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EXCITATION AND IONIZATION OF LASER-PUMPED Ba VAPOUR

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Résumé — L'excitation et l'ionisation de vapeur dense de Baryum (10¹⁰ à 10²¹ m⁻³) par irradiation de lumière laser résonnante (λ = 553.5 nm) est décrite. On discute les processus qui transforment l'ionisation de l'énergie d'excitation des atomes. On trouve que l'efficacité de l'ionisation dépend de la densité et qu'une ionisation collisionnelle est dominante. Il a été établi que des électrons sont chauffés dans des collisions super-élastiques avec des atomes excités par laser et que subseqüemment l'excitation et l'ionisation — ainsi que la photoionisation des niveaux élevés — entraîne la création d'un nombre croissant d'électrons. Le transfert d'énergie aux électrons par collisions super-élastiques prend place non seulement du niveau de résonance pompé par laser, mais aussi des niveaux métastables de Ba qui se trouvent en dessous. Ces niveaux étaient peuplés immédiatement via le niveau de résonance.

Abstract — We describe the excitation and ionization of dense Ba vapour (10¹⁰ to 10²¹ m⁻³) by resonant (λ = 553.5 nm) laser radiation and discuss the processes responsible for the transfer of energy from the laser-excited atoms into ionization. Ionization was found to be density-dependent and this pointed to collision-dominated ionization mechanisms. It has been established that seed electrons were heated in superelastic collisions with laser-excited atoms, and that subsequent electron-impact excitation and ionization — as well as photoionization of high-lying levels — lead to the creation of more electrons. The observed transfer of the excitation energy to the electrons by superelastic collisions requires contributions not only from the laser-pumped resonance level, but also from the lower-lying metastable Ba levels. These were, in fact, found to be very efficiently populated via the resonance level.

I — INTRODUCTION

A dense metal vapour ionizes when irradiated by resonant laser light. This process, discovered by Lucatorto and McIlrath in 1976, has been the object of intensive investigations in the past few years.

A number of elements — Li, Na, Ca, Sr and Ba — have been studied at densities ranging from 10¹⁸ to 10²² m⁻³ and several recent studies focused on the time-resolved excitation and ionization of laser-pumped Ba vapour.²⁻⁸ Some authors³,⁶,⁷ favour radiative processes as the dominant ionization mechanism; others²,⁵ consider collisional processes as being mainly responsible for the ionization and thus confirm the original conjecture⁵ that the dominant energy transfer from laser-excited atoms takes place by superelastic collisions with seed electrons. The effective energy transfer by superelastic collisions had originally been pointed out by Measures⁶, and the effect of such collisions has recently been demonstrated in the electron spectrum originating from laser-excited Ba-atoms in a beam.¹¹

We describe here the results of the most complete study of the excitation and ionization of resonantly irradiated Ba vapour⁵, and discuss — in the light of these results — several further studies.

II — EXPERIMENT

The experimental arrangement used in ref. 5 is shown in Fig. 1. Ba vapour with densities ranging from 10¹⁰ to 10²¹ m⁻³ in ca. 5 × 10⁻³ Pa (40 Torr) of Ar buffer gas was contained in a furnace and was irradiated.
by a flash-lamp pumped dye laser. This pump laser, which was tuned to the Ba resonance line ($\lambda = 553.5$ nm), generated a 1-\musec long pulse with a peak irradiance of 7 GWM$^{-2}$. The transient populations of the Ba levels in the laser-excited vapour could then be sampled by a 1-\musec long pulse from a broad-band dye laser, which illuminated a Mach-Zehnder interferometer — comprising the furnace in its test arm — and a stigmatic spectrograph. The delay of the 1-\musec sampling pulse could be chosen in the range from zero to 20 \mu s after the start of the 1-\musec long pump-laser pulse.

The combination of the broad-band laser, the Mach-Zehnder interferometer and stigmatic spectrograph enabled us to record the anomalous dispersion near spectral lines by use of the so-called hook method in a single 1-\musec exposure on Polaroid film. The hook method yields the product $f \int N(z) dz$, where $f$ is the oscillator strength of the spectral line in question and $\int N(z) dz$ is the column density of the lower level of the transition of interest, i.e. the local density $N(z)$ integrated over the column length $l$. (For the sake of simplicity we will from now on write the column density as $N_l$.) Given the oscillator strengths $f$ for the spectral lines observed in the spectrograph, column densities and — with some geometrical assumptions — estimates of the densities of several levels could be obtained.

The hook-method diagnostics comprised the Ba ground state $6s^2 \, ^1S_0$, the metastable levels $5d \, ^3D_{1,2,3}$, $5d \, ^1D_2$, $6p \, ^3P_{0,1,2}$ and the resonance level $6p \, ^1P_1$, as well as the ground state $6s \, ^3S_{1/2}$, the metastable level $5d \, ^3D_{0/2}$, and the resonance level $6p \, ^3P_{3/2}$ of Ba$^+$. (cf. Fig. 2). Accordingly, the population histories as shown in Figs. 3 through 6 were recorded by varying the delay of the 1-\musec broad-band laser pulse with respect to the 1-\musec pump-laser pulse. It was also possible to photoelectrically monitor the emission spectrum, and thus the populations of high-lying levels. In the case of the emission measurements, however, optical thickness and radiation trapping had an influence and consequently were taken into account.

It is worthwhile noting that anomalous dispersion measurements show no saturation and that the sampling light-pulse is not subject to an appreciable delay (beyond that imposed by the speed of light in vacuum), because the refractive index at the wavelengths where the dispersion is measured (i.e. away from the line centers) is close to unity.

III — RESULTS

The resonance radiation from the pump laser burnt through the 0.23-m long vapour column of $1.46 \times 10^{20}$ Ba atoms m$^{-2}$ within ca. 40 ns and thus created a uniformly excited column within that time. The metastable terms $5d \, ^1D$ and $^3D$ were found to be populated almost simultaneously with the resonance level $^1P_1$; this can be explained by stimulated radiative processes at the infrared wavelengths 1.13 and 1.5 \mum. Given some seed electrons, the copious laser-excited and metastable Ba-atoms were then able to
transfer their stored energy by superelastic collisions. In turn, the energy gained by an electron in such collisions was soon sufficient so that it could further excite and ionize the Ba atoms. The additional free electrons thus created then contributed to a chain reaction which resulted in a nearly complete ionization of the Ba-vapour within ca. 1 µs.

These basic processes as well as the curious, fast population of the metastable 6p 3P0 term, which is not accessible by dipole-transitions from the laser-excited 6p 1P1 level, are demonstrated and discussed in more detail in the following paragraphs.

Fig. 2 - Simplified level diagram (from ref. 5).

Fig. 3 (left) - Logarithmic population histories of the Ba ground state (6s 1S0), the laser-pumped resonance level (6p 1P1) and of a metastable level (5d 1D2). As shown in the later case, ca. 30 data points have been taken for each curve representing a population history. The groundstate column density before the laser pulse was \( N_0 = 1.48 \times 10^{20} \text{ m}^{-2} \) \((\text{corresponding to } N_0 = 6.3 \times 10^{10} \text{ m}^{-2})\). Argon buffer gas \((5.3 \times 10^3 \text{ Pa}, \text{i.e. } 40 \text{ Torr})\) was present. The pump laser, whose pulse shape is shown (with linear right hand ordinate), had a peak irradiance of 7 GW m\(^{-2}\).

Fig. 4 (right) - Logarithmic population histories for the metastable Ba levels 5d 3D1,2,3 and 6p 1P0,1,2, recorded under the same conditions as those of Fig. 3.
In Fig. 3, which shows a semi-logarithmic plot of the laser pulse and logarithmic population histories of the ground state $6s^2 \, 1S_0$, the laser-excited level $6p \, 1P_1^0$ and of the metastable $5d \, 1D_2$ level, we see that the density ratio $N(6p \, 1P_1^0)/N(6s^2 \, 1S_0)$ becomes constant at $t \simeq 40 \, ns$; at that time the ratio assumes the value 3 (corresponding to 0.5 dex), i.e. the ratio of the statistical weights of the two levels. Thus it was concluded that the laser irradiance was sufficient to burn through the 0.23-m long vapour column within ca. 40 ns; from that time on we are dealing — to a reasonable approximation — with a uniformly excited column.

We also note from Fig. 3 that the population of the metastable level $5d \, 1D_2$ follows closely that of the laser-excited level $6p \, 1P_1^0$: the density ratio again corresponds to 5/3 (or 0.22 dex), i.e. to the ratio of the statistical weights, at $t \simeq 45 \, ns$. The stimulated infrared emission that leads to the rapid equipartition between $1P_1^0$ and $1D_2$ has been observed in a related experiment.\(^6\) (In this context we might mention that the 1.5- and 1.13-\mu m radiation has earlier been observed in Ba lasers.\(^13,14\))

The similar, fast rise of the density of $5d \, 3D_2$ can also be ascribed to the rapid transition $5d \, 3D_2 \rightarrow 6p \, 1P_1^0 (\lambda = 1.13 \, \mu m)$ caused by amplified spontaneous emission.\(^5\) The corresponding infrared light—pulse has been observed.\(^6,8\) The population of the other two fine-structure levels $J = 1$ and $J = 2$ of the $3D$ term is achieved by fine-structure mixing collisions involving the Ar buffer-gas atoms. Such collisions are energetically possible at the prevailing temperature ($T = 870 \, C$) because of the fine-structure splitting $\Delta E < kT$; in addition, the fine-structure components $J = 1$ and 3 have been shown to be more rapidly populated both at higher Ba and at higher buffer-gas pressures.\(^6\)

The behaviour of the population of the $6p \, 3P_0^0, 1, 2$ levels is more complex, as seen in Fig. 4. The fast and efficient population of the level $6p \, 3P_2^0$ has not yet been explained quantitatively. A fast decay via stimulated dipole-radiation from the laser—excited $6p \, 1P_1^0$ level (as was the case for the other metastables $5d \, 1D_2$ and $3D$) is not possible here, since the $3P_0^0$ term has the same parity as the laser—excited level. A number of energy—pooling collisions that are energetically conceivable at $kT \simeq 0.1 \, eV$, namely $6p \, 1P_1^0 + 5d \, 3D_{1,2} \rightarrow 6p \, 3P_2^0 + 6p \, 3P_0^0$ with $\Delta E = 0.008$ and 0.031 eV, respectively, had to be excluded — especially for the extremely short delays ($t \simeq 5 \, ns$) when appreciable population of $3P_2^0$ was observed, but sufficient $3D_{1,2}$ populations were absent.\(^5\)

A similar energy—pooling collision ($5d \, 1D_2 + 5d \, 1D_2 \rightarrow 5d \, 3D_2 + 6p \, 3P_2^0, \Delta E = 0.040 \, eV$) would have required a cross—section exceeding by at least a factor of ten those observed for similar collisions in Na and Cs.\(^6\) It has been suggested therefore that stimulated electronic Raman scattering (SERS) from the laser—pumped level $6p \, 1P_1^0$ to the $6p \, 3P_0^0$ term via the quasi—resonant levels $5d \, 3D_{1,2}$ could be responsible for the fast ($t \simeq 5 \, ns$) excitation of the level $6p \, 5P_0^0$.\(^5\) This view has been disputed: it has been claimed, albeit without quantitative upper limit, that the corresponding anti—Stokes radiation could not be measured.\(^6,7\)

The difference in the behaviour of the fine—structure components $6p \, 3P_0^0, 1, 2$ can be explained more easily: the near—coincidence of the transition $6p \, 3P_1^0 - 6d \, 3D_2$ at $\lambda = 553.586 \, nm$ (cf. Fig. 2) with the 553.548-nm resonance line results in some depopulation of the $6p \, 3P_0^0$ level by the pump—laser radiation. To prevent excessive excitation in this additional channel, the 15-\mu m laser bandwidth was centered on the blue side of the resonance line, namely at $ca. 553.52 \, nm$.\(^5\) (The equivalent width of the resonance line at our column densities was about 0.5 nm, so that this detuning had no influence on the efficiency of the pump laser.)

Künne Meyer and Kock have shown that tuning the pump laser onto the coincident $3P_1^0 - 3D_1$ line does, in fact, appreciably increase the ionization efficiency.\(^7\) (It should be noted however that the maximum ionization observed in that experiment was only 2% of the total Ba density — presumably because the laser did not burn through, when tuned near resonance.)

In summary, we note that the earth—alkali Ba shows a different behaviour from that of the alkalis, not only because of the fortuitous wavelength coincidence between the resonance line and a transition from a metastable, but because the presence of metastable levels in general leads to a much larger reservoir of excited atoms. It will be seen below that the energy stored in the metastable levels is indeed needed to explain the fast ionization by superelastic collisions.

The pronounced dip around 1 \mu s, that is common to all level densities of neutral Ba in Figs. 3 and 4, reflects the peaking of the densities of the Ba$^+$ levels at the same time, as shown in Fig. 5.

In Fig. 6 we show the population histories for an initial Ba density which is 50% higher than that used for Figs. 3 through 5. Here the ionization is faster than at the lower density.
Correspondingly, the ionization was found to occur more slowly at lower initial densities, and — as seen in Fig. 7 — the peak ionization efficiency is also strongly density dependent. This behaviour points toward a collision-dominated ionizing process. Nevertheless, it cannot be excluded that seed electrons are created by radiative processes. In particular, it has been suggested that the 20% ionization seen in Fig. 7 for the lowest density \(10^{19} \text{ m}^{-3}\) may almost entirely be due to two-photon ionization and to population of high-lying levels by SERS with subsequent photoionization by the laserlight.6

First, the time dependence of the energy density in the Ba vapour is compared with that calculated with superelastic collisions. In short, one finds that the energy transfer from the laser-excited level \(6p^2P_{3/2}\) alone is insufficient, so that superelastic collisions with the numerous metastable atoms are also needed.5 In analogy to the case of He, it was assumed that the electron collision cross-sections for the resonance level...
and for metastable levels have a similar size. The important rôle played by the metastables in transferring the energy to the electrons was confirmed with Sr in an experiment by Bréchignac and Cahuzac\textsuperscript{18}, where the laser was tuned to the intercombination line $5s^2 1S_0 - 5p^3 P^0_1$: quite similar ionization signals (in that case ion fluorescence) had been observed as with resonance excitation. It was also established\textsuperscript{5} that the observed energy density in the Ba vapour was proportional to $N^+ N_e$, i.e. to the product of the density of excited levels and electron density, rather than to $N^+^2$, as it were the case, if energy-pooling collisions were dominant.

Second, the observed ionization time was shown to be compatible with dominant electron-impact excitation. In this case, a population of the levels lying above the laser-excited resonance level, as it is present with local thermodynamic equilibrium (LTE), is essential. An LTE population has been confirmed from emission measurements.\textsuperscript{5} In fact, a coarse indication for the population of high-lying levels is obvious to the naked eye: the green resonance fluorescence of the laser-excited vapour changes to white radiation, when the initial Ba density is high enough for efficient ionization to occur.

The plasma parameters needed for modeling the ionization processes are shown in Fig. 8: the electron density $N_e$ was obtained from the sum of the densities of the Ba$^+$ configurations $6s$ and $5d$, and the two Boltzmann temperatures $kT_B$ and $kT_{H_a}$ could be derived from the relative populations of the Ba$^+$ ground state $6s^2 S_{1/2}$ and, respectively, the metastable and resonance levels $5d^2 D_{5/2}$ and $6p^3 P_{1/2}$. The two Boltzmann temperatures agree with each other within their error limits, and it can be shown that for electron densities $N_e$ exceeding $10^{20}$ m$^{-3}$, i.e. for delay times $t \geq 140$ ns under the present conditions, LTE is established.\textsuperscript{5}

It was concluded from a model that a combination of excitation of high-lying levels by "ladder climbing" as well as collisional and radiative ionization from high-lying Ba levels was sufficient to achieve the observed ionization in the required time.

In a more recent, unpublished investigation, Bachor, Bone and Sandeman have used a circular furnace and an interferometric arrangement with a streak camera to obtain spatially resolved data on the laser-excitation of Ba at similar densities and laser irradiances as those used in ref. 5. The most striking finding was that all processes — excitation as well as ionization — extend well beyond the diameter of the exciting laser beam, with all observed excited levels occupying volumes of order 10$^2$ times the laser-pumped volume. While it is not too surprising that a combination of spontaneous decay (fluorescence) of the laser-excited resonance level and radiation trapping will create an appreciable number of atoms beyond the volume occupied by the pump beam, it is rather remarkable that Ba atoms in the "curiously" excited $6p^3 P_{1/2}$ level were also found to be present in the volume surrounding that of the pump-laser beam. It remains to be seen, whether the extensive excitation of $3P_{1/2}$, which occurs considerably slower than that of the resonance level, can be explained by energy-pooling collisions of the kind mentioned above.

An entirely unexpected result of the described experiment on the laser excitation and ionization of Ba was the need for a correction of the oscillator strengths for some of the Ba I lines used in the hook-method diagnostics. It was observed that the apparent total column density $\sum N_i f_i$ of barium (which is derived from measures of the products $N_i f_i l_i$ for the different lower levels $l$ with the aid of supposedly well-known oscillator strengths $f_i$) was reduced by a factor of about two at ca. 50 ns, from that before the excitation started. Furthermore, it appeared that the full initial Ba density was restored at ca. 500 ns, i.e. after the probed vapour had been fully ionized. Since at a delay time of 50 ns the atoms were almost exclusively in the low-lying levels of neutral Ba (i.e. in levels up to the laser-excited resonance level, cf. Fig. 2), it was reasonable to assume a scale shift of the oscillator strengths for absorption lines originating from these levels as being responsible for the apparent loss of Ba atoms.

In a detailed study, which is being prepared for publication, it has been shown that the surmised scale shift of the oscillator strengths in question can be substantiated and be brought into accordance with the results of recent lifetime measurements for excited levels of Ba.

**IV — CONCLUSIONS**

The strong density dependence of the ionization efficiency found in the described experiments strongly suggests that a collision-dominated process is responsible for the fast ionization observed in resonantly excited dense vapours.
A model involving superelastic collisions of the laser-excited atoms (in both the laser-pumped as well as in the metastable levels) with seed electrons can explain the observed growth of energy density in excitation, ionization and electron motion. It could also be quantitatively verified that further electron impact excitation leads to ladder climbing of the excitation up to the ionization limit with subsequent collisional ionization and photo-ionization by laser light.

The mechanism (or mechanisms) responsible for the rapid (5 ns) population of the \( 6p \ 3P \) level, still poses questions. Spatially resolved studies of this excitation are expected to yield further insights.

A further result of this work is a correction of several Ba-I oscillator strengths by a factor of two.

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12 dex designates intervals in powers of ten.