Emission in transitions to the ground and metastable states of Ba^+ during two-photon ionization of barium vapor by a XeCl* laser beam

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Emission has been achieved in transitions to the ground state of Ba^+ during two-photon ionization of barium atoms by the beam from a XeCl* laser. The threshold conditions for emission have been found. The degree of ionization and the pumping efficiency are calculated.

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Progress in the development of ultraviolet lasers is leading to some interesting new experiments on photoionization. A case of particular interest is the photoionization of some element accompanied by the simultaneous excitation of the resulting ion, i.e., the case with $\Delta E = nE_f - E_i \geqslant E_R$, where n is the number of photons causing the ionization, E_f is the energy of a laser photon, E_i is the ionization potential of the atom, and E_R is the excitation energy of the resonant state of the ion. In this case, many-photon ionization of the medium could be observed by monitoring the ion emission spectrum, and in the case of a resonant photoionization it would be possible to achieve lasing action in transitions of the ion. Lasers using monohalides of inert gases would make it possible to arrange this process for many elements with as few as $n = 2$ photons.

In this letter we are reporting emission in transitions to the ground and metastable states of the barium ion during the two-photon ionization of barium by the beam from a XeCl* laser. We have chosen this combination (barium vapor and the XeCl* laser) because the value of ΔE for the XeCl* laser with only $n = 2$ is 22 927 cm^{-1}, which is some ~900 cm^{-1} larger than E_R for Ba^+. The positive value of this energy difference slightly away from resonance should promote the preferential excitation of the $6p^2P^0_{3/2}$ state of Ba^+. Furthermore, the beam from a XeCl* laser is at a quasi-resonance with the $6s^2\ ^1S_0 - 6s7p^1P^0_1$ transition of the Ba atom (the beam is ~70 cm^{-1} from the resonance for the 0, 1 band and ~50 cm^{-1} for the 0, 0 band), so that there should be a resonant increase in the ionization efficiency. At the typical power levels of XeCl* discharge lasers, ~10^6 W, this transition is saturated. Figure 1 illustrates the excitation of resonant levels of Ba^+ through two-photon ionization of Ba by a XeCl* laser.

The apparatus used in the present experiments consisted of an excimer laser, a cell holding a column of barium vapor, a grating monochromator, a photomultiplier, and an oscilloscope. We used the ÉLAN-01 excimer laser developed by the ISÉ institute of the Siberian Branch of the Soviet Academy of Sciences. The ordinary resonator in this laser was replaced by an unstable resonator with $M = 4$ in order to increase

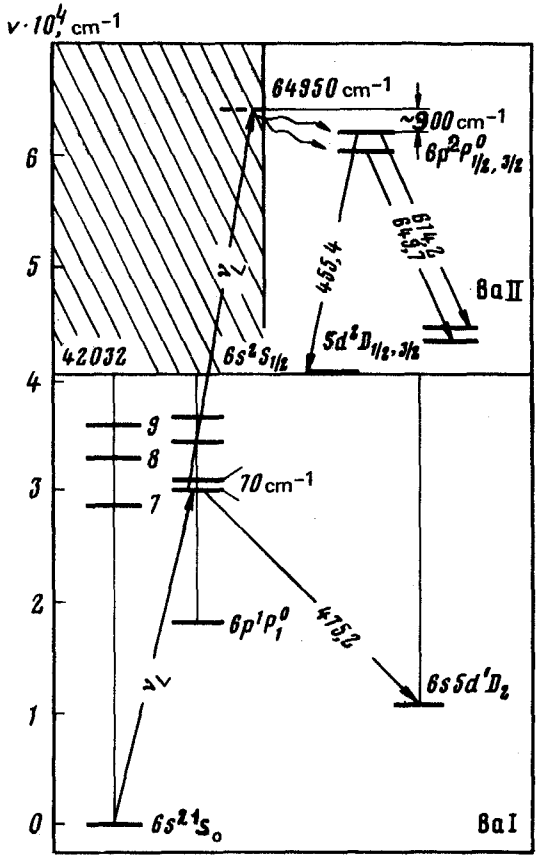

FIG. 1.

the power density when the beam was focused into the cell holding the barium vapor. The laser produced an output of 20–30 mJ at $\lambda = 308$ nm in $t \sim 10$ ns with a divergence of 0.3×10^{-3} rad. The cell holding the column of barium vapor was of the usual type for the active elements of metal-vapor lasers. A column of the vapor was produced inside a beryllium ceramic tube 1.1 cm in diameter and 40 cm long. The laser beam was focused into the central part of the cell by a lens with $f = 60$ cm. The buffer gas was He at a pressure 0.1–0.5 atm. The laser beam which was transmitted through the cell and also the radiation emitted inside the cell, in both the "forward" and "backward" directions, were incident on the monochromator.

When the cell temperature reached 600° C we observed spontaneous emission. When the temperature was raised further we observed a superradiance at the lines with $\lambda = 614.2$ nm and $\lambda = 649.7$ nm, which are the familiar Ba^+ red lines in work on metal-vapor lasers, and also a superradiance corresponding to a transition to the ground state, with $\lambda = 455.4$ nm. To the best of our knowledge, emission to the ground state has previously been observed only in atomic spectra during the photodissociation of molecules, e.g., in the transition $^2P_{1/2} - {}^2P_{3/2}$ of I during the photodissociation of fluorine- and iodine-substituted hydrocarbon molecules. This is the first observation of emission in a transition to the ground state of an ion.

The superradiance intensity increases rapidly with increasing temperature up to 850° C, where it stabilizes. This stabilization of the radiation energy correlates with a previous observation[1] of the appearance of intense stimulated Raman scattering at $\lambda = 475$ nm (Fig. 1). The efficiency of the stimulated Raman scattering in terms of the number of photons reaches 30%, so that the saturation of the output power for the emission corresponding to transitions of the ion results from a lowering of the pump power density due to the stimulated-Raman loss.

The observation of these lasing effects is evidence that the photoionization in the present case is selective. The selectivity is extremely high, since the $^2P_{3/2}$ sublevel is excited more effectively than the other sublevel of the $6p^2P^0_{1/2,\,3/2}$ state, with $\Delta E = 1691$ cm^{-1}. The $^2P_{3/2}$ level is closer to resonance with the pump. At present, we cannot definitely say which of the photoionization mechanisms—direct photoionization to the continuum or photoionization through an autoionization state—is operating in the present case. Although autoionization resonances are narrow,[2] they typically have a cross section of 10^{-15} cm^2, so that the autoionization mechanism appears extremely probable.

From the observed lasing action we can draw some inferences regarding the two-photon ionization. Visual observation of the emission from the end of the cell near the threshold pump level revealed that the emission builds up near a "neck" of the pump beam and that the maximum cross-sectional area of the emission region is 1–2 mm^2 (for the red lines). In this case the barium vapor density was 2.2×10^{14} cm^{-3}, and the pump power density was 6×10^7 W/cm^2. Using the empirical condition $k_0 l \geqslant 30$ for the onset of superradiance, we can estimate the degree of ionization of the medium and the electron density n_e. When the cross-sectional area of the emission region was 2 mm^2, the length of the active zone was ~9 cm. If we assume a Doppler shape for the Ba^+ lines, we can estimate the difference between the densities of particles in the $^2P_{3/2,\,1/2}$ and $^2D_{5/2,\,3/2}$ states: $\Delta N = 2 \times 10^{12}$ cm^{-3}. Using this value of ΔN as a lower estimate of the Ba^+ state density, summed over all the Ba^+ states considered, we find a lower estimate ~10^{13} cm^{-3} for the electron density n_e. The degree of ionization of the Ba is thus ~10^{-1}, and the efficiency of the two-photon ionization in terms of the pump is $n_e/N_f \cong 10^{-3}$.

In connection with this observed selectivity of the two-photon photoionization we should point out that this process leads to a rapid emission of electrons (in $t \sim 10$ ns) with energies of the order of that corresponding to the distance from the resonance and with an energy distribution which reflects the degree of selectivity. In the present case, electrons with an energy ~0.1 eV are produced. This type of ionization is quite different from the conventional ionization methods such as electric discharges and electron beams. It is not difficult to see that the recombination of the plasma produced during the selective photoionization should also have some distinctive features; in particular, the recombination should occur more rapidly than for a gas-discharge plasma.

In addition to the emission in the ion spectrum, we observed emission at six visible lines of barium atoms belonging to a triplet system of levels. The pumping mechanism for these transitions is apparently of a recombination nature.[3]

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