Comment on "Optical pumping to the $6s5d$ states in atomic barium"

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In a barium vapor pumped with laser light on the resonance transition at 553.5 nm the level $6p\ ^1P_1$ is depopulated mainly by collisional processes. With neglect of this item, radiative lifetimes derived from fluorescence measurements come out systematically too short.

In a recent paper Myers *et al.*[1] reported upon the time-dependence of $6s^2\ ^1S_0$-$6s6p\ ^1P_1$ (553.5 nm) resonance fluorescence in atomic barium using switch-on cw laser excitation. As a buffer gas Myers *et al.* used argon at a pressure of 133 hPa. Despite constant laser irradiation Myers *et al.* found a decay in the fluorescence signals, whereby the falloff of the fluorescence was faster at higher laser intensities.

The effect can be explained by a transfer of laser-excited barium atoms into lower-lying metastable states. For a more quantitative interpretation Myers *et al.* applied a simplified rate-equation model in which only radiative transfer from the pumped $6p\ ^1P_1$ level to the $5d\ ^1D_2$ level (1.5 μm) and to the $5d\ ^3D_2$ level (1.13 μm) was taken into account. With an extrapolated fluorescent decay constant of $(1.6\pm0.3)\times10^6\ s^{-1}$ Myers *et al.* derived a total spontaneous decay rate of $(2.1\pm0.4)\times10^6\ s^{-1}$ to the metastable states mentioned above.

In our opinion collisional processes cannot be neglected at all.[2] Several authors, e.g., Refs. 3-6, have seen a strong population of the $6p\ ^3P_2$ level in comparable laser-pumping experiments. Kallenbach *et al.*[7] concluded that spin-changing collisions with buffer-gas atoms (A) are responsible for the population of the $6p\ ^3P_2$ level

$$\mathrm{BaI}(^1P_1)+\mathrm{A}\rightarrow\mathrm{BaI}(^3P_2)+\mathrm{A}+\delta E\ .$$

For argon as a collision partner they extrapolated a cross section of $(1.5\pm0.5)\times10^{-20}\ m^2$ from their experimental curves. Applying this value to the results of Myers *et al.* and taking into account a repopulation of the ground state by quenching collisions from the $6p\ ^3P_2$ level as well as spontaneous emission from the $6p\ ^3P_1$ level we find a total decay rate of $1.5\times10^7\ s^{-1}$ from the pumped $6p\ ^1P_1$ level when applying a more extensive rate-equation model. To this total decay rate, spontaneous emission contributes less than 10%. Hence we consider the procedure Myers *et al.* applied to be inadequate for an explanation of the decaying fluorescence curves. Especially, the sum of the transition probabilities Myers *et al.* derived from their model calculations should be too high by about a factor of 5, which is consistent with other experimental and theoretical results Myers *et al.* cited in their paper.

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