# Measurements of $4s4p\ ^1P$ lifetimes in the Zn I isoelectronic sequence: Ga II, Ge III, As IV

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The oscillator strengths for the $4s\ ^2{}^1S$-$4s4p\ ^1P$ resonance line in the Zn I isoelectronic sequence have been obtained for Ga II, Ge III, and As IV by means of lifetime measurements utilizing beam-foil spectroscopy at low initial ion energies. The energy dependence of the emission cross sections for the resonance transition and for the main cascades populating the resonance state in Ga II exhibit significantly different energy dependence, which allows an elimination or strong reduction in the influence of the cascades by performing the lifetime measurements at low initial ion energies, for Ga II at 100–150 keV. The experimental oscillator strengths are in good agreement with recent calculations by Froese-Fischer and Hansen.

## I. INTRODUCTION

The oscillator strengths for the principal resonance line in the Zn I isoelectronic sequence have recently become of significant interest for fusion studies since many high-temperature magnetically confined plasmas may contain heavy element impurities, sufficiently ionized to belong to the Zn I sequence.

Recently, five theoretical studies[1-5] have yielded oscillator strengths for resonance lines belonging to the Zn I sequence. For the ionic members of this sequence the theoretical oscillator strengths are all significantly larger than the experimental values available,[6-10] whereas the agreement is good for Zn I (Ref. 11). Weiss[1] calculated the oscillator strengths using intermediate coupling multiconfiguration wave functions. Cowan[2] performed relativistic Hartree-Fock calculations, whereas Shorer and Dalgarno[3] applied the relativistic random-phase approximation. All of these calculations only included correlations within the $n=4$ shell. Froese-Fischer and Hansen[4] included core polarization in addition to $n=4$ correlation, and Shorer[5] extended the random-phase approximation calculations by taking perturbation of the $3d$ core electrons into account. The experimental values reported[6-11] have all been obtained by means of the beam-foil technique.

The discrepancies between the experimental and theoretical oscillator strengths have been suggested[1,4] to originate from the fact that the lifetime measurements of the $4s4p\ ^1P$ levels could be strongly affected by severe cascading from higher-lying levels.[1,4] It is, however, in most cases possible to take the cascades populating the resonance level into account if they can be identified, but the level schemes for some of the members belonging to the Zn I isoelectronic sequence have been incompletely known. Very recently, Denne *et al.*[12] have revised the Ga II level scheme. The previously assumed $4s4d\ ^1D$ level is now assigned $4p^2\ ^1D$, and the missing $4s4d\ ^1D$ level is identified. Since it has not been possible for Sørensen[6] in 1973 to take the lifetime of the $4s4d\ ^1D$ level ($0.73 \pm 0.07$ ns)[12] in Ga II into account in the analysis of the $4s4p\ ^1P$ resonance lifetime, reported to be $0.65 \pm 0.08$ ns,[6] it is reasonable to assume that the experimental $^1P$ lifetime[6] is considerably affected by cascading from the $4s4d\ ^1D$ level.

In order to obtain a reliable lifetime for the $4s4p\ ^1P$ level in Ga II by means of the beam-foil technique, it is necessary to reduce the cascade contributions considerably, particularly from levels with lifetimes of the same order as the resonance level. Since the lifetime of the $4p^2\ ^1D$ level in Ga II is two orders of magnitude longer[12] than the lifetime of the resonance level, the main cascade problems are due to the decays from the $4s5s\ ^1S$ and $4s4d\ ^1D$ levels.

We have performed experimental investigations of the excitation functions for foil-excited $4s4p\ ^1P$, $4s5s\ ^1S$, and $4s4d\ ^1D$ Ga II levels and observed that it is possible to eliminate or very strongly reduce the cascades populating the $4s4p\ ^1P$ resonance levels at excitation energies as low as 100–150 keV. We have utilized this "threshold excitation" technique to reinvestigate the lifetimes of the resonance levels in Ga II, Ge III, and As IV. The advantages of performing beam-foil spectroscopy at low initial ion energies have been pointed out before,[13] but the low spectral intensity obtained under these conditions has so far prohibited an extensive use of this excitation method.

## II. EXPERIMENT

The experimental setup is standard for beam-foil spectroscopy. 100–500 keV $Ga^+$, $Ge^+$, or $As^+$

 

ions from the 600-kV heavy-ion accelerator at Aarhus University were passed through thin carbon foils (~5 $\mu$g/cm$^2$). The beam current was kept below 0.05 $\mu$A in order to extend the lifetime of the exciter foils. The spectral measurements below 1100 Å were performed using a 2.2-m McPherson 247 grazing incidence monochromator which is equipped with an ellipsoidal, gold-coated mirror, enabling a spatial resolution of ~90 $\mu$m in the light source. In the region 1100–2000 Å, a 0.5-m McPherson Seya-Namioka spectrometer, adjusted to a spatial resolution of ~110 $\mu$m, was applied.

In the lifetime measurements, the carbon foil was moved along the fast ion beam with typical stepping widths of 50 $\mu$m. The resulting decay curves were analyzed using a multiexponential least-squares fitting procedure. The shortcomings of this technique will be discussed later.

The energy loss suffered by the ion beams passing through the carbon foil was estimated on the assumption that only a minor part of the nuclear energy loss $\Delta E_n^*$ should be added to the electronic energy loss $\Delta E_e$ to yield the proper energy loss for the velocity evaluation in beam-foil experiments for heavier elements.[13,14] The energy loss used for 100–500 keV $Ga^+$, $Ge^+$, and $As^+$ range from 18–30 keV.

To uncover the role of cascading in populating the $4s4p\ ^1P$ level in Ga II, spectral intensity measurements were performed at 1414 Å($4s^2\ ^1S$-$4s4p\ ^1P$), 1802 Å($4s4p\ ^1P$-$4s4d\ ^1D$) as well as at 2780 Å($4s4p\ ^1P$-$4s5s\ ^1S$), all within ≃50 ps after excitation. The emission cross sections were normalized, in the case of $4s4d\ ^1D$ and $4s4p\ ^1P$, using the quantum efficiency of the detector (EMI26E315) and assuming the grating response (blaze wavelength 1500 Å) to be the same at 1414 and 1802 Å. The cross sections for $4s5s\ ^1S$ and $4s4p\ ^1P$ were normalized to one another, using the cascade contributions in the 1414-Å emission at (2.0± 0.3) ns. The energy dependence of the emission cross section thus obtained agrees well with direct spectral measurements at 2780 Å.

## III. RESULTS AND DISCUSSION

The emission cross-section curves shown in Fig. 1 for the $4s^2\ ^1S$-$4s4p\ ^1P$, $4s4p\ ^1P$-$4s4d\ ^1D$ and $4s4p\ ^1P$-$4s5s\ ^1S$ Ga II multiplets are recorded immediately after the foil. The population of the $4s4p\ ^1P$ level by direct excitation is dominant in the investigated energy region, but the cascades become important at increasing energies. At 100 keV, the cascades contribute only 4% to the population of the $4s4p\ ^1P$ level, but increase to 20% near 250 keV, close to the maximum in the emission cross-section curve, and reach 45% at 500 keV. The lifetime of the $4s4d\ ^1D$ level was measured to be 0.76± 0.07 ns and found to be cascade free. The lifetime of the $4p^2\ ^1D$ level was measured to be 55± 5 ns. These two values agree well with the recently published[12] values of 0.73 ± 0.07 ns and 54± 5 ns, respectively. The cascade contribution of 2.0± 0.3 ns found in the $4s4p\ ^1P$ decay was assigned to the $4s5s\ ^1S$ level.

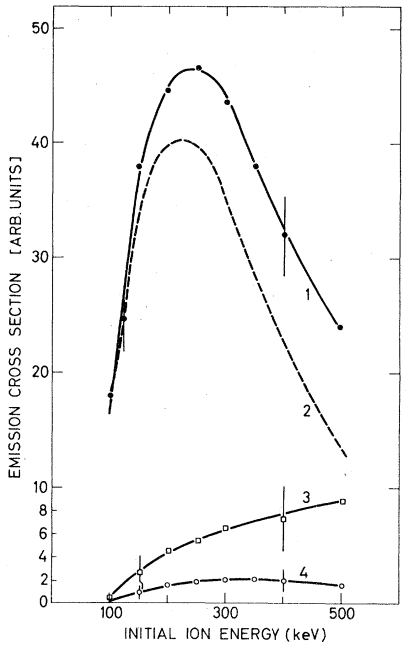


FIG. 1. Emission cross sections as function of initial ion energy for the $4s^2\ ^1S$-$4s4p\ ^1P$ (curve 1), $4s4p\ ^1P$-$4s4d\ ^1D$ (curve 3), and $4s4p\ ^1P$-$4s5s\ ^1S$ (curve 4) Ga II multiplets for the carbon-foil-excited $Ga^+$ ions. The emission cross sections are determined ≃ 50 ps behind the foil. Curve 2 represents the emission cross section for the resonance transition with the cascade contributions for the $4s4d\ ^1D$ (curve 3) and $4s5s\ ^1S$ (curve 4) subtracted.

For the $4s4p\ ^1P$ level, the situation is slightly more complex with the strong cascading present. As shown in Fig. 1, these cascades can be controlled by changing the incident ion energy. This is illustrated in Fig. 2, where (b) shows a typical decay (1414 Å) at 400 keV, yielding a primary lifetime of 0.56 ns as well as cascade contributions at 2 and 55 ns. On the contrary, a 100-keV incident $Ga^+$ decay (1414 Å) exhibiting no detectable cascades is shown in Fig. 2(a). A single exponential fit yields (0.49± 0.05) ns. Figure 3 shows the apparent primary lifetime $\tau_1$ of the $4s4p\ ^1P$ level in Ga II, as a function of incident ion energy, analyzing the experimental data, using (1) a fitting function $A_1 \exp(-t/\tau) + \text{const}$ background

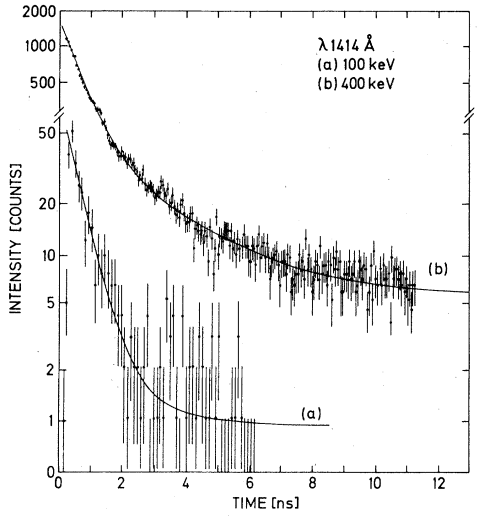


FIG. 2. Typical decays of 1414 Å at (a) 100-keV and (b) 400-keV incident $Ga^+$ ion energy. In the 100-keV data, no cascades can be found, and a single-exponential analysis yields $\tau\,(4s4p\ ^1P) = (0.49 \pm 0.05)$ ns. In the 400-keV data, several cascade components are present. A multiexponential analysis cannot separate the $4s4p\ ^1p$ and $4s4d\ ^1D$ contributions, whereas the $4s5s\ ^1S$ (2.0 ± 0.3) ns and $4p^2\ ^1D$ (55 ± 5) ns are found. Owing to the short foil lifetimes at low ion energies (≃ 2 min at 50-nA beam current), the recording times available at 100 keV and 400 keV differ considerably, hence the difference between 100- and 400-keV amplitude [(a) and (b)]. Figure 1, however, shows that the relative emission cross sections only differ ≈ 30%.

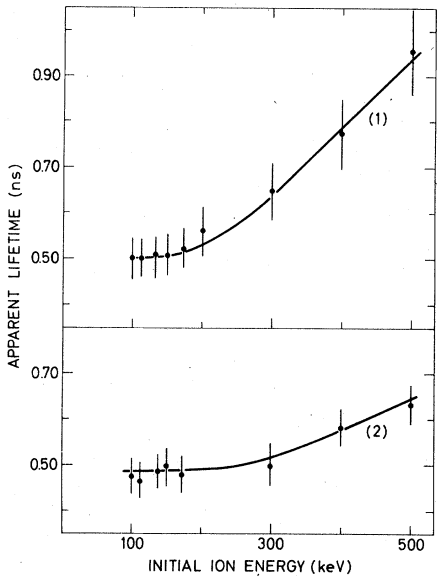


FIG. 3. Apparent lifetimes $\tau_1$ for the $4s4p\ ^1P$ state in Ga II. (1) summarizes the results of analysis of experimental decay curves by means of the function $A_1 \exp(-t/\tau_1) + \text{background}$, whereas (2) shows the apparent lifetimes $\tau_1$ using the function $A_1 \exp(-t/\tau_1) + A_2 \exp(-t/2.0) + A_3 \exp(-t/55) + \text{background}$, in both cases as function of initial ion energy. The solid lines do not represent any fit but are only intended to display the trend in the data.

and (2) a fitting function $A_1 \exp(-t/\tau) + A_2 \exp(-t/2.0) + A_3 \exp(-t/55) + \text{const}$ background, i.e., by taking the cascades from $4s5s\ ^1S$ and $4p^2\ ^1D$ but not from the $4s4d\ ^1D$ level into account.

The present case clearly displays the difficulty in using multiexponential fitting, when trying to take the $4s4d\ ^1D$ cascade of (0.73 ± 0.07) ns into account, i.e., a lifetime close to the primary lifetime of 0.49 ns, especially with this cascade contributing heavily. For incident beam energies below 170 keV, this problem was reduced as no cascades were detectable in the $4s4p\ ^1P$ decay. The situation was drastically different at higher energies, where the cascade contributions were so large that a meaningful analysis was impossible. Using only the low-energy data (<170 keV), where a pure-single-exponential decay was observed, verified by the low-cascade contributions of a few percent as directly measured, a $4s4p\ ^1P$ lifetime of (0.49 ± 0.04) ns is deduced.

This threshold excitation technique thus seems useful in revealing the true lifetime of a particular level in a clean and direct fashion. It should be possible to extend it to other members of the Zn I isoelectronic sequence, even if the level schemes are only incompletely known.

Figure 3 indicates that, independent of the data analysis used, the lifetimes obtained are identical and constant in this threshold region. This fact supports the assumption applied in the evaluation of the energy losses, that the ions emitting light at the resonance transition have predominantly been excited in soft collisions, i.e., by electronic stopping processes. If hard collisions played a role, the scattering of the fast ions would lead to a steadily decreasing lifetime with decreasing initial ion energies.

Since the level schemes for Ge III and As IV may be incompletely known[12] with respect to levels populating the $4s4p\ ^1P$ level through cascading, it is necessary to perform lifetime measurements at low initial ion energies in order to eliminate the cascade contributions as already discussed for Ga II. In the case of Ge III and As IV, this means to perform the measurements below 300 keV. We have determined the $4s4p\ ^1P$ lifetimes in Ge III and As IV, respectively, by performing the measurements in the energy range 250–500 keV initial ion energy. The experimental data exhibit the same trends as seen in Fig. 3. At low

TABLE I. Lifetimes for the $4s4p\,^1P$ state.

| | This work (ns) | Previous values (ns) (Ref. 6) |
|---|---|---|
| Ga II | 0.49 ± 0.04 | 0.65 ± 0.08 |
| Ge III | 0.29 ± 0.03 | 0.39 ± 0.06 |
| As IV | 0.23 ± 0.03 | 0.32 ± 0.03 |

TABLE II. Oscillator strengths for the $4s^2\,^1S$-$4s\,4p\,^1P$ resonance transition in the Zn I isoelectronic sequence.

| | Experiment | Theory (Ref. 4) |
|---|---|---|
| Ga II | 1.85 ± 0.15 | 1.75 |
| Ge III | 1.85 ± 0.20 | 1.74 |
| As IV | 1.56 ± 0.23 | 1.71 |

initial energies, i.e., below 300 keV single-exponential decays are observed, the lifetimes remaining constant. Above 300 keV, the lifetimes increase, reaching apparent values ≃40% longer at 500 keV, due to the growing cascade contributions. The lifetimes obtained are given in Table I.

Table II shows a comparison between the recent theoretical oscillator strengths calculated by Froese-Fischer and Hansen[4] and the experimental values. The theoretical values by Shorer[5] agree with the results by Froese-Fischer and Hansen[4] to within a few percent. Within the experimental accuracy, the experimental and theoretical values agree, which may support the assumption[1] that the oscillator strengths of the resonance transitions for more ionized members of the Zn I isoelectronic sequence are better predicted on the basis of the available theoretical calculations[4,5] than on the extrapolation from the experimental values.[6-10]

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