Formation of singly and doubly charged ions by nonlinear laser-induced strontium and barium atom ionization at frequencies 16800–18000 cm^{-1}

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Investigations of the yields of singly and doubly charged ions produced by ionization of Sr and Ba by dye-laser radiation are reported. The frequency dependences of the yields of singly charged Sr and Ba ions reveal resonance maxima due to two-photon excitation of a number of singlet and triplet one- and two-electron states. The probability of two-photon excitation of triplet and singlet states are of the same order of magnitude. The frequency dependences of the strontium and barium doubly-charged ion yields have a large number of resonance maxima that are readily identified in the spectra of the singly charged Sr^+ and Ba^+ ions. This points to realization of a cascade mechanism of doubly-charged ion production. At the same time, the frequency dependences of the yield of doubly charged ions reveal facts that cannot be unambiguously explained by the cascade mechanism.

1. INTRODUCTION

Multiphoton ionization of Sr and Ba atoms was previously investigated in many experiments,[1-13] which revealed formation of both singly and multiply charged ions of these atoms. A common feature of such experiments was the relatively narrow range of the laser emission frequency (~ 100 cm^{-1}). The aggregate of the presently available experimental data does not lead to unambiguous conclusions concerning the mechanism whereby the doubly charged ions are produced. Thus, some of the experimental results point to the feasibility of a cascade mechanism,[11,12] whereas others indicate realization of a direct two-electron mechanism—simultaneous detachment of two electrons from the atom.[1-6] One possibility of explaining the formation of doubly charged ions is to lengthen the time during which the emission frequency is varied and observing thereby a larger number of resonance maxima. Another way is to perform the experiments at the lowest possible field intensity, when the identification of the resonance maxima is most reliable. We have attempted to implement this program in the present study.

2. EXPERIMENT

The general experimental procedure was conventional, viz., intersection of an atom beam with a laser beam. The ions produced in the focusing region were drawn out by a dc field and subsequently analyzed with a time-of-flight mass spectrometer. The ionization was produced by a dye laser with a dispersive cavity. The dye was pumped by the second harmonic of a neodymium laser. The dye used was rhodamine-6G (yellow). The dispersing elements were three prisms of heavy optical glass with large refractive indices. The emission frequency was varied in the range 16800–18000 cm^{-1}. The frequency-variation interval in these experiments was thus appreciably larger than in the preceding studies.[1-13] The emission-spectrum with was $\Delta\omega \approx 6$ cm^{-1}. Note that the detachment of one electron from an Sr or Ba atom calls for absorption of three laser photons, detachment of a second electron requires five photons, and simultaneous detachment of two electrons—eight photons. The laser operated on one transverse and many longitudinal modes. A Glan-Foucault prism was used to measure the linear polarization of the radiation. The laser beam was focused into the center of the atom beam by a long-focus lens. The experimental data were obtained at a field intensity not higher than $\varepsilon \approx 2\cdot 10^6$ V/cm.

3. RESULTS

We measured the frequency dependences of the yields (N_i^+, N_i^{2+}) of singly and doubly charged Sr and Ba ions obtained by using linearly polarized radiation. Note that singly and doubly charged ions were produced in each laser pulse, but the number of doubly charged ions was always lower than that of singly charged. For an unambiguous interpretation of the data, the dependences of N_i^+ and N_i^{2+} on the frequency were measured therefore at various emission intensities. Thus, for example, the $N_i^+(\omega)$ dependences were recorded at low field strengths ($\varepsilon \approx 10^5$ V/cm) to prevent saturation of the yield of the singly charged ions. The frequency dependences of $N_i^{2+}(\omega)$ were measured in stronger fields ($\varepsilon \approx 10^6$ V/cm). In this case saturation was observed for the singly charged ions.

The experimental results are shown in Figs. 1–4. Each point on a curve is the average of several (~ 10) measurements of the ion-signal amplitudes.

1. Formation of singly charged ions

The experimental data obtained for singly charged ions in fields $\varepsilon \approx 10^5$ V/cm are shown in Figs. 1 and 2. The plots of the singly charged strontium- and barium-ion yields vs frequency show many resonance maxima. The order of magnitude of their widths is determined by the width $\Gamma = K^{1/2}\Delta\omega$ of the laser-emission spectrum, where K is the number of photons whose absorption causes the resonance. Estimates

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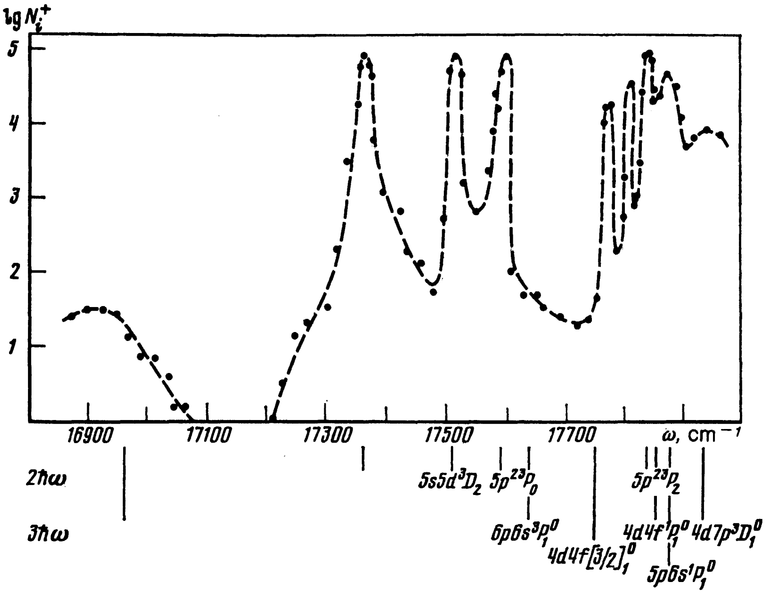

FIG. 1. Frequency dependence of three-photon ionization of strontium atoms. The first and second maxima correspond to the states $4d\,6p\,^1P_1^0$ and $5s5d\,^1D_2$.

show that the low field strength at which these data were obtained permit the identification to be carried out disregarding the laser-emission field. Figures 1 and 2 show the bound and autoionizing states of the Sr and Ba atoms, with which two- and three-photon resonances are allowed by the selection rules,[14] and indicate also the emission frequencies corresponding to these transitions. The energies of these states are known from Refs. 15–17. The resonance transitions in the Sr and Ba spectra, which cause the resonance maxima in the frequency dependences of the yields of doubly charged strontium and barium ions, are listed in Tables I and II. These tables show that all the identified resonance maxima are due only to two-photon excitation of one- and two-electron bound states. We note that resonance maxima in the ionization of the Sr atom in the frequency interval 17300–17900 cm^{-3} were observed and identified earlier.[13] The resonance maxima observed by us in this frequency region, and their identification, agree with the results of Ref. 13. The most interesting fact is that most resonance maxima are due to photon transitions with change of spin (intercombination transitions). The probabilities of two-photon transitions with and without change of spin are approximately equal.

As to the Sr and Ba autoionizing states marked in Figs. 1 and 2, with which three-photon resonances are possible, no corresponding maxima are observed in the $N_i^+(\omega)$ plots. In the case of the barium-atom ionization this may be due to the large width of the autoionizing states.[16] When the Sr atom is ionized, however, both wide and relatively narrow autoionizing states can be produced.[16] The possible cause of the absence of resonances with autoionizing states may be the strong broadening of these states by the radiation field, as observed previously.[18]

It is noteworthy that for three-photon ionization of the Sr atom, the $N_i^+(\omega)$ plot has only three maxima, at the frequencies $\omega = 17\,772$, $17\,805$ and $17\,765$ cm^{-1}, which cannot be unambiguously identified. It was suggested in Ref. 13 that these maxima can be due to three-photon excitation of autoionizing states with total angular momentum $J = 3$.

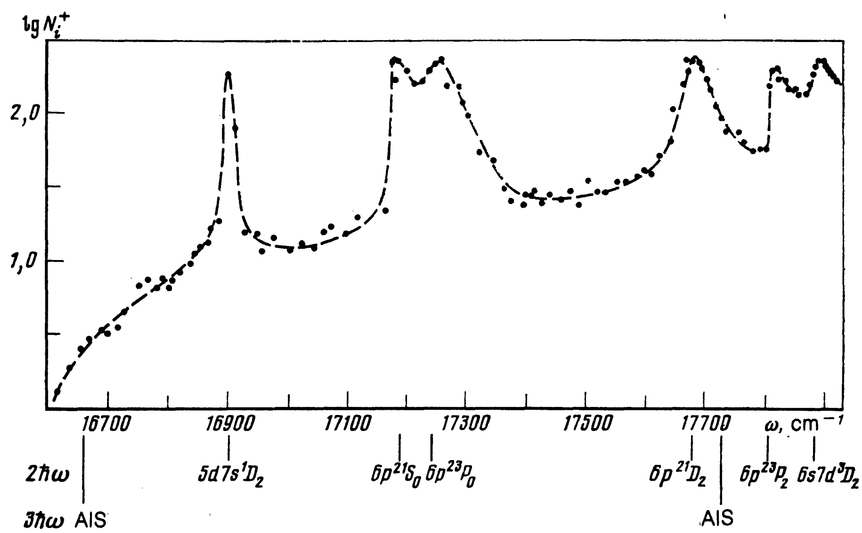

FIG. 2. Frequency dependence of three-photon ionization of barium atoms. AIS stands for the Ba-atom autoionizing states, for which only the energies and widths, but not the configuration, are known.[17]

TABLE I. Identification of resonance maxima in the frequency dependence of the yield of singly charged Sr^+ ions.

ω_r, cm^{-1}	ω_{tr}, cm^{-1}	Resonance transition
17 363	17 363	$5s^2\,{}^1S_0+2\hbar\omega \to 5s5d\,{}^1D_2$
17 511	17 511	$5s^2\,{}^1S_0+2\hbar\omega \to 5s5d\,{}^3D_2$
17 596	17 596	$5s^2\,{}^1S_0+2\hbar\omega \to 5p^2\,{}^3P_0$
17 768	–	Not identified
17 800	–	»
17 837	17 837	$5s^2\,{}^1S_0+2\hbar\omega \to 5p^2\,{}^3P_2$
17 864	–	Not identified

Note: ω_r—emission frequencies corresponding to maxima in the $N_i^+(\omega)$ plots, Fig. 1; ω_{tr}—emission frequencies corresponding in the spectrum of the Sr atom to the resonance transitions that cause the maxima of the $N_i^+(\omega)$ dependence.

The energies of these states, however, are unknown at present. The question of the nature of the observed resonances in the yield of the Sr^+ ions at the indicated frequencies remains therefore open.

Summarizing the results of the investigations of the formation of Sr^+ and Ba^+ ions, we note that the major role is played by the two-photon transitions with change of spin, observed in the spectra of the Sr and Ba atoms. The probabilities of such transitions are of the same order as those of transitions without change of spin. We note that an investigation of the ionization of Ba and Ca atoms by the second harmonic of a neodymium laser also revealed a high probability of two-photon intercombination transitions, whereas neither four- nor five-photon intercombination transitions were observed for these atoms.[6,9,20] It appears that the high probability of the two-photon intercombination transitions is a distinctive feature of the multiphoton ionization of all alkaline-earth atoms.

It should be noted that single-photon intercombination transitions in alkaline-earth atoms are a known fact. The probability of these transitions, however, is significantly lower than that of transitions without change of spin.[21,22] Thus, for example, the probabilities of the one-photon transitions $ns^2\,{}^1S_0 - nsnp\,{}^3P_1^0$ are smaller by approximately 150 times inthe case of the Ba atom, and by 1700 times in the case of the Sr atom, than the probabilities of the one-photon transitions $ns^2\,{}^1S_0 - nsnp\,{}^1P_1^0$.[21] This raises the question of the cause of the high probability of two-photon intercombination transitions. It may be that the strong field leads to a substantial weakening of the *LS* bond and, by the same token, lifts the spin hindrance. An unambiguous answer to this question calls for further theoretical and experimental research.

The investigations of the production of Sr^+ and Ba^+ show that a theoretical description of multiphoton ionization of alkaline-earth metals must include intercombination transitions along with the allowed ones.

2. Formation of doubly charged ions

The experimental data for doubly charged ions are shown in Figs. 3 and 4 together with the results for singly charged ions. For a reliable observation of doubly charged ions, the measurements were made at a field strength $\varepsilon \sim 2\cdot 10^6$ V/cm. In this case the yield of the singly charged ions saturates. The positions of the resonance maxima in the yields of Sr^+ and Ba^+ remain unchanged when the field strength is increased from 10^5 (Figs. 1, 2) to $2\cdot 10^6$ V/cm (Figs. 3, 4), and accordingly the identification of these maxima remains likewise unchanged.

The frequency dependences of the doubly charged ions Sr^{2+} and Ba^{2+} have, just as in the case of singly charged ions, clearly pronounced resonance maxima. In no case do these maxima occur at the same frequencies as the maxima of the singly charged ion yields.

The frequency dependences of the yield of doubly charged Sr^{2+} ions were investigated earlier in the ranges $\omega = 17760$–17900 cm^{-1} (Ref. 11) and $\omega = 17640$–17900 cm^{-1} (Ref. 12). Note that our results agree well with the results of these references.

We examine now the results shown in Figs. 3 and 4 from the viewpoint of production of the doubly charged ions by two mechanisms, two-electron and cascade. In the two-elec-

TABLE II. Identification of resonance maxima in the frequency dependence of the yield of singly charged Ba^+ ions.

ω_r, cm^{-1}	ω_{tr}, cm^{-1}	Resonance transition
16 897	16 897	$6s^2\,{}^1S_0+2\hbar\omega \to 5d7s\,{}^1D_2$
17 185	17 185	$6s^2\,{}^1S_0+2\hbar\omega \to 6p^2\,{}^1S_0$
17 246	17 246	$6s^2\,{}^1S_0+2\hbar\omega \to 6p^2\,{}^3P_0$
17 672	17 672	$6s^2\,{}^1S_0+2\hbar\omega \to 6p^2\,{}^1D_2$
17 808	17 808	$6s^2\,{}^1S_0+2\hbar\omega \to 6p^2\,{}^3P_2$
17 881	17 881	$6s^2\,{}^1S_0+2\hbar\omega \to 6s7d\,{}^3D_2$

Note: ω_r and ω_{tr} are the same as in the note of Table I, but for the barium-ion yield (Fig. 2).

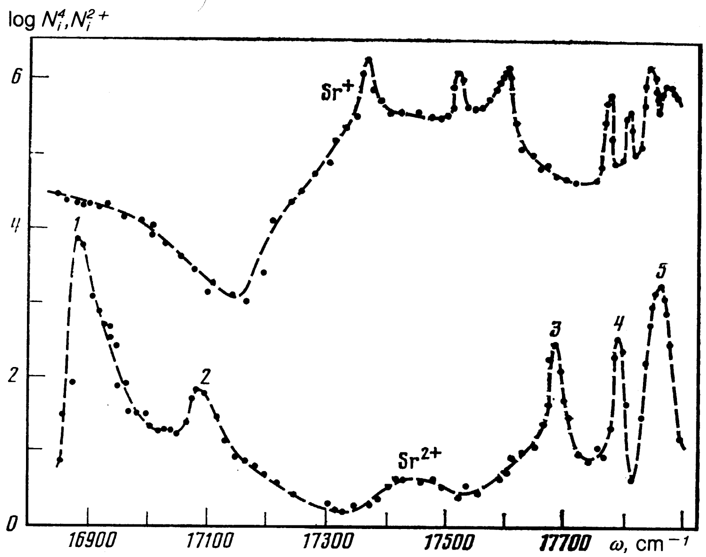

FIG. 3. Frequency dependences of the yields of single and double ionization of the Sr atom.

tron mechanism, two electrons are simultaneously detached from the neutral atom. The resonance maxima can be due in this case to two-electron bound or autoionization states of the atom. Recall that when Sr and Ba atoms are ionized at frequencies $\omega = 16800$–$18000\ \mathrm{cm}^{-1}$ multiphoton excitation is possible of a number of two-electron bound and autoionization states (the frequencies of the transitions to these states are; marked on Figs. 1 and 2). Analysis has shown that the resonance frequency is equal to the three-photon-excitation frequency of the autoionizing state $4d\,4f\,^1P_1^0$ only for the Sr^{2+}-yield resonance maximum numbered 5 in Fig. 3. The state $4d\,4f\,^1P_1^0$, however cannot be responsible for this resonance maximum since, as shown in Ref. 12, this maximum of the $N_i^{2+}(\omega)$ maximum is preserved also for circular polarization of the radiation, whereas three-photon excitation of this state in a circular field is forbidden by the selection rules. The resonance maxima observed on the $N_i^{2+}(\omega)$ plots are thus due to neither two-electron bound states nor autoionizing states of the Sr and Ba atoms. This precludes the feasibility of the two-electron mechanism of formation of doubly charged ions.

We consider now the possibility of realizing the cascade mechanism of formation of doubly charged ions. In this case, doubly charged ions A^{2+} are the result of multiphoton ionization of singly charged ions A^+. The resonance maxima observed in the frequency dependences of doubly charged ion formation can be due in this case to the onset of resonances in the spectrum of the singly charged ions. Furthermore, in individual cases the $N_i^{2+}(\omega)$ plots can have also maxima due to the resonant increase of the probability of formation of singly charged ions, and can therefore occur at the same frequency as the maxima of ion yield N_i^+. Note that in the cascade mechanism doubly charged ions can be produced not only from ground states of singly charged ions, but also from excited ionic states due to excitation and further decay of the autoionizing states of the atom, and also due to ionization of atoms from two-electron states.

Figure 5 shows the possible realizations of the cascade mechanism of formation of Sr^{2+} and Ba^{2+} ions at frequencies $\omega = 16800$–$18000\ \mathrm{cm}^{-1}$. In these realizations, a number of resonances can appear in the spectra of Sr^+ and Ba^+. Analysis of the experimental data of Figs. 3 and 4 shows that the large number of resonance maxima on the $N_i^2(\omega)$ plots can be well identified in the spectra of the Sr^+ and Ba^+ ions.

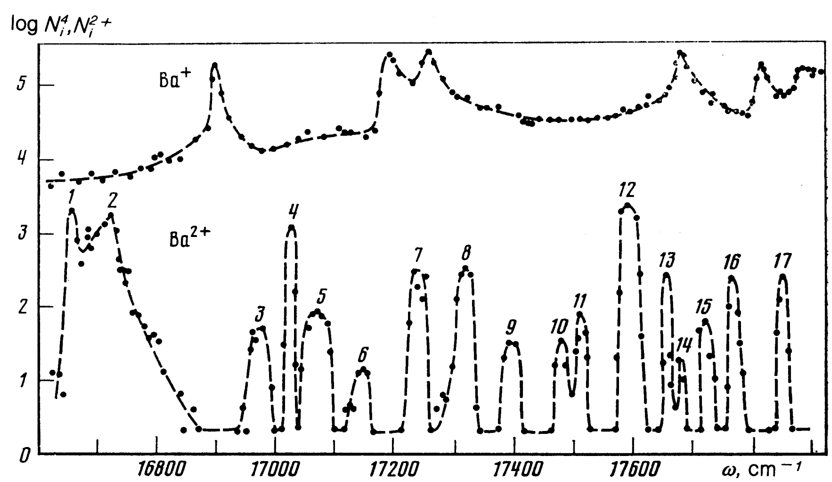

FIG. 4. Frequency dependences of the yields of single and double ionization of the Ba atom. The value $\log N_i^{2+} = 0.3$ corresponds to the lower limit of the recording-apparatus sensitivity in this experiment.

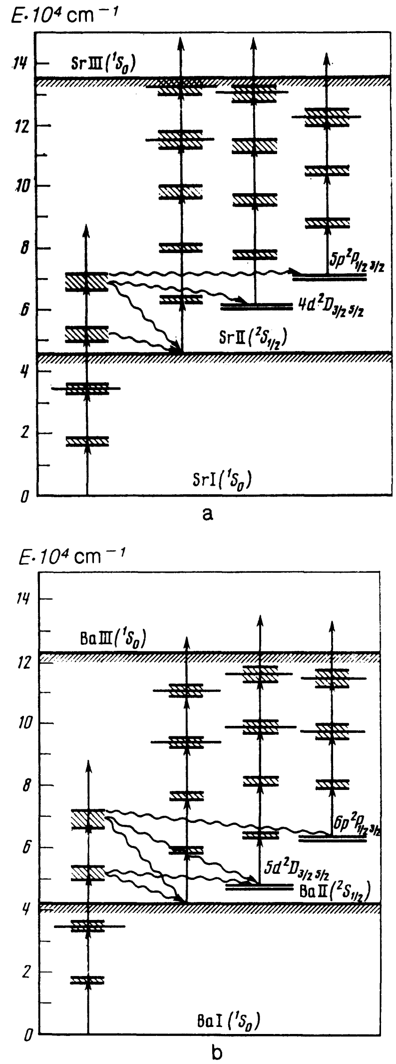

FIG. 5. Scheme of realization of the cascade mechanism of formation of doubly charged Sr^{2+} (a) and Ba^{2+} (b) ions when Sr and Ba atoms are ionized by irradiation at $\omega = 16800$–$18000\ \mathrm{cm}^{-1}$. The shaded sections correspond to the regions where the energy is changed when both the Sr and Ba atoms and the Sr^+ and Ba^+ ions absorb the number of photons needed for ionization. The horizontal lines mark the possible onset of resonances in the spectra of the atoms and ions. The wavy lines with arrows mark the possible channels for decay of the autoionization states of the neutral atoms to different states of singly charged ions. The decay of autoionization states to both terms of a doublet is represented by a single line.

Certain resonance maxima are due to the onset of several resonances at close frequencies, which are not resolved because of the large width of the employed laser radiation. Multiphoton resonance transitions in the spectra of singly charged ions, which cause the resonance maxima in the $N_i^{2+}(\omega)$ plots, are listed in Tables III and IV. These tables include only those transitions whose frequencies land in the width the resonance maxima of the Sr^{2+} and Ba^{2+} yields.

Note that besides the resonances indicated in Table IV, five-photon resonances with high-lying Rydberg states, having terms $^2P_J^0$, $^2F_J^0$, and $^2H_J^0$, can appear when the Ba^+ ion is ionized from the ground state. Transitions to these states are not included in Table IV, since there are no published data on their energies. The density of these Rydberg states is expected to be very high, and their excitation by radiation with a spectrum width $\Delta\omega \sim 6\ \mathrm{cm}^{-1}$ will not yield clearly pronounced resonances on the $N_i^{2+}(\omega)$ plot.

Thus, the identifications, in Tables III and IV, of the resonance maxima of the plots of $N_i^{2+}(\omega)$ point to the feasibility of the cascade mechanism of formation of doubly charged Sr^{2+} and Ba^{2+} ions.

A quantitative argument favoring the feasibility of formation of doubly charged ions by the cascade mechanism is the yield ratio of the singly and doubly charged ions in the intervals between the resonances. This ratio accords with the probability ratio of three- and five-photon processes. Typical data[23,24] on the probabilities of multiphoton ionization of atoms at the radiation intensities realized in these experiments lead to an ion-yield ratio $N_i^+/N_i^{2+} \sim 10^5$. This value is in satisfactory agreement with the experimental data shown in Figs. 3 and 4.

The feasibility of the cascade mechanism of doubly charged ion formation is likewise not contradicted by the fact that the $N_i^{2+}(\omega)$ plots have no resonance maxima due to the resonant increase of the probability of formation of singly charged ions. As mentioned above, the data illustrated in Figs. 3 and 4 were obtained under conditions of possible saturation of the yield of singly charged ions. Our estimates have shown that the resonant increase of the yield of singly charged ions, which is caused under saturation conditions by the increase of the interaction volume, does not increase substantially the yield of doubly charged ions, since the effective formation of the latter takes place only in that region of the interaction volume where the field strength is a maximum, and in this region the density of the singly charged ions remains practically unchanged.

Thus, the experimental data of Figs. 3 and 4 point to the feasibility of the cascade mechanism of formation of the doubly charged ions Sr^{2+} and Ba^{2+}.

At the same time, notice must be taken of two experimental facts that cannot be unambiguously explained at present within the framework of the proposed realizations of the cascade process (Fig. 5).

1. Ionization of the Sr^+ and Ba^+ ions from the ground and excited *nd* and *np* states in the frequency range $\omega = 16800$–$18000\ \mathrm{cm}^{-1}$ can excite, besides the states listed in Tables III and IV, also a large number of states (22 and 18 upon ionization of Sr^+ and Ba^+, respectively). These states have the same quantum numbers as the states listed in Tables III and IV, and their excitation requires absorption of the same number of photons as excitation of the states listed in the very same tables.[14] However, the $N_i^{2+}(\omega)$ plots do not show the resonance maxima corresponding to excitation of these states. One possible reason for the absence of these maxima may be that the Sr^+ and Ba^+ ions are not produced in initial states needed to excite the states in question. This may be due to the difference between the probabilities of decay of the autoionizing states of the Sr and Ba atoms into different ionic states. A conclusive confirmation of this assumption, however, calls for further research, particularly

TABLE III. Identification of resonance maxima of the temperature dependence of the yield of doubly charged Sr^{2+} ions.

№	ω_r, cm^{-1}	Γ, cm^{-1}	ω_{tr}, cm^{-1}	Resonance transition
1	16 884	20	16 881	$5s^2S_{1/2} + 4\hbar\omega \to 6d^2D_{3/2}$
			16 883	$4d^2D_{3/2} + 4\hbar\omega \to 7g^2G_{7/2,\,9/2}$
			16 890	$5s^2S_{1/2} + 4\hbar\omega \to 6d^2D_{5/2}$
2	17 090	40	17 099	$4d^2D_{3/2} + 4\hbar\omega \to 10d^2D_{3/2,\,5/2}$
3	17 682	10	–	Not identified
4	17 790	10	–	»
5	17 850	20	–	»

Note: The number of the resonance maxima on the $N_i^{2+}(\omega)$ plot corresponds to the numbers indicated in Fig. 3. ω_r—emission frequencies corresponding to the maxima of the $N_i^{2+}(\omega)$ plots; ω_{tr}—emission frequencies corresponding to those resonance transitions in the spectrum of Sr^+ to which the maxima of the $N_i^{2+}(\omega)$ plot are due.

TABLE IV. Identification of the resonance maxima of the frequency dependence of the yield of doubly charged Ba ions.

№	ω_r, cm^{-1}	Γ, cm^{-1}	ω_{tr}, cm^{-1}	Resonance transition
1	16 660	15	16 668	$6s^2\,S_{1/2} + 4\hbar\omega \to 8d^2\,D_{3/2}$
			16 670	$6s^2\,S_{1/2} + 3\hbar\omega \to 7p^2\,P^0_{3/2}$
2	16 720	40	16 702	$5d^2\,D_{3/2} + 4\hbar\omega \to 7g^2\,G_{7/2,\,9/2}$
3	16 970	25	16 958	$5d^2\,D_{3/2} + 4\hbar\omega \to 11s^2\,S_{1/2}$
4	17 025	10	17 030	$5d^2\,D_{5/2} + 4\hbar\omega \to 8g^2\,G_{7/2,\,9/2}$
5	17 070	45	17 049	$6p^2\,P^0_{3/2} + 3\hbar\omega \to 10d^2\,D_{3/2}$
			17 056	$6p^2\,P^0_{3/2} + 3\hbar\omega \to 10d^2\,D_{5/2}$
			17 057	$5d^2\,D_{3/2} + 4\hbar\omega \to 10d^2\,D_{3/2}$
			17 062	$5d^2\,D_{3/2} + 4\hbar\omega \to 10d^2\,D_{5/2}$
6	17 145	25	17 140	$6p^2\,P^0_{1/2} + 3\hbar\omega \to 7g^2\,G_{7/2,\,9/2}$
7	17 240	20	17 238	$5d^2\,D_{5/2} + 3\hbar\omega \to 5f^2\,F^0_{5/2}$
8	17 320	20	17 319	$5d^2\,D_{5/2} + 3\hbar\omega \to 5f^2\,F^0_{7/2}$
9	17 395	20	17 393	$5d^2\,D_{5/2} + 4\hbar\omega \to 9g^2\,G_{7/2,\,9/2}$
			17 404	$5d^2\,D_{3/2} + 4\hbar\omega \to 12s^2\,S_{1/2}$
10	17 475	20	17 473	$5d^2\,D_{3/2} + 4\hbar\omega \to 11d^2\,D_{3/2}$
			17 476	$5d^2\,D_{3/2} + 4\hbar\omega \to 11d^2\,D_{5/2}$
			17 481	$6p^2\,P^0_{1/2} + 3\hbar\omega \to 11s^2\,S_{1/2}$
11	17 508	15	17 503	$6s^2\,S_{1/2} + 4\hbar\omega \to 10s^2\,S_{1/2}$
			17 513	$6p^2\,P^0_{3/2} + 3\hbar\omega \to 11s^2\,S_{1/2}$
12	17 590	20	17 592	$5d^2\,D_{3/2} + 4\hbar\omega \to 9g^2\,G_{7/2,\,9/2}$
			17 604	$6p^2\,P^0_{3/2} + 3\hbar\omega \to 11d^2\,D_{3/2}$
13	17 654	15	17 651	$5d^2\,D_{5/2} + 4\hbar\omega \to 10g^2\,G_{7/2,\,9/2}$
			17 655	$6s^2\,S_{1/2} + 4\hbar\omega \to 9d^2\,D_{3/2}$
			17 662	$6s^2\,S_{1/2} + 4\hbar\omega \to 9d^2\,D_{5/2}$
14	17 678	10	–	Not identified
15	17 720	10	17 719	$6p^2\,P^0_{3/2} + 2\hbar\omega \to 5f^2\,F^0_{5/2}$
16	17 765	15	17 764	$6p^2\,P^0_{3/2} + 3\hbar\omega \to 9g^2\,G_{7/2}$
			17 768	$5d^2\,D_{3/2} + 4\hbar\omega \to 12d^2\,D_{3/2,\,5/2}$
17	17 850	15	17 843	$5d^2\,D_{5/2} + 4\hbar\omega \to 11g^2\,G_{7/2,\,9/2}$
			17 844	$6p^2\,P^0_{1/2} + 3\hbar\omega \to 8g^2\,G_{7/2}$
			17 852	$5d^2\,D_{3/2} + 4\hbar\omega \to 10g^2\,G_{7/2,\,9/2}$

Note: The numbers of the resonance maxima in the $N_i^{2+}(\omega)$ dependence accord with the numbers indicated in Fig. 4 ω_r and ω_{tr} are the same as in the Note of Table III, but for the yield of the Ba^+ ions (Fig. 4).

measurement of the energy of the electrons produced upon ionization of the Sr and Ba atoms.

2. The frequency plots of the yield of doubly charged strontium and barium ions have resonance maxima that cannot be identified either in the spectrum of the singly charged ions or in the spectrum of the neutral atoms (see Tables 3 and 4). Note that an attempt was made in Refs. 11 and 12 to identify the resonance maxima of the yield $N_i^{2+}(\omega)$ of doubly charged strontium ions at the frequencies $\omega = 17682$, 17799, and 17850 cm^{-1}, due to multiphoton transitions in the Sr^+ spectrum. This identification, however, can hardly be regarded as final, since differences that remain unchanged with changing field[12] exist between the frequencies at which these maxima are observed and the frequencies with which attempts were made[11,12] to identify them. It should be noted that in an earlier investigation of multiphoton ionization by the second harmonic of a neodymium laser we have likewise observed in the yields of doubly charged ions resonance maxima that cannot be identified in the spectrum of either the singly charged ion or of the neutral atom.[6] Unambiguous identification of the foregoing resonance maxima will reveal other channels for formation of doubly charged ions. The facts noted indicate that in the frequency region $\omega = 16800$–18000 cm^{-1} investigated by us the Sr^{2+} and Ba^{2+} ions are produced not only by resonant multiphoton ionization of the singly charged ions Sr^+ and Ba^+.

Summarizing the results for the productions of Sr^{2+} and Ba^{2+} ions we note that the most important is the detection of the cascade mechanism of formation of doubly charged ions.

4. CONCLUSION

It is of interest to compare our results with other experimental data on multiphoton ionization of alkaline-earth atoms in a radiation field of lower frequency. We have in mind the data obtained in Refs. 2, 3, 5, and 19, where a neodymium laser ($\hbar\omega \approx 1.2$ eV) was used. Such a comparison reveals substantial differences between the character of the ionization process at various irradiation frequencies. An example of such a difference is the already mentioned absence of resonance with the intermediate triplet states in five-photon ionization of the Ba atom.[19] On the basis of the data on the spectrum and the selection rules, such resonances should be observed in four-photon excitation.

The second difference is that at low frequencies ($\hbar\omega \approx 1.2$ eV) no resonances maxima are produced by the cascade mechanism in the N_i^{2+} yield.[2,3,5] This circumstance can be qualitatively understood if it is recognized that the degrees of nonlinearity of atom ionization ($K^+ \approx 5$) differs greatly from that of singly charged ions ($K^{2+} \approx 10$). This points to little likelihood of realization of the cascade process.

It is natural to assume that the character of the process of formation of doubly charged ions depends on the degree of nonlinearity of different transitions in the spectra of the atom and ion, and depends by the same token on the emission frequency. The process of formation of doubly charged ions may be different in different frequency ranges. In particular, when the emission frequency is increased the probability of formation of doubly charged ions by the cascade mechanism should increase. There are at present, however, not enough experimental data at frequencies $\hbar\omega \approx 1$ eV for a final confirmation of this viewpoint.

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[1] I. S. Aleksakhin, N. B. Delone, I. P. Zapesochnyi, and V. V. Suran, Zh. Eksp. Teor. Fiz. **76**, 887 (1979) [Sov. Phys. JETP **49**, (1979)].
[2] I. I. Bondar', I. P. Zapesochnyi, N. B. Delone, and V. V. Suran, Pis'ma Zh. Tekh. Fiz. **7**, 243 (1981) [Sov. J. Tech. Lett. **7**, 104 (1981)].
[3] D. Delone, I. Bondar, I. Suran, and B. Zon, Opt. Commun. **40**, 268 (1982).
[4] I. I. Bondar' and V. V. Suran, Opt. Spektrosk. **55**, 615 (1983).
[5] I. I. Bondar', N. B. Delone, I. P. Zapesochnyĭ, and V. V. Suran, Preprint, Inst. Gen. Phys. USSR Acad. Sci., No. 133, 1984.
[6] I. I. Bondar', A. I. Gomonaĭ, and V. V. Suran, in: Nonlinear processes in two-electron atoms [in Russian], Council on Spectroscopy, Moscow, 1984, p. 26.
[7] D. Sh. Akramova, D. T. Akimov, V. K. Medvedeva, *et al.*, Opt. Spektrosk. **56**, 987 (1984).
[8] D. Sh. Akramova, D. T. Alimov, V. K. Medvedeva, *et al.*, Ref. 6, p. 48.
[9] D. Sh. Akramova, D. T. Alimov, N. B. Delone, *et al.*, Opt. Spektrosk. **55**, 1062 (1983).
[10] D. Feldman, I. Krautwald, and K. Welge, J. Phys. **D15**, L529 (1982).
[11] P. Agostini and G. I. Petite, *ibid.* **B18**, L281 (1985).
[13] B. Feldman and K. Welge, *ibid.* **B15**, 1651 (1982).
[14] V. I. Sobelman, *Introduction to the Theory of Atomic Spectra*, Pergamon, 1973.
[15] C. E. Moore, *Atomic Energy Levels*, CNBC, Washington, Vols. 2 and 3.
[16] M. G. Kozlov, Absorption Spectra of Metal Vapors in the Vacuum Ultraviolet [in Russian], Nauka, 1981.
[17] C. M. Brown and M. L. Ginter, J. Opt. Soc. Am. **68**, 817 (1978).
[18] S. L. Chin, D. Feldman, J. Krautwald, and K. Welge, J. Phys. **B14**, 2353 (1981).
[19] I. I. Bondar, A. I. Gomonai, N. B. Delone, *et al.*, *ibid.* **17**, 2048 (1984).
[20] I. I. Bondar', N. B. Delone, M. A. Preobrazhenskiĭ, and V. V. Suran, Abstracts, 12th All-Union Conf. on Nonlinear and Coherent Optics [in Russian], part 1, 1985, p. 262.
[21] L. A. Vaĭnshteĭn and I. A. Poluektov, Opt. Spektrosk. **12**, 460 (1962).
[22] A. A. Radtsig and B. M. Smirnov, Handbook of Atomic and Molecular Physics [in Russian], Atomizdat, 1980.
[23] Multiphoton Ionization of Atoms. Trudy FIAN, Vol. **115**, Nauka, 1980.
[24] J. Morellec, D. Normand, and G. Petite, Adv. J. Phys. **B18**, 97 (1983).

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