Z. Physik A 279, 1–6 (1976)

Zeitschrift
für Physik A

A High Precision Beam-Foil Meanlife Measurement of the $1s\,3p\,{}^1P$ Level in He I

G. Astner, L.J. Curtis*, L. Liljeby, S. Mannervik and I. Martinson**
Research Institute for Physics, Stockholm, Sweden

Received June 28, 1976

A beam-foil measurement of the meanlife of the $1s\,3p\,{}^1P$ level in He I has been made and yields the value 1.7225 ± 0.0046 ns. The measurement was made with standard beam-foil techniques and equipment, but special attention was devoted to minimizing sources of uncertainty. The precision far exceeds that of previous beam-foil meanlife measurements and demonstrates that the beam-foil technique is capable of high precision and is competitive with and more flexible than methods such as resonant laser excitation.

1. Introduction

The beam-foil technique is probably the most versatile and most prolific method of atomic meanlife measurement available today, and an impressive amount of accurate data has been obtained by this technique. One can usually be 95% confident that modern beam-foil results are reliable to within 10% [1]. Recently, however, considerable efforts have been expended to develop alternative techniques to measure lifetimes to within a few parts per thousand. To date one beam-laser experiment [2] has succeeded in achieving this accuracy, but it appears that this method does not yet have the flexibility of beam-foil excitation and thus cannot be considered a substitute for it. We have therefore attempted to probe the ultimate precision of a modern beam-foil measurement to determine whether accuracies in the parts per thousand range are attainable with reasonable effort. The level selected, $1s\,3p\,{}^1P$ in He I, has already been well studied both experimentally and theoretically; we have now obtained results with an estimated uncertainty of only 0.26% which far exceeds the precision of previous measurements and rivals that of existing theoretical calculations.

The advantages of the beam-foil source are many. Nearly any charge state of any atom is accessible, and highly excited and multiply excited states are often copiously populated. Mass analysis of the beam ensures purity, and the low particle density in the beam and high vacuum conditions in the chamber eliminate problems of collisional deexcitation and radiation imprisonment. The primary disadvantages involve the low light intensities, the velocity dispersion introduced by scattering processes in the foil, and the cascade repopulation of the levels. However, optical systems which utilize the properties of a moving light source to greatly enhance the light intensity without loss of spatial resolution have recently been developed [3], and the effects of velocity dispersion and cascade repopulation can generally be accounted for by a suitably designed experiment. Thus the capabilities of the beam-foil source are very substantial and its disadvantages should not be overstated.

2. Experimental Procedure

Our measurement was performed with standard equipment used currently in many modern beam-foil laboratories, but with special attention to the details which can affect the accuracy of the measurement. The experimental arrangement is shown schematically in Figure 1. A nominally 130 keV He^+ beam was provided by a Nucletec accelerator which consists of a 30 kV isotope separator followed by a 350 kV Deltatron post accelerator. The energy stability of the accelerator is within $\pm 0.1\%$. The current was maintained at 3.5 ± 0.5 μA (a gate circuit suspended

* Permanent address: Department of Physics and Astronomy, University of Toledo, Toledo, Ohio 43606
** Permanent address: Institute for Physics, Lund University, S-223 62 Lund, Sweden

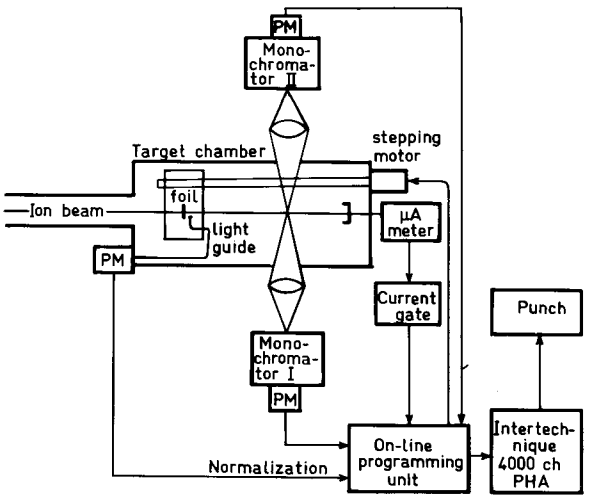

Fig. 1. Schematic diagram of the experimental arrangement. The foil-excited beam is simultaneously viewed by two optical monochromators, in practice at 90° geometry to each other. The motion of the foil is achieved through a precision screw, driven by a stepping motor, triggered by the accumulation of a fixed number of counts from a monitor phototube which uses a fiber optics light guide to view the beam at a fixed position from the foil. If the beam current varies outside of present limits data accumulation is suspended

measurements if the current through the foil varied outside this range). The nominally 5–10 μg/cm^2 foils were prepared in our laboratory and mounted on 4 mm diameter holders and placed in a carriage which moved parallel to the beam past 90° viewing optics. The carriage is driven by a stepping motor with a precision machine screw drive which is resettable to within 0.05 mm. The linearity of the screw is better than 0.05%. A decay curve could thus be obtained by stepping the foil carriage past the viewing optics, with the step size and total number of steps set by an on-line programming unit. The data accumulation time at each position of the foil was determined by a light monitoring system, in which a photomultiplier tube viewed the beam at fixed distance from the foil through a fiber optics system which moved with the carriage. Data were thus accumulated for a fixed number of monitor counts, which compensated for any small fluctuations in the beam current. The optical system viewed a 0.2 mm segment of the beam through a lens. The lens was focussed and the spatial resolution was determined by a knife edge test. Calculations were made to determine the effect of loss of beam particles from the viewing volume as a function of distance downstream due to the divergence of the foil emergent beam. A universal expression developed by Meyer [4] for the angular distribution of particles scattered from thin amorphous foils was applied to a 100 keV helium beam incident upon a 10 μg/cm^2 carbon foil. A numerical integration indicated that for a 10 mm high observation slit, the loss of the viewed beam at a distance 50 mm downstream from the foil was about 0.5%. This is the worst case, and 40 mm downstream the loss was less than 0.1%. The pressure within the excitation chamber was maintained at less than 10^{-5} Torr. Andrä [2] has estimated the effect of rest gas collisions in lifetime measurements, and using a cross section of 10^{-14} cm^2, found it to be 0.2% at 10^{-5} Torr. Light from the 5,015 Å He I $1s2s\,^1S-1s3p\,^1P$ transition was selected with a 35 cm Heath EUE 700 monochromator, and single photons were detected by a Peltier-cooled EMI 6256 photomultiplier with dark counts less than 5 per second.

One of the most serious problems in a beam-foil measurement is the velocity determination for the foil-excited particles. Two methods were used in this experiment: simultaneous observation of the decay of two excited terms by a two-spectrometer technique, and electrostatic energy analysis.

The first method, which is shown in Figure 1, takes advantage of the zero-field quantum beats [5, 6] superimposed on the decay curve of the He I 3,889 Å multiplet ($1s2s\,^3S_1-1s3p\,^3P_{0,1,2}$). One of the optical spectrometers, usually the Heath monochromator, views the 5,015 Å line, while another, a Jarrell-Ash 25 cm grating monochromator, mounted with its axis mutually perpendicular to the first monochromator and to the beam, selects the 3,889 Å multiplet. The decay curves of the 5,015 Å and 3,889 Å transitions are thus simultaneously recorded. The time scale after foil excitation is given by the $^3P_1-{}^3P_2$ quantum beats which have an accurately known frequency of 658.55 ± 0.15 MHz [7]. Thus the effects of velocity straggling and fluctuations in central velocity are averaged over the decay curve (5,015 Å) and the time calibration curve (3,889 Å) in nearly the same way.

The other method for velocity determination employed a Danfysik 50 cm radius 90° electrostatic analyzer which was mounted at the end of the target chamber. As in Reference 6, we calibrated the analyzer, which is capable of 0.1% precision in velocity determination [8], by means of the 3,889 Å quantum beat pattern.

In the present case the first method is judged to be more reliable because the time scale is obtained simultaneously with the lifetime measurement. However, the electrostatic analyzer allows precision measurements also in other atomic systems which do not possess a convenient quantum beat time standard.

We found it unnecessary to compensate for the earth's magnetic field in the target chamber, because shifts in the 658 MHz frequency, due to this field, must be less than the corresponding electron Larmor frequency (0.4 MHz). Andrä [2] has estimated that the lifetime uncertainties due to the earth's magnetic field should be less than 0.025%. It is also conceivable that the

velocities of helium atoms excited to triplet states could differ slightly from those excited to singlet states. Here, also, Andrä [2] has found that He^+ and He^{++} atoms after the foil have the same velocity to within 0.03%, so we can safely assume that any velocity differences within the same ionization state are negligible.

A well known problem in beam-foil velocity determinations arises from the fact that the foil becomes thicker during ion bombardment [9, 10]. To first order this foil thickening is averaged over time in the same way for both the decay curve and the quantum beat curve, but there is a second order effect. The meanlives of the $1s3p\,^1P$ and $1s3p\,^3P$ terms are quite different (1.7 and 96 ns, respectively) and whereas the beat pattern is measured with about the same statistical accuracy over the full region, the decay curve determination is most heavily weighted in the region of high statistical accuracy, near the beginning. We may thus argue that the quantum beat points are taken on the average some 100 channels (~100 s) later than the strongest meanlife points. However, if we use a value for the foil thickening of 2.7 μg/cm²/h, [9] this effect on lifetimes would be less than 0.04%.

During measurements the number of counts from each of the photomultiplier tubes and the accumulation time for each position of the foil was routed and stored in an Intertechnique 4000 channel analyzer. Typical accumulation times were 1 s/point. The foil was stepped in 0.25 mm (0.11 ns) increments for a total of 200 steps (~23 ns). A new foil was inserted every three complete sweeps. Sweeps in which a foil deteriorated measurably were discarded. An accumulation of 12 sweeps contained about 15,000 counts in the peak of the decay curve, and was considered a data set for purposes of analysis. A typical data set including both the decay curve and the quantum beat time calibration curve is shown in Figure 2. A total number of 10 such data sets were measured and analyzed, and the results combined to yield the final meanlife determination.

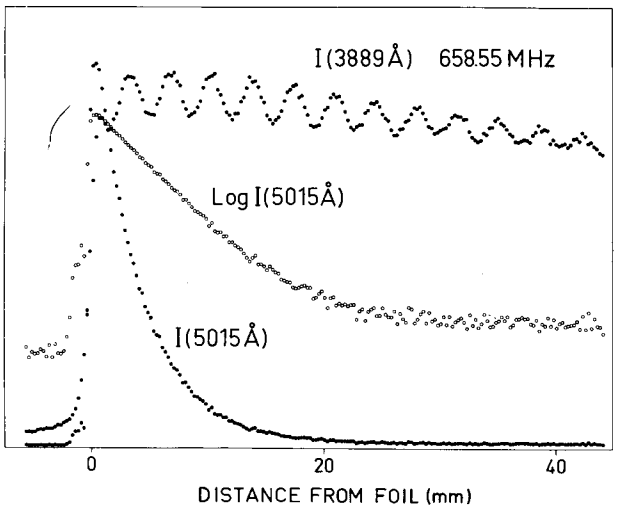

Fig. 2. Example of a decay curve for the $1s3p\,^1P$ level (5,015 Å), velocity-normalized by a simultaneous quantum beat measurement on the 3,889 Å line

3. Data Treatment

The analysis of the decay curves began with the subtraction of beam-independent background. Measurements were made of the background count rate in all three phototubes (those counting the 5,015 and 3,889 Å radiation and the total light near the foil). Using the measured accumulation time in each channel, all backgrounds were subtracted and the corrected intensities at 5,015 and 3,889 Å per corrected monitor count were computed. The time per channel, accumulated over 12 sweeps, was generally the same for all channels to within ±5%. A subtraction of the beam-dependent background was also performed. This was done by beginning the measurement slightly upstream of the foil and subtracting the prefoil value from all post foil channels. A typical value for this background was 60 corrected counts, compared with a peak value of 15,000. The actual meanlife determination was, however, rather insensitive to the background subtraction, and shifting it deliberately outside its possible error range, e.g. by 20%, did not affect the meanlife value to within our quoted limits. Dead time corrections to the decay curve were also necessary, since the count rate at the peak of the decay curve was two orders of magnitude higher than that on the tails. The storage time of the multichannel analyzer was 21 μs, which for a typical peak rate of 500/s amounts to a 1% correction. We estimate that the uncertainty in this correction is 5%, introducing at most a 0.05% error in the measurement. The corrected decay curve was then fitted to multiple exponential functions by a standard non-linear least squares χ^2 minimization program, using statistical weights. Several possible fitting functions were used. One example was a two exponential sum with both coefficients and both meanlives as free fitting parameters. Another example was a nine exponential sum with two coefficients and one meanlife free, and the other meanlives fixed to theoretical values of likely cascades with coefficients as suggested by the He I excited state population studies of Davidson [11]. Several other fitting functions were also tested, but the primary lifetime extracted was remarkably insensitive to the form of the mathematical function used to account for cascades. When the same data set was analyzed with different fitting functions the values obtained for the primary meanlife rarely differed by more than 0.1%. When different data sets were analyzed with the same fitting function the cascade-

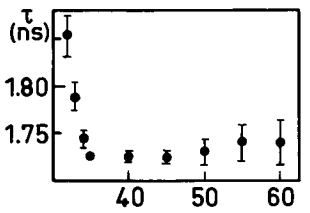

Fig. 3. The influence of the "start channel" in the analysis of the decay curve (such an analysis helps in including the maximum amount of high-quality data), showing that in this particular case points from channel 35 onward can be included, while vignetting causes systematic errors for lower channels

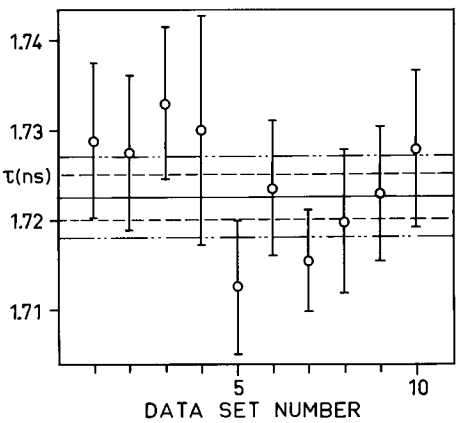

Fig. 4. The results of ten independent lifetime measurements

associated parameters could vary by several percent, but the value obtained for the primary lifetime was the same to within 0.5%. When the results of the analysis of several data sets were combined, the results were generally independent of fitting function to within 0.02%, indicating that the inclusion of more cascade contributions does not systematically shorten or lengthen the extracted primary meanlife, and that cascades are a rather small source of uncertainty in the measurement.

The reduced χ^2 value was close to unity for all of the fits, implying that the fitting functions and the statistical weighting adequately represented the data. Tests were also made of the uniformity of contributions to χ^2 from regions of high and low statistical accuracy on the same decay curve, which provided further evidence for this assumption. As a part of the fitting process it was also necessary to examine the first points on the decay curve after the peak, which may be affected by vignetting by the foil holder. On the other hand, these points have the best statistical accuracy, and should not be discarded unnecessarily. To check this data were analyzed with a varying start channel. A typical result can be seen in Figure 3, which clearly indicates that points from channel 35 onward can be safely included. Figure 4 shows the results of the analysis of the individual data sets. The error bars on the points are the standard deviations returned by the fitting program under the assumption of statistical uncertainties. The weighted average of these measurements is 1.7225 ± 0.0025 with a reduced χ^2 of 0.74, indicating consistency between fluctuations in the sample distribution and a parent Poisson distribution.

Table 1 provides a survey of our estimates of the various sources of uncertainty in the measurement. The largest sources of error are the statistical uncertainties in the decay curve fit, which could be reduced still further by additional data accumulation, and the possible rest gas excitation, which could be reduced by use of ultra-high vacuum techniques. It is interesting to note that, although cascade corrections are much discussed in the context of beam-foil measurements, they are in the present case an insignificant source of uncertainty in the primary meanlife determination. This is largely due to the fact that all the levels responsible for direct cascading into $1s\,3p\,^1P$, e.g. $1s\,4s\,^1S$, $1s\,4d\,^1D$ as well as higher ^1S and ^1D terms have lifetimes much longer than the $1s\,3p\,^1P$ decay time. Decompositions of the $1s\,3p\,^1P$ decay curve into exponentials should therefore be quite unambiguous.

Table 1. Measurement uncertainties (standard deviation) in per cent

A. Instrumental uncertainties	
Deadtime correction uncertainties	0.05
Nonlinearities in the screw drive	0.05
Loss of viewed particles by diverging beam	0.01
Possible rest gas excitation	0.2
B. Decay curve analysis uncertainties	
Statistical uncertainties	0.14[a]
Cascade corrections	0.02
Background corrections	0.02
C. Velocity determination	
Statistical uncertainties in fitting and Fourier analysis of beat pattern	0.03[a]
Calibration frequency	0.02
Effect of foil thickening	0.04
Earth's magnetic field	0.03
Total, assuming uncorrelated	0.26

[a] Statistical uncertainties in the fitting of the lifetimes and the quantum beats were combined for each data set. Thus these figures represent the merged statistical uncertainties of all 10 data sets broken down into categories *B* and *C* according to their average contribution

4. Result and Discussion

Table 2 compares our result with a selection of other measurements and theoretical values. Only more recent results are included, for a complete list see the compilation of Wiese et al. [12]. Prior to the advent of

techniques such as beam-foil excitation most measurements of the $1s\,3p\,^1P$ lifetime were strongly influenced by the imprisonment of the $1s^2\,^1S-1s\,3p\,^1P$ resonance radiation, as already discussed in the paper of Heron et al. [13]. When this radiation is completely imprisoned in He gas, the $1s\,3p\,^1P$ term mainly decays by the $1s\,2s\,^1S-1s\,3p\,^1P$ branch at 5,015 Å, which has a theoretical inverse transition probability of 75 ns, a result obtained in most measurements prior to 1969. All modern experimental results, based on beam-foil [14–16], on zero-field level crossing, or on beam-gas [17] or atomic beam [18] techniques, are in agreement with each other and with our results. In fact their variance is only 2%, much better than their quoted uncertainties would suggest.

Precise theoretical calculations of the $1s\,3p\,^1P$ lifetime have been available since the classic work of Hylleraas [19] but we have only included the more recent studies. Schiff et al. [20] have examined the agreement between results obtained by using the dipole-length and dipole-velocity formulae for f-value calculations, together with the convergence of their results as an increasing number of terms (up to 364) are included in the variational calculations, and they conclude that their values are accurate to within one part per thousand for the $1s\,3p\,^1P$ level in He I. The frozen-core approximation has been employed by Cohen and McEachran [21] to calculate f-values for the He I isoelectronic sequence. These calculations, being much less laborious than those of Schiff et al. [20] have estimated uncertainties of less than 5% for the $S-P$ transitions. The frozen-core Hartree-Fock approximation was used by Devine and Stewart [22] who included perturbation corrections up to second order. These values, as well as earlier data of Weiss [23], who performed variational calculations with up to 50 parameters in the wavefunctions, and the results of configuration-interaction calculations of Green et al. [24] are all included in Table 2. The calculations of Weiss have estimated uncertainties below 3%. Most of these authors make detailed comparisons between the dipole length, dipole velocity and dipole acceleration approximations. The theoretical lifetimes, included in Table 2, are based on the arithmetic means of the dipole-length and dipole-velocity f-values. Anderson and Weinhold [25] discuss situations in which length-velocity agreement and numerical convergence can give erroneously small error estimates. Instead they were able to determine rigorous upper and lower bounds to their f-values, calculated by using variational wavefunctions with about 140 terms. Their result for the $1s\,3p\,^1P$ level is only quoted to be accurate to within 5%, most of the uncertainty being due to the $1s^2\,^1S-1s\,3p\,^1P$ f-value, but it agrees with Schiff et al. [20] to within 0.5%. It should be noted, however, that the error limits in Reference 25 are obtained in a much simpler way than most previously estimated theoretical uncertainties.

Table 2. The $1s\,3p\,^1P$ lifetime (ns) in He I

This work	Other measurements (since 1969)	Theory
1.7225 ± 0.0046	1.78 ± 0.10[a]	1.74[f]
	1.72 ± 0.10[b]	1.726 ± 0.002[g]
	1.70 ± 0.04[c]	1.73[h]
	$1.8\ \pm0.1$[d]	1.76[i]
	1.73 ± 0.11[e]	1.73[j]
		1.72 ± 0.09[k]

[a] Martinson and Bickel [14], beam-foil
[b] Martinson et al. [15], beam-foil
[c] Heine et al. [16], beam-foil
[d] Carré et al. [17], beam-gas, level crossing
[e] Burger and Lurio [18], atomic beam, level crossing
[f] Green et al. [24]
[g] Schiff et al. [20]
[h] Weiss [23]
[i] Cohen and McEachran [21]
[j] Devine and Stewart [22]
[k] Anderson and Weinhold [25]

Our experimental result is clearly consistent with all these experimental and theoretical results but the precision is higher than previously quoted, with the exception of the work of Schiff et al. [20]. We conclude that the beam-foil method of atomic meanlife measurement is capable of very high accuracy, the errors being a few parts per thousand in favourable cases.

We wish to thank Professor J.O. Stoner, Jr. and Dr. A. Gaupp for their valuable advice and assistance during the early phases of this work. The experience of Dr. L. Lundin in high-precision measurements has provided a source of guidance throughout the experiment. We are finally grateful to Professor I. Bergström for enlightening discussions and to Professor B. Edlén for critically reading the manuscript.

References

1. Pinnington, E.H., Livingston, A.E., Kernahan, J.A.: Phys. Rev. A **9**, 1004 (1974)
2. Andrä, H.J.: In: Proceedings of the 4th International Conference on Beam-Foil Spectroscopy, Gatlinburg, September 1975, Vol. 2, p. 835, ed. by I.A. Sellin and D.J. Pegg, Plenum Publ. Corp. 1976
3. Bergkvist, K.-E.: J. Opt. Soc. Am. (in press)
4. Meyer, L.: Phys. Stat. Sol. **44**b, 253 (1971)
5. Andrä, H.J.: Phys. Rev. Letters **25**, 325 (1970)
6. Wittmann, W., Tillmann, K., Andrä, H.J., Dobberstein, P.: Z. Physik **257**, 279 (1972)
7. Wieder, I., Lamb, W.E. Jr.: Phys. Rev. **107**, 125 (1957)
8. Herb, R.G., Snowdon, S.C., Sala, O.: Phys. Rev. **75**, 246 (1949)
9. Bickel, W.S., Buchta, R.: Physica Scripta **9**, 148 (1974)
10. Dumont, P.D., Livingston, A.E., Baudinet-Robinet, Y., Weber, G., Quaglia, L.: Physica Scripta **13**, 122 (1976)

11. Davidson, J.F.: Phys. Rev. A **12**, 1350 (1975)
12. Miles, B.M., Wiese, W.L.: Bibliography on Atomic Transition Probabilities, 1916–1969
Fuhr, J.R., Wiese, W.L.: Bibliography on Atomic Transition Probabilities 1969–1971, 1971–1973
NBS Spec. Publ. 320 and Suppl. 1 and 2. (U.S. Govt. Printing Office, Washington, D.C., 1970, 1971, 1973)
13. Heron, S., McWhirter, R.W.P., Rhoderick, E.H.: Proc. Roy. Soc. (London) A **234**, 565 (1956)
14. Martinson, I., Bickel, W.S.: Phys. Letters **30** A, 524 (1969)
15. Martinson, I., Bickel, W.S., Bromander, J., Berry, H.G., Lundin, L., Buchta, R., Bergström, I.: J. Opt. Soc. Am. **60**, 352 (1970)
16. Heine, G., Reinke, M., Bukow, H.H.: To be published
17. Carré, M., Desesquelles, J., Dufay, M., Gaillard, M.L.: Phys. Rev. Letters **27**, 1407 (1971)
18. Burger, J.M., Lurio, A.: Phys. Rev. A **3**, 64 (1971)
19. Hylleraas, E.A.: Z. Physik **106**, 395 (1937)
20. Schiff, B., Pekeris, C.L.: Phys. Rev. **134** A, 638 (1964)
Schiff, B., Pekeris, C.L., Accad, Y.: Phys. Rev. A **4**, 885 (1971)
21. Cohen, M., McEachran, R.P.: Can. J. Phys. **50**, 1363 (1972)
22. Devine, K.R., Stewart, A.L.: J. Phys. B **5**, 2132 (1972)
23. Weiss, A.W.: J. Res. NBS **71** A, 163 (1967)
24. Green, L.C., Johnson, N.C., Kolchin, E.K.: Astrophys. J. **144**, 369 (1966)
25. Anderson, M.T., Weinhold, F.: Phys. Rev. A **9**, 118 (1974)

G. Astner
L.J. Curtis
L. Liljeby
S. Mannervik
I. Martinson
Research Institute of Physics
Roslagsvägan 100
S-104 05 Stockholm, Sweden