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Production of doubly charged ions in the ionisation of Ba atoms in two laser fields

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***Abstract*. The production of doubly charged ions is studied upon multiphoton ionisation of Ba atoms exposed simultaneously to two radiation fields: the fundamental radiation of a colour centre laser ($\omega = 8800 - 8880$ cm^{-1}) and its second harmonic. A two-electron mechanism was shown to be responsible for the production of these ions.**

Keywords: *multiphoton ionisation, doubly charged ions, two-electron mechanism, dynamic Stark effect.*

1. Introduction

Beginning with studies [1, 2], in which the production of doubly charged A^{2+} ions upon multiphoton ionisation of atoms was discovered for the first time, the mechanism of their production is extensively investigated. Previous studies showed that A^{2+} ions can be produced upon multiphoton ionisation of alkaline-earth elements (Sr and Ba atoms) by two mechanisms: a stepwise mechanism and a two-electron one. When the stepwise mechanism is realised, the A^{2+} ions are produced upon multiphoton ionisation of singly charged A^{+} ions produced by the same laser pulse. In other words, this A^{2+} ion production mechanism involves two stages: initially, the A^{+} ions are produced upon multiphoton ionisation of neutral A atoms and next the A^{2+} ions are produced upon multiphoton ionisation of the A^{+} ions. This mechanism is relatively simple, and its general features are now well understood (see, for instance, Refs [3 – 7]).

In the case of the two-electron mechanism, the A^{2+} ions are produced due to the simultaneous detachment of two electrons directly from neutral atoms [8 – 11]. This mechanism is more complicated and, unlike the stepwise mechanism, its special features have not been studied in detail. Some of these features were investigated in our earlier papers [10 – 18].

We have found in papers [16 – 18] that the production of A^{2+} ions by the two-electron mechanism is caused by ionisation of perturbed atoms. The resonance structure in the A^{2+} ion yield in this case is caused by excitation of strongly perturbed states of neutral atoms. The perturbation of the states is caused by the dynamic Stark effect under the conditions when their dynamic polarisabilities α are high in modulus.

The studies also showed that the realisation of one or the other of the two above mechanisms of the A^{2+} ion production depends on the spectral range of the laser radiation employed. In particular, upon multiphoton ionisation of atoms of alkaline-earth elements in the visible and UV spectral ranges, the stepwise mechanism dominates, whereas upon ionisation in the IR range, the two-electron mechanism plays a key role.

The fact that the two-electron A^{2+} ion production mechanism is realised in the IR and not in the visible range is explained as follows. A substantially larger number of photons is required for the multiphoton ionisation of alkaline-earth atoms (to produce the A^{+} ions) in the IR spectral range than for their ionisation in the visible range. Therefore, the probability of ionisation of atoms in the IR range is lower than in the visible range. The efficient ionisation of the atoms in the former case necessitates higher strengths ε of the laser radiation field. In this case, the conditions for a significant perturbation of the spectrum of neutral atoms are produced automatically.

As for the ionisation of the atoms in the visible spectral range, because the probability of this process is high, its saturation (the ionisation of all atoms in the interaction region) in one laser pulse begins much earlier than a significant perturbation of their spectrum can occur. Therefore, no neutral atoms will be found in the interaction region during the laser pulse when the conditions for the realisation of the two-electron mechanism are produced.

In this paper, we studied the production of doubly charged ions when neutral atoms are exposed to two laser fields – IR and visible.

2. Experimental

We studied Ba atoms, which were excited by a tunable colour centre laser (CCL) and by its second harmonic. The excitation was performed in the range $\omega_1 = 8800 - 8880$ cm^{-1} and the second harmonic (SH) range $2\omega_1 = 17600-17760$ cm^{-1}. The duration of both laser pulses was $\tau \approx 4 \times 10^{-8}$ s.

We used in the experiment one CCL pumped by an yttrium aluminium garnet laser. The fundamental CCL radiation was directed to a KDP crystal to be converted to the second harmonic. Both beams (the second harmonic and the CCL radiation transmitted through the KDP crystal) were made spatially coincident and were focused at the

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centre of the beam of Ba atoms. Both beams were linearly polarised, the angle between their light vectors was 45°. The CCL and SH radiation field strengths were $\varepsilon_1 \approx 3 \times 10^6$ V cm^{-1} and $\varepsilon_2 \approx 2 \times 10^5$ V cm^{-1}, respectively. Otherwise the experimental procedure did not differ from that typical for the investigations of excitation and ionisation in two laser radiation fields. It was described in detail in our paper [19].

The tuning range of the CCL was chosen to provide efficient excitation of Ba atoms. In particular, several frequencies ω_{nm} corresponding to the $5d6p\,^3P_1^0 \rightarrow 6p^2\,^3P_0$ ($\omega_{nm} =$ 8790 cm^{-1}), $6s5d\,^3D_2 \rightarrow 6s6p\,^1P_1^0$ ($\omega_{nm} = 8845$ cm^{-1}), and $5d6p\,^3P_2^0 \rightarrow 6p^2\,^3P_1$ ($\omega_{nm} = 8867$ cm^{-1}) single-photon transitions between the states n and m of atomic Ba fall within the tuning range of ω_1. It is known that upon irradiation of an atom at frequencies ω_{nm}, its spectrum should be strongly perturbed due to the dynamic Stark effect. This occurs because the dynamic polarisabilities $\alpha_{\rm n}$ and $\alpha_{\rm m}$ of the states n and m have high moduli at these frequencies.

As for the SH radiation, it was used to excite and ionise the states perturbed by the fundamental CCL radiation. Note that a part of these perturbed states could also be excited by the fundamental CCL radiation. However, the probability of excitation by the SH radiation is significantly higher, because their excitation by the SH radiation requires a smaller number of photons than their excitation by the fundamental CCL radiation. Under combined exposure to the CCL and SH radiation, Raman excitation is also possible. Excited in this case are those perturbed states which cannot be excited by exposing atomic Ba to only the CCL radiation or only the SH radiation.

Furthermore, the frequency of two-photon excitation of the unperturbed $6p^2\,^1D_2$ state (17672 cm^{-1}) also falls within the tuning range of the SH frequency. Therefore, different processes of level perturbation, as well as of excitation of these perturbed and unperturbed levels, can be realised under the combined exposure of atomic Ba to the CCL and SH radiation. The corresponding process are shown in Fig. 1.

It follows from the Introduction that the excitation of perturbed states should create the conditions for two-electron production of Ba^{2+} ions. As for the high probability of excitation and ionisation of the unperturbed state, it should cause a reduction in the number of neutral Ba atoms in the interaction region upon the ionisation saturation, resulting in a lowering of the Ba^{2+} yield in the case of two-electron production mechanism of doubly charged ions.

3. Experimental results

We measured the yields of Ba^+ and Ba^{2+} ions exposed to the CCL and SH radiation simultaneously and separately (Fig. 2). Note that the results of investigations of Ba^+ and Ba^{2+} ion production by exposing atomic Ba to only the CCL radiation and also to only the SH radiation were reported in our earlier papers [5, 6, 16, 17], so that we will not describe them in detail. Note only that to ionise atomic Ba by the CCL radiation and the SH, five and three photons should be absorbed, respectively. Under our experimental conditions, the ionisation of Ba atoms by the SH was saturated in the vicinity of the resonance peak in the Ba^+ ion yield. At other SH frequencies, the ionisation of Ba atoms was unsaturated, like the ionisation by the CCL radiation.

Consider first the results obtained for Ba^+ ions. One can see from Fig. 2 that the resonance peak in the ionisation of

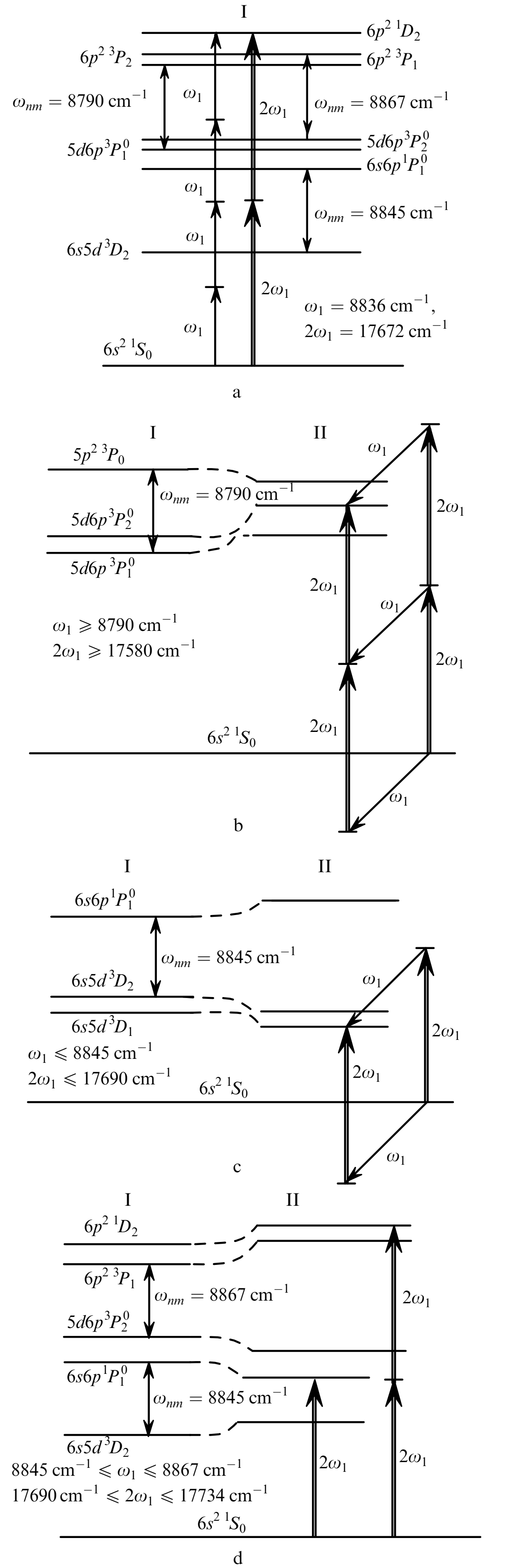

Figure 1. Schemes for excitation of Ba atoms in unperturbed (I) and perturbed (II) states. The dashed lines show the energy variation caused by the increase in the CCL radiation intensity.

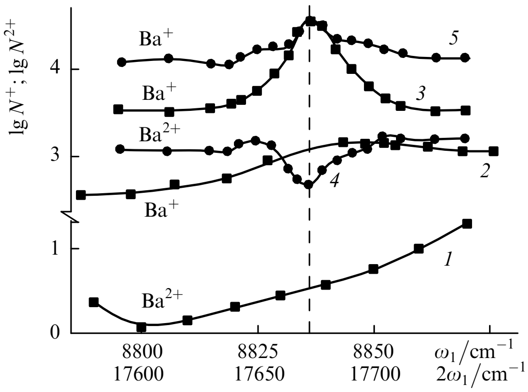

Figure 2. Frequency dependences of Ba^{2+} (*1*) and Ba^{+} (*2*) ion yields upon ionisation of barium atoms by the CCL radiation, of Ba^{+} ion yield upon ionisation of Ba atoms by the SH radiation (*3*), and of Ba^{2+} (*4*) and Ba^{+} (*5*) ion yields upon simultaneous ionisation by the CCL and SH. The vertical dashed line indicates the excitation frequencies of the unperturbed $6p^2\,{}^1D_2$ ($\omega_1 = 8836$ cm^{-1} and $2\omega_1 = 17672$ cm^{-1}).

Ba atoms by only the CCL radiation is asymmetric and is shifted relative to the resonance frequency $\omega_r = 8836$ cm^{-1}. This is caused by the excitation of perturbed states. A detailed description of the perturbation corresponding to this peak will be given below.

By contrast, the resonance peaks in the Ba^{+} ion yield, which are observed both upon irradiation by only the SH and upon simultaneous irradiation by the SH and CCL, are symmetric. Their frequencies ω_1 and ω_2 are independent of the irradiation type (the SH only or the SH and the CCL radiation together) and coincide exactly with the frequencies corresponding to the four- and two-photon excitation of the unperturbed $6p^2\,{}^1D_2$ state ($\omega_r = 8836$ cm^{-1} and $2\omega_r = 17672$ cm^{-1}).

It also follows from Fig. 2 that the Ba^{+} ion yield at the exact resonance is approximately the same both upon ionisation by only the SH radiation and upon ionisation by both beams, and is significantly higher than upon ionisation by the CCL radiation only. This means that the ionisation of atomic Ba upon simultaneous irradiation by CCL and SH beams in the range of resonance frequencies is primarily performed by the SH via the two-photon resonance with the unperturbed $6p^2\,{}^1D_2$ state.

As for the frequencies that differ from the resonance frequencies $\omega_r = 8836$ cm^{-1} and $2\omega_r = 17672$ cm^{-1}, the Ba^{+} ion yield in the case of simultaneous CCL and SH irradiation is significantly higher than in the case of individual irradiation by either of the two beams. This means that the ionisation of atomic Ba in this case results from the combined action of CCL and SH radiation. Our analysis shows that the Ba^{+} ion production in this case is not caused by direct ionisation: when the difference of direct ionisation probabilities for two different radiation beams is large, the ionisation under their combined action should be performed primarily by the radiation for which the probability is higher. In particular, this effect was observed in our paper [19] upon ionisation of atomic Ba exposed to two laser fields.

Fig. 2 implies that the Ba ion yield away from the resonances ($\omega_1 < 8836$ cm^{-1}, $2\omega_1 < 17672$ cm^{-1}) upon ionisation by only the SH is substantially higher than upon ionisation by only the CCL radiation. In other words, the probability of three-photon ionisation of Ba atoms by the SH far exceeds the probability of five-photon ionisation by the CCL radiation. Therefore, if the ionisation of Ba atoms upon simultaneous irradiation by the CCL and SH were direct in the above frequency ranges, the Ba^{+} ion yield would be the same as upon ionisation by the SH only.

However, as noted above, the Ba^{+} ion yield in this case is significantly higher than upon ionisation by not only the CCL radiation, but also by the SH. Therefore, the Ba^{+} ion yield upon simultaneous irradiation by two laser beams in the frequency range $\omega_1 \neq 8836$ cm^{-1} and $2\omega_1 \neq 17672$ cm^{-1} is caused by the resonance ionisation. However, the unperturbed states cannot be excited when Ba atoms are exposed to the combined action of two laser beams with frequencies $\omega_1 \neq 8836$ cm^{-1} and $2\omega_1 \neq 17672$ cm^{-1}. At the same time, one can see from Fig. 1 that in this case, both Raman excitation and conventional two-photon excitation of perturbed states of atomic Ba can occur.

Therefore, Ba^{+} ions are produced upon simultaneous irradiation by two beams with frequencies $\omega_1 \neq 8836$ cm^{-1} and $2\omega_1 \neq 17672$ cm^{-1} due to excitation and subsequent ionisation of perturbed atomic Ba states. Note that different excitation processes shown in Fig. 1 are dominant (give the highest Ba^{+} ion yield) at different frequencies in the spectral region employed. However, because all these processes involve the absorption of a small number of photons, their probability is relatively high and the corresponding ionisation processes are saturated.

Note another feature of the perturbation and excitation of the states of atomic Ba occurring in the case under study. As a rule the Stark shift is equal to or greater than the difference between the energy of the unperturbed state and the energy of the neighbouring unperturbed state. Consider, for example, the state perturbation in the region $\omega_1 < \omega_{nm} = 8867$ cm^{-1} (Fig. 1d). The frequency $\omega_{nm} = 8867$ cm^{-1} corresponds to the $5d6p^3P_2^0 \to 6p^2\,{}^3P_1$ single-photon transition. The dynamic polarisabilities of the $5d6p^3P_2^0$ and $6p^2\,{}^3P_1$ states are large in modulus at this frequency, the dynamic polarisability of the $5d6p^3P_2^0$ state being positive and that of the $6p^2\,{}^3P_1$ state being negative. Due to the Stark shift, the $6p^2\,{}^3P_1$ state will approach the $6p^2\,{}^1D_2$ state.

Note that the frequencies of CCL radiation employed are significantly different from the frequencies ω_{nm} corresponding to single-photon transitions from the $6p^2\,{}^1D_2$ state. For this reason, the dynamic polarisability of the $6p^2\,{}^1D_2$ state should be significantly lower than that of the $6p^2\,{}^3P_1$ state in the spectral range of CCL radiation employed. Therefore, the energy of the $6p^2\,{}^3P_1$ state will approach that of the $6p^2\,{}^1D_2$ state due to the Stark shift, their energies should become equal for some intensity of the CCL radiation.

This case was described in detail in Ref. [20]. After the approach of the two states for which $\Delta J \leqslant 2$ (ΔJ is the difference of the total angular momenta corresponding to these states), the energy of each of them becomes approximately equal to the energy of the neighbouring state it would have in the absence of the interaction between the states. In particular, after the approach of the $6p^2\,{}^3P_1$ state to the $6p^2\,{}^1D_2$ state, the $6p^2\,{}^1D_2$ state begins to shift. This state is excited in the range $2\omega_1 < 2\omega_{nm} = 17734$ cm^{-1}.

Our estimates show that the above-described perturbation of the $6p^2\,{}^3P_1$ and $6p^2\,{}^1D_2$ states can be produced under our experimental conditions when the dynamic polarisability α of the $6p^2\,{}^3P_1$ state is $\sim 10^4$ au. This is quite a realistic dynamic polarisability for the states in the vicinity of the resonance frequencies ω_{nm} (see, for instance, Ref. [21]), so that the perturbation described above is quite possible under our experiment conditions.

Similar perturbations take place in other cases (see Fig. 1). The change in the level energy with increasing the CCL radiation intensity is shown schematically by the dashed lines in Fig. 1. Note that such perturbation of the $6p^2\,^1D_2$ state also occurs upon ionisation of Ba atoms by the CCL radiation only. This, in particular, accounts for the asymmetry of the corresponding resonance in the yield of Ba^+ ions (see Fig. 2). In this case, the perturbed $6p^2\,^1D_2$ state is excited due to absorption of four CCL radiation photons. Upon simultaneous irradiation by the CCL and SH, the $6p^2\,^1D_2$ state is excited due to absorption of two SH photons. Naturally, the excitation probability is significantly higher in the latter case than in the former one. This accounts for a substantial increase in the Ba^+ ion yield upon simultaneous ionisation by the CCL and SH compared to the yield obtained upon excitation by the CCL laser only.

Note that the type of perturbation of the remaining states, which correspond to the cases presented in Fig. 1, is also the same for simultaneous irradiation by the CCL and SH and excitation by the CCL laser only. However, selection rules allow the excitation of these perturbed states from the ground state of atomic Ba exposed to two radiation beams and forbid it in the case of single-beam irradiation. This also accounts for the strong rise in the Ba^+ ion yield in the corresponding frequency ranges upon simultaneous irradiation by the CCL and SH compared to the yield of these ions upon exposure to the CCL radiation only.

Therefore, the multiphoton excitation of Ba atoms and the subsequent ionisation of several of its excited states, and also of the unperturbed $6p^2\,^1D_2$ state in the vicinity of the $\omega_1 = 8836$ cm^{-1} ($2\omega_1 = 17672$ cm^{-1}) frequency, was realised in our conditions under simultaneous exposure of Ba atoms to the CCL and SH radiation.

Consider now the production of doubly charged ions. The results of this study are presented in Fig. 2. One can see that the simultaneous exposure of the atomic beam to the CCL and SH radiation results, throughout the frequency range employed, in a significantly higher (by about a factor of $10^2 - 10^3$) Ba^{2+} ion yield than in the case of separate irradiation. Note that Fig. 2 does not show the results of investigation of Ba^{2+} ion production upon ionisation of Ba atoms by the SH, because their yield did not exceed the sensitivity limit of the recording system.

The investigation of Ba^{2+} ion production upon ionisation of Ba atoms by only the CCL radiation and only the SH radiation was described in detail in our papers [5, 6, 16, 17] and hence will not be analysed here. Note only that, as shown in papers [5, 6], upon ionisation of atomic Ba in the frequency range corresponding to the SH tuning range employed in our work, the Ba^{2+} ions are produced by the stepwise mechanism. Upon ionisation by the CCL radiation only [16, 17], the Ba^{2+} ions are produced by the two-electron mechanism. In this case, as shown in Refs [16, 17], the Ba^{2+} ions are produced due to excitation and ionisation of perturbed states of neutral Ba atoms.

An analysis of the results obtained in our work shows that the Ba^{2+} ions are also produced upon simultaneous irradiation of Ba atoms by the CCL and SH due to the two-electron mechanism. In this case, the Ba^{2+} ion production results from the excitation and subsequent ionisation of perturbed states of neutral Ba atoms. This is confirmed by two facts.

First, a higher probability of the excitation of strongly perturbed states of atomic Ba was obtained upon simultaneous irradiation by the CCL and SH throughout the frequency range employed, and throughout this range the Ba^{2+} ion yield was far greater than the yield upon separate irradiation.

Second, as follows from Fig. 2, in the vicinity of the frequencies $\omega_1 = 8836$ cm^{-1} and $2\omega_1 = 17672$ cm^{-1} a dip in the Ba^{2+} ion yield is observed, whose position coincides exactly with that of the resonance peak in the Ba^+ ion yield. As shown above, this peak appears due to the excitation of the unperturbed $6p^2\,^1D_2$ state by the SH. This means that the high excitation probability for unperturbed states does not result in the enhancement of the Ba^{2+} ion yield.

Perturbed states can also be excited in the frequency range of the dip mentioned above. In particular, the frequency $\omega_{nm} = 8845$ cm^{-1} corresponding to the $6s5d^3\,D_2 \to 6s6p^1\,P_1^0$ one-photon transition also falls within this range. The type of levels perturbation in this range is the same as in the case of the $6p^2\,^3P_1$ level perturbation described above (see Figs 1b and 1c). The excitation probabilities for these perturbed states should also be high, because they are associated with the absorption of a small number of photons. Nevertheless, the Ba^{2+} ion yield obtained upon simultaneous irradiation by the CCL and SH in the above frequency range is lower than the yield observed upon the same simultaneous irradiation in other regions of the spectrum employed.

This effect is explained as follows. The probability of the three-photon ionisation of Ba atoms by the SH radiation via the two-photon resonance with the unperturbed $6p^2\,^1D_2$ state is relatively high under our conditions. Due to the high ionisation probability, the saturation begins in a time period during the laser pulse during which the CCL radiation field strength does not become high enough to significantly perturb the corresponding states. In other words, when the conditions exist for a significant perturbation of the corresponding levels of atomic Ba by the CCL radiation, the neutral atoms themselves will no longer be found in the interaction region upon simultaneous irradiation by the CCL and SH at frequencies $\omega_1 = 8836$ cm^{-1} and $2\omega_1 = 17672$ cm^{-1}. Because doubly charged ions are produced due to excitation and ionisation of the perturbed states of neutral atoms, the ionisation saturation described above should result in a decrease of the yield of Ba^{2+} ions compared to their yield in the vicinity of other frequencies of excitation and ionisation of the perturbed states of atomic Ba.

Therefore, doubly charged Ba^{2+} ions are produced upon simultaneous irradiation by the CCL and SH due to excitation and ionisation of the strongly perturbed states of neutral Ba atoms, i.e., via the two-electron mechanism.

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