DOUBLY-CHARGED IONS FORMATION DURING MULTIPHOTON IONISATION OF Ba ATOMS

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The results of measurements of the yield of singly and doubly charged ions against laser radiation frequency in non-linear ionisation of Ba atoms are presented. It was shown that in the investigated spectral range (17700-18700 cm^{-1}) doubly charged Ba ions are formed in result of stepwise mechanism realization.

This work continues the cycle of our studies of a process of non-linear ionisation of alkaline-earth atoms. One of the main problems, which are solved in studying the non-linear ionisation of alkaline-earth atoms, is the analysis of the mechanism of doubly charged ion formation [1]. Two mechanisms have been proposed for explaining this process, namely the two-electron one (in which the doubly charged ions are formed directly from neutral atoms) and step-wise one (in which the doubly charged ions are formed from singly charged ions formed in the same laser radiation pulse). We present the results of experimental studies of multiphoton ionisation of Ba atoms within 17700-18700 cm^{-1} spectral range and discuss the mechanism of doubly charged ion formation.

The experimental procedure was typical for studying the non-linear ionisation of atoms: the laser beam crossed the beam of neutral atoms; the ions formed in the beams' interaction region were pulled in by constant field and analized by a time-of-flight mass-spectrometer. Experimental techniques are not described here, since the details are given in earlier papers [2, 3]. We used the radiation of a dye laser with the dispersion resonator for ionisation of Ba atoms. The dye was pumped by the second harmonic of the Nd-glass laser. Rhodamine-110 was used as dye. The laser generation varied within 17700–18700 cm^{-1} spectral range. The laser generation spectrum width was 2–3 cm^{-1}. The laser pulse duration was $\tau \cong 5\times10^{-8}$ s. We used the linearly polarized laser radiation.

Experimental results

We have measured the yield of singly charged and doubly charged ions as the functions of laser radiation frequency ω for fixed values of field strength $\varepsilon \cong 2\times10^{6}$ V/cm (the radiation intensity $F = 3.5\times10^{28}$ $cm^{-2}s^{-1}$). The yields of singly and doubly charged ions as functions of laser radiation frequency are shown in Fig.1.

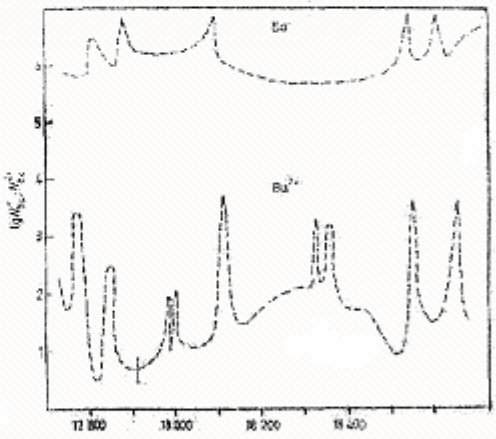

Figure 1. Singly charged (N^{+}) and doubly charged (N^{2+}) ion yield against laser radiation frequency in the ionization of Ba atoms

The procedure for measurements of the dependencies shown in Fig.1 was as follows. For fixed values of laser radiation and intensity several measurements (near five) of ion signal amplitudes were carried out and then the mean values of amplitudes were found from these measurements. The radiation frequency varied with a step of 3–4 cm^{-1} in the intraresonance intervals of de-

pendences and 2–3 cm^{-1} in the region of resonance maxima. The accuracy of determination of radiation frequency was 3 cm^{-1}. We must note that the sensitivity of recording equipment was as much as approximately two times higher for doubly charged ions than for singly charged ones.

As one can see from Fig.1, both $N^{2+}(\omega)$ and $N^{+}(\omega)$ dependences have resonance maxima. The investigations have shown that when the field strength changes slightly the positions of the resonance maxima in these dependencies do not change. This fact allows disregarding the perturbation of the levels of atoms and ions when the data obtained are interpreted.

The formation of singly charged ions will be considered in this paper as additional information for explaining the doubly charged ion formation mechanism only. Therefore, the peculiarities of the singly charged ion formation will not be considered here in detail.

For the removal of one electron from Ba atom three photons must be absorbed. The identification of resonance maxima done on the basis of available data about the bound and autoionising states of Ba atom [4,5] in the $N^{+}(\omega)$ dependence is given in Table 1. As one can see from Table 1 all resonance maxima, except one at the frequency 18620 cm^{-1}, are identified by two photon transitions into the bound atomic states. The nature of this unidentified maximum is unknown yet.

Table 1. Identification of resonance maxima in the $N^{+}(\omega)$ dependence. ω_r is the frequency at which a resonance maximun in the $N^{+}(\omega)$ dependence exists; ω_t is the frequency corresponding to the resonance transition in the Ba atom, which identifies this maximum.

ω_r, cm^{-1}	ω_t, cm^{-1}	Resonance transitions in the Ba atom
17810	17808	$6s^2\,{}^1S_0 + 2\hbar\omega \rightarrow 6p^2\,{}^3P_2$
17880	17881	$6s^2\,{}^1S_0 + 2\hbar\omega \rightarrow 6s7d\,{}^3D_2$
18100	18100	$6s^2\,{}^1S_0 + 2\hbar\omega \rightarrow 5d6d\,{}^3D_2$
18545	18544	$6s^2\,{}^1S_0 + 2\hbar\omega \rightarrow 5d6p\,{}^3F_2$
18620	—	—

Let us consider now the results obtained on our study of the $N^{2+}(\omega)$ dependence. The identification of resonance maxima was provided in the spectrum of neutral atom BaI and singly charged ion BaII and is given in Table 2. This identification was performed on the basis of existing data on spectra BaI and BaII [4,5]. All resonance maxima are identified by transition in the spectrum of BaII. What give these results from the viewpoint of the doubly charged ions formation?

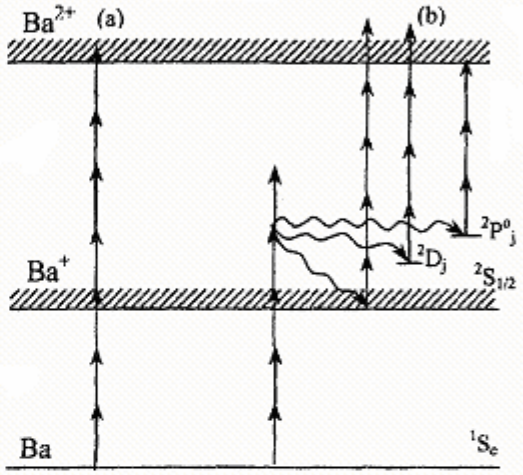

Fig. 2. Implementation schemes for the two-electron (a) and stepwise (b) mechanism of doubly charged ion formation in the ionization of Ba atoms.

As has been already noted, to explain the doubly charged ion formation two mechanisms were considered: the two-electron and stepwise mechanisms. Their implementation schemes are shown in Fig. 2. In the model of two-electron mechanism of doubly charged ions formation the resonance maxima in the $N^{2+}(\omega)$ dependence must be caused by the spectrum of neutral atom – its two-electron bound and autoinising states, as doubly charged ions in this case are formed directly from neutral atoms. In the case of stepwise mechanism the formation of doubly charged ions occurs in two stages: first, singly charged ions are formed and then, in result of multiphoton ionisation of singly charged ions, doubly charged ions form. In this case resonance maxima in the

$N^{2+}(\omega)$ dependence must be caused by a growing concentration of singly charged ions (these maxima in the $N^{2+}(\omega)$ dependence must coincide in frequency with maxima in the $N^{+}(\omega)$ dependence), as well as by resonance transitions in the spectrum of singly charged ion BaII.

However, the maxima in the $N^{2+}(\omega)$ dependence do not occur at any frequency at which there exist resonance maxima in the $N^{+}(\omega)$ dependence. In some cases this circumstance may be explained as follows. As noted above, in the conditions of our experiment the saturation takes place in the yield of singly charged ions. The yield of singly charged ions in the region of resonance frequencies increases in these conditions due to increasing the interaction volume. Since the degree of non-linearity of doubly charged ions formation from singly charged ones is greater than the degree of non-linearity of singly charged ion formation, then due to inhomogeneity in the distribution of laser radiation in the region of the beams interaction, the formation of doubly charged ions occurs in the volume which is less than the volume in which singly charged ions are formed.

Table 2. Identification of resonance maxima in the $N^{2+}(\omega)$ dependence. ω_r is the frequency at which a resonance maximun in the $N^{2+}(\omega)$ dependence exists; ω_t is the frequency corresponding to the resonance transition in the BaII spectrum, which identifies this maximum.

ω_r, cm^{-1}	Γ, cm^{-1}	ω_t, cm^{-1}	Resonance transitions in the Ba^+ ion
17765	10	17764	$6p\ ^2P^o_{3/2} + 3\hbar\omega \rightarrow 9g\ ^2G_{7/2,\,9/2}$
		17768	$5d\ ^2D_{3/2} + 4\hbar\omega \rightarrow 12d\ ^2D_{3/2,\,5/2}$
17850	15	17839	$6p\ ^2P^o_{3/2} + 2\hbar\omega \rightarrow 5f\ ^2F^o_{7/2}$
		17843	$5d\ ^2D_{5/2} + 4\hbar\omega \rightarrow 11g\ ^2G^o_{7/2,\,9/2}$
		17844	$6p\ ^2P^o_{1/2} + 3\hbar\omega \rightarrow 8g\ ^2G_{7/2}$
		17852	$5d\ ^2D_{3/2} + 4\hbar\omega \rightarrow 10g\ ^2G_{7/2}$
17990	5	17988	$5d\ ^2D_{5/2} + 4\hbar\omega \rightarrow 12g\ ^2G_{7/2,\,9/2}$
18000	5	17998	$6p\ ^2P^o_{3/2} + 3\hbar\omega \rightarrow 11d\ ^2D_{3/2,\,5/2}$
18110	10	18109	$6p\ ^2P^o_{3/2} + 3\hbar\omega \rightarrow 10g\ ^2G_{7/2,\,9/2}$
18330	10	18327	$6p\ ^2P^o_{1/2} + 3\hbar\omega \rightarrow 9g\ ^2G_{7/2,\,9/2}$
18360	10	18364	$6p\ ^2P^o_{3/2} + 3\hbar\omega \rightarrow 11g\ ^2G_{7/2,\,9/2}$
18560	15	18561	$6p\ ^2P^o_{1/2} + 3\hbar\omega \rightarrow 12d\ ^2D_{3/2}$
		18564	$6p\ ^2P^o_{1/2} + 2\hbar\omega \rightarrow 5f\ ^2F_{5/2}$
18670	5	18673	$6p\ ^2P^o_{1/2} + 3\hbar\omega \rightarrow 10g\ ^2G_{7/2}$

But as one can see from Table 2, all resonance maxima in the $N^{2+}(\omega)$ dependence are identified in BaII spectrum. As one can see from the Table 2 we observed resonance maxima from excited $6p\ ^2P^0_j$ and $5d\ ^2D_j$ states. The stepwise mechanism implementation scheme (see figure 2) presents the cases of doubly charged ion formation from singly charged ions which are both in the ground and in the first excited states. The formation of Ba^+ ions in the excited $6p\ ^2P^0_j$ and $5d\ ^2D_j$ states may occur as a result of decay of autoionising states excited in result of the above-threshold absorption of photons. We must note that the concentration of singly charged ions in the excited $6p\ ^2P^0_j$ and $5d\ ^2D_j$ states may be comparable to the concentration of the same ions in the ground $6s\ ^2S_{1/2}$ state.

Thus, the resonance structure of the $N^{2+}(\omega)$ dependence, obtained in the ionisation of the Ba atom throughout the spectral

range studied may well be explained in the model of stepwise mechanism of doubly charged ions formation.

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УТВОРЕННЯ ДВОЗАРЯДНИХ ІОНІВ ПРИ БАГАТОФОТОННІЙ ІОНІЗАЦІЇ АТОМІВ Ba

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Наведено результати вимірювань залежностей виходу одно- і двозарядних іонів від частоти лазера при нелінійній іонізації атомів Ba. Показано, що в досліджуваній області спектру (17700-18700 $см^{-1}$) двозарядні іони Ba утворюються в результаті реалізації каскадного механізму.