Three-photon ionization of a Ba atom from the excited $6s6p\,^1P_1^0$ state in the IR spectral range

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We study the three-photon ionization of the barium atom from the excited $6s6p\,^1P_1^0$ state. In the dependence of the Ba^+ ion yield on the frequency of the radiation from a color-center laser ($\omega_c = 8300$–$9100\ \mathrm{cm}^{-1}$) we discover resonance peaks of various amplitudes and widths and can identify some unambiguously. The identified large-amplitude peaks are caused by two-photon excitation of singlet and triplet bound states. We find a high probability of two-photon excitation of odd triplet bound states and a low probability of three-photon excitation of autoionization states. In addition, we establish the existence of large-amplitude resonance peaks that could not be identified by known processes. Apparently, these peaks can be attributed to the peculiarities in the ionization of barium atoms in two resonant laser fields of different frequencies. © *1996 American Institute of Physics.* [S1063-7761(96)00602-5]

1. INTRODUCTION

Over the years extensive experimental study has been conducted involving one- and two-electron singlet and triplet states and autoionization states in the resonant process of multiphoton ionization of alkali-earth atoms.[1-6] It was discovered that often the excitation of these states accompanied by the absorption of a small number (two or three) photons is highly probable. Note that in these experiments the investigated objects were alkali-earth atoms in their ground state ($ns^2\ ^1S_0$). An interesting problem arising in this connection is the effect of the above states on the multiphoton ionization of alkali-earth atoms in an excited state. Since the parity of the excited state can be opposite to that of the ground state, states that cannot be excited from the ground state in the dipole approximation can be studied. One of the simplest ways to excite an atom is provided by the method of optical excitation involving laser radiation.[7] The ionization of excited alkali-earth atoms in the IR spectral range has not previously been studied. In this paper we give the results of such a study involving the Ba atom.

2. THE EXPERIMENTAL METHOD

In our studies we used a method involving the crossing of a laser beam with a beam of barium atoms. The Ba atom concentration in the region where the beams interacted was $n \simeq 10^{10}\mathrm{cm}^{-3}$. To ionize the barium atoms we used the radiation of an F_2^--center laser (the LiF crystal) with $\omega_c = 8300$–$9100\ \mathrm{cm}^{-1}$. To obtain barium atoms in the excited $6s6p\,^1P_1^0$ state we used the linearly polarized radiation of a fixed-frequency dye laser with $\omega_d = 18\,060\ \mathrm{cm}^{-1}$, which is equal to the transition frequency from the $6s^2\ ^1S_0$ ground state to the excited $6s6p\,^1P_1^0$ state (Fig. 1a). The dye laser makes it possible to excite nearly 50% of the atoms.[7] Note that these excited atoms were also aligned.[8] The beams of the color-center and dye lasers were combined in space by a special mirror and focused by a single lens onto the atomic beam. Note that the beams of the lasers were synchronized, and pulse length was $\tau = 40$ ns. The spectral widths of the dye laser and color-center laser beams was $\Delta\omega_d = 2\ \mathrm{cm}^{-1}$ and $\Delta\omega_c = 4\ \mathrm{cm}^{-1}$, respectively. In our experiments both laser beams were linearly polarized, though in a few cases both were circularly polarized.

Ionization of Ba atoms from the excited $6s6p\,^1P_1^0$ state and the $6s^2\ ^1S_0$ ground state requires the absorption, respectively, of three and five photons from the color-center laser radiation (see Figs. 1b, c, and d). We chose optimal intensities of the light from the color-center and dye lasers. For instance, the intensity of the light from the color-center laser must be such that the process of five-photon ionization of Ba atoms from the ground state (when the $6s6p\,^1P_1^0$ level is not excited; see Fig. 1d) does not lead to noticeable formation of Ba^+ ions and the process of three-photon ionization from the excited $6s6p\,^1P_1^0$ state (see Figs. 1b and c) is not saturated, i.e., the condition $W\tau < 1$, where W is the probability of ionization of a Ba atom from the $6s6p\,^1P_0^1$ state, must be met in the entire range of ω_c. The experiments were done with a laser field intensity $\varepsilon = 4.8\times 10^5$ V/cm, so that the above condition was met.

The optimum intensity of the dye-laser light was selected in such a way that there was effective excitation (population) of the $6s6p\,^1P_1^0$ state (see Fig. 1a), but the three-photon ionization of Ba atoms from the ground state (see Fig. 1e) did not lead to a noticeable formation of Ba^+ ions. Effective excitation of a resonant state occurs at laser field intensities at which saturation sets in, i.e., the populations of the ground and excited states become equal.[7,9] According to Ref. 9, such equalization occurs for $\Omega = d\varepsilon \gg \gamma$, where Ω is the Rabi frequency, d is the dipole moment, and γ is the width of the excited state. In our experiment Ω was approximately $25\ \mathrm{cm}^{-1}$ ($\varepsilon \simeq 6\times 10^4$ V/cm and $d \simeq 8.7$ a.u.), which is much larger than the width of the $6s6p\,^1P_1^0$ state ($\gamma \simeq 4\times 10^{-3}\ \mathrm{cm}^{-1}$). Thus, in our experimental conditions saturation set in and there was effective excitation (population) of the $6s6p\,^1P_1^0$ state and formation of aligned Ba atoms.

The effectiveness of excitation of the resonant

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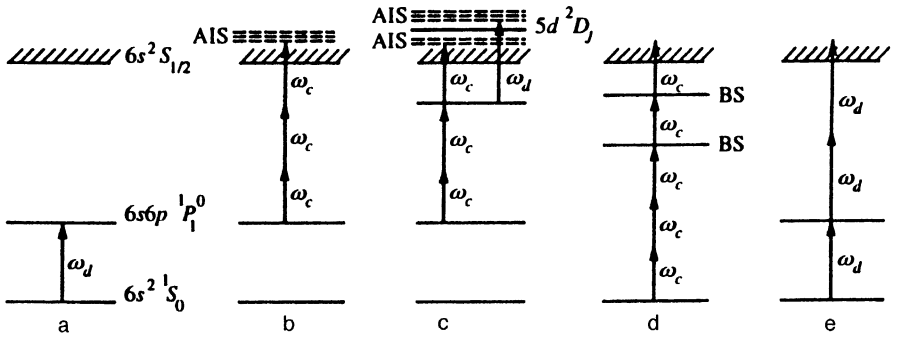

FIG. 1. Diagrams of the various processes.

$6s6p\,^1P_1^0$ state was verified experimentally. In this case, at a fixed frequency of the color-center laser radiation we measured the Ba^+ ion yield as the frequency ω_d of the dye laser was varied. The frequency ω_c of the color-center laser was selected in such a way that the Ba atoms were ionized nonresonantly from the ground and excited states. In our experiments this frequency was $8620\ cm^{-1}$. The results are depicted in Fig. 2. Clearly, when the dye-laser frequency is $\omega_d \simeq 18\,060\ cm^{-1}$ (when one-photon excitation of the $6s6p\,^1P_1^0$ state occurs), there is a sharp increase in the Ba^+ ion yield, while when ω_d is higher or lower than the above value, the Ba^+ ion yield is approximately at the same level as when the Ba atoms are ionized solely by the laser radiation at ω_c. This fact proves unambiguously that in the conditions of our experiment there is effective excitation (population) of the $6s6p\,^1P_1^0$ states, and the ionization of a Ba atom at ω_c transforms from a five-photon process to a three-photon. No further details of the experimental method will be given since they are thoroughly described in our previous paper (see, e.g., Ref. 3).

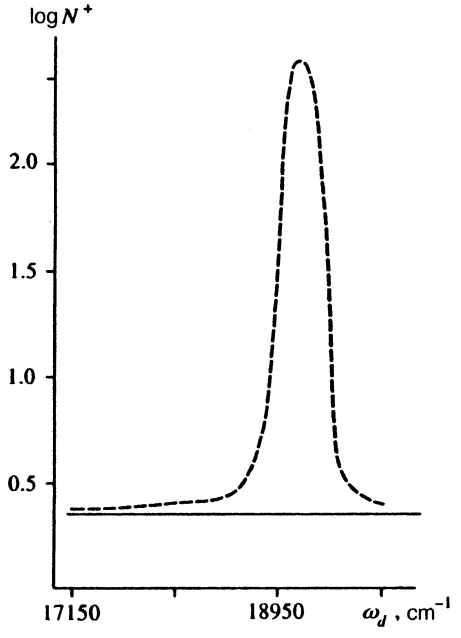

FIG. 2. The Ba^+ ion yield when two laser fields act on the Ba atoms simultaneously: one of a tunable dye laser, and the other of a fixed-frequency color-center laser with $\omega_c = 8620\ cm^{-1}$ (the dashed curve). The solid line represents the Ba^+ ion yield when only the radiation of the fixed-frequency color-center laser with $\omega_c = 8620\ cm^{-1}$ acts on the Ba atoms.

3. EXPERIMENTAL RESULTS

We studied the dependence of the Ba^+ ion yield on the frequency ω_c of the color-center laser (N^+ vs ω_c) when Ba atoms are ionized from the excited $6s6p\,^1P_1^0$ state. The results of studying this dependence at a fixed field strength ($\varepsilon = 4.8\times 10^5$ V/cm) generated by the color-center laser are depicted in Fig. 3a. In this experiment we used linearly polarized radiations of both the dye laser and the color-center laser, with the polarization vectors being parallel in space. The N^+ vs ω_c dependence exhibits resonance peaks of various heights and widths. Since there is no saturation in the Ba^+ ion yield, the height of the resonance peaks may serve as an indicator of the probability of multiphoton excitation of various states and may be used to find the real width of the resonance peaks. In our studies the width of the resonance peaks was determined at half-maximum. Note that here the minimum width of the resonance peaks is limited from below by the effective width of the laser radiation spectrum, $\Gamma = \Delta\omega_c K^{1/2}$, where K is the number of absorbed photons.[9]

The results of identifying the resonance peaks are listed in Table I. To identify the peaks we used tabulated data on the spectra of bound and autoionization states.[10,11] In analyzing the data obtained we allowed for the possibility of the formation of Ba atoms not only in the excited $6s6p\,^1P_1^0$ state but also in the excited metastable $6s5d^3D_{1,2,3}$ and

TABLE I. Identification of the resonance peaks in the N^+ vs ω_c dependence in the three-photon ionization of Ba atoms from the excited $6s6p\,^1P_1^0$ state.

Nos.	K	ω_m, cm^{-1}	ω_t, cm^{-1}	State
1	2	8270	8271	$6s4f^3F_2^0$
2	2	8278	8278	$6s4f^3F_2^0$
3	2	8308		unidentified
4	2	8338	8338	$6s4f^1F_2^0$
5		8375		unidentified
6		8752		unidentified
7		8760		unidentified
8		8803		unidentified
9		8848		unidentified
10	2	8915	8916	$6s8p\,^1P_1^0$
11		9042		unidentified
12	3	9075	9076	AIS, $J=1$
13	2	9087	9087	$5d7p^3F_2^0$

Note: K is the number of photons of the laser radiation needed to excite the resonant states of the Ba atom, and ω_t and ω_m are the frequencies of the resonant transitions in the spectrum of the Ba atom and the frequencies corresponding to the resonance peaks in the Ba^+ ion yield.

$6s5d\,^1D_2$ states and the presence of atoms in the $6s^2\,^1S_0$ ground state. We found that the N^+ vs ω_c dependence contains no resonance peaks caused by ionization of the Ba atom from the above metastable states. This suggests that initially the atomic beam has no atoms in such states. Also, the N^+ vs ω_c dependence has no peaks associated with the resonant process of ionizing Ba atoms from the ground state; such peaks were studied in Ref. 12. An exception is the peak at $\omega_c = 8938\ \text{cm}^{-1}$, which is especially evident when the radiation is circularly polarized (see Fig. 3b). According to the data of Ref. 12, this resonance peak has the highest probability and, therefore, becomes apparent in our investigations. As Table I implies, we were able to unambiguously identify some resonance peaks in the N^+ vs ω_c dependence. A peak is assumed identified if the frequency of multiphoton excitation of the resonance state and the frequency corresponding to the resonance peak coincide within the width of the peak. At the same time resonance peaks were observed in the dependence that could not be identified. We begin with the identified peaks.

The identified peaks (except the peak at $9075\ \text{cm}^{-1}$) are caused by excitation of bound states. The spectrum of bound states of the Ba atom has been well studied and the widths of these states are known to be approximately $10^{-3}\ \text{cm}^{-1}$, which is much smaller than the effective width of laser radiation ($\Delta\omega_c \simeq 6\ \text{cm}^{-1}$). The resonance peaks in the N^+ vs ω_c dependence manifest themselves for all bound states that can be excited from the excited $6s6p\,^1P_1^0$ state in the frequency range studied. As the data listed in Table I imply, when the peaks are identified, the frequency of excitation of a bound state practically coincides with that of the resonance peak. This suggests that in our conditions the excitation by laser radiation is insignificant in comparison to the width of the resonance peaks.

From Table I it follows that the large-amplitude identified peaks are caused by two-photon excitation of one- and two-electron (singlet and triplet) bound states by laser radiation whose frequency is ω_c. But Ba^+ ions may form near a resonance peak as a result of ionization of the atom in these excited states by laser radiation at a frequency ω_d or ω_c or by the simultaneous action of laser light at these frequencies (see Fig. 1c).

Two-photon excitation of the two-electron $5d7p\,^3F_2^0$ state arises as a result of a one-electron process. But the formation of Ba^+ ions near a resonance peak caused by the excitation of this state can occur as a result of either a one-electron or a two-electron process (a correlation process). Indeed, a one-electron process can occur in the ionization of the $5d7p\,^3F_2^0$ state at the frequency ω_d, since in this case energy conservation implies that Ba^+ ions can form in $5d^2D_J$ states because the electron with orbital angular momentum p becomes detached from the atom while the electron with orbital angular momentum d remains with the core. At the same time, a correlation process becomes possible at the same frequency ω_d, when the Ba^+ ions are formed in the $6s^2S_{1/2}$ state. However, this process has a lower probability than the one-electron process.

A pure correlation process can be accomplished only in the ionization of the $5d7p\,^3F_2^0$ state by radiation with frequency ω_c, since in this case, due to energy conservation, the Ba^+ ions can be formed only in the $6s\,^2S_{1/2}$ state. Here one of the electrons remaining in the core (a d- or p-electron) must change its orbital angular momentum to s and the other must detach itself from the atom with the orbital angular momentum which it has.

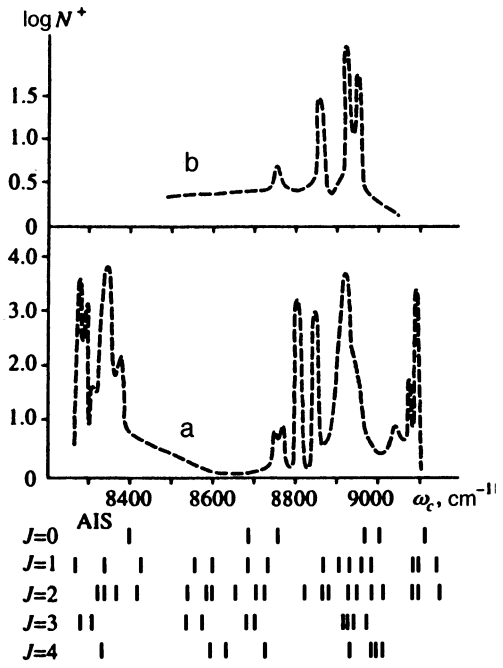

FIG. 3. The Ba^+ ion yield as a function of the frequency of the color-center laser: (a) both dye and color-center lasers emit linearly polarized radiation, and (b) both dye and color-center lasers emit circularly polarized radiation.

When, however, the resonance peaks are caused by excitation of one-electron bound states (singlet or triplet),Ba^+ ions form through the ionization of these states due to the radiation with a frequency ω_d or ω_c (or due to simultaneous action of both radiations) because of one-electron processes.

Let us now discuss the effectiveness of two-photon excitation of triplet states. We see that in the N^+ vs ω_c dependence the amplitudes of the resonance peaks caused by two-photon excitation of singlet and triplet states are approximately the same. This suggests that the probability of two-photon excitation of singlet and triplet states in the IR spectral range is approximately equal, too. Note that these triplet states are odd. Earlier we discovered that the probability of two-photon excitation of even triplet states in the IR spectral range for the Sr atom is also high.[13] We also discovered that the even triplet states of the Ba atom[1,3,4] and the Sr atom[1,4] probability are excited with a high probability by two photons in the visible spectral range as well. Thus, in investigations we discovered that in both the visible range and the IR spectral range there is a high probability of two-photon excitation of triplet states of different parities. At the same time it was discovered that the excitation of triplet states of the Ba atom by four photons[14] and the Sr atom by four or five photons[13] in the IR spectral range has a low probability. Note that the same triplet states of the Sr and Ba atoms are excited with a high probability in the visible range[1-4] and are not excited by four photons in the IR range.[13,14] The

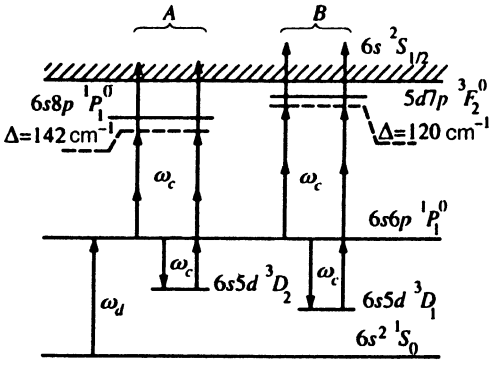

FIG. 4. The diagram of transitions realized in Ba atoms by ionizing the atoms with color-center laser light from the excited $6s6p\,^1P_1^0$ state at $\omega_c = 8845$ cm^{-1} (A) and 9027 cm^{-1} (B).

reason for such behavior of the probability of excitation of triplet states in the absorption of a different number of photons is yet to be found.

Interestingly, the widths of the resonance peaks caused by two-photon excitation of singlet states are greater (especially at the base) than those of the resonance peaks caused by two-photon excitation of triplet states. For instance, the widths of the resonance peaks related to the excitation of triplet states are approximately 5 cm^{-1} and are caused by the effective width of the laser radiation spectrum, while the similar widths of the peaks caused by excitation of singlet states are considerably greater and approximately 15 cm^{-1}. The greater width of the latter resonance peaks may be caused by excitation at the closer frequencies of autoionization states, and in some cases of other bound states. Let us discuss this in greater detail.

For instance, in the 42 660–45 360 cm^{-1} energy range there is a large number of even autoionization states with a total angular momentum $J = 0-4$ that can be excited by three photons of the radiation from a color-center laser.[11] At present the exact values of the widths of these states are unknown. It follows from Ref. 7 that the widths of autoionization states vary within an extremely broad range, from fractions of a cm^{-1} to several hundreds of cm^{-1}. The excitation frequencies for these autoionization states are designated in Fig. 1. From Fig. 1 it is clear that the autoionization states of different series are grouped at random. There are regions where the spectrum of autoionization states is dense and regions where it is sparse. A fairly dense spectrum of autoionization states exists in the frequency range corresponding to resonance peaks caused by excitation of singlet states. As noted earlier, it is the widths of these resonance peaks that are great. But in the frequency range corresponding to the resonance peaks caused by excitation of triplet states, the density of autoionization states is low ($\omega_c \simeq 9088$ cm^{-1}) or such states do not exist ($\omega_c \simeq 8270$ and 8278 cm^{-1}). Hence we can assume that a possible reason why the resonance peaks caused by excitation of singlet states are wide is the resonant or quasiresonant excitation of autoionization states through these bound states. Earlier we observed such quasiresonant excitation of autoionization states in the ionization of Ba atoms in the visible spectral range.[3]

It should also be noted that the great width of the resonance peak at 8915 cm^{-1} may have another reason, one related to the appearance of two resonance peaks at close frequencies (see Fig. 3b) from different initial states. One peak is caused by the resonance process of ionization of the Ba atom from the excited $6s6p\,^1P_1^0$ state ($\omega_m = 8915$ cm^{-1}; see Table I), and the other ($\omega_m = 8938$ cm^{-1}) by the resonance process of ionization of the Ba atom from the $6s^2\,^1S_0$ ground state of the atom[12] (via the three-photon excitation of $5d6p\,^1F_3^0$ or via the following sequence of transitions: $6s^2\,^1S_0 + 3\omega_c \rightarrow 5d6p\,^1F_3^0$ and $5d6p^2\,^1F_3^0 + \omega_c \rightarrow 6s7d^3D_2$).

Let us now consider the features of multiphoton excitation of autoionization states in the investigated frequency range. We believe that in the N^+ vs ω_c dependence only the resonance peak at 9075 cm^{-1} can be assumed to be caused by excitation of autoionization states with $J = 1$ (see Table I). At the same time the N^+ vs ω_c dependence does not exhibit many resonance peaks caused by excitation of autoionization states. These peaks are absent in regions where the density of autoionization states is high (where these peaks cannot be resolved in the frequency ω_c) and in regions where the density of autoionization states is moderate (where they are positioned quite far apart and where the corresponding peaks could be accurately resolved). There may be several reasons for this: the great width of the autoionization states, or the low probability of multiphoton excitation of autoionization states, or a strong perturbation (broadening) of the autoionization states in the laser field. Note that the large broadening of autoionization states in a laser field has been observed in the ionization of Sr atoms in the visible spectral range.[15]

We also note that our earlier investigations have shown that multiphoton excitation of autoionization states of alkali-earth atoms from ground states has a small effect on the yield of singly charged ions.[1,3] The resonance peaks caused by excitation of these states are much lower than those caused by excitation of bound states, and usually there are no such peaks. An exception is some autoionization states of the Sr atom whose excitation in the visible range leads to resonant peaks in the Sr^+ ion yield whose heights are comparable to those of peaks caused by excitation of bound states.[4] At the same time we never observed the multiphoton excitation of autoionization states of the Sr atom in the IR range.[13]

Thus, the results of our study prove that the low probability of excitation of autoionization states of different parities is characteristic of the ionization of alkali-earth atoms not only from the ground state[1,3,4] but from the excited $6s6p\,^1P_1^0$ state as well.

As noted earlier, in addition to identified resonance peaks, the N^+ vs ω_c dependence contains unidentified peaks of various amplitudes. The most interesting are the peaks (see Fig. 3a) at 8803 and 8848 cm^{-1}, since their amplitudes are comparable to those of peaks caused by two-photon excitation of singlet and triplet bound states. To acquire extra information about these resonance peaks we carried out additional investigations. In order to excite and ionize the Ba atom we used the same laser as in the previous investiga-

tions, but the radiation of the dye laser and that of the color-center laser were circularly polarized. The result can be seen in Fig. 3b.

Figure 3b shows that only the resonance peak at 8848 cm^{-1} remains when the radiations are circularly polarized. This suggests that the nature of the unidentified peaks is different or that these peaks may be caused by excitation of states with different values of the total angular momentum J. Indeed, according to selection rules, with circular polarization the only states that can be excited are those in which the variation of total angular momentum between the initial and final states is $\Delta J = \mp K$, where K is the number of absorbed photons, while in the case of linear polarization we have $\Delta J \leqslant \pm K$ (see Ref. 9).

To explain the nature of the peaks at 8803 and 8848 cm^{-1}, let us examine the various processes that may cause them. There are two types of processes: one involves only one laser whose light acts on the Ba atoms, the other both lasers. We start with processes related to the action of only one laser on the Ba atom. Analysis has shown that the peaks at 8803 and 8848 cm^{-1} are not caused by dipole and quadrupole excitation of bound states and autoionization states in the ionization of the Ba atom from the $6s^2\ ^1S_0$ ground state and the excited $6s6p\ ^1P_1^0$ state. Nor are they caused by resonant ionization of Ba atoms from the $6s5p\ ^1D_2$ state.

Now let us examine the various processes that emerge when the two laser fields act on the Ba atom. Analysis shows that the peaks at 8803 and 8848 cm^{-1} are not caused by processes arising as a result of the sum of photons of the two fields, i.e., processes of the type $\omega_c+\omega_d=E_0$ and $\omega_d+2\omega_c=E_0$, where E_0 is the level energy. Analysis shows that in the energy range under investigation there are no levels involved in resonant processes of the first type. As for processes of the second type, in this energy range there can be levels whose excitation could lead to such processes. But they cannot explain the occurrence of the two peaks since their frequencies do not coincide with those of the analyzed peaks.

The unidentified peaks mentioned are not caused by various Raman excitation processes either. Analysis shows that ionization of the Ba atom from the excited $6s6p\ ^1P_1^0$ state can lead to many such processes. The energies of the states that can become excited can be found from the following conditions: $\omega_d+(4-n)\omega_c-\omega_d+n\omega_c=E_0$ $(n=1-3)$ and $\omega_d+(3-n)\omega_c-\omega_d+n\omega_c=E_0$ $(n=1,2)$. However, the frequencies ω_c at which such Raman processes can be realized do not coincide with the frequencies ω_m corresponding to the unidentified resonance peaks just noted, and the N^+ vs ω_c dependence does not contain the respective resonance peaks.

Neither are autoionization-like resonances the cause of these peaks. Such resonances can emerge if the radiation at one frequency induces a transition between an unpopulated bound state and a state in the continuum populated by radiation of another frequency.[16] The energies E_0 of the resonance states in such processes are found from the conditions that $\omega_d+2\omega_d-3\omega_c=E_0$ and $\omega_d+3\omega_c-\omega_d=E_0$. One such autoionization-like resonance described by the first condition can be realized at $\omega_c=8842$ cm^{-1} and must involve the $5d7p\ ^3D_1^0$ level. Note that this frequency is close to that of one unidentified peak ($\omega_m=8848$ cm^{-1}). We believe, however, that this coincidence is purely accidental, since in our case a large number of such resonances do not manifest themselves at other frequencies and with other states.

Analysis shows that the second condition can also result in many autoionization-like resonance peaks. However, these resonances cannot be the cause of the peaks under discussion because their frequencies do not coincide with those of the peaks.

A possible reason for the emergence of the above-noted resonance structure at 8803 and 8848 cm^{-1} could be the effects associated with the complicated nature of the processes of perturbation, excitation, and ionization of the Ba atom in two laser fields, which take place in the case at hand (Fig. 4). Let us consider this problem in greater detail.

The nature of perturbation and excitation of Ba atoms as the frequency of the color-center laser varies within a range corresponding to the localization of the resonance structure under discussion (8803 and 8848 cm^{-1}) is indeed complicated and differs from that of the excitation of the Ba atom at other frequencies ω_c. The reason is that in this range we find the frequency $\omega_c=8845$ cm^{-1} corresponding to the one-photon transition $6s6p\ ^1P_1^0 \rightarrow 6s5d^3D_2$ (see Fig. 4*A*). In our experiments the color-center laser field was rather high for the one-photon transition, so that saturation was possible. As noted earlier, in this case one state, say, the $6s5d^3D_2$ state, may be populated very effectively from the $6s6p\ ^1P_1^0$ state. Our estimates did indeed show that such a one-photon transition occurs in conditions of saturation, when $\Omega > \gamma$. For these estimates we took the data on the probability of the $6s6p\ ^1P_1^0 \rightarrow 6s5d^3D_2$ transition from Ref. 17. Hence in contrast to the other frequencies ω_c, at which the perturbation of the Ba atom is largely determined by the one-photon transition $6s^2\ ^1S_0 \rightarrow 6s6p\ ^1P_1^0$, the perturbation near $\omega_c=8845$ cm^{-1} can occur owing to two one-photon transitions: $6s^2\ ^1S_0 \rightarrow 6s6p\ ^1P_1^0$ and $6s6p\ ^1P_1^0 \rightarrow 6s5d^3D_2$.

Moreover, near this frequency there may be ionization of the Ba atom not only from the $6s6p\ ^1P_1^0$ state but also from the $6s5d^3D_2$ state. And the ionization from these states occurs through a quasiresonance with the same state $6s8p\ ^1P_1^0$. The continuum energy in the realization of these two ionization channels is the same, which may lead to channel interference. It is this complicated pattern of perturbation, excitation, and ionization of the Ba atom that could give rise to the resonance structure at 8848 cm^{-1}.

Note that a similar nature of perturbation, excitation, and ionization of the Ba atom can be realized in the vicinity of $\omega_c=9027$ cm^{-1}, which corresponds to the one-photon transition $6s6p\ ^1P_1^0 \rightarrow 6s5d^3D_1$ (see Fig. 4*B*). However, the N^+ vs ω_c dependence exhibits no resonance structure near 9027 cm^{-1}. The reason may be that the one-photon transition $6s6p\ ^1P_1^0 \rightarrow 6s5d^3D_1$ is unsaturated. Indeed, according to Bizzarri and Huder,[17] this transition is approximately 30 times less probable than the $6s6p\ ^1P_1^0 \rightarrow 6s5d^3D_2$ transition, which means that the condition $\Omega_2 < \gamma_2$ may be met, with the result that saturation does not set in and there is no effective population of the $6s5d^3D_1$ state.

Thus, there is a qualitative explanation for the resonance peak at 8843 cm^{-1}, but this cannot be assumed final. At the same time the question of the origin of the resonance peak at 8803 cm^{-1} remains open. Note that the resonance peaks at 8803 and 8848 cm^{-1} can be caused by different processes or by excitation of different resonance states, since (as noted earlier) they manifest themselves differently when the radiation of the color-center laser is circularly polarized. A possible explanation of why there are resonance peaks at 8803 cm^{-1} and, perhaps, at 8848 cm^{-1} and other unidentified peaks may lie in the processes related to the special features of aligned atoms that form in the excitation of Ba atoms into the $6s6p\,^1P_1^0$ state by linearly polarized radiation,[8] or in the peculiarities of the ionization of Ba atoms in two laser fields, one of which is in resonance with the atomic transition. Note also that at certain laser frequencies ($\omega_d = 18\,060$ cm^{-1} and $\omega_c = 8845$ or 9027 cm^{-1}) there can be two resonant transitions in the Ba atom. Further studies are needed to interpret the unidentified resonance peaks in the N^+ vs ω_c dependence and the laws governing the emergence of such peaks unambiguously, to establish the patterns of multiphoton excitation of triplet bound states and autoionization states, and to clarify the features of ionization of the Ba atom in two laser fields.

4. CONCLUSION

In our studies of the ionization of the Ba atom from the $6s6p\,^1P_1^0$ state we discovered

(1) resonance peaks of unknown origin that at present cannot be explained unambiguously;

(2) a high probability of two-photon excitation of triplet odd states;

(3) a low effectiveness of autoionization states in multiphoton ionization of the Ba atom by three photons.

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