M1, M2 and hyperfine-induced decay rates in Mg-like ions of Co, Ni and Cu measured at a heavy-ion storage ring

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Abstract. The optical decay rates of the 3s3p$^3P^0_2$ level in Mg-like ions of $^{59}$Co, $^{58}$Ni and $^{63,65}$Cu have been measured at a heavy-ion storage ring. The measurement is sensitive to M1, M2 and hyperfine-induced decay rates. The measurement precision is just about high enough to detect the difference in the isotope effect on the level lifetime in the ions $^{63,65}$Cu$^{17+}$. We also discuss our findings when trying to measure the hyperfine-induced decay of the lowest triplet level, 3s3p$^3P^0_6$, in the two Cu isotopes.
1. Introduction

Selection rules for transitions in atoms combine the geometry of the wave functions and properties of the radiation field, reflecting their symmetries. Some selection rules are valid only as useful approximations, whereas others are strict. For example, in the many cases where the total angular momentum $J$ is a valid quantum number, there can be no $J = 0 - J' = 0$ transition, because the photon carries nonzero angular momentum. Hence the lowest excited level in Be- or Mg-like ions, $nsnp\ ^3P^o_0$, cannot by single-photon emission decay to the $ns^2\ ^1S^0_0$ ground state, unless there is a nonvanishing nuclear spin $I$. $J$ and $I$ couple to $F$, and since now the ground state and the excited level have $F$ values that are different from zero, a single-photon transition becomes possible. The process is usually modeled as an effect of hyperfine interaction. Hyperfine interaction mediates the mixing of $^3P^o_0$ hyperfine levels with the hyperfine levels of $^3P^o_1$, which in turn is (spin–orbit) mixed with $^1P^o_1$. Hyperfine sublevels of the $nsnp\ ^3P^o_2$ level can similarly mix with the hyperfine levels of $^3P^o_1$, but in this case hyperfine interaction provides an additional, not the only, decay channel, and hence causes a modification of the level lifetime that in the absence of hyperfine interaction is already finite and largely determined by M1 and M2 decay branches.

This basic concept of hyperfine quenching was developed in the 1930s [1, 2], when detection had to rely on photographic emulsions and time resolution was not yet available. Time-resolving spectroscopy offered by the beam-foil technique in combination with photoelectric detection came about in the late 1960s. Since the 1970s, this combination has been employed to study the hyperfine quenching of the $1s2p\ ^3P^o_0$ level in He-like ions, both to test theory (starting from the known level structure and measured decay rates) and to alternatively derive the $^3P^o_0 - ^3P^o_1$ level splitting from the lifetime measurement in the cases when a level crossing as a function of $Z$ occurs along the isoelectronic sequence, making this splitting unobservable by x-ray spectroscopy [3]–[9]. In the He isoelectronic sequence, there are two crossings of these levels, and in the vicinity of such a crossing, the mixing (and thus the effect on the $J = 0$ level lifetime) is particularly large [10]. There are also experiments that have induced by laser irradiation the hyperfine-interaction enabled $1s2s\ ^1S^0_0 - 1s2p\ ^3P^o_0$ transition in He-like nitrogen in a precision measurement of the $N^{5+}$ ion level structure [11]. The level lifetimes measured in those hyperfine quenching experiments are in the range from nanoseconds to picoseconds, and largely determined by the hyperfine mixing of the $1s2p\ ^3P^o_0$ and $1s2p\ ^3P^o_1$ levels, the other important contribution to the hyperfine quenching effect being the multiplet mixing (of the order...
of $10^{-4}$–$10^{-2}$ of the transition rates) between the $1s2p\,{}^3P^0_1$ level and the very-short-lived $1s2p\,{}^1P^0_1$ level.

In Be- or Mg-like ions, the decay rate of the $nsp\,{}^1P^0_1$ resonance level (with a $\Delta n = 0$ transition) is many orders of magnitude lower than in He-like ions with their $\Delta n \neq 0$ transitions and scales only linearly with the nuclear charge $Z$ (instead of the $Z^4$ dependence in He-like ions). Consequently, the hyperfine-quenched levels in such ions are many orders of magnitude more long-lived than those in comparable He-like ions. Because of this longevity, beam-foil spectroscopy in its traditional configuration is not suitable to study the hyperfine lifetime quenching effect in ions with a filled K shell. Instead, these lifetimes lie in the typical range accessible in measurements using an ion storage ring.

The long lifetimes of hyperfine-quenched levels in Be- or Mg-like ions are of interest in the diagnostics of very-low-density plasmas, for example those encountered in planetary nebulae [12]. Indeed, the line intensities in the observations of one such nebula have been interpreted in terms of the hyperfine-induced lifetime quenching in the Be-like ion $N^{3+}$, with a $2s2p\,{}^3P^0_1$ level lifetime result of the order of 20 min and an uncertainty of about 30% [13], otherwise assuming that the atomic structure calculations are reliable.

Hyperfine quenching affects not only the $J = 0$ level, but also the other levels of the same term. However, the effect is noticeable only if it changes the decay rate substantially (that is, not for the $J = 1$ level with its dominating fast intercombination decay channel). The quenching effect on the $J = 2$ level was demonstrated decades ago by Gould et al [14] for the example of the $1s2p\,{}^3P^0_2$ level of He-like $^{51}$V$^{2+}$ ions, in beam-foil experiments on the M2 x-ray decay branch. In Be- and Mg-like ions, however, except at the low-$Z$ end of the isoelectronic sequence, the dominant decay branch is not the M2 branch, but the M1 decay to the $3P^0_1$ level of the same multiplet, as has been calculated by Lin et al [15]. In fact, the NIST on-line data base [16] and the otherwise very useful compilation by Kaufman and Sugar [17] list only the M1 decay. However, the M2 intercombination branch should still alter the total decay rate by a few per cent, even in the absence of hyperfine interaction. With hyperfine interaction, the corresponding intercombination decay of the $3s3p\,{}^3P^0_2$ level should increase by about 10% of the total decay rate (for Mg-like Cu$^{17+}$). The low probability of the direct decay to the ground state, and the importance of hyperfine interaction in opening that decay channel, have been discussed for the astrophysical context of neutral Hg as early as in the 1930s by Huff and Houston [18], who refer to a comment by Bowen [19]. The Be- and Mg-like ion levels relevant here again have lifetimes in the typical range accessible by storage rings.

We report $3P^0_2$ level lifetime measurements in Mg-like Cu ($Z = 29$, two odd isotopes), Ni ($Z = 28$) (even isotope, no hyperfine structure) and Co ($Z = 27$) (single odd isotope), with the aim of disentangling the various contributions to the decay rate. The analysis of these measurements is supported by our own new calculations in addition to recently published work from other sources. The new calculations are moreover motivated by possible inconsistencies in previous treatments of the Mg- and Be-like isoelectronic sequences. For example, the early hyperfine quenching calculations by Marques et al [20, 21] yielded rather similar lifetime results for Be- ($n = 2$) and Mg-like ($n = 3$) ions of the same isotopes, a surprising similarity that may or may not be valid. These older calculations have two aspects that have already been tested, but with contradictory results. On the one hand, the energy values of the $3s3p\,{}^3P^0_1$ level as obtained by Marques et al [22] are among the best available for mid- to high-$Z$ Mg-like ions. This can be seen as a sign of quality of the wave functions or the algorithms applied. On the other hand, for low-$Z$ ions, the intercombination transition rate of the $2s2p\,{}^3P^0_1$ level in Be-like C$^{2+}$ ions
(which, as mentioned before, implies multiplet mixing due to spin–orbit coupling and enters the calculation of the hyperfine-induced decay rate) comes close to the findings of an early radiofrequency ion trap experiment [23]. Those lifetime results deviate considerably (by about 20%) from later, much more accurate results obtained at a heavy-ion storage ring [24], as well as from the result of very extensive relativistic configuration interaction (RCI) calculations [25] with more than a hundred thousand configurations. So something is far from being well with the same calculations that excel for the energies of high-$Z$ ions. This mismatch indicates the need for revisiting the hyperfine-induced decay rates that have been predicted by the same approach.

A recent storage ring measurement by Schippers et al [26] has achieved an uncertainty of as little as 5% on the hyperfine-induced $2s2p\,{}^3P_0$ level decay rate in Be-like $^{47}$Ti$^{18+}$ ions. Interestingly, a sizeable discrepancy of almost 60% appears between these experimental findings and the calculational predictions by Marques et al [20]. The latest results of RCI calculations by Cheng et al [27] differ from the results of the earlier calculations by more than 80%, and thus by some 20% from the new experimental data. The agreement with the measurement is thus much improved, but not yet good. Cheng et al discuss various approximations together with the ranges of nuclear charge $Z$ in which each of them might be used to optimal effect. Considering this activity, the need for an improvement of our understanding of hyperfine interaction and its implementation in atomic structure codes also for Mg-like ions is evident.

Therefore, we have recalculated the hyperfine-induced decay rate of the $3s3p\,{}^3P_0$ level in Mg-like ions as well as attempted a determination by passive atomic lifetime measurements of $^{63,65}$Cu$^{17+}$ ions in a heavy-ion storage ring, applying well-established techniques [24, 28]. The hyperfine-induced decay rates of the $3s3p\,{}^3P_0$ level (figure 1) in the two isotopes are expected to differ by about 10%. Hence both, the absolute decay rates and their difference, should yield information about the hyperfine interaction and the wave functions of the participating electrons. Making use of the much expanded computing power that is available nowadays, the multi-configuration Dirac–Fock (MCDF) calculations by Marques et al [21] have been expanded and improved upon, and further calculations by other techniques have been instigated.

2. Calculations

Figure 1 illustrates the schematics of the lowest $n = 3$ levels in an Mg-like ion and of the transitions between these levels. The ground state is $3s^2\,{}^1S_0$, and there are four $3s3p$ levels, $^1P_1$ and $^3P^o_{0,1,2}$. The $^1P_1$ level decays to the ground state by an electric dipole (E1) transition and has the shortest lifetime of these four levels. For the four nuclides of interest here, $^{59}$Co ($I = 7/2$), $^{63,65}$Cu$^{17+}$, $^{39}$Ca ($I = 3/2$), and $^{69}$Cd ($I = 11/2$), the $^3P^o_0$ level has the longest lifetime.
\(^{58}\text{Ni} (I = 0)\) and \(^{63,65}\text{Cu} (I = 3/2)\), the lifetimes are nearly 30 ps \[29\]. By the relativistic effect of spin–orbit coupling (the leading term of the Breit interaction), the \(1P\) and \(3P\) levels mix. This mixing opens a spin-changing E1 decay channel of the \(3P_1\) level to the ground state and thereby reduces the lifetime of the \(3P_0\) level to the range near 10 ns \[30\]. The M1 decay to the \(3P_0\) level is negligible in comparison. The strongest decay branch of the \(3P_2\) level is the M1 decay to its neighboring \(3P_0\) fine structure level. There are also very weak E2 branches to the \(3P_1\) level and to the \(3P_0\) level. The second strongest decay branch is an M2 decay to the ground state.

Recent calculations by Aggarwal et al \[31\] appear to be the first ones that systematically include the electric-dipole forbidden transitions in Mg-like ions of some iron group elements (Fe, Co, Ni). According to their calculations with the CIV3 code, the M2 decay rate of the \(3s3p\) \(3P_2\) level amounts to 8.9% of the M1 decay rate in Fe XV, 6.3% in Co XVI, 4.6% in Ni XVII and 3.4% in Cu XVIII. This is compatible with our own estimate (neglecting hyperfine interaction) of 2.7% in Cu XVIII. (The E2 decay rate to \(3s3p\) \(3P_0\) and the E2 and M3 decay rates to \(3s3p\) \(3P_1\) are negligible in comparison.)

We have performed new MCDF calculations, to improve upon older work by Marques et al \[21\]. The present calculation differs in the way correlation was included. Here we have used a newly implemented configuration generator in the latest version of the MDFGME code \[32\] to include all single and double excitations from all the electrons in the \(1s^2 2s^2 2p^6 3s^2 \) and \(1s^2 2s^2 2p^6 3s3p\) cores to all free levels up to either \(5p\) or \(4f\), depending on the size of the calculation \[33\]. It required up to 32 GB of RAM to fit such large-scale calculations, with up to 15 000 \(jj\) configurations. Opening all occupied shells in the core is very important to obtain more accurate hyperfine interaction matrix elements. It is also important to obtain the correct nonrelativistic limit for transition energies \[34\]. In the determination of the wave functions, all orbitals, including highly excited correlation orbitals, were fully relaxed, and the Breit interaction was made fully self-consistent (i.e. contributions from the Breit operator were included). The second improvement was to take into account nonorthogonalities between initial and final state orbitals for both transition probabilities and nondiagonal hyperfine matrix elements. Higher-order multipoles contributing to the transition rates were all calculated, and appear not to play any role in this particular case (in some cases, higher-order multipoles can be larger than the first nonzero one in relativistic calculations, see e.g. \[35\]). The hyperfine interaction matrix elements have been determined for all possible total angular momenta \(F = I + J\). Once all of the matrix elements and transition rates between all possible \(3s3p\) \(25+1 P_J\) levels and the ground state or other possible \(3s3p\) \(25+1 P_J\) levels were calculated, a complex matrix was built, following \[10\], using the total transition rate (total level width) for the imaginary part of the matrix, and \(F\)-dependent nondiagonal and diagonal matrix elements. A matrix was built for each \(F\) value, and diagonalized to obtain the hyperfine-quenched rate from each excited level.

In the absence of hyperfine quenching, the \(3s3p\) \(3P_0\) level can decay only by an E1M1 two-photon transition and must hence be extremely long-lived. If one considers only magnetic dipole hyperfine interaction matrix elements, only the \(3s3p\) \(3P_1\) and \(3s3p\) \(1P_0\) levels with \(F = I\) can interact with the (single) \(3s3p\) \(3P_0\) level. In contrast, there are several (anyway much shorter lived) sublevels of \(3s3p\) \(3P_1\) level that can have very different lifetimes, depending on their \(F\) values. The two strongest decay branches combine to give the \(J = 2\) level in the Mg-like ions of present interest a level lifetime of the order of 4 ms (Cu) to 15 ms (Co) \[31\]. If there is a nonvanishing nuclear spin, only three out of the four hyperfine sublevels of the \(J = 2\) level in Cu (three out of five sublevels in \(^{59}\text{Co}\)) take part in the hyperfine mixing with the \(J = 1\) levels.
Table 1. Decay rates $A_{ki}$ of the $3s3p^3P_0^o$ level in Mg-like ions from the present experimental and theoretical work. Previous theoretical results, including those for Fe$^{14+}$ in order to illustrate the charge scaling, are given for comparison. HFI signifies the presence of hyperfine interaction.

| Ion     | Decay to level | Branch fraction | Total decay rate | Life time | Experiment lifetime |
|---------|----------------|-----------------|------------------|-----------|---------------------|
| Fe$^{14+}$ | $3p^0_0$ (s$^{-1}$) | $3p^0_1$ (s$^{-1}$) | $1S_0$ (s$^{-1}$) | $1S_0$ (s$^{-1}$) | (s$^{-1}$) | (ms) | (ms) |
| HFI     | 3.0$^a$ | 3.3$^a$ | 8.9$^a$ | 40.3$^a$ | 24.8$^a$ | – |
| No      | 37.4$^b$ |
| Co$^{15+}$ | 0.019$^a$ | 68.0$^a$ | 4.30$^a$ | 6.3$^a$ | 72.3$^a$ | 13.8$^a$ |
| HFI     | 70.1$^b$ |
| $^{59}$Co$^{15+}$ | 3/2 | 66.2 | 4.25 | 6.04 | 70.4 | 14.2 |
| 11/2    | 66.2 | 4.25 | 6.04 | 70.4 | 14.20 |
| 5/2     | 66.2 | 63.2 | 48.8 | 129.4 | 40.3 |
| 7/2     | 66.2 | 108.8 | 62.2 | 175.0 | 5.71 |
| 9/2     | 66.2 | 104.8 | 61.3 | 171.0 | 5.85 |
| Ni$^{16+}$ | 0.039$^a$ | 121$^a$ | 5.56$^a$ | 4.65$^a$ | 127$^a$ | 7.87$^a$ | 7.672 |
| 123$^b$ |
| 125$^c$ |
| Cu$^{17+}$ | 0.078$^a$ | 212$^a$ | 7.13$^a$ | 3.26$^a$ | 219$^a$ |
| HFI     | 216$^b$ |
| Cu$^{63}$ | 0.0952 | 219 | 7.12 | 3.26 | 226 |
| All F   | 219 | 33 | 12.9 | 256 | 3.91 |
| Cu$^{65}$ | 219 | 35 | 13.5 | 259 | 3.86 |
| All F   | 4.02 ± 0.02$^f$ |

$^a$Aggarwal et al [31].
$^b$Kaufman and Sugar [17], M1 decay only.
$^c$Slow decay component.
$^d$Fast decay component.
$^e$NIST ASD on-line database [16], M1 decay only.
$^f$Representing sliding average at early time after the begining of trapping cycle.

Thus the four (five) sublevels feature different lifetimes, one (two) remaining unchanged from the case without hyperfine interaction, whereas the others are reduced by different amounts. For the two Cu isotopes, the order of magnitude of the decay-rate increase (lifetime reduction) is roughly 10% (almost 50% in $^{59}$Co). We thus obtain the various decay rates for the individual $F$ levels of the $^3P_2$ level in $^{59}$Co$^{15+}$, as listed in table 1, while lifetime results for individual $F$ levels in Cu are listed in table 2.

For nickel, we have performed no new calculation, but in table 1 we list the previous results. For Cu (table 2), both isotopes of interest have a nuclear spin $I = 3/2$. Of the $3s3p^3P_2$ sublevels ($F = 1/2 \ldots 7/2$), the $F = 7/2$ sublevel has no mixing partner; hence its total decay rate is...
Table 2. Lifetimes (in ms) calculated in this work for the $3s^3p\ ^3P^o_2$ hyperfine sublevels in Mg-like Cu isotopes, together with the $(2F + 1)$ weighted average.

| Ion       | $F$ level | 1/2  | 3/2  | 5/2  | 7/2  | Average |
|-----------|-----------|------|------|------|------|---------|
| $^{63}\text{Cu}^{17+}$ | 4.12      | 3.58 | 3.38 | 4.42 | 3.91 |
| $^{63}\text{Cu}^{17+}$ | 4.26$^a$  | 3.84$^a$ | 3.66$^a$ | 4.48$^a$ | 4.08$^a$ |
| $^{65}\text{Cu}^{17+}$ | 4.08      | 3.49 | 3.27 | 4.42 | 3.86 |
| $^{65}\text{Cu}^{17+}$ | 4.265$^b$ | 3.833$^b$ | 3.650$^b$ | 4.491$^b$ | 4.08$^b$ |

$^a$Andersson et al [38].

$^b$Kang et al [37].

not affected by hyperfine quenching. For the other three sublevels, the hyperfine interaction increases the decay rate by 24.26, 10.97 and 7.09% ($F = 5/2, 3/2, 1/2$), respectively. Such decay rates (and hence the level lifetimes) are too closely spaced for a meaningful statistical analysis. Assuming a level population proportional to $(2F + 1)$ (statistical weights), a lifetime prediction (for $^{63}\text{Cu}$) would run to 3.91 ms. However, this average would itself be time dependent, because the decay components from the shorter-lived sublevels would disappear first—as has been seen in the experiment (see below).

In the case of copper, we have also calculated E2 matrix elements for the transition to $^3P^o_0$, which arise due to a nonzero electric quadrupole moment of the nucleus (all magnetic and electric quadrupole moments have been taken from [36]). The E2 hyperfine interaction matrix elements obey selection rules different from those for M1 transitions and can couple e.g. the $3s3p\ ^3P_2$ and $3s3p\ ^3P^o_0$ states. Yet the effect is very limited, because the E2 matrix elements are typically between one and two orders of magnitude smaller than the M1 ones. For example, the $3s3p\ ^3P^o_2\ F = 7/2$ level E2 decay rates change the level lifetime from 3.379 to 3.377 ms. Very recently, Kang et al [37] and Andersson et al [38] have also calculated the M1, M2 and hyperfine-induced decay rates of the $3s^3p\ ^3P^o_2$ level in a number of ions, including $^{63}\text{Cu}$ and $^{65}\text{Cu}$, respectively. Their M1 and M2 transition rates are close to ours.

Coupling a nonvanishing nuclear spin $I$ to the $3s^3p\ ^3P^o_0$ level results in a single ($F = I + J = 3/2$) level that is no longer subject to the ‘no 0–0 transition’ selection rule. For the Mg-like ions of $^{63,65}\text{Cu}$, the hyperfine-induced decay reduces the $^3P^o_0$ level lifetime from close to infinitely long to the range of 40–30 ms—a substantial effect. Although both isotopes have the same nuclear spin $I = 3/2$, the resulting calculated hyperfine-quenched lifetimes differ by about 10%. The new calculations, listed in table 3, change the predictions for the $3s^3p\ ^3P^o_0$ level lifetime from 39.0 and 34.1 ms for $^{63}\text{Cu}$ and $^{65}\text{Cu}$, respectively, as obtained in the earlier work [21], to 33.1 and 29.0 ms now. We note that Chen and Cheng [44] have obtained rather similar values of 31.66 and 27.69 ms from their recent RCI calculations (see table 3). The calculations by Kang et al [45] relate to $^{65}\text{Cu}$ only, whereas Andersson et al [38] calculate $^{63}\text{Cu}$ only; both results are close to those obtained by Chen and Cheng.

In an attempt to prevent either the experimental data analysis or theory from taking guidance through the other results, the final experimental results were determined independently of the theoretical calculations of this study, and also without any communication of preliminary experimental results to the theory side of the collaboration.
Table 3. Decay rates and lifetimes of the $3s3p^3P^o_0$ level in Mg-like ions from this work and previous calculations, as indicated.

| Ion       | Decay rate (s$^{-1}$) | Lifetime (ms) |
|-----------|-----------------------|----------------|
| $^{59}$Co$^{15+}$ | 42.55                 | 17.3$^a$       | 23.50 | 57.8$^a$ |
| $^{63}$Cu$^{17+}$ | 30.2                  | 25.64$^a$      | 33.1  | 39.0$^a$ |
|           |                       | 31.6$^b$        |       | 31.7$^b$ |
|           |                       | 34.9$^c$        |       | 28.7$^c$ |
| $^{65}$Cu$^{17+}$ | 34.5                  | 29.3$^a$       | 29.0  | 34.1$^a$ |
|           |                       | 36.5$^b$        |       | 27.4$^b$ |
|           |                       | 36.76$^d$       |       | 27.2$^d$ |

$^a$Marques et al [21].
$^b$Chen and Cheng [44].
$^c$Andersson et al [38].
$^d$Kang et al [45].

3. Experiment

Our experiment employed the TSR heavy-ion storage ring at the Max Planck Institute for Nuclear Physics, at Heidelberg, Germany, using the procedures described previously [24, 28], [39]–[41]. Negative Cu or Ni ions from a sputter-type ion source were accelerated to an energy of about 10 MeV in the first half of a tandem accelerator, stripped to charge state $q = 6+$ in a gas stripper, and accelerated further to a final energy of about 70 MeV. This ion beam was stripped again, this time by being passed through a thin foil. At our beam energies, charge states of $q = 17+$ (Cu) or $q = 16+$ (Ni) are favored. Correspondingly, a beam of Co ions at $q = 15+$ was obtained at an ion beam energy of 47.6 MeV. A beam of ions of the selected charge state only was transported to and injected into the storage ring. Multiturn injection and stacking of the ions over about 30 turns increased the number of stored ions, so that ion currents in the ring reached up to about 40 µA for $^{63}$Cu$^{17+}$, $^{65}$Cu$^{17+}$ and $^{58}$Ni$^{16+}$ ions, respectively, and 30 µA for $^{59}$Co$^{15+}$. The ions were left coasting for 200 ms, and then the stored ion beam was dumped and the procedure repeated.

A few per cent fraction of the ion beam was expected to be in excited levels from the foil stripping and excitation processes that take place between the injector and the storage ring. The ion beam travels about 100 m from the injector to the ion storage ring, which at these ion energies takes about 6 µs, corresponding to about two revolution periods of the ions in the storage ring (circumference 55 m). Injection extended over $\approx 0.3$ ms; the pulsed magnetic field used to inflect the ions settles down at $\approx 0.8$ ms after the start of the injection. The full injection and settling time is $\approx 1$ ms, which is short compared to the shortest of the expected radiative lifetimes studied here, but very long compared to all relevant cascade transitions from higher-lying levels. After the end of the settling time, the ions are stored on stable orbits. The storage time constants (limited by collisional losses) depend on the background gas pressure (here a few times $10^{-11}$ mbar); the associated ion beam lifetime was measured as 13.6 s for the Co ion beam, 23–27 s for the Ni ions and about 22–65 s for the Cu ions, depending on beam focusing and beam energy as well as on the vacuum conditions. The actual ion beam current.
was monitored on-line by a beam profile monitor (BPM) that detects residual gas ions that are collisionally produced by the circulating ion beam. The detector is also sensitive to extreme ultraviolet light; hence on top of the ground state ion signal the BPM signal shows a (relatively small) decay curve component that relates to a superposition of the 3s3p level decays, of which the 3s3p $^3P^o_2$ level stands out (see the discussion below). The finite ion storage time reflects the collisional ion loss rate from the stored ion cloud, which was subtracted from the total decay rate of (visible or EUV) emission in order to derive the radiative decay rate.

We used optical observation in a side-on geometry for two separate photon measurements. The experiment used optical observations in the visible range for the M1 decays of the 3s3p $^3P^o_2$ levels to 3s3p $^3P^o_1$, and EUV observations for the M1 decay of the 3s3p $^3P^o_0$ level to the $3s^2\,^1S_0$ ground state. Although the techniques used in both cases are similar in many aspects, we split the presentation according to the two levels studied.

3.1. 3s3p $^3P^o_2$ level

The near-UV or visible light from the 3s3p $^3P^o_2$–3s3p $^3P^o_1$ M1 transition passed through a sapphire window 5 cm from the average ion trajectory. In order to boost the signal rate, a light collection system was employed [24]. This simple trough-shaped reflector (of elliptical cross section, with the cylinder axis oriented along the beam trajectory) enhances light collection by about a factor of two. The light (for Co$^{15+}$ at $\lambda = 574.4$ nm, for Ni$^{16+}$ at $\lambda = 475.01$ nm [42] and for Cu$^{17+}$ at $\lambda = 394.275$ nm [43]) was detected by a 25 mm diameter end-on window photomultiplier tube with an inherent dark rate of some 15 counts per second (bi-alkali-type EMR 541 N for visible and near-UV light), combined with an interference filter with a central wavelength of $\lambda_0 = 577$ nm and bandpass $\Delta\lambda = 10$ nm $^5$ for Co, $\lambda_0 = 476$ nm and $\Delta\lambda = 10$ nm for Ni and $\lambda_0 = 394$ nm and $\Delta\lambda = 4$ nm $^6$ for Cu). In these transmitted wavelength intervals, only the respective lines of interest are expected to show.

Each detection cycle was started about 1 ms before injection, and events were sorted into 200 bins of 1 ms width each. Every minute the accumulated data were stored, and the (many hundreds of) 1 min data sets were afterwards checked for periods of ion beam loss or similar events, and then the corresponding (few) sets discarded. Total data accumulation time was $\approx 15$ h for Co$^{15+}$, $\approx 13$ h for Ni$^{16+}$, $\approx 13$ h for $^{63}$Cu$^{17+}$ and $\approx 25$ h for $^{65}$Cu$^{17+}$. The 570 nm wavelength of interest in Co$^{15+}$ is very close to the daylight spectral intensity distribution maximum near 550 nm, while the corresponding lines in Ni$^{16+}$ and Cu$^{17+}$ are successively farther away on the blue side. This situation is reflected in a variation of the photomultiplier background level (worst for Co), which clearly dropped after sunset and with clouds overhead, indicating that daylight in the storage ring building could enter the vacuum vessel in spite of the precautions taken. In a check for systematic errors, also the photomultiplier voltage was varied, finding no effect on the lifetime results. While the higher signal rate and lower background of the nighttime data runs were beneficial for achieving small statistical uncertainties of the fit results, the results did not vary systematically with the signal-to-background ratio. Examples of the data recorded on the M1 transition signal are shown in figures 2–4.

The data sets for the $^3P^o_2$ levels of each isotope were combined into groups, which were then evaluated individually. For Cu and Ni, the data analysis proceeded by straightforward fitting of a single exponential plus background to the data; the fit curve approximates the data so

$^5$ Manufactured by Omega Optical Inc., Brattleboro, VT, USA.

$^6$ Manufactured by BK Interferenzoptik, Nabburg, Germany.
well that no second exponential component is recognized. The extracted decay time constant was corrected for relativistic time dilation ($\gamma \approx 1.0013$) and for the ion loss rate from the stored ion beam. The latter correction amounts to 0.03% for Ni and to 0.01% for Cu, partly canceling the minute relativistic correction. The errors of the corrections were much smaller than the smallest statistical uncertainty of the data. Besides the negative test for an additional exponential component, we have truncated early data channels (up to 10 ms) from the decay curves in order to check for perturbations from the injection process into the storage ring or the presence of blending transitions with a slightly different time constant. In the Ni data, no such effect was seen beyond the first two channels (2 ms) of the data recording; consequently, the statistical variation of the fit results was taken into account for the determination of the lifetime measurement error. A very precise result of $(7.672 \pm 0.015)$ ms is obtained for the Ni$^{16+}$ 3s3p $^3P_2$ level in an Mg-like ion species without hyperfine effects.

For the 3s3p $^3P_2$ level in Co$^{15+}$, the decay curve (figure 2) reveals the presence of several decay components. Our own calculation predicts a level lifetime of 14.2 ms, while Aggarwal et al predict 13.8 ms for the $F = 3/2, 11/2$ levels, which are not affected by hyperfine quenching. In contrast, the $F = 5/2, 7/2$ and $9/2$ levels are quenched by hyperfine mixing, and our calculations predict lifetimes of 7.73, 5.71 and 5.84 ms, respectively. Hence the decay curve is expected to be dominated by two components of about 14 and 6 ms, respectively, which cannot reliably be separated in unconstrained multi-exponential fits. However, when truncating early data channels, the relative weight of the slower decay component increases. In this way, a result of $(13.6 \pm 1.8)$ ms was determined from ‘late’ fits. This lifetime was then set fixed in constrained fits to data that included earlier channels, and a faster component with an effective lifetime of $5.5 \pm 2.5$ ms was found to represent the hyperfine-quenched levels. The uncertainty...
Figure 3. Photomultiplier signal (logarithmic scale) obtained with $^{58}\text{Ni}^{16+}$, observing the M1 decay wavelength of the 3s3p $^3\text{P}_0$ level decay to the 3s3p $^3\text{P}_1$ level, and a single exponential plus background fit to the data. Only the first half of the 200 ms measurement cycle is shown. The statistical error bars are smaller than the data symbols. The total data represent a collection time of about 240 s per 1 ms wide data channel. The signal-to-background ratio is better for the night data because of both a lower stray light level and a higher photomultiplier voltage. The fit results for the level lifetime (raw nighttime data 7.67 ± 0.02 ms, raw daytime data 7.62 ± 0.04 ms) agree within the error bars and yield a combined lifetime result (after the corrections discussed in the text) of 7.672 ± 0.015 ms.

of this component is especially large, because only a subset of the recorded data could be used when a bright fast signal component of unidentified origin appeared in a period of about half the data accumulation time.

In Cu, single lifetime values comprising all $F$ components were derived for each of the two nuclides studied. A systematic increase in the fit results with later start channels was observed; this has to be expected from the presence of several hyperfine levels with different individual lifetimes (see table 2). (These results represent level averages, where the three levels affected by hyperfine interaction decay slightly faster than the $F = 7/2$ level that is not affected by hyperfine interaction.) For example, fits return a lifetime result of about 4.04 ms for very early starting points and about 4.10 ms for starting about 5 ms later. If the systematic trend towards longer lifetimes with the cutting off of early channels was disregarded, the analysis by fitting a single exponential would justify quoting a Cu$^{17+}$ 3s3p $^3\text{P}_2$ level lifetime of 4.05 ± 0.02 ms (neglecting isotopic differences). However, the statistical errors in the results for both individual nuclides are low enough to find a significant difference between them. When comparing the lifetime results for the two isotopes for corresponding starting channels of the evaluation (see figure 5), the fit results consistently display a 1% difference in apparent decay time constant. This difference is larger than the combined error bars and we identify it with the predicted isotope effect difference between the two nuclides $^{63}\text{Cu}$ and $^{65}\text{Cu}$. However, from the start channel-dependent (sliding) fit results for each isotope, we derive representative results with somewhat larger error bars.
Figure 4. Photomultiplier signal (logarithmic scale) obtained with $^{63,65}$Cu$^{17+}$, observing the M1 decay of the 3s3p $^3P_0^0$ level decay to the 3s3p $^3P_1^0$ level (lifetime close to 4 ms), and a single exponential plus background fit to the data. Only the first half of the 200 ms measurement cycle is shown. The error bars are included, but are smaller than the data symbol size. The data represent a collection time of about 260 s per 1 ms wide channel for $^{63}$Cu and 450 s for $^{65}$Cu. The slight lifetime difference between the isotopes is too small to be recognized by eye.

(of 0.5%) that comprise a measure of the variation with starting channel, namely lifetimes of 4.06 ± 0.02 ms and 4.02 ± 0.02 ms for $^{63}$Cu and $^{65}$Cu, respectively. All experimental lifetime results are included in table 1.

3.2. 3s3p$^3P_0^0$ level

The Cu$^{17+}$ ion is the only one we studied for the $J = 0$ level decay. For the 3s3p $^3P_0^0$ level decay to the ground state, the wavelength of 35.74 nm [43] is in the EUV, and no filter can be used. In earlier experiments, the sensitivity to EUV light of the BPM, part of the beam diagnostics system of the heavy-ion storage ring, has been noted [39]. This monitor consists of microchannel plates (MCP) and mainly detects the spatial distribution of residual gas atoms ionized by collisions with the energetic beam particles. An electric field helps to move the low-energy recoil ions to the actual MCP. The signal of this detector is dominated by the ionization effected by the most abundant ion fraction, which is in the ground state. Ions in excited states tend to have slightly larger collision cross sections and thus may be recognized by the time dependence of their contribution, corresponding to the excited level lifetime. Moreover, if these excited ions emit EUV or X radiation, it will also be detected by the BPM. Figure 6 shows examples of BPM signals accumulated with the two Cu isotopes. The fast initial component has a time constant very close to the lifetime of the 3s3p $^3P_0^0$ level. However, the far stronger residual gas ionization signal associated with the ground state ions dominates any analysis and severely limits any meaningful determination of excited level lifetimes from such data.

We have therefore constructed a separate detector for EUV light, placed inside the storage ring vessel, which does not attract recoil ions from the residual gas and thus provides a better ratio of EUV signal to ionization-related background. EUV detection in the present experiment
Figure 5. 3s3p $^3P_2$ level lifetime fit results for $^{63,65}$Cu$^{17+}$ ions for evaluations beginning at ten consecutive starting channels. The upper graph shows the individual results; the lower graph shows the deviation of the ratio of corresponding fit results for the same starting channel from unity (in per cent). Over the range of starting channels (one every millisecond), the amplitude of the optical decay signal decreases by about a factor of 10, which causes a corresponding increase in the error bar size. If there was no isotope effect, the ratio should be equal to one, and the deviation zero. The observed deviation is in better agreement with a value of 1% (indicated by the horizontal bar), as is predicted by theory.

employed two open channeltrons positioned within a few centimeters of the ion beam. The channeltrons were operated with the front end at ground potential, inside a grounded shield with a thin wire mesh between the ion beam and the detector. In this way, the detector was screened from a large part of the stray particles or recoil ions set free by collisions of the fast ions with atoms of the residual gas. The detector has been described in more detail elsewhere [41].

*New Journal of Physics* **13** (2011) 023017 (http://www.njp.org/)
Figure 6. Examples of BPM signals accumulated for a number of hours over the 200 ms measurement cycle used for Cu$^{17+}$ ions, injecting one isotope species at a time. The first two data channels have been discarded as they show the period of ion dumping and new injection that cannot be evaluated for the lifetimes sought. The initial fast component of the curve corresponds to the 3s3p $^3P_0^o$ level. Any contribution specific to the 3s3p $^3P_0^o$ level would be expected at an amplitude about 40–50 times smaller.

With a roughly 30 ms lifetime to be measured, the data acquisition proceeded as outlined before. Sample curves are shown in figure 7. Owing to the technical precautions, the EUV signal-to-ion background ratio was much more favorable than that of the BPM. Nevertheless, the ion background was still relatively high and dominated the overall signal. This is a major impediment, even though the slope of the background (due to ion loss from the stored beam) is very small and can be measured independently (but only over much longer time intervals).

In the observed decay curves (figure 7), isotopic differences appear most clearly between $\approx 10$ and 70 ms, but no individually recognizable decay components of the expected time constant near 30 ms can be found. We consider this to be caused by the superposition of signals from various EUV decay channels (see figure 1). While the $^3P_1^o$ level has a lifetime of only 9 ns [30], much shorter than the transit time of the ions between stripper foil and storage ring, the $^3P_2^o$ level feeds the $J = 1$ level by its 85% M1 decay branch, and the subsequent spin-changing intercombination decay is seen by the EUV detector, along with the 15% direct M2 + hyperfine-induced decay of the same $^3P_2^o$ level. Thus 100% of the $^3P_2^o$ level decay appears superimposed on the $^3P_0^o$ level decay curve. A presumably statistical population of the fine structure levels gives a five times higher weight to the $J = 2$ level than to the $J = 0$ level; the initial amplitude of the signal curve is moreover proportional to the decay probability, which adds another factor of eight to the short-lived decay. Hence the component of interest, the decay signal of the $^3P_0^o$ level, can be expected with an initial amplitude that is about a factor of 40 lower than that of
Figure 7. Signal (logarithmic scale) obtained with the windowless channeltron detectors when $^{63,65}\text{Cu}^{17+}$ ions were injected. An isotopic difference is evident. The fast decay curve component is relatively much more prominent than with the BPM measurement of figure 4, owing to a suppression of charged particles from detection. The early part of the curve is dominated by the (direct and indirect) $3s3p\,^3\text{P}_0^0$ level decay with its 4 ms time constant, followed by the $3s3p\,^3\text{P}_0^2$ level decay with an expected time constant near 30 ms. (Fitted results here are 20 and 24 ms, respectively.) If only the $3s3p\,^3\text{P}_0^0$ levels played a role, the curves should be fitted well by three exponentials (the third representing ion loss from the stored ion cloud, on a time scale that is three orders of magnitude longer) and a background. The two-exponential fitted curves (black) in the example show a less than perfect fit with the fitted curve overshooting near 30–40 ms and beyond 160 ms, and undershooting the data in the range 60–120 ms (most clearly in the $^{63}\text{Cu}$ data), which suggests a more complex physical situation and precludes a definite fit result for the $3s3p\,^3\text{P}_0^0$ level lifetime. However, fits with three exponentials showed no significant improvement.

In principle, the two level lifetimes of about 4 and 30 ms are sufficiently different, so that multi-exponential analysis should be no problem. Unfortunately, however, the wanted EUV photon signal also sits on top of a high ion beam related background with its own statistical scatter. Fits to the data by two or three exponentials (plus a background contribution) failed to describe the data well enough. Any such fits in which early channels were successively truncated up to 40 ms after injection returned results with a scatter that exceeded the statistical uncertainties by far. Beginning a fit of two exponentials at the early channels returns one component with a good approximation to the roughly 4 ms $^3\text{P}_0^0$ level lifetime, and a slower
component of about 20 ms for $^{65}$Cu (24 ms for $^{63}$Cu). Beginning the fit at later start channels, the slower component lifetimes gradually increase (towards 24 ms and a range of 30–40 ms, respectively), but they do not stabilize near the expected $^3P_0^0$ level lifetime values from our own predictions or those made by Chen and Cheng [44]. The situation is rather similar for fits using three exponential components. One component represented the 4 ms $^3P_0^2$ level lifetime, while two components represented the $^3P_0^0$ level decay and whatever other contribution to the overall decay curve. However, no stable fit result for the $^3P_0^0$ level lifetime emerged. When fixing one component lifetime to various test values of the expected $^3P_0^0$ level lifetime value, the result for the third component remained unstable.

Apparently, the decay curves are more complex than expected, and they are not yet fully understood. The lack of stability of the fit results obtained under different assumptions makes us refrain from actually quoting lifetime results from this part of our experiment. Nevertheless, the data suggest $^3P_0^0$ level lifetimes some 25 ± 10% shorter than most recently predicted. Although the lifetime of the $J = 0$ level is longer than that of the $J = 2$ level discussed above, the recognized systematic errors are not much larger. The ion beam lifetime (inverse of the ion loss rate) correction remains in the range of $<0.2\%$ and cannot account for the disagreement of the fit results with predictions.

After subtraction of the fast decay component and the perceived background, the ratio of the signals of the two isotopes, also expected to be an exponential, is compatible with the expected 10% isotope effect on the decay rates (analogous to the 1% isotope effect for the $^3P_0^2$ level lifetime demonstrated in figure 5), for fits starting within the first about 20 ms of the data, before the $J = 0$ signal amplitude becomes so small that the fit results scatter too much to be useful.

4. Discussion and outlook

Figure 8 presents predictions and our experimental results for the $3s3p \, ^3P_0^0$ level total decay rates for the elements listed in table 1, using a rough Z-scaling to magnify the data range of interest and to bring the data for different elements onto a common scale. Our measured lifetime for the Ni$^{16+}$ $3s3p \, ^3P_0^0$ level of $(7.672 \pm 0.015)$ ms differs from the NIST listing (M1 decay branch only, M2 decay not considered) by 4%. The calculations by Aggarwal et al [31] estimate the M2 branch fraction at 4.5%, which fully corroborates the shortcoming of the NIST database entry. Figure 8 demonstrates the size of this contribution by displaying predictions with and without the M2 decay branch. The Aggarwal et al lifetime prediction of 7.87 ms differs by 2.5% from our measurement result. This deviation appears to mark the calculational precision in the absence of hyperfine effects.

Our measured total decay rate for the $^3P_0^0$ level in Cu$^{17+}$ deviates by 15% from the calculated M1 decay rate. Explaining the difference by M2 and hyperfine-induced decay, our calculations show that the hyperfine-induced decay rate is about three times higher than the M2 rate. This finding corroborates the observation by the astronomer I S Bowen, who realized already in the 1920s that the $^3P_0^0$ level in Hg I had a surprisingly strong direct decay to the ground state, and his suggestion that the hyperfine-induced decay was more important for this than the M2 decay. The 15% deviation also highlights how misleading the notion of the M1 decay channel as the only important decay channel would be.

At about 0.5% precision of our lifetime experiment (taking into account the need for averaging over several hyperfine structure sublevels), our statistical uncertainty is small enough to determine the overall hyperfine quenching to about 5% and the isotope difference of the...
Figure 8. Scaled lifetimes of the $3s3p \, ^3P_2$ level in Mg-like ions. The data entries have been scaled by $(Z-10)^{10}$ to account roughly for the underlying $Z$-dependences of gross and fine structure. The measurement for Ni demonstrates the need to include not only the M1 decay, but also the M2 decay branch, as well as the quality of the calculations by Aggarwal et al [31] (red) and our own (blue) in the absence of hyperfine interaction. The data point for Cu demonstrates the extent of hyperfine-induced quenching and how well the calculations match the experimental finding; the hyperfine interaction contribution in both cases is the one calculated in our work. The measurement for Co is not shown, because its error bar spans the full height of the diagram. The calculational data provided by Kaufman and Sugar [17] and by NIST [16] (not shown) are based on the M1 decay branch only; the corresponding partial lifetimes fall in between the M1 and the M1 + M2 + M2 predictions shown in the figure.

hyperfine quenching to within some 30%. The measurement tests the prediction for the M2 rate at the 13% level while the dominant M1 decay rate is tested to better than 1%. The experimental results for Ni and Cu are compared to predictions in figure 8.

For the $3s3p \, ^3P_2$ level in Co$^{15+}$, the calculations by Aggarwal et al estimate the M2 branch fraction at 6.3%; their lifetime prediction is 13.8 ms without the hyperfine-induced decay contribution. Our own calculation including hyperfine interaction predicts a level lifetime of 14.2 ms, close to the $(13.6\pm 1.8)$ ms we measure with limited precision and therefore not included in figure 8, as a component of a multi-exponential decay. This component represents the $F = 3/2$, $11/2$ levels that are not affected by hyperfine quenching. The $F = 5/2$, $7/2$ and $9/2$ levels are quenched by hyperfine mixing, our calculations predicting lifetimes of 7.73, 5.71
and 5.84 ms, respectively. One cannot expect to resolve these three closely spaced components by multi-exponential fits. However, a second decay component in our data, 5.5±2.5 ms, is consistent with this lifetime range of the hyperfine-quenched levels. All in all, the 3s3p 3P_2 level lifetime data for the four isotopes studied demonstrate the high degree of understanding reached in atomic structure theory even as the isoelectronic trends of the predictions (see figure 8) differ slightly. The M1 decay branch is tested to rather high precision in Ni, the importance of the M2 decay branch is tested to a few per cent in Ni and Cu (and somewhat less precisely in Co), and the hyperfine quenching contribution to radiative decay, including the differential effect in two isotopes, is measured with notable precision in Cu.

Concerning the 3s3p 3P_0 level, the general assumption is that hyperfine quenching provides the only mechanism that enables a single-photon decay and thus a decay rate amenable to laboratory studies. In various He-like ions studied decades ago [3]–[9], [11], hyperfine quenching calculations seemed to be validated, and our own results on the 3s3p 3P_0 level corroborate the same type of calculations with notable accuracy. Hence it is somewhat surprising to find that our experiment on the 3s3p 3P_0 level yields fit results that differ drastically (roughly 25%) from prediction. These fit results deviate from expectation by about as much as the recent heavy-ion storage ring lifetime measurements on the hyperfine-quenched 2s2p 3P_0 level in the Be-like ion of 47Ti. However, the fit results for the present data are not stable enough as a function of the start channel for being reliable. The insufficiently understood decay curves in our case deter us from claiming that we have already determined the lifetimes of interest in Mg-like Cu ions. Our observations nevertheless point to lifetime values that are significantly shorter than are currently predicted. It should be interesting to apply the DR technique used by Schippers et al [26] for Be-like ions also to the present example of Mg-like ions. Another option would be a measurement by time-resolved EUV spectroscopy at an electron beam ion trap [46].

For any future measurement using photon detection (as in the present study), it would be advantageous to further improve the signal-to-background ratio by better suppressing the beam-related background. The dark rate of the EUV detecting channeltrons is negligibly small. Practically all of the background contribution to the decay curve is related to the presence of the stored ion beam. This radiation background may be from residual gas being ionized and/or excited by the ion beam, from the charge exchange of recoil ions with residual gas atoms and molecules, from recoil ions recombining upon reaching the inner surface of the vacuum vessel and sending out VUV light, or from residual gas ions actually striking the channeltron. Suppressing this background would require an optical enclosure of the detection volume, but must not interfere with the ion beam during focusing, injection and storage. Such an engineering effort, however, has not yet been undertaken.

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