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Cascade-Induced Alignment Changes of Intensity-Decay Curves

H. G. BERRY, L. J. CURTIS,* AND J. L. SUBTIL

Laboratoire de Physique de l'Atmosphère, Université de Lyon 1, 43, boulevard du 11 Novembre 1918, 69-Villeurbanne, France

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We study the effects of changes of alignment of an atomic level due to cascading as a function of time after excitation. For many of the decays observed in beam-foil spectroscopy, we expect the cascade-induced alignment to be close to zero. Accordingly, we have found that the difference between multiexponential decay curves measured in light polarized parallel and perpendicular to the beam may contain only a single exponential characteristic of the level lifetime, thus providing a cascade-free lifetime measurement. By similar considerations, we have also used alignment information to reduce or eliminate unresolved blending in lifetime measurements.

INDEX HEADINGS: Spectra; Source; Helium; Lithium.

The precision of lifetime measurements from beam-foil decay curves is generally limited by cascading effects from other excited terms. We introduce a technique for reducing or eliminating this cascading, which makes use of the nonisotropic nature of this excitation source.

Radiation emitted after beam-foil excitation may exhibit appreciable polarization,[1] which implies that magnetic substates are not equally populated by the source. This partial alignment can produce modulations in time-resolved multiplets,[2] but those from singlets will not be modulated as long as the decays take place in the absence of external electric and magnetic fields. Our consideration will be restricted to unmodulated decay curves. Cascade repopulation subsequent to the initial excitation will, in general, produce a change in alignment of the level concerned. For many situations, cascade-induced alignment can be expected to be close to zero. Thus, since cascade effects often complicate the determination of a level lifetime (which is the same for substates differing only in magnetic quantum number), the polarization-analyzed decay curves can be used to reduce cascade effects in lifetime measurements. Further, since the polarization of a desired transition and that of its unresolved blends usually differs, polarization information can be used to reduce or eliminate blending.

Accordingly, we herein report a lifetime measurement that was made by subtracting the two (relatively normalized) polarization components of the multiexponential decay curve of a level from each other, to obtain a single-exponential decay curve characteristic of the level lifetime. By similar considerations, we have also used alignment information to reduce or eliminate unresolved blending in lifetime measurements.

EXPERIMENTAL ARRANGEMENT

Beams of He^+ and Li^+ ions were obtained from a 2-MeV Van de Graaff accelerator and directed through a 10-μg cm^{-2} carbon foil. Optical radiation was collected at 90° to the beam direction, passed through a rotatable polarizer, and focused on to the entrance slit of a 60-cm Czerny–Turner monochromator. Single photons of the desired frequency were detected with an EMI 6256 photomultiplier at the exit slit, amplified, and pulse counts were recorded with a scaler. For each transition studied, two decay curves were measured: $I^{\parallel}(t)$, measured with light polarized with the electric-field vector parallel to the beam axis, and $I^{\perp}(t)$, measured with light polarized with the electric-field vector perpendicular to the beam axis. The ratio of detection efficiencies for parallel and perpendicularly polarized light, denoted k, was measured through the study of unpolarized S-state transitions in the wavelength regions of interest. The instrumental polarization was small, less than 1% over a wide range of wavelengths.[3]

THEORY

The instantaneous population of a beam-foil-excited atomic state contains two separable contributions: One

is the remnant of its initial foil-excited population, and a second results from its continuous repopulation by cascades from higher states. These two contributions are independent, and can be considered individually and the results superposed. For a substate M of a degenerate level J, repopulated by cascades from the states $J'M'$, the instantaneous population $N_{JM}(t)$ can formally be written[4]

$$N_{JM}(t)=N_{JM}(0)\exp(-t/\tau_J)+\sum_{J'M'}N_{J'M'}(0)\sum_{J''}C([A])\exp(-t/\tau_{J''}). \quad (1)$$

The initial foil-excitation contribution decays with a single exponential lifetime τ_J, which is common to all substates of the level. The cascade contribution involves a multiexponential sum for each cascade, where J'' is summed over J, J', and any indirect cascade levels that connect J with J'. The coefficients C are calculable from the various transition probabilities $[A]$ of the contributing states.

We wish to consider cases for which the foil-excited population differs among the individual M substates, but for which the cascade repopulation is the same for each M substate. In such cases, the decay curve of level J would consist of a polarized (single exponential) component and an unpolarized (multiexponential) component. Thus cascade effects could be eliminated by consideration of the experimental quantity

$$I^{\parallel}(t)-kI^{\perp}(t)\propto\exp(-t/\tau_J)+\text{cascade cancellations}. \quad (2)$$

These conditions would clearly be met for an aligned level repopulated by nonaligned cascade levels, or by aligned cascade levels for which the alignment is washed out through the $J'M'$ summation. The latter possibility can be examined theoretically.

The alignment of a level is usually specified in terms of the polarization fraction $P(t)$, defined at time t by

$$P(t)=\frac{I^{\parallel}(t)-kI^{\perp}(t)}{I^{\parallel}(t)+kI^{\perp}(t)}. \quad (3)$$

However, a related quantity, the fractional alignment T, is normalized to the total radiated intensity, and is useful in studying the transfer of alignment. It is defined

$$T=(I^{\parallel}-kI^{\perp})/(I^{\parallel}+2kI^{\perp}). \quad (4)$$

Macek[5] has given a general formulation for the transfer of alignment by cascade after excitation in the beam-foil process. Nedélec[6] has shown that the transfer of alignment can be written in 6-j notation as

$$\frac{T(J_2,J_3)}{T(J_1,J_2)}=\frac{\begin{Bmatrix}1&1&0\\J_1&J_1&J_2\end{Bmatrix}\begin{Bmatrix}1&1&2\\J_2&J_2&J_3\end{Bmatrix}\begin{Bmatrix}2&J_1&J_1\\1&J_2&J_2\end{Bmatrix}}{\begin{Bmatrix}1&1&2\\J_1&J_1&J_2\end{Bmatrix}\begin{Bmatrix}1&1&0\\J_2&J_2&J_3\end{Bmatrix}\begin{Bmatrix}0&J_1&J_1\\1&J_2&J_2\end{Bmatrix}}, \quad (5)$$

where $T(J_1,J_2)$ is the fractional alignment in the cascade $(J_1\rightarrow J_2)$, and $T(J_2,J_3)$ is the fractional alignment in the second transition $(J_2\rightarrow J_3)$. This ratio is unity for decreasing or increasing sets of angular momentum states (e.g., $F\rightarrow D\rightarrow P$ or $P\rightarrow D\rightarrow F$), but is generally less than unity. Fine structure and hyperfine structure will tend to decrease this ratio, thus reducing the fractional polarization induced in the second decay.

We observe that, in the beam-foil excitation mechanism, transitions from higher-angular-momentum states tend to exhibit less polarization, so that cascade-induced alignment along these branches will be small. Cascades from S states can induce no polarization. Thus, there are many cases in which cascade repopulation introduces only an unpolarized component into an already polarized decay curve. Then, a point-by-point subtraction of cascade-affected decay curves of the two polarizations, corrected for instrumental polarizations, should result in a detailed cancellation of the cascade contributions.

ALIGNMENT OF CASCADE-FREE LEVELS

If there is no cascade repopulation of a level, both polarization components will decay alike with the same single exponential, so that $P(t)$ should be constant, unless there are stray fields that depolarize the sample. To verify that our source was free of such depolarizing influences, we measured the transition at 5015 Å from the $3p$ ^{1}P term in He I, after observing from our spectra that its principal cascades from $4s$ ^{1}S and $4d$ ^{1}D were not strongly populated in our source. The absence of long cascade tails on this decay curve also permitted a study of possible polarization effects in the background subtraction. The background matched exactly the dark-count level of the phototube, and exhibited no polarization. After the background was subtracted, the decay curves of both polarizations could be fitted with a single exponential of lifetime 1.80 ns, in good agreement with previous measurements and theory.[7] The -1% instrumental correction was made; Fig. 1 shows that $P(t)$ for this transition is constant and equal to 10% for all statistically significant values of t. At the extreme tail, the polarization becomes masked by background fluctuations, but it is clear that polarization is not being reduced in the absence of cascading.

ALIGNMENT OF LEVELS REPOPULATED BY CASCADES

A measurement was made of the polarization-decay curves of the transition at 4787 Å from the $4d$ ^{1}D term in Li II. Cascading was considerable, and arises from higher ^{1}P and ^{1}F terms. The higher ^{1}P cascades should contribute essentially zero alignment.[8] Although we were unable to resolve the $3d$ ^{1}D–$5f$ ^{1}F° and $3d$ ^{3}D–$5f$ ^{3}F° transitions, their blend was measured to have a polarization of less than 1%. After subtracting

the unpolarized background and correcting for the instrumental polarization, a point-by-point subtraction of two multiexponential curves resulted in a single exponential curve, corresponding to the 2.5-ns mean life of the $4d$ ^{1}D term. Figure 2 shows the two polarization decay curves, their subtracted difference, and the subsequent change in the polarization function $P(t)$.

This technique should work quite generally to discriminate against nonpolarized cascade contributions to an initially polarized level, provided instrumental polarizations can be determined.

POLARIZATION FROM LINES WITH UNRESOLVED BLENDS

Blending effects have also been reduced by measuring decays with a polarizer. An extreme example exists for hydrogenic decays where the fine structure is unresolved, and the almost degenerate L states have different lifetimes. However, different L-state transitions have different polarizations; for example, S-state transitions are never polarized, while levels of P and D terms, etc., may be polarized to varying degrees. Thus, application of Eq. (2) to a blended line will certainly remove S-state contributions, and will change relative contributions of the other fine-structure levels, as well as removing unpolarized cascades. We have performed such a measurement for the He II $n=3$–5 3204-Å transition. Thus, a subtraction with Eq. (2) should contain at most three exponentials. At low energy (300 keV), the result was a single exponential corresponding to the lifetime of the $5f$ level and at higher energies the decay is faster, reaching the $5p$ decay time at 1.5-MeV beam energy. A similar result was obtained for the He II $n=3$–4 transition at 4686 Å.

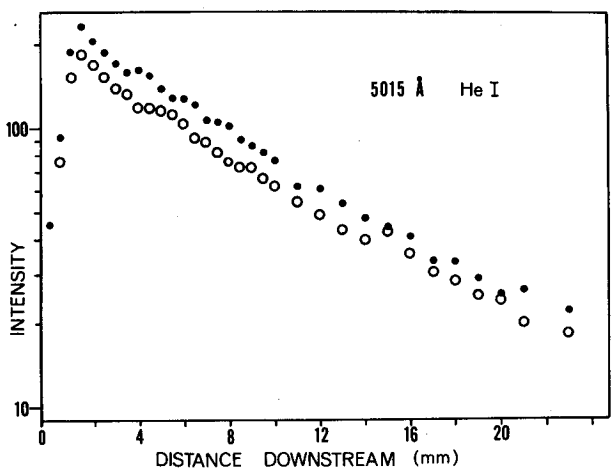

FIG. 1. Decay curves for the He I, $2s$ ^{1}S$-3p$ ^{1}P transition at 5015 Å, at a beam energy of 300 keV, measured in polarizations parallel (●) and perpendicular (○) to the beam axis. The polarization is 10% and constant for all times after excitation.

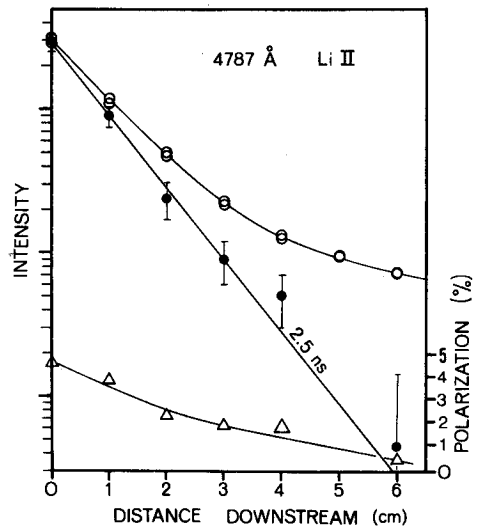

FIG. 2. Decay curves for the Li II, $3p$ ^{1}P$-4d$ ^{1}D transition at 4787 Å, at a beam energy of 500 keV, measured in polarizations parallel and perpendicular (○) to the beam axis, and their subtracted difference (●), multiplied by 10 for common normalization. The lower part of the figure (△) shows the change of polarization as the cascading fraction increases.

CONCLUSION

We have shown that polarization induced by cascading is generally small or zero in beam-foil decay-time measurements. It is then possible, by subtracting decay curves of two different polarization directions, to obtain single-exponential decay curves. The blending in hydrogenic decay curves can be similarly reduced or eliminated by use of these techniques.

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* Permanent address: The University of Toledo, Toledo, Ohio 43606.

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