Proof of a dielectronic mechanism for the formation of Ba^{2+} ions in the IR region ($\omega = 9395\ cm^{-1}$)

I. I. Bondar' and V. V. Suran
Uzhgorod State University, 294000, Uzhgorod, the Ukraine

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The mechanism for the formation of Ba^{2+} ions during ionization of Ba atoms by light from a YAG laser has been studied. A beam of Ba atoms was exposed to the light from a dye laser, as well as the light from the YAG laser, in order to identify the Ba^{2+} formation mechanism. The light from the dye laser caused an additional excitation of the neutral atoms in the course of their ionization. The yield of Ba^{2+} ions was found to be much lower when the dye laser excited the Ba atoms than when the Ba atoms were not excited. This result indicates that the formation of Ba^{2+} ions at the frequency of the YAG-laser beam occurs as the result of a dielectronic mechanism.

The processes by which doubly charged ions A^{2+} form in the course of ionization of alkaline-earth atoms by laser light have been studied previously (Refs. 1 and 2, for example). This work has shown that in the visible and UV parts of the spectrum these ions are formed primarily by a cascade mechanism, in which the A^{2+} ions form in two steps: In one laser pulse, singly charged A^{+} ions first form during ionization of the neutral atoms A; then the A^{2+} ions form upon ionization of the A^{+} ions. In the IR part of the spectrum, on the other hand, studies[3,4] show that the formation of A^{2+} ions cannot be explained by the cascade mechanism. It has been suggested that a dielectronic mechanism operates to form the A^{2+} ions in this part of the spectrum. In that mechanism, the A^{2+} ions would form as a result of the simultaneous detachment of two electrons from the neutral atom. However, the reality of that mechanism has yet to be unambiguously proved. We have accordingly undertaken a search for proof that a dielectronic mechanism operates to form A^{2+} ions in the IR part of the spectrum.

In particular, we studied the mechanism for the formation of doubly charged ions during ionization of Ba atoms by the light from a YAG laser, with $\omega = 9395\ cm^{-1}$. To identify the mechanism for the formation of the Ba^{2+} ions in this case, we exposed a beam of Ba atoms to not only the beam from the YAG laser but also the beam from a tunable dye laser. The beams from the two lasers were focused in the same region.

The intensity of the light from the YAG laser was chosen in such a way that the ionization of the Ba atoms would be saturated by this beam (this intensity was $F = 3.5 \times 10^{29}\ s^{-1} \cdot cm^{-2}$ in our case). Both Ba^{+} ions and Ba^{2+} ions were formed. The yield of Ba^{2+} ions was only 1/40 that of the Ba^{+} ions.

The intensilty ($F = 9.4 \times 10^{24}\ s^{-1} \cdot cm^{-2}$) of the light from the dye laser was chosen such that only Ba^{+} ions would form during ionization of Ba atoms by this light. The frequency of the dye laser was varied over the interval $\omega = 17\,860–18\,200\ cm^{-1}$. This interval includes several frequencies at which one- and two-photon excitation of the Ba atom is possible.[5] The supplemental application of the light from the dye laser during ionization of the Ba atom by the YAG beam thus results in an additional excitation of this atom.

In the experiments we measured the yields of the Ba^+ and Ba^{2+} ions in cases in which the Ba atoms were subjected to (a) only the beam from the YAG laser, (b) only the beam from the dye laser, and (c) the two beams simultaneously. The experimental results are shown in Fig. 1.

We look first at part *a* of this figure, which shows results found on the Ba^{2+} ions. We see that the Ba^{2+} yield is lower when the Ba atoms are subjected to both laser beams, and the frequency of the dye laser corresponds to excitation of the Ba atom, than when the atoms are exposed to only the beam from the YAG laser. The additional excitation of the Ba atom in the course of its ionization by the light from the YAG laser thus reduces the Ba^{2+} yield. The greatest decrease in the yield of these ions occurs during one-photon excitation of the Ba atom into the $6s\,6p\,^1P_1^0$ state ($\omega = 18\,060$ cm^{-1}).

We turn now to the results on the Ba^+ ions, which are shown in parts *b* and *c* of Fig. 1. Part *b* shows results found at the same laser beam intensities as in part *a*. It can be seen from this figure that the yield of Ba^+ ions is lower when the two laser beams are applied simultaneously than when only the beam from the YAG laser is applied.

The additional excitation of the Ba atom in the course of its ionization by the YAG beam thus leads to an increase in the yield of the Ba^+ ions. The maxima in the yield of Ba^+ ions in this case coincide in frequency with minima in the yield of Ba^{2+} ions.

The increase in the Ba^+ yield upon the additional excitation of the Ba atom naturally suggests an increase in the probability for the formation of Ba^+ ions in this case. From the results in Fig. 1b we cannot draw a conclusion regarding the extent to which the probability for the formation of Ba^+ ions increases at the various frequencies. The reason is that these results were obtained during saturation of the ionization process, so the maxima in the yield of Ba^+ ions are roughly the same in size. We accordingly measured the yield of Ba^+ ions at lower intensities of the two laser beams. These results are shown in part *c* of Fig. 1. It follows from these results that the greatest increase in the Ba^+ formation probability occurs when the dye laser excites the $6s\,6p\,^1P_1^0$ state; i.e., it occurs at the frequency corresponding to the greatest decrease in the yield of Ba^{2+} ions.

The experimental results thus show that the additional excitation of the Ba atom in the course of its ionization by the YAG laser beam results in an increase in the probability for the formation of Ba^+ ions and a decrease in the yield of Ba^{2+} ions. The greater the increase in the Ba^+ formation probability, the greater the decrease in the Ba^{2+} yield. This effect cannot be explained by the model of a cascade mechanism for the formation of Ba^{2+} ions. In that model, the Ba^+ ions serve as the target in the formation of Ba^{2+} ions, so any increase in the probability for the formation of Ba^+ ions would naturally lead to an increase in the yield of Ba^{2+} ions. At the very worst, the yield of Ba^{2+} ions should not decrease.

Looking at the dielectronic mechanism, we note that the decrease in the yield of

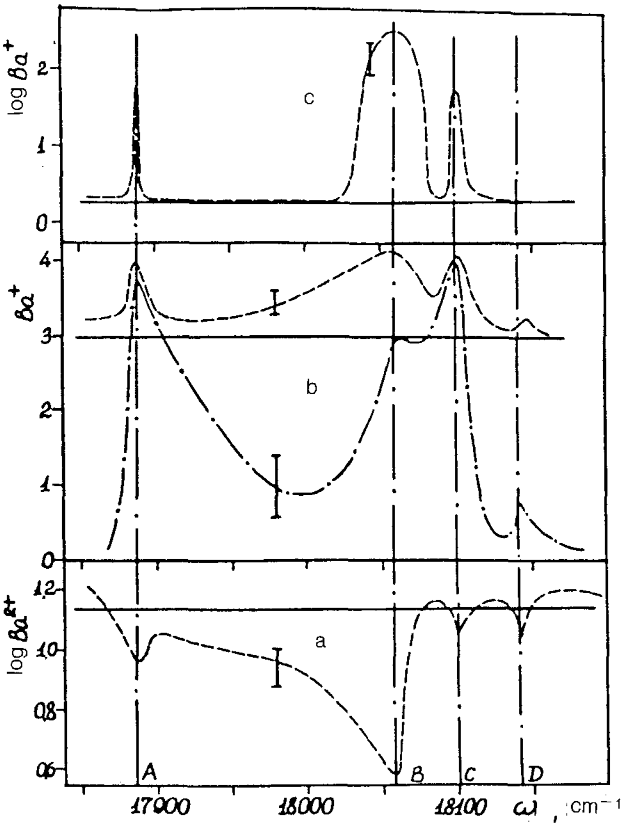

FIG. 1. Measured yields of Ba^+ and Ba^{2+} ions versus the frequency of the dye laser. Dashed lines—The ionization process in which the Ba atom is subjected to light from the YAG and dye lasers simultaneously; dot-dashed lines—only the beam from the dye laser is applied; horizontal solid lines—only the beam from YAG laser is applied. The vertical dot-dashed lines with letters near their bottom show the frequencies of the dye laser corresponding the transitions in the spectrum of the neutral Ba atom: A) $6s^2\,{}^1S_0 + 2\hbar\omega \rightarrow 6s7d\,{}^3D_2$ (17 881 cm^{-1}); B) $6s^2\,{}^1S_0 + \hbar\omega \rightarrow 6s6p\,{}^1P_1^0$ (18 060 cm^{-1}); C) $6s^2\,{}^1S_0 + 2\hbar\omega \rightarrow 5d\,6d\,{}^3D_2$ (18 100 cm^{-1}); D) $6s6p\,{}^1P_1^0 + \hbar\omega \rightarrow 5d\,6d\,{}^3D_2$ (181 140 cm^{-1}). The results in parts *a* and *b* were obtained at intensities $F_1 = 3.5 \times 10^{29}$ and $F_2 = 9.4 \times 10^{24}$ $s^{-1} \cdot cm^{-2}$ of the YAG laser and the dye laser, respectively; the results in part *c* were obtained at intensities $F_2 = 1.5 \times 10^{29}$ and $F_1 = 2.0 \times 10^{24}$ $s^{-1} \cdot cm^{-2}$.

Ba^{2+} ions which we observed upon an increase in the probability for the formation of Ba^+ ions can be explained well by the model in which this mechanism is operating. For example, if this mechanism is operating, the neutral Ba atoms should serve as the target for the formation of both Ba^+ ions and Ba^{2+} ions, as we just mentioned. Let us now assume that during the ionization of the neutral Ba atoms the dielectronic mechanism becomes supplemented by an additional excitation of atoms which raises the probability for the formation of Ba^+ ions. The introduction of this excitation, under conditions such that the ionization of the Ba atom has reached saturation (these are the conditions under which our results were obtained), has the result that saturation sets in more quickly than in the case without excitation. The saturation of an ionization process can, as we know, be summarized as a situation in which, because of multiphoton ionization, there are no neutral atoms in the region in which they would interact with the laser light. The introduction of an additional excitation of Ba atoms in the course of their ionization thus has the consequence that the Ba atoms disappear from the interaction region earlier than in the absence of excitation. Since the neutral Ba atoms serve as the target for the formation of Ba^{2+} ions in the case of the dielectronic mechanism, this change in the time at which the Ba atoms disappear naturally leads to a decrease in the yield of Ba^{2+} ions.

In summary, our results indicate that the formation of Ba^{2+} ions during ionization of Ba atoms by the light from the YAG laser occurs as a result of the dielectronic mechanism. These results constitute the first proof that this mechanism actually occurs. Precisely how two electrons are simultaneously removed from neutral atoms in the realization of this mechanism, on the other hand, remains an open question.

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