# Influence of radiative trapping on precision measurements of nonresonance transition lifetimes in helium

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Radiative lifetimes of the $n\,^1S$ ($n=3$–$6$) and $n\,^3P$ ($n=3$–$5$) states in He I have been measured using pulsed excitation by a high-energy electron beam over a broad pressure range of the He target. The observed pressure dependences reveal that these nonresonance transitions are subjected to strong radiative trapping effects for higher pressures that may have distorted earlier precision measurements of He I lifetimes. This effect is particularly large for the $2\,^3S$–$3\,^3P$ transition for which published experimental values are, on the average, about 10% higher than accurate theoretical values.

## I. INTRODUCTION

Emission from excited levels $i$ to lower levels $f$ in atoms and molecules may be subject to repeated absorption and reemission processes before it finally escapes the light source. This reduces the effective decay rate of the transition from the spontaneous emission transition probability $A_{if}$ to an apparent value $A_{if}^{\mathrm{eff}}$ where

$$A_{if}^{\mathrm{eff}}=\gamma A_{if}\ ,\quad 0\leq\gamma\leq 1\ . \tag{1}$$

Here $\gamma$ is called by various names in the literature such as "trapping factor," "escape factor," or "imprisonment factor". If the trapping is assumed to take place within an infinite cylinder with radius $R$, an approximate formula[1] for $\gamma$ gives

$$\gamma=1-\frac{\pi}{2\sqrt{2}}k_0R\quad(k_0R\ll 1)\ , \tag{2}$$

where $k_0$ is the absorption coefficient at the center of the transition $i\rightarrow f$ given by

$$k_0=\frac{1}{8\pi^{3/2}}\left[\frac{M}{2kT}\right]^{1/2}\frac{g_i}{g_f}A_{if}\lambda_{if}^3N_f\ . \tag{3}$$

Here $N_f$ denotes the density of the lower state $f$, while the remaining quantities have their usual meanings.

The presence of radiative trapping has a profound influence on lifetime measurements of resonance transitions and too long $\tau$ values will always be registered unless the ground-state density of the atoms is very low. Repeated emission and absorption of resonance radiation may also signal its presence as a self-reversal of the emission lines. Thus, radiative trapping and self-absorption are intimately connected processes and if a considerable lengthening of a lifetime is observed for a spectral line because of trapping, the same line could then show self-absorption or self-reversal effects and vice versa.

While trapping and self-reversal of resonance transitions have been treated in a great number of papers, this is not the case concerning nonresonance transitions. However, it was demonstrated in our laboratory[2] that trapping effects also could yield a considerable distortion of lifetimes of nonresonance transitions. More specifically, lifetimes $\tau$ of the $1s_f$-$2p_i$ transitions in Ar I were measured using high-power electron excitation and the high-frequency deflection (HFD) technique[3] and a strong increase of $\tau$ was observed with increasing pressure of the argon target gas. The largest effects were observed for transitions terminating on the metastable ($J=0,2$) $1s_3$ and $1s_5$ levels and a considerable effect was also observed for the remaining transitions terminating on the ($J=1$) $1s_2$ and $1s_4$ levels. Thus the experiments revealed trapping effects in the (nonresonant) $1s_f$-$2p_i$ radiation indicating the creation of considerable population of the $1s_{3,5}$ levels (due to metastability) and the $1s_{2,4}$ levels (due to strong pumping following trapping of the $1p_0$-$1s_{2,4}$ resonance radiation). Similar effects should appear in all experimental situations where the ground-state density of the atoms is high enough and a sufficiently effective (nonselective) excitation is applied. In particular, analogous situations should occur in other rare gas atoms excited in various kind of light sources involving particle bombardment. As emphasized in Ref. 2 trapping of the nonresonance radiation may then appear in a treacherous way and escape the attention of the observer.

Subsequently, other authors have also discussed trapping of nonresonance radiation and some of them have been sceptical to the interpretation of our results (cf. Refs. 4 and 5). For instance, it has been suggested[4] that our observed lifetime prolongations are primarily due to disalignment effects and not to trapping effects. Accordingly, we have repeated our studies of the $1s_f$-$2p_i$ transitions in Ar I and searched for self-reversal effects using a conventional hollow-cathode lamp.[6] At a high spectral resolution (1:$10^6$) accomplished by a newly constructed scanning monochromator (the so-called MEGA spectrometer[7]) it was indeed found that all the studied $1s_f$-$2p_i$ lines in Ar I (and in Ne I) showed self-reversal effects when the lamp was operated at high enough power. Moreover, studies of a particular sequence $1s_5$-$2p_i$ showed that the self-reversal increased with increasing values of $k_0$ as calculated using relation (3). Thus, the

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observed self-reversal of the Ar I $1s$-$2p$ transitions support our original suggestion[2] that these transitions may also be subjected to trapping effects in lifetime measurements.

In the present work we have investigated trapping effects on nonresonance radiation in He I. This is a particularly convenient case for studies of this kind since the spectrum contains extended sequences of easy accessible lines with different transition probabilities to a common lower state. Moreover, the He I lifetimes have earlier been subjected to a great number of experimental investigations as well as highly accurate calculations.

## II. EARLIER LIFETIME MEASUREMENTS OF THE $n\,^1S$ AND $n\,^3P$ LEVELS IN He I

As follows from the partial level scheme of He I (Fig. 1), two sequences of transitions $2\,^1P-n\,^1S$ and $2\,^3S-n\,^3P$ are particularly well suited for studies of radiative trapping of nonresonance transitions. Thus, the $2\,^1P-n\,^1S$ sequence has transition probabilities ranging from $(18-0.18)\times10^6\ \mathrm{s}^{-1}$ for $n=3-6$ and considerable populations of the $2\,^1P$ state could be built up as a result of pumping following trapping of the very intense $1\,^1S-2\,^1P$ radiation if the ground-state population is high enough. Even larger populations should be created of the $2\,^3S$ state in view of the metastability of this state yielding a substantial trapping of, in the first place, the $2\,^3S-3\,^3P$ radiation ($\lambda=388.9$ nm). Figure 2 shows a compilation of 16 different experimental estimates of the lifetime for this transition (cf. Ref. 8 and references therein). Obviously, the spread in the results are much larger than the individually quoted errors and the average result $\tau=104\pm3$ ns (Table I) is larger than all theoretical results (cf. Refs. 8–10) that are in the range 94.7–98.1 ns with a stated accuracy of 1% or better.

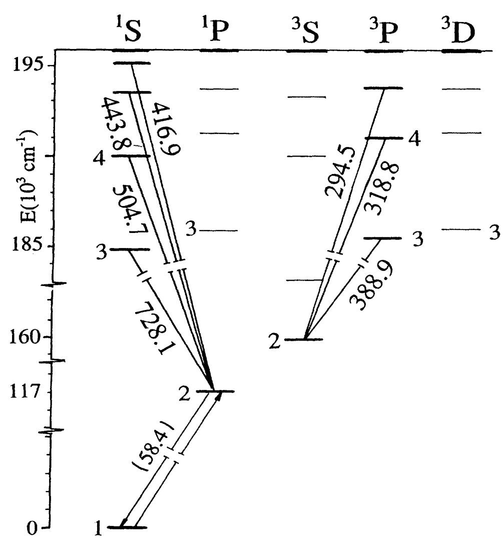


FIG. 1. Partial level scheme of He I indicating the nonresonance transitions ($\lambda$, nm) studied in the present lifetime investigations. At electron excitation of a helium target the intense 58.4-nm radiation might pump the $2\,^1P$ state up to a considerable population as a consequence of resonance trapping. Even larger populations of $2\,^3S$ may be created in view of metastability. In this way the indicated transitions might be subjected to trapping effects which should lengthen the lifetimes at higher pressures.

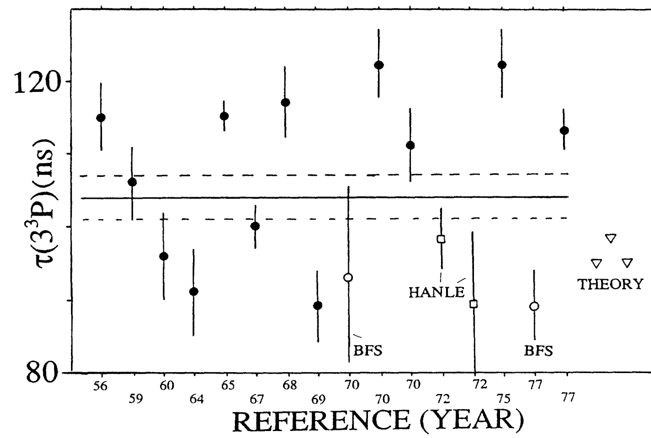


FIG. 2. Compilation of 16 different experimental determinations of $\tau(3\,^3P)$ in He I (see Ref. 8 and references therein). Solid circles denote measurements using electron excitation work using a static gas target. The average value, 104±3 ns is significantly higher than the indicated theoretical values (Refs. 8–10) and the results of the present work suggest that the discrepancy is due to radiative trapping. However, it may be noted that measurements using beam-foil technique (BFS) and the Hanle effect overlap the theoretical values.

The great majority of the considered $\tau(3\,^3P)$ lifetime measurements have been performed using electron excitation at various pressures and in our opinion the discrepancy between the average experimental value and theory is significant and caused by radiation trapping. This discrepancy has been pointed out earlier, for instance in Ref. 11, but the authors[11] suggest that the calculated values are not sufficiently accurate for a comparison with their experimental value ($=113\pm3$ ns) obtained via electron beam excitation of a 5–400-mTorr helium target.

In beam-foil excitation radiation trapping should in principle be absent in view of the very low densities, but unfortunately the $3\,^1P$ lifetime is a little too long for accurate determinations using this technique. Nevertheless, it is interesting to note that the two measurements in Fig. 2 denoted BFS have been estimated using beam-foil technique yielding 89±5 ns (Ref. 12) and 93±12 ns (Ref. 13), i.e., overlapping the theoretical values.

## III. EXPERIMENTS AND RESULTS

Lifetimes of the He I transitions displayed in Fig. 1 were measured using the high-frequency deflection technique using a 20-keV, 30-mA beam of electrons exciting a helium gas target. The emission was studied at various spectral resolution ranging from $5\times10^3$ up to $10^5$ using a 2-m Czerny Turner scanning monochromator equipped with different cooled photomultipliers, EMI 9789 SG (for $\lambda<600$ nm) and Hamamatsu R 943-02 (for $\lambda>600$ nm).

TABLE I. Lifetimes of low-lying $n\ ^1S$ and $n\ ^3P$ levels in He I (ns). A, this work. B, average experimental (see Ref. 8 and references therein). The figure within parentheses describes the number of considered measurements. C, theory, Ref. 8. D, theory, Ref. 9. E, theory complication 10 (with $A_{6s5p}$ and $A_{5p5s}$ scaled from Ref. 9). Sources: $3\ ^1S$, $4\ ^3P$; A. W. Weiss (unpublished). $4\ ^1S$,$6\ ^1S$; E. Trefftz, A. Scluter, K. H. Dettmar, and K. Jörgens, Z. Astrophys. **44**, 1 (1957). $5\ ^1S$, Coulomb approximation. $3\ ^3P$, B. Schiff and C. L. Pekeris, Phys. Rev. **134**, A368 (1964). $5\ ^3P$, A. Dalgarno and A. E. Kingston, Proc. Phys. Soc. London Ser. A **72**, 1053 (1958).

| Level | A | B | C | D | E |
|---|---|---|---|---|---|
| $3\ ^1S$ | 56.3±2.0 | 55.0±1.3 (6) | 55.3 | 54.7 | 55.2 |
| $4\ ^1S$ | 88.7±3.0 | 85.0±2.0 (11) | 88.8 | 86.4 | 89.8 |
| $5\ ^1S$ | 149±5 | 135±6 (9) | 148 | 147 | 150 |
| $6\ ^1S$ | 235±8 | 220±8 (4) | 234 | 233 | 240 |
| $3\ ^3P$ | $105^{+5}_{-10}$ | 104±3 (16) | 98.1 | 94.8 | 94.7 |
| $4\ ^3P$ | 164±7 | 144±7 (4) | 143 | 152 | 170 |
| $5\ ^3P$ | 245±15 | 186±14 (3) | 225 | 252 | 268 |

A very intense He I spectrum was observed and lifetime studies of the $n\ ^1S$ and $n\ ^3P$ levels are in principle straightforward using this technique even for high $n$ values ($n \leq 13$) as presented in Ref. 14. Since the aim of the present work primarily is to establish trapping effects, lifetimes of the $2\ ^1P-n\ ^1S$ and $2\ ^3S-n\ ^3P$ transitions were measured for lower $n$ values and the pressure dependence was carefully checked within a broad range (0.2–100 mTorr). The decay curves were registered at various sweep frequencies in order to look for possible cascade components, and the final determinations were made at 455 kHz. Examples of typical decay curves are displayed in Fig. 3 showing $\tau(3\ ^3P)$ at two very different target pressures (0.5 and 100 mTorr).

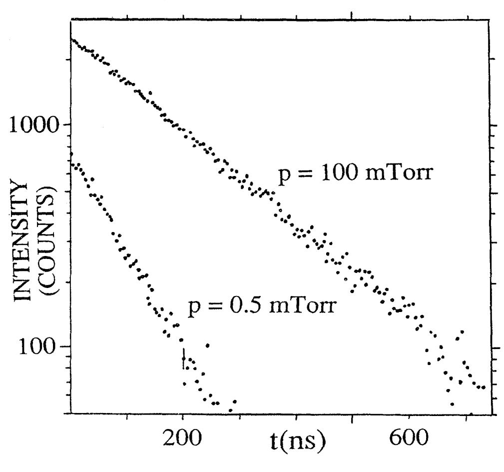


FIG. 3. The decay curve of the $3\ ^3P$ level in He I recorded at two different pressures and 455-kHz sweep frequency using the high-frequency deflection technique. While the recorded value at 0.5 mTorr is in close agreement with the average result of other measurements (see Fig. 2 and Table I), a 100% lengthening of the lifetime is observed at 100 mTorr which indicates the presence of strong radiation trapping effects of the $2\ ^3S-3\ ^3P$ radiation.

No traces of cascades are discernible and this is the case also for the remaining transitions. This negligible cascade feedings of the presently studied transitions have also been noticed by earlier investigators (cf. Refs. 12 and 15) and this is a consequence of the positions and lifetimes of the possible cascading states in He I. Thus, cascades of the $n\ ^1S$ states are negligible since they may only occur from the $(n'>n)\ ^1P$ states which have much shorter lifetimes and dominating branches to the $1\ ^1S$ state. In the case of a given $n\ ^3P$ state, the possible cascading states are $(n' \geq n+1)\ ^3S,D$ that have much smaller electron excitation cross sections and shorter lifetimes for the dominating $n'=n+1$ components.

As shown in Fig. 3 we find that $\tau(3\ ^3P)$ increases a factor of 2 when the He target pressure is increased from 0.5 to 100 mTorr. Since thermal collision deexcitation effects would yield an opposite effect (i.e., a shortening of $\tau$ with increased pressure), the observed lengthening should be due to strong trapping effects.

Figures 4 and 5 display our measured pressure dependences of the lifetimes of the $n\ ^1S$ and $n\ ^3P$ levels, respectively. The solid lines represent the expected pressure dependence if only thermal collision effects affect this dependence. The collision cross sections are extracted from Ref. 16 and the curves are matched to the associated experimental points at low pressures. It follows from Fig. 4 that $\tau(n\ ^1S)$ show trapping effects which gradually decrease with increasing $n$ being barely observable for $n=5$ at 100 mTorr, while for $n=6$ the pressure dependence follows the Stern-Volmer curve (i.e., it is dominated by thermal collision effects). This $n$ dependence is expected in view of relation (3) since the product $A_{if}\lambda_{if}^3$ has the values 7.0, 0.84, 0.27, and 0.13 ($10^{-12}$ $m^3s^{-1}$) for $n=3$, 4, 5, and 6, respectively. It is difficult to perform a more accurate estimate of the expected $n$ dependence since trapping of the branching transitions to the $n'\ ^1P$ states then have to be considered which would require knowledge of the populations of these levels. For $3\ ^1S$, however, there is only the $2\ ^1P-3\ ^1S$ branch and we find from Eq. (2) that the 30% lengthening of $\tau(3\ ^1S)$ observed at $p=100$ mTorr corresponds to $k_0R=0.21$ which ac-

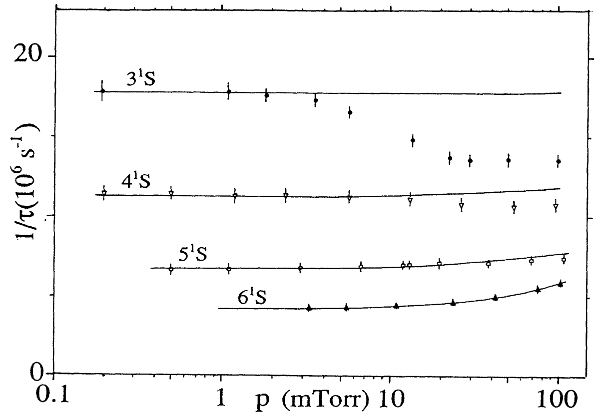


FIG. 4. Observed pressure dependences of the $n\ ^1S$ ($n=3-6$) lifetimes in He I compared to the expected behavior (solid lines) which are calculated under the assumption that only thermal collision effects are present (a Stern-Volmer plot). The large discrepancy between the experimental points and the Stern-Volmer plot is particularly pronounced for $n=5$ but decreases with increasing $n$ as expected in view of the relations (1)–(3) describing the trapping process.

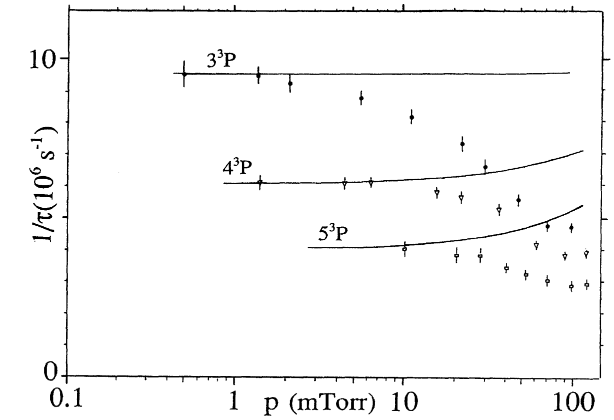


FIG. 5. Observed pressure dependences of the $n\ ^3P$ ($n=3-5$) lifetimes in He I compared to the expected Stern-Volmer behavior. The observed trapping effects are even larger than for the $n\ ^1S$ levels indicating that the population density of the metastable $2\ ^3S$ state is about 3 times larger than that of the $2\ ^1P$ state formed by pumping following resonance trapping of the $1\ ^1S-2\ ^1P$ radiation.

cording to (3) requires a population ratio $N(2\ ^1P)/N(1\ ^1S)$ of the order of $10^{-5}-10^{-4}$ for $R$ values in the range 1–10 cm. It seems very reasonable that such high densities of $2\ ^1P$ may be built up following pumping from the ground state via trapping of the very intense $1\ ^1S-2\ ^1P$ radiation.

The population density of the metastable $2\ ^3S$ state is about 3 times larger than that of $2\ ^1P$ at similar conditions as suggested from the pressure dependence of $\tau(3\ ^3P)$ displayed in Fig. 5 (indicating a 100% lengthening at 100 mTorr) and similar considerations using Eqs. (2) and (3). Since the product $A_{if}\lambda_{if}^3$ decreases only a factor of 7 between $n=3$ and 5, the trapping is still very pronounced for $\tau(5\ ^3P)$.

The zero pressure extrapolated lifetimes are given in Table I. As seen from Fig. 4 this extrapolation does not introduce more than a 3–4 % error in spite of the trapping at higher pressures. Our results are also in satisfactory agreement with theory as well as the average result of a number of experimental estimates. However, in the case of $\tau(3\ ^3P)$ the trapping is so large that it introduces a considerable uncertainty in the zero pressure extrapolation. Thus, if the intensities would allow measurements at still lower pressures it cannot be excluded that we would register even a few percent shorter lifetime. Accordingly we prefer to quote "asymmetric" error limits, i.e., $\tau(3\ ^3P)=104^{+5}_{-10}$ ns which is in close agreement with the average value of sixteen other measurements but some ten percent longer than the theoretical values. Having the demonstrated trapping effects in mind we prefer to recommend the theoretical $\tau(3\ ^3P)$ values as a "best estimate" rather than any of the experimental values. Our measured $\tau(4\ ^3P)$ and $\tau(5\ ^3P)$ agree satisfactory with theory and $\tau(4\ ^3P)$ also with other experiments, while other experimental determinations of $\tau(5\ ^3P)$ seem to yield a 25% shorter value than theory on the average. It may not be excluded that this shortening is due to cascades from the $6\ ^3S$ and $6\ ^3D$ levels. However, as stated above, no traces of such cascades were observed in the present measurements.

## IV. CONCLUSIONS

The present investigations of the pressure dependences of the lifetimes of the nonresonance $2\ ^1P-n\ ^1S$ and $2\ ^3S-n\ ^3P$ transitions in He I clearly demonstrate that they may be disturbed by radiation trapping effects which have to be considered in precision measurements. The suggestion emphasized in Ref. 4 that our measurements[2] were affected by disalignment effects and not by trapping cannot be correct since we have observed very strong trapping also of radiation from alignment-free $J=0$ levels in Ar I (Ref. 2), Ne I (Ref. 6) as well as the $n\ ^1S(J=0)$ levels in the present case of He I. We conclude that trapping of nonresonance radiation might occur at all possible kinds of light sources where a sufficiently efficient nonselective excitation is applied and the ground-state density of atoms is high enough. Unfortunately, these experimental conditions are necessary to create in high-resolution work in many cases where neither laser nor atomic beam techniques may be applied.

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