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Mechanism of formation of doubly charged ions upon nonlinear ionisation of Ba atoms in the frequency range from 9390 to 9470 cm^{-1}

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Abstract. **A mechanism of Ba^{2+} ion formation upon nonlinear ionisation of Ba atoms in the frequency range between 9390 and 9470 cm^{-1} is studied. The ions are shown to be produced by the two-electron mechanism.**

Starting with the first experiments in which the formation of doubly charged ions upon multiphoton ionisation of atoms has been discovered [1] and till the present time, the regularities and specific features of the formation of these ions and the mechanism of their formation are actively studied in different spectral regions at different durations of laser radiation [2]. In Ref. [3], two possible mechanism of formation of doubly charged A^{2+} ions, namely, the cascade and two-electron mechanisms, have been predicted.

In the case of the cascade mechanism, doubly charged ions are formed in two stages. At the first stage, atoms are ionised through absorption of K_0 quanta, and singly charged A^+ ions are formed. At the second stage, singly charged ions are ionised through absorption of K_1 quanta, and doubly ionised A^{2+} ions are formed. These two stages are realised during a single laser pulse. In the case of the two-electron mechanism, doubly charged ions are formed through simultaneous detachment of two electrons from an atom that absorbs K_2 quanta. Note that a model describing the simultaneous detachment of two electrons from an atom has not yet been developed.

The studies of multiphoton ionisation of atoms of alkaline-earth elements in the visible spectral region between 15000 and 19000 cm^{-1} at linear and circular polarisation of laser radiation showed that in most cases the resonance structure in the dependences $N_{2+}(\omega)$ (N_{2+} is the yield of doubly charged ions and ω is the laser radiation frequency), which is typical of the formation of doubly charged ions, can be unambiguously assigned to the resonance transitions in the spectrum of singly charged ions of these atoms (see, e.g., [4 – 7]). This fact unequivocally proves that the cascade mechanism of formation of doubly charged ions of the aforementioned atoms is realised in the visible region. Note that singly charged ions are ionised not only from their ground $ns\,^2S_{1/2}$ state, but also from the first excited states $nd\,^2D_{3/2,5/2}$ and $np\,^2P^0_{1/2,3/2}$.

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Similar studies in the IR region of the spectrum (8100 – 9100 cm^{-1} and 9390 – 9500 cm^{-1}) showed that, when the dependences $N_{2+}(\omega)$ exhibited the resonance structure, the two-electron mechanism of formation of the Sr^{2+} and Ba^{2+} ions was realised [8 – 10].

The aim of this paper is to study the dependence of the probability of formation of Ba^+ and Ba^{2+} ions on the laser radiation intensity F and elucidate the mechanism of formation of Ba^{2+} ions in the IR region when the yield of these ions shows no resonance structure. Note that the similar study performed earlier in the visible region of the spectrum between 18790 and 19000 cm^{-1} [11] proved the cascade mechanism of formation of doubly charged ions.

We used a neodymium laser operating on a single transverse mode, whose frequency was tuned between 9390 and 9470 cm^{-1}. The laser radiation was linearly polarised, and the laser pulse length was 40 ns. The dependences $\lg N_+(\lg F)$ and $\lg N_{2+}(\lg F)$ were measured by the traditional experimental technique, which is described in detail in our previous papers (see, e.g., [4]).

Note that the dependences $\lg N_+(\lg F)$ and $\lg N_{2+}(\lg F)$ measured by us were interpreted using the results of the study of the dependences $N_+(\omega)$ and $N_{2+}(\omega)$ in the frequency range from 9390 to 9470 cm^{-1} [8, 12]. The dependence $N_{2+}(\omega)$ measured in these studies for Ba^{2+} ions contained only one resonance peak at 9410 cm^{-1}. Its position on the frequency scale and the width were virtually the same as the position and the width of the resonance peak in the dependence $N_+(\omega)$ for Ba^+ ions.

Note that the dependence $N_+(\omega)$ for Ba^+ ions exhibits one more resonance peak at 9465 cm^{-1} in addition to the peak mentioned above. One of the typical results of the study of the dependences $\lg N_+(\lg F)$ and $\lg N_{2+}(\lg F)$ upon ionisation of Ba ions is presented in Fig. 1. To obtain a more complete information, we studied these dependences both at the frequencies in the region of the aforementioned resonance maxima of the yield of Ba^+ and Ba^{2+} ions and at the frequencies lying outside this region. The results of our study of these dependences suggest a number of conclusions.

The Ba^{2+} ions are formed not only at those laser radiation intensities at which the multiphoton ionisation of Ba atoms is saturated, but also at the lower intensities. Note that the saturation of the multiphoton ionisation of atoms is accompanied by their full ionisation in the effective volume of interaction of laser radiation with an atomic beam.

The degree of nonlinearity of the Ba^{2+} ion formation is changed at the laser radiation intensity F_s that corresponds to the beginning of saturation of the multiphoton ionisation of Ba atoms. In our experiments, $F_s = 2.1 \times 10^{29}$ cm^{-2} s^{-1}.

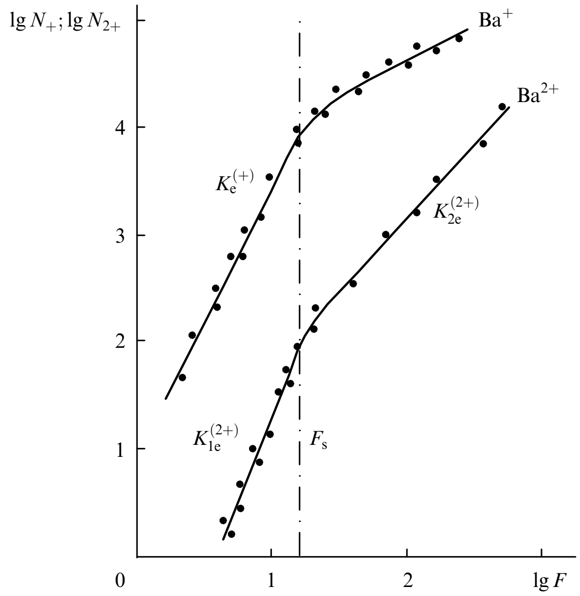

Figure 1. Dependences of the yields of singly and doubly charged barium ions ($\omega = 9395$ cm^{-1}) on the intensity of linearly polarised laser radiation; F_s (the vertical dashed line) is the intensity at which the saturation of multiphoton ionisation of barium atoms begins.

The dependences lg N_+(lg F) and lg N_{2+}(lg F) measured upon multiphoton ionisation of Ba atoms in the IR region substantially differ from the dependences obtained for these atoms in the visible region between 18790 and 19000 cm^{-1}, where the cascade mechanism of formation of doubly charged ions is realised [11]. In particular, in the case of the cascade mechanism, doubly charged ions are formed with a high efficiency only at the laser radiation intensity corresponding to the saturation of multiphoton ionisation of atoms.

The results of the measurements of the degrees of nonlinearity of the Ba$^+$ ion formation at $F < F_s$ (K_e^+) and the degrees of nonlinearity of the Ba^{2+} ion formation at $F < F_s$ ($K_{1e}^{(2+)}$) and $F > F_s$ ($K_{2e}^{(2+)}$) at different laser radiation frequencies are presented in Table 1. Here, $K_e^{(+)}$ and $K_{1e}^{(2+)}$, $K_{2e}^{(2+)}$ are the degrees of nonlinearity of formation of singly and doubly charged ions, respectively.

Consider first the possibility of interpretation of the data on the degree of nonlinearity of the Ba^{2+} ion formation in the cascade model of formation of these ions. Note that the multiphoton ionisation of Ba$^+$ ions from the ground $6s\,^2S_{1/2}$ state in the frequency region used by us requires absorption of the largest number of quanta ($K_1 = 9$), and the ionisation of these ions from the excited $6p\,^2P^0_{3/2}$ state requires the smallest number of quanta ($K_1 = 7$). Note also that the multiphoton ionisation of Ba atoms in this frequency region requires absorption of five quanta ($K_0 = 5$).

Table 1. Degrees of nonlinearity of the formation of singly and doubly charged barium ions.

ω/cm^{-1}	$K_e^{(+)}$	$K_{1e}^{(2+)}$	$K_{2e}^{(2+)}$	K_3	K_3'
9395	4.9 ± 0.2	9.1 ± 0.3	2.7 ± 0.2	12 – 14	7 – 9
9412	4.1 ± 0.2	7.2 ± 0.3	2.5 ± 0.2	11 – 13	7 – 9
9465	2.1 ± 0.1	9.2 ± 0.3	2.6 ± 0.2	9 – 11	7 – 9

If the cascade mechanism of formation of doubly charged ions is realised, Ba$^+$ ions represent a target for the formation of Ba^{2+} ions. Because of this, the dependence of the Ba^{2+} yield on the laser radiation intensity F in the region $F > F_s$ should be extrapolated in the form $N_{2+} \sim F^{K_1}$. This is caused by the fact that when $F > F_s$, the concentration of singly charged ions n in the effective volume $V_{\rm eff}$, where doubly charged ions are efficiently produced, is almost independent of the radiation intensity because of the full ionisation of atoms and, therefore, it is equal to the initial concentration of neutral atoms ($n \approx n_0$).

As for the intensity region $F < F_s$, where the ionisation of atoms is not saturated, the Ba^{2+} yield should be extrapolated by the dependence $N_{2+} \sim F^{K_e^+ + K_1}$. This is caused by the fact that when $F < F_s$, the concentration of singly charged ions in the effective volume $V_{\rm eff}$ depends on the radiation intensity as $n \sim F^{K_e^{(+)}}$. The type of the dependences $N_{2+}(F)$ discussed above should be observed for the nonresonance multiphoton ionisation of singly charged ions. Indeed, only in this case the degree of nonlinearity of ionisation of these ions that is observed in the experiment should coincide with the number of quanta K_1. Moreover, it is also assumed that the singly charged ions formed upon multiphoton ionisation of atoms are found only in a certain state (in the ground state or in one of the excited states).

In the frequency range under study, no data is available on the distribution of Ba$^+$ ions, which are formed upon multiphoton ionisation of atoms, over different states. Because of this, we will assume that these ions appear only in the ground $6s\,^2S_{1/2}$ state or only in the excited $6p\,^2P^0_{3/2}$ state, whose energy is larger than the energies of the $5d\,^2D_{3/2,5/2}$ states. Note that in the study of multiphoton ionisation of Ba atoms in the visible region, a high efficiency of formation of Ba$^+$ ions was observed only for the above-mentioned states [4 – 7].

Upon the nonresonance ionisation of Ba$^+$ ions from these states, the degree of nonlinearity of their ionisation K_1' should lie in the range from $K_1 = 7$ (when Ba$^+$ ions are ionised only from the excited $6p\,^2P^0_{3/2}$ state) to $K_1 = 9$ (when Ba$^+$ ions are ionised only from the ground $6s\,^2S_{1/2}$ state), i.e., $K_1' = 7 - 9$. The above discussion suggests that in the case of the cascade mechanism of formation of doubly charged ions, the dependence of the Ba^{2+} yield on the laser radiation intensity at $F < F_s$ and $F > F_s$ should be extrapolated by the dependences with the degrees of nonlinearity $K_3 = K_e^{(+)} + K_1'$ and $K_3' = K_1'$, respectively.

It follows from Table 1 that the degrees of nonlinearity $K_{2e}^{(2+)}$, which are observed in the region of saturation of multiphoton ionisation of atoms (at $F > F_s$), are almost independent of the laser radiation frequency. As for the degrees of nonlinearity $K_{1e}^{(2+)}$, which are observed at intensities below the saturation value (at $F < F_s$), they are almost the same for those frequencies at which the resonant structure in the yield of Ba^{2+} ions is not observed. Moreover, we have $K_{1e}^{(2+)} < K_3$ and $K_{2e}^{(2+)} < K_3'$ at all the frequencies.

The experimental observation of the degrees of nonlinearity $K_{1e}^{(2+)}$ and $K_{2e}^{(2+)}$ that do not coincide with K_3 and K_3', respectively, is inconsistent with the model of the cascade mechanism of formation of Ba^{2+} ions. Primarily, this concerns the degree of nonlinearity $K_{2e}^{(2+)}$ (at $F > F_s$) observed in the experiment. Indeed, our studies [11] and the studies of other authors [13, 14] in the visible region of the spectrum showed that in the case of the cascade mechanism of formation of doubly charged ions, one observes in the intensity region $F > F_s$ the degrees of nonlinearity $K_{2e}^{(2+)}$ that coincide with K_1, i.e., with the number of quanta that should be absorbed to produce the multiphoton ionisation of singly charged ions from certain initial states.

Moreover, as noted above, doubly charged ions are efficiently formed under the action of visible radiation only in the ionisation saturation region. This is caused by the fact that when $F > F_s$, the effective volume V_{eff} contains only singly charged ions, which represent a target for the formation of doubly charged ions.

The observation of the degrees of nonlinearity of Ba^{2+} ion formation ($K_{2e}^{(2+)}$) that are smaller than K_3 is also inconsistent with the cascade mechanism of formation of these ions. An anomalously large ratio of the yields of singly and doubly charged ions ($N_+/N_{2+} \leqslant 10^2$) in the intensity region where the saturation of multiphoton ionisation of Ba atoms is not realised (at $F < F_s$) is also inconsistent with the cascade mechanism of Ba^{2+} ion formation. In particular, the estimates based on the technique [5] show that in the case where the cascade mechanism of formation of Ba^{2+} ions is realised, the calculated ratio of the yields of singly and doubly charged ions N_+^c/N_{2+}^c for the nonresonant multiphoton ionisation of Ba^+ ions from the $6p\,^2P_{3/2}^0$ and $6s\,^2S_{1/2}$ states should be about 10^7 and 10^{10}, respectively. Note that the ratio N_+^c/N_{2+}^c was estimated using the typical ionisation cross sections for atoms [15] because the data of Ref. [11] show that the cross sections for multiphoton ionisation of Sr^+ and Ba^+ ions (at $K_1 = 4$ and $K_1 = 5$) are approximately the same as the cross sections for multiphoton ionisation of atoms.

One can see that there exists a large difference between the experimental ratio N_+/N_{2+} and the calculated ratio N_+^c/N_{2+}^c that is expected for the cascade mechanism. Note also that the same estimates of the ratio N_+^c/N_{2+}^c for the visible region, where the cascade mechanism of formation of doubly charged ions is realised, agree well with the ratio N_+/N_{2+} that is observed in the experiments [5] performed in the same spectral region.

Thus, the results of the study of the dependences $\lg N_{2+}(\lg F)$ for Ba^{2+} ions cannot be explained by the cascade mechanism of their formation.

Consider the possibility of explanation of the experimental results within the framework of the two-electron mechanism of formation of Ba^{2+} ions. In this case, neutral atoms represent a target for the formation of doubly charged ions. If the two-electron mechanism is realised, the simultaneous detachment of two electrons from a Ba atom requires that $K_2 = 13$ or 14 quanta be absorbed depending on the laser radiation frequency. Therefore, in the case of realisation of this mechanism under conditions of nonresonance detachment of two electrons from an atom at the laser radiation intensity $F < F_s$, the yield of Ba^{2+} ions should be extrapolated by the dependence $N_{2+} \sim F^{K_2}$.

As follows from Table 1, $K_{1e}^{(2+)}$ and K_2 considerably differ, and this difference is observed in the frequency region where the frequency dependences $N_+(\omega)$ and $N_{2+}(\omega)$ (for Ba^+ [12] and Ba^{2+} ions [8], respectively) exhibit no resonance structure. This fact contradicts to the realisation of nonresonant detachment of two electrons from an atom.

As in the analysis of realisation of the cascade mechanism of formation of doubly charged ions, we estimated the ratio N_+^c/N_{2+}^c for the nonresonance detachment of two electrons from an atom. These estimates show that the ratio N_+^c/N_{2+}^c should substantially exceed the ratio expected for the cascade mechanism (because $K_2 > K_1$) and the ratio N_+/N_{2+} obtained in the experiment. Thus, an anomalously large experimental ratio of the yields of singly and doubly charged ions ($N_+/N_{2+} \leqslant 10^2$) at $F < F_s$ also contradicts to the realisation of nonresonant detachment of two electrons from an atom.

The two facts considered above may suggest that a certain 'resonance' detachment of two electrons from an atom occurs.

As noted in the beginning of the paper, a model describing this resonance process, i.e., the mechanism of simultaneous detachment of two electrons from an atom, is unavailable in the literature. Because of this, we are unable to give an unambiguous answer to the question: what is the degree of nonlinearity $K_{1e}^{(2+)}$ that should be observed in the experiment in the case of the two-electron mechanism of formation of doubly charged ions?

Nevertheless, the results of our study suggest that the two-electron mechanism of formation of Ba^{2+} ions is realised in the experiments. This is primarily supported by the fact that Ba^{2+} ions are efficiently formed at the laser radiation intensities when no saturation of multiphoton ionisation of Ba atoms is observed (at $F < F_s$), i.e., when the atomic concentration in the effective volume V_{eff} is considerably higher than the concentration of singly charged ions. This fact suggests that neutral atoms represent a target for the formation of Ba^{2+} ions. As noted above, in the case of the cascade mechanism, doubly charged ions are efficiently formed only upon the saturation of ionisation of atoms, when the concentration of singly charged ions in the effective volume V_{eff} is considerably higher than the atomic concentration [11, 13, 14].

Thus, the results of our study prove the two-electron mechanism of formation of Ba^{2+} ions upon nonlinear ionisation of atoms.

Using the dependences $\lg N_+(\lg F)$ and $\lg N_{2+}(\lg F)$, one can estimate the probability of the two-electron mechanism of formation of Ba^{2+} ions in the intensity region where no saturation of ionisation of Ba atoms is observed. The estimates made for the intensities $F = (1.3 - 2.1) \times 10^{29}$ cm^{-2} s^{-1} show that the probabilities of formation of Ba^+ and Ba^{2+} ions lie in the ranges $W_1 = 10^{6.9} - 10^{7.9}$ s^{-1} and $W_2 = 10^{4.1} - 10^{5.8}$ s^{-1}, respectively.

References

1. Suran V V, Zapesochnyi I P *Pis'ma Zh. Tekh. Fiz.* **1** 973 (1975)
2. Delone N B, Krainov V P, Suran V V *Laser Phys.* **2** 815 (1993)
3. Alesakhin I S, Zapesochnyi I P, Suran V V *Zh. Eksp. Teor. Fiz.* **76** 887 (1979)
4. Bondar' I I, Dudich M I, Suran V V *Zh. Eksp. Teor. Fiz.* **90** 1952 (1986)
5. Bondar I I, Delone N B, Dudich M I, Suran V V *J. Phys. B* **21** 2763 (1988)
6. Suran V V *Laser Phys.* **7** 665 (1997)
7. Suran V V *Laser Phys.* **7** 1220 (1997)
8. Delone N B, Bondar I I, Suran V V, Zon B A *Opt. Commun.* **40** 268 (1982)
9. Bernat T T, Bondar' I I, Suran V V *Opt. Spektrosk.* **71** 22 (1991)
10. Bondar' I I, Suran V V *Pis'ma Zh. Eksp. Teor. Fiz.* **68** 796 (1998)
11. Suran V V *Laser Phys.* **7** 637 (1997)
12. Bondar I I, Gomonay A I, Delone N B, Zapesochny I P, Suran V V *J. Phys. B* **17** 2049 (1984)
13. Feldmann D, Krautwold J, Chin S L, Hellfeld A, Welge K H *J. Phys. B* **15** 1663 (1982)
14. DiMauro L F, Dalwoo Kim, Courtney M W, Anselment M *Phys. Rev. A* **38** 2338 (1988)
15. Ammosow M V, Delone N B, Ivanov M Yu, Bondar I I, Masalov A V *Advances in Atomic, Molecular, and Optical Physics* **29** 33 (1992)