Multiphoton ionization of Ba atoms in two fields of laser radiation

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We study multiphoton ionization of Ba atoms initiated by the combination of two laser beams, one from a YAG laser ($\omega_1 = 9395\ \text{cm}^{-1}$) and the other from a tunable dye laser ($\omega_2 = 16\,200$–$17\,000\ \text{cm}^{-1}$). Of the many known processes that can result in the ionization of Ba atoms in two laser fields, we detected only the excitation due to the sum of the frequencies of the two beams ($\omega_1 + \omega_2$) and the Raman excitation described by the difference $2\omega_2 - \omega_1$. We also detected a resonance structure that has proven impossible to identify with any known process. © *1995 American Institute of Physics.*

1. INTRODUCTION

Studies have recently begun of processes involving multiphoton ionization of atoms initiated by two fields of laser radiation. Interest in such studies is due to the fact that when several fields of laser radiation act on an atom, various processes emerge that are impossible when only one field acts on the atom. One manifestation of such processes in multiphoton ionization is a resonant increase in the ion yield. These processes include, for example, resonances at the sum of frequencies of the beams used in the experiment, various Raman excitation processes, and autoionization-like resonances.[1,2]

The relationship describing the first process is

$$E = n_1\omega_1 + n_2\omega_2,$$

where E is the energy of the excited state, ω_1 and ω_2 are the lasing frequencies of the first and second lasers, and n_1 and n_2 are integers that specify the number of absorbed photons of the respective radiation. The relationships describing the Raman excitation processes are

$$E = n_1\omega_1 - n_2\omega_2, \quad E = n_2\omega_2 - n_1\omega_1.$$

Autoionization-like resonances are described by

$$E = k_1\omega_1 - \omega_2, \quad E = k\omega_2 - \omega_1,$$

where k_1 and k_2 are the number of photons necessary for the ionization of an atom with beams whose frequencies are ω_1 and ω_2, respectively.

Thus far, multiphoton ionization in two laser fields has largely been studied with alkali metal atoms (see, e.g., Refs. 1, 3, and 4). In this paper we present the results of the first studies involving Ba atoms over a broad spectral range. For instance, we conducted experiments in multiphoton ionization of Ba atoms that were simultaneously in two laser fields: one from a YAG laser ($\omega_1 = 9395\ \text{cm}^{-1}$), and the other from a tunable dye laser ($\omega_2 = 16\,200$–$17\,000\ \text{cm}^{-1}$). Note that the ionization of Ba atoms only by the YAG laser and only by the dye laser requires $k_1 = 5$ and $k_2 = 3$ photons, respectively. We also note that when both beams act on Ba atoms simultaneously, all three processes mentioned above are possible.

2. THE EXPERIMENT

The experimental layout is shown in Fig. 1. In our experiments the beam from the YAG master oscillator *1* is amplified and split into two beams. One is frequency-doubled by a KDP crystal *5*, the output of which pumps the dye laser *6*. Mirror *8* is used to spatially combine the beam from this laser with the second beam from the YAG laser, and the two are focused onto a beam of Ba atoms *11*.

The linewidth of the dye laser was 2–3 cm^{-1}, and the two lasers had the same the pulse width, 4×10^{-8} s. The field strength generated within the focal spot by the beam from the YAG laser was $\mathscr{E} \approx 3\times10^6\ \text{V/cm}^{-1}$. The field strength generated by the dye laser varied with frequency; the maximum value was $\mathscr{E} \approx 4\times10^5\ \text{V/cm}^{-1}$.

We measured the yield of Ba^+ ions when both lasers act on the Ba atomic beam simultaneously and when only one of the two beams acts on the atoms. Both linear and circular polarizations of the two beams were tested, and the experiments were carried out with different combinations of the polarizations of the two lasers.

Note that when the Ba atoms were ionized only by the dye laser, the yield of Ba^+ in the interresonance gaps under the present conditions was at the level of the detector background noise, while in ionization by the beam from the YAG laser this yield was above the noise level. When both beams were applied, the yield of Ba^+ ions was only slightly above the yield registered in ionization by the YAG laser alone.

3. EXPERIMENTAL RESULTS

Figure 2 depicts the results of our experiments under conditions in which both beams were linearly polarized and their electric field vectors were parallel or perpendicular to each other. Clearly, when the two beams (dye laser and YAG laser) act on the atomic beam simultaneously, the yield of Ba^+ ions exhibits an additional resonance structure that is lacking when the dye laser alone acts on the atomic beam. The nature of this additional resonance structure varies, depending on the orientation of the two electric field vectors.

For example, when Ba atoms are ionized only by the dye laser, the yield of Ba^+ ions exhibits two resonance peaks due to ordinary two-photon excitation of the $5d7s\ ^3D_2$ ($\omega_2 = 16\,470\ \text{cm}^{-1}$) and $5d7s\ ^1D_2$ ($\omega_2 = 16\,900\ \text{cm}^{-1}$)

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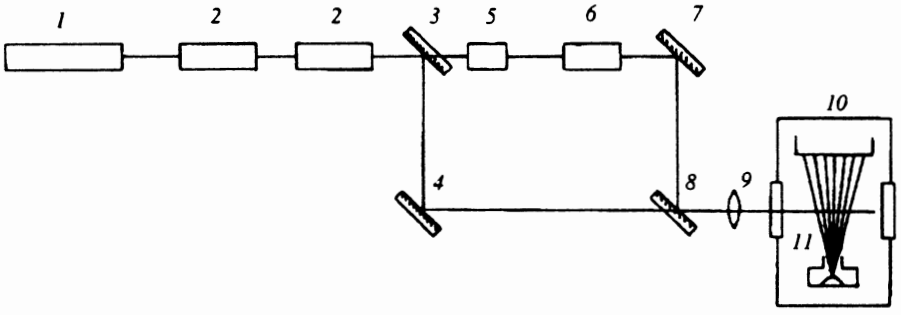

FIG. 1. Experimental layout: *1*—YAG master oscillator; *2*—laser amplifiers, *3*—YAG laser beamsplitter; *4*—total internal reflection mirror for the YAG laser beam; *5*—KDP crystal; *6*—dye laser; *7*—total internal reflection for the dye laser beam; *8*—dichroic mirror reflecting the dye laser beam and transmitting the YAG laser beam; *9*—lens; *10*—vacuum chamber; and *11*—beam of Ba atoms.

bound states. In Fig. 2 these peaks are labeled *A* and *B*. The excitation diagram for these levels is shown in Fig. 3a. As Fig. 2a implies, adding the beam from YAG laser to the one from the dye laser does not increase the height of these peaks compared with the case in which there is only the beam from the dye laser. Note that earlier[5] we thoroughly studied the resonance structure emerging in the ionization of Ba atoms by a dye laser covering the same range.[5]

Figure 2 demonstrates that when the two lasers act simultaneously on the Ba atom beam and the electric field

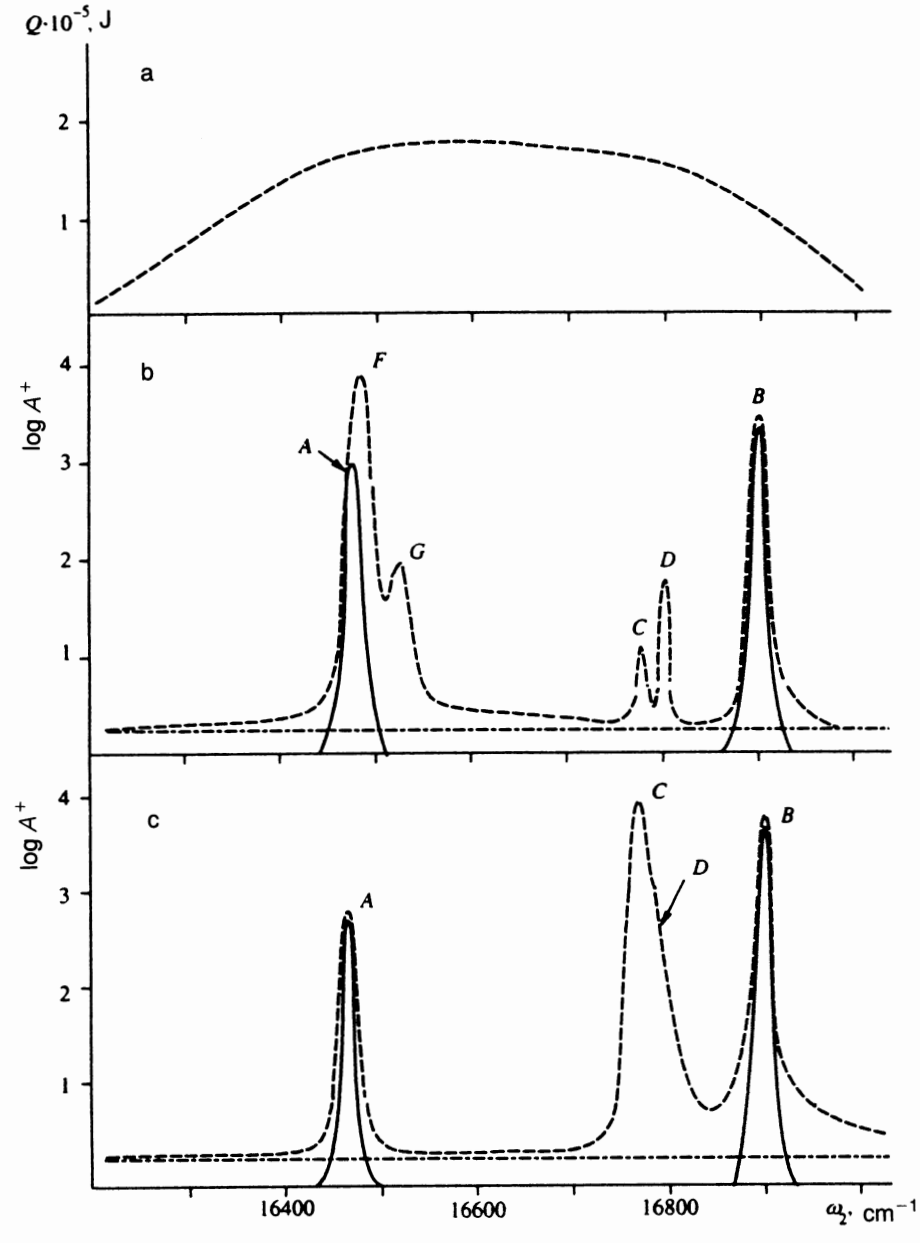

FIG. 2. Yield of Ba^+ ions as a result of ionizing Ba atoms with a YAG laser and a dye laser when both beams are linearly polarized. The electric field vectors are perpendicular (a) and parallel (b) to each other. Solid curves represent the yield of Ba^+ ions when only the dye laser ionizes the Ba atoms, dot-dash lines represent the yield of Ba^+ ions when only the YAG laser ionizes the Ba atoms, and dashed curves represent the yield of Ba^+ ions when both beams ionize the Ba atoms. Figure 2c shows the frequency dependence of dye laser energy.

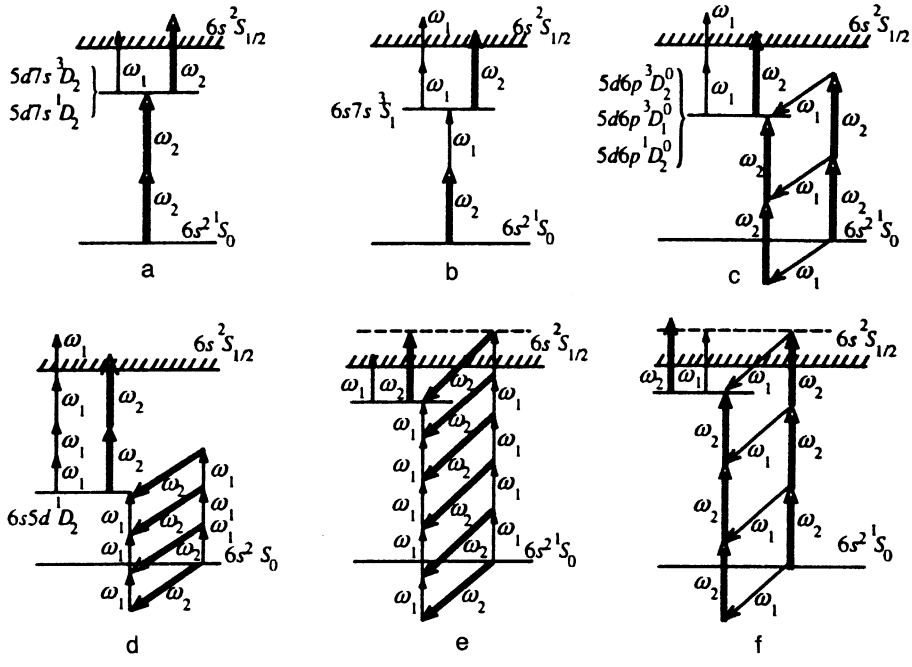

FIG. 3. Diagrams of the relevant processes. Explanations are given in the main text.

vectors are perpendicular, the Ba^+ ion yield displays a broad, high-amplitude, asymmetric peak at $\omega_2=16\,765$ cm^{-1} (C in the figure). When the electric field vectors are parallel, two narrow peaks appear in the vicinity of this peak. One coincides in frequency with peak C ($\omega_2=16\,765$ cm^{-1}), and the other occurs at $\omega_2=16\,795$ cm^{-1} (peak D in the figure). Under these conditions, peak C is lower than peak D. This information makes it possible to explain the asymmetry and large width of the additional peak that appears when the electric field vectors are perpendicular to each other. Obviously, this peak is the sum of two maxima of different amplitude, the higher peak at $\omega_2=16\,765$ cm^{-1} and the lower at $\omega_2=16\,795$ cm^{-1}. In the case of parallel electric field vectors, the first peak (C) is suppressed (the slight increase in the yield of Ba^+ ions in this case is apparently due to the electric field vectors not being perfectly parallel), while the second peak remains.

Furthermore, for parallel electric fields, a new resonance structure not present in the "perpendicular" case emerges. For instance, a high resonance peak appears at $\omega_2=16\,475$ cm^{-1} (peak F in Fig. 2a) and a small resonance peak appears at $\omega_2=16\,520$ cm^{-1} (peak G).

Note that the additional resonance structure emerging in the ionization of Ba atoms in two laser fields has both a lower amplitude and a larger amplitude than the resonances that appear in the ionization of Ba atoms by the dye laser alone (i.e., that result from ordinary two-photon excitation of bound states).

We have also studied the behavior of the additional resonance structure in the yield of Ba^+ ions for other polarization combinations of the two lasers. We found that when the YAG laser is circularly polarized and the dye laser is linearly polarized, peak C is approximately five times lower than the peak in Fig. 2a, and peak F is ten times lower than in Fig. 2b. Peaks D and G are suppressed almost completely.

When both beams are circularly polarized, peak C is 25 times lower than in Fig. 2a, and the other peaks (D, F, and G) are entirely suppressed.

Analysis has shown that there are only two peaks in the additional resonance structure that can be uniquely identified when two laser fields act on a Ba atom. For instance, peak C ($\omega_2=16\,765$ cm^{-1}) results from the excitation of the $6s7s\ ^3S_1$ level via resonance with the frequency sum: $E\ (6s7s\ ^3S_1)=\omega_1+\omega_2$ (Fig. 3b); peak D ($\omega_2=16\,795$ cm^{-1}) comes from the excitation of the $5d6p\ ^3D_1^0$ level via the Raman process, with $E\ (5d6p\ ^3D_1^0)=2\omega_2-\omega_1$. Both processes start from the $6s^2\ ^1S_0$ ground state of the Ba atom. These identifications are based on the behavior of the maxima for various combinations of the beam polarizations in the experiments, and the selection rules for the transitions that occur in the two laser fields.

Let us now consider excitation of the $6s7s\ ^3S_1$ triplet state as a result of the frequency-sum resonance. As Fig. 2 implies, the height of the peak corresponding to this process is comparable to the height of the peak resulting from ordinary excitation of the $5d7s\ ^1D_2$ singlet state (peak B). Thus, our results show that the probability of exciting triplet states of alkaline-earth atoms is comparable to the probability of exciting singlet states, not only when two identical photons are absorbed (as follows from the relative heights of peaks A and B in Fig. 2 and from the results of Ref. 6), but also when the two photons are different.

The peak produced by Raman excitation of the $5d6p\ ^3D_1^0$ triplet state is lower than the one just considered, probably due to the Raman excitation process requiring more photons than the frequency-sum process.

Note that in addition to the foregoing Raman excitation of the $5d6p\ ^3D_1^0$ level under our experimental conditions, two more Raman excitation processesare possible, and are described by the same relations that describe the excitation of the $5d6p\ ^3P_1^0$ level, $E=2\omega_2-\omega_1$ (Fig. 3c). For instance,

when $\omega_2=16\,222$ cm^{-1}, excitation of the $5d6p\ ^1D_2^0$ level becomes possible; at $\omega_2=16\,963$ cm^{-1} the $5d6p\ ^3D_2^0$ level may become excited. However, there are no corresponding peaks in the Ba^+ ion yield. A possible reason for their absence is the fact that the two frequencies land on the edge of the dye-laser lasing band, and the intensity of radiation at these frequencies is lower than at $\omega_2=16\,795$ cm^{-1}, the frequency at which the Raman excitation of the $5d6p\ ^3D_1^0$ level is manifested.

Note that three photons must be absorbed for the aforementioned Raman excitation processes to occur. Under our experimental conditions, however, Raman excitation with the absorption of four photons also becomes possible. In particular, Raman excitation of the $6s5d\ ^1D_2$ level, as described by $E(6s5d\ ^1D_2)=3\omega_1-\omega_2$, becomes possible at $\omega_2=16\,790$ cm^{-1} (Fig. 3d). There is no distinct peak, however, in the Ba^+ ion yield at that frequency.

The reason may be that ionization of atoms from the $6s5d\ ^1D_2$ states requires the absorption of more photons (four photons from the YAG laser or two photons from the dye laser) than required for ionization from the $6s7s\ ^3S_1$ state considered above (two photons from the YAG laser or one photon from the dye laser), where the peak actually shows up.

We also note that the Ba^+ ion yield exhibits no peaks that can be ascribed to a large number of autoionization-like resonances described by $E=3\omega_2-\omega_1$ and $E=5\omega_1-\omega_2$ (Figs. 3e and f). The reason for their absence is still unknown.

Thus, of the many processes that can result in multiphoton ionization of Ba atoms in two laser fields, we have detected only frequency-sum excitation processes and a single Raman excitation process involving the absorption of three photons.

One more important result of our investigations was the detection of an additional resonance structure in the yield of Ba^+ ions generated by two laser fields, which cannot be identified with any of the foregoing processes known to take place in two laser fields. The peaks in question are labeled G and F in Fig. 2. Peak F turns out to be much higher than the peaks resulting from ordinary two-photon excitation of bound states. Note that in establishing the nature of these peaks, we analyzed the possibility of the aforementioned processes originating not just in the $6s^2\ ^1S_0$ ground state, but in the first excited $6s5d\ ^3D_1$, $6s5d\ ^3D_2$, and $6s5d\ ^3D_3$ metastable states as well. We were unable, however, to identify the processes emerging from these states. Further studies are needed to establish the nature of these peaks.

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