution of the laser radiation [Eq. (6)].

perturbation of neutral atoms. The assumed probabilities of A^+ and A^{2+} ion formation in this case were $W_+ = 6\times10^8$ s^{-1} and $W_{2+} = 10^5$ cm^{-1}. The dashed curves in Figs. 2a–2c correspond to ionization with additional perturbation of neutral atoms. Curves *1–3* differ in the values of the probability W'_{2+} of A^{2+} ion formation. In all three cases the probability W'_+ of A^+ ion formation was assumed equal to 1.5×10^{11} s^{-1}.

The results shown in Figs. 2a–2c serve as a good illustration of the fact that although introducing an additional excitation of neutral atoms in the ionization of these atoms within the two-electron mechanism of A^{2+} ion formation does raise the probability of A^{2+} ion formation, the yield of these ions may not only increase (in Fig. 2a this increase is depicted by Δ') but remain constant or even decrease (the decrease is denoted by Δ).

The most exotic is the third possibility, where an increase in the probability of A^{2+} ion formation is accompanied by a decrease in the ion yield. Qualitatively this phenomenon can be explained as follows. The increase in the probability of A^+ ion formation (due to additional excitation of atoms), which is predominant in the ionization process, leads to faster saturation of the ionization than when there is no additional excitation. As noted earlier, the fact that saturation has set in in the ionization process indicates that there are no neutral atoms in the interaction volume. Thus, introducing additional excitation of atoms when ionization is saturated results in a situation in which neutral atoms disappear sooner than when there is no such excitation. If, in addition, the probability of A^{2+} ion formation, while increasing, remains such that the effective formation of these ions starts much later than the onset of ionization saturation (that is, when there are no neutral atoms in the interaction region), the yield of these ions naturally decreases.

Recall that in the above reasoning concerning the two-electron mechanism we assumed that additional excitation of atoms increases the probabilities of formation of both A^+ ion and A^{2+} ions. The effect of a decreasing A^{2+} ion yield, obviously, will also manifest itself when only the probability of A^+ ion formation grows while that of A^{2+} ion formation remains constant or decreases (here condition (5) is satisfied automatically).

We also assumed that the spatial distribution of laser radiation in the interaction volume is homogeneous, although in real experiments this distribution is always inhomogeneous. Allowing for this inhomogeneity modifies the above picture. The reason is that when the radiation distribution is inhomogeneous, the integral ion yield is affected not only by the region where the ionization of the atoms is saturated but also by regions where this is not so. In view of this, in contrast to our calculations, the increase, due to the additional excitation, of the probability of A^+ ion formation leads to an increase in the ion yield (as observed in our experiment). As for the A^{2+} ions, the inhomogeneities in the distribution will only reduce the extent to which the ion yield varies but will not change the pattern of this variation.

This analysis shows that the observed increase in the probability of Ba^+ ion formation and decrease in the Ba^{2+} ion yield, due to the introduction of additional excitation in the process of ionizing barium atoms, can be explained fairly well by the two-electron mechanism. Since, as noted earlier, there is no way in which this effect can be realized in the cascade model, its manifestation unambiguously points to the two-electron mechanism of formation of doubly charged ions Ba^{2+}.

5. CONCLUSION

The main result of our study is that for the first time it has been proved unambiguously that the two-electron mechanism of formation of doubly charged ions can be realized. For one thing, we discovered that the mechanism manifests itself in the ionization of barium atoms by YAG-laser radiation.

Note that the two-electron mechanism is, in all likelihood, predominant in the formation of doubly charged ions of alkali-earth atoms in the infrared range. This assumption rests on the fact that studies conducted up to now (Refs. 3–11) have shown that formation of doubly charged ions of alkali-earth atoms in the optical and ultraviolet ranges takes place basically by the cascade mechanism. As for the infrared range, studies have shown (see Refs. 12 and 13) that the formation of A^{2+} ions in this range cannot be explained by that mechanism.

A qualitative explanation of this goes as follows. When the cascade mechanism is realized in the infrared range, there is a greater difference between the number of photons

whose absorption is necessary for the formation of singly charged ions, k_1, and that of photons whose absorption is necessary for the formation of doubly charged ions, k_2 ($k_1 = 5$–6 and $k_2 = 9$–10), than there is when the mechanism is realized in the optical range ($k_1 = 3$–4 and $k_2 = 6$–7). In view of this in the infrared range the probability of A^{2+} ion formation is considerably suppressed by that of A^+ ion formation. This is not the case in the optical range.

As for the two-electron mechanism, in its realization in the infrared range there is also a great difference between the number of photons whose absorption is necessary for the formation of singly charged ions, k_1, and that of photons whose absorption is necessary for the formation of doubly charged ions, k_3 ($k_1 = 5$–6 and $k_3 = 14$–16). Notwithstanding this fact, however, in this mechanism the probability of A^{2+} ion formation may not differ very much from the probability of A^+ ion formation not only in the optical range but also in the infrared. The reason is that the two-electron mechanism manifests itself in the neutral-atom spectrum, that is, in the spectrum of the bound autoionization states between the first and second ionization potentials. It is known that the spectrum of such autoionization states of alkali-earth atoms is very dense,[18] which makes one-photon resonant transitions between these states possible for each photon. This is the reason why the probability of A^{2+} ion formation does not differ very much from that of A^+ ion formation.

As for the cascade mechanism, in view of the anharmonicity of the spectrum of the singly charged A^+ ions acting as the target in the formation of A^{2+} ions, there is practically no way in which a sequence of one-photon transitions can form. Hence, as the photon energy decreases, the probability of the cascade mechanism realizing itself will be suppressed by the probability of realization of the two-electron mechanism. At higher photon energies, dependence on the actual probability values, the cascade mechanism will prevail over the two-electron.

Of course, this explanation of why different mechanisms of doubly charged ion formation are realized in different spectral ranges is only qualitative and cannot be considered final.

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