LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-JRNL-226985

Observation of hyperfine mixing in measurements of a magnetic octupole decay in isotopically pure nickel-like 129Xe and 132Xe ions

E. Träbert, P. Beiersdorfer, G. V. Brown

December 22, 2006

Physical Review Letters

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

Observation of hyperfine mixing in measurements of a magnetic octupole decay in isotopically pure nickel-like ^{129}Xe and ^{132}Xe ions

E. Träbert,* P. Beiersdorfer, and G. V. Brown

High Temperature and Astrophysics Division,

Lawrence Livermore National Laboratory,

Livermore, CA 94550-9234, U.S.A.

Abstract

We present measurements of high statistical significance of the rate of the magnetic octupole (M3) decay in nickel-like ions of isotopically pure ^{129}Xe and ^{132}Xe. On ^{132}Xe, an isotope with zero nuclear spin and therefore without hyperfine structure, the lifetime of the metastable level was established as (15.06±0.24) ms. On ^{129}Xe, an additional fast (2.7±0.1 ms) decay component was established that represents hyperfine mixing with a level that decays by electric quadrupole (E2) radiation.

PACS numbers: 32.30.Rj; 32.70.Cs; 32.10.Fn

*Electronic address: traebert@ep3.rub.de; Also at Ruhr-Universität Bochum, D-44780 Bochum, Germany

The hyperfine interaction, although a small effect, provides a powerful window on atomic-nuclear interactions and results in manifestations exploited in multiple branches of physics. Hyperfine mixing affects the line intensities in atomic and singly ionized heavy elements [1, 2] seen in astrophysics [3, 4], as well as of susceptible light ions observed in planetary nebulae [5]. The hyperfine levels of the ground state of atomic hydrogen give rise to the 21-cm line used in radioastronomy to image our galaxy and were the basis for the first maser (see [6]). Hyperfine line splitting is of importance in photopumped x-ray lasers [7], and hyperfine interaction affects the polarization of spectral lines used in plasma diagnostics [8]. Hyperfine interaction is sufficient to break symmetry in atoms and ions with a J=0 excited level above a J=0 ground level (typically in two-electron ions), and this can turn a very long-lived level (for example, 1s2p $^3\mathrm{P}^\mathrm{o}_0$ in heliumlike ions) into a shorter one [9], in some cases drastically so [10]. Hyperfine mixing also affects the lifetimes of other levels, such as the 1s2p $^3\mathrm{P}^\mathrm{o}_2$ level of He-like ions first measured in the 1970s [11]. More recently, such hyperfine-mixing induced decays have been studied in the quest for extremely accurate atomic clocks [12], as have been levels that can decay only via high multipole order transitions [13]. The property that these decays have in common is their low transition probability, which translates into a long upper level lifetime and a very narrow natural line width. The same longevity renders these levels of interest in plasma diagnostics; depending on the actual lifetime, they serve as density diagnostics in terrestrial and astrophysical plasmas [14, 15]. In some cases, only the small transition amplitude lets an emission line reach Earth while all other lines appear only as absorption features [16] after passage through the interstellar medium.

Recently Yao *et al.* [17] have postulated that the hyperfine interaction should affect the lifetime of the lowest excited level in nickel-like ions. That level ($3\mathrm{d}^9 4\mathrm{s}\ ^3D_3$; cf. Fig. 1) can decay to the closed-shell $3\mathrm{d}^{10}\ ^1S_0$ ground level only via a magnetic octupole (M3) transition [18]. The lifetime of that level had been measured recently for several nickel-like ions [19]. The value found for the $3\mathrm{d}^9 4\mathrm{s}\ ^3D_3$ level in natural Xe^{26+} was (11.5±0.5) ms [19] and significantly shorter than calculated without taking hyperfine structure into account. Moreover, Yao *et al.* noted that in this atomic system the mixing is so strong that not only the usual mixing coefficient, but also interference terms play a massive role and cause a considerable variation of the magnetic sublevel (m_F, with quantum number $F = J + I$) lifetimes. In short, for a Xe^{26+} ion without hyperfine structure (HFS), they compute a level lifetime of 15.1 ms. The same lifetime is valid in isotopes with HFS for those sublevels that

do not mix (for example, $F = 7/2$ in ^{129}Xe, which has a nuclear spin $I = 1/2$). The standard mixing coefficient describing the mixing with the $^3D_2$ level, which has a comparatively fast E2 decay channel, would reduce the lifetime of the other sublevels in ^{129}Xe ($F = 5/2$) to 2.84 ms. The interference term would introduce a variation of the actual sublevel lifetimes by up to ± 30% in the case of ^{129}Xe, so that they would range from 2.19 ms to 4.02 ms [17]. The existence of this interference effect has been disputed [20]. Because of the larger nuclear spin, there are more m_F sublevels in ^{131}Xe ($I = 3/2$) than in ^{129}Xe. Without the interference effect, the four F levels are predicted to have lifetimes of 15.12 ms (F=9/2), 6.15 ms (F=7/2), 6.30 ms (F=5/2), and 8.92 ms (F=3/2), respectively. Here the purported interference effect would cause the 19 calculated sublevel lifetimes to spread from 3.80 ms to 23.30 ms.

Modelling a decay curve on the basis of their calculations, with level populations proportional to the statistical weights $(2F + 1)$ and a natural isotope mixture, Yao *et al.* obtained a curve that closely resembled a single exponential with an effective decay time near the experimental finding in [19]. Discerning any individual decay component from what could be fitted well by a single exponential was not possible from the relatively low-statistics experimental data, as the multitude of decay components present in natural xenon were unresolvable.

In this Letter we present measurements that clearly demonstrate the effects of hyperfine mixing between M3- and E2-decaying levels for the simpler, isotopically pure case of ^{129}Xe. In addition, we obtain reference data on ^{132}Xe, an isotope of zero nuclear spin and thus without any hyperfine structure. We have also repeated the measurement using natural xenon. A high-statistics measurement indicates the presence of several decay components, but without revealing reliable lifetime values.

The experiments were carried out on the SuperEBIT electron beam ion trap [21] coupled with a position sensitive proportional photon counter (PSPC) [22]. The PSPC has a 1 μm thick 100 mm × 8 mm polyimide window on a 90% open area gold support mesh that separates the active detector volume (at atmospheric pressure) from the $< 10^{-5}$ Torr pressure of the spectrometer. The transmission of the polyimide window at 590 eV, the energy of interest, is ∼ 22%. The viewing port of SuperEBIT restricted the collection area of the detector (placed about 35 cm from the ion trap) to 30 mm × 8 mm. The detector was operated with a P10 gas mixture of 90% argon and 10% methane and detects radiation in an energy band

from roughly 300 eV to 1200 eV. The time difference of pulses arriving at the ends of the anode wire was used to determine the position of the x-ray event along the detector window; the pulse height distribution was analyzed to discriminate against electronic noise and higher energy x rays. While the PSPC's energy resolution of only ~ 1 keV is much poorer than that of the microcalorimeter used in the earlier experiment [19], the very much larger solid angle of observation made it possible to accumulate data with such good statistical significance that multiple decay components in the composite decay curves of highly charged Xe ions could be recognized and resolved.

For these measurements, SuperEBIT was run in a cyclic mode where the electron beam is switched on at an electron current of 13 mA and an energy of 1.5 keV for 80 ms to create and excite Xe^{26+} ions, and then turned off for up to 900 ms while photons produced by radiative decay of the metastable levels are measured. The ion trap depth was varied in the course of the experiment from 500 to 700 eV. Xe was continuously injected as a neutral material using a ballistic gas injector during the entire SuperEBIT cycle time. When the beam is off, X-ray emission from charge exchange (CX) recombination also occurs, and, consequently, the decay curves (Fig. 2) are contaminated by the CX signal. We also note that at our beam energies we create fully ionized nitrogen and oxygen, which often appear as background contaminants, and we also detected the (weak) presence of Xe^{27+}. Xe^{27+} can be created at an energy below the 1.5 keV ionization energy of Xe^{26+}, by ionizing an electron in a metastable level in Xe^{26+}. All the data were recorded in event mode using a data acquisition system described earlier [23]. The time base of the experiment referred to an atomic clock via the Loran-C network of navigational radio beacons. For our run conditions, the typical data rate was of the order of several thousand counts per second with the beam on, and 5 to 10 counts per second with the beam off. With the beam off, only a fraction of the signal resulted from the radiative decay of interest, while most of the rate was generated by the charge-exchange "tail" of the decay curve.

Decay curves were recorded at various Xe gas injection pressures from $5{\cdot}10^{-8}$ Torr to $3{\cdot}10^{-7}$ Torr; the CX tail of the decay curves then varied in slope from a decay time of 240 ms for the highest injection pressure to some 800 ms for the lowest. Control measurements were carried out with CO_2 injection instead of Xe. In this case, a radiative decay with a 1 ms lifetime was observed on top of the CX tail. This 1 ms decay is the M1 decay of the lowest triplet level, 1s2s $^3S_1$, in He-like oxygen. Its near-1 ms lifetime has been measured before

with high accuracy at the Livermore electron beam ion traps [25]; here it merely served as a confirmation of experimental arrangement. In order to find out about possible dead time effects of the signal processing electronics, fast cascades, electron beam switching or data fit problems, we evaluated the decay curve data after successively cutting out initial data channels up to 20 ms after the electron beam was switched off. The event data yielded consistent fit results irrespective of the starting channel of the evaluation.

The CX signal results from the presence of a multitude of highly charged ion species. Its time dependence reflects the ion loss from the trapped ion cloud and thus yields a measure of this systematic error. We would like to know the ion loss rate for the Xe charge state of interest (Xe^{26+}), but we only have the superposition of CX data of many charge states. Taking guidance from the superposition and the ion storage time of 500 to 800 ms (the inverse of the ion loss rate), a systematic correction to the apparent atomic lifetime values of the Xe^{26+} ions of 2 to 3% (corresponding to the individually observed storage time values) is appropriate. This ion loss correction can be determined with a statistical uncertainty of 2%, an uncertainty which has a negligible effect on the uncertainty of the radiative lifetime value sought. However, more highly charged ions are experiencing a deeper trap from which they evaporate with lower probability. At the same time, their CX cross sections are the largest. Not knowing the individual ion loss rate of the charge state of interest, we increase the associated error on the ion loss rate correction by an order of magnitude to one quarter of the systematic correction, i. e. to 0.5% of the lifetime result.

The first set of experiments used natural Xe, which comprises several isotopes of which the two odd ones (^{129,131}Xe) make up almost 50%. If the Xe contribution to the decay curves was approximated by a single exponential, the effective lifetime was of between 9 and 11 ms, in agreement with [19]. However, the improved statistics revealed that the decay curve shape was better approximated by two exponentials representing Xe (on top of the CX signal). The lifetimes derived were about 3.2 ms, with some 10% statistical uncertainty, and 13.2 ms, with a statistical uncertainty of 2%, that is less than the scatter from different data sets. These observations lent qualitative support to the hypothesis by Yao *et al.* [17] about hyperfine interaction playing an important role here, but they were not sufficient to determine reliably any of the individual level lifetimes involved.

In a second set of experiments, mono-isotopic ^{132}Xe, an isotope with zero nuclear spin, and therefore without hyperfine structure, was employed. With such an even isotope, only

a single decay component was expected besides the contribution from CX, and this was, indeed, found. The results of each day of measurement scattered by $\sim 5\%$, more than the statistical reliability of the lifetime evaluation which was of the order of 1% to 2%. A combination of a week's worth of data yielded a lifetime value of the $^3D_3$ level of 15.06±0.24 ms, which is very close to the 15.1 ms prediction by Yao *et al.* Our overall error estimate is larger by a factor of four than the purely statistical error of a fit to the sum of all data, because it accommodates the scatter observed with subsets of the data.

In a third experiment, mono-isotopic ^{129}Xe (I=1/2) was used. Here, we found two major decay components, of 15.1±0.5 ms; and of 2.7±0.1 ms. The relative prominence of the fast component was clearly higher than with a natural isotope mix. The value for the slow component agrees well with that expected for the m_F level associated with $F = 7/2$, that is, with the unmixed level. The fast component can readily be associated with the superposition of the decays of the six magnetic sublevels of the $F = 5/2$ level, confirming the effect of hyperfine mixing on the M3 transition.

A summary of the results is given in Table I. The measured Xe^{26+} M3 decay rate of 66.4±1.1 s^{-1} in the absence of hyperfine effects differs significantly from earlier predictions [19, 26], but is in excellent agreement with the GRASPUVU code calculations by Yao *et al* [17]. Similarly, the hyperfine-induced fast decay component agrees well with the hyperfine mixing prediction by Yao *et al.* [17], i.e., with the superposition of the decays of the six $F = 5/2$ magnetic sublevels, whose individual lifetimes are centered at 2.84 ms. Despite the greatly improved statistical significance of the data (below the 0.4% level) our experiment was not sensitive to the postulated interference effect, i.e., to the predicted 30% variation of the magnetic sublevel lifetimes around this mean, and no such variation was discerned. Nevertheless, hyperfine mixing has been clearly established as having a strong effect on the lifetimes of metastable levels in highly charged ions and must be considered when undertaking future measurements or interpretation of the results.

Acknowledgments

We are happy to acknowledge the dedicated technical support by Ed Magee and Phil D'Antonio, as well as help with the experiment by H. Chen, J. T. Clementson, and D. B. Thorn. E. T. acknowledges travel support by the German Research Association (DFG). The

work at the University of California Lawrence Livermore National Laboratory was performed under the auspices of the Department of Energy under Contract No. W-7405-Eng-48.

---

[1] S. Mrozowski, Zs. Physik **108** 204 (1938).

[2] S. Mrozowski, Phys. Rev. **57** 207 (1940).

[3] D. Huff, W. V. Houston, Phys. Rev. **36** 842 (1930).

[4] I. S. Bowen, (unpublished work cited by Huff and Houston)

[5] T. Brage, P. G. Judge, and C. R. Proffitt, Phys. Rev. Lett. **89**, 281101 (2002).

[6] A. L. Schawlow and C. H. Townes, Phys. Rev. **112**, 1940 (1958).

[7] J. H. Scofield, J. Nilsen, Phys. Rev. A **49**, 2381 (1994).

[8] J. R. Henderson, P. Beiersdorfer, C. L. Bennett, *et al.* , Phys. Rev. Lett. **65**, 705 (1990).

[9] B. Denne, S. Huldt, J. Pihl, R. Hallin, Phys. Scr. **22** 45 (1980).

[10] B. B. Birkett, J.-P. Briand, P. Charles P, *et al.*, Phys. Rev. A **47** R2454 (1993).

[11] H. Gould, R. Marrus, and P. J. Mohr, Phys. Rev. Lett. **33**, 676 (1974).

[12] J. von Zanthier, T. Becker, M. Eichenseer, *et al.*, Opt. Lett. **25**, 1729 (2000).

[13] M. Roberts, P. Taylor, G. P. Barwood, P. Gill, H. A. Klein, and W. R. C. Rowley, Phys. Rev. Lett. **78**, 1876 (1997)

[14] J.-U. Ness, R. Mewe, J. H. M. M. Schmitt, *et al.*, Astron. Astrophys. **367**, 282 (2001).

[15] M. Klapisch, J. L. Schwob, M. Finkenthal, *et al.*, Phys. Rev. Lett. **41**, 403 (1978).

[16] K. C. Steenbrugge, J. S. Kaastra, D. M. Crenshaw, *et al.*, Astron. Astrophys. **434**, 569 (2005).

[17] K. Yao, M. Andersson, T. Brage, R. Hutton, P. Jönsson, and Y. Zou, Phys. Rev. Lett. **97**, 183001 (2006).

[18] P. Beiersdorfer, A. L. Osterheld, J. Scofield, B. Wargelin, and R. E. Marrs, Phys. Rev. Lett. **67**, 2272 (1991).

[19] E. Träbert, P. Beiersdorfer, G. V. Brown, *et al.*, Phys. Rev. A **73**, 022508 (2006).

[20] T. Walker, University of Wisconsin (private communication).

[21] R. E. Marrs, P. Beiersdorfer, and D. H. Schneider, Physics Today **47**, issue 10, 27 (1994).

[22] P. Beiersdorfer, G. V. Brown, R. Goddard, and B. J. Wargelin, Rev. Sci. Instrum. **75**, 3720 (2004).

[23] P. Beiersdorfer, G. V. Brown, L. Hildebrandt, K. L. Wong, and R. Ali, Rev. Sci. Instrum. **72**,

TABLE I: Lifetime of the $3d^9 4s\ ^3D_3$ level in Xe^{26+}, without and with hyperfine interaction (HFS).

Level	Calculated	Measured
	^{132}Xe ($I = 0$, without HFS)	
	16.0 ms [a]	
	18.6 ms [b]	
	14.0 ms [c]	
	15.12 ms [d]	15.06±0.24 ms *
	^{129}Xe ($I = 1/2$, with HFS)	
F=7/2	15.12 ms [d]	15.1±0.5 ms *
F=5/2	2.84 ms [d]	2.7±0.1 ms *

a [30]

b Relativistic many-body perturbation theory [26, 27]

c GRASP code [19, 28, 29]

d GRASPVU code [17], but disregarding interference

* This work

508 (2001).

[24] P. Beiersdorfer, L. Schweikhard, J. Crespo López-Urrutia, and K. Widmann, Rev. Sci. Instrum. **67**, 3818 (1996).

[25] J. R. Crespo López-Urrutia, P. Beiersdorfer, D. W. Savin, and K. Widmann, Phys. Rev. A **58**, 238 (1998).

[26] U. I. Safronova, A. S. Safronova, S. M. Hamasha, and P. Beiersdorfer, At. Data Nucl. Data Tables **92**, 47 (2006).

[27] U. I. Safronova (private communication)

[28] I. P. Grant, in Methods in Computational Chemistry, Vol. 2 (ed. S. Wilson), Plenum Press, New York, 1988, p. 1

[29] F. Parpia, C. Froese Fischer, and I. P. Grant, Comp. Phys. Comm. **94**, 249 (1996).

[30] J. H. Scofield (private communication)

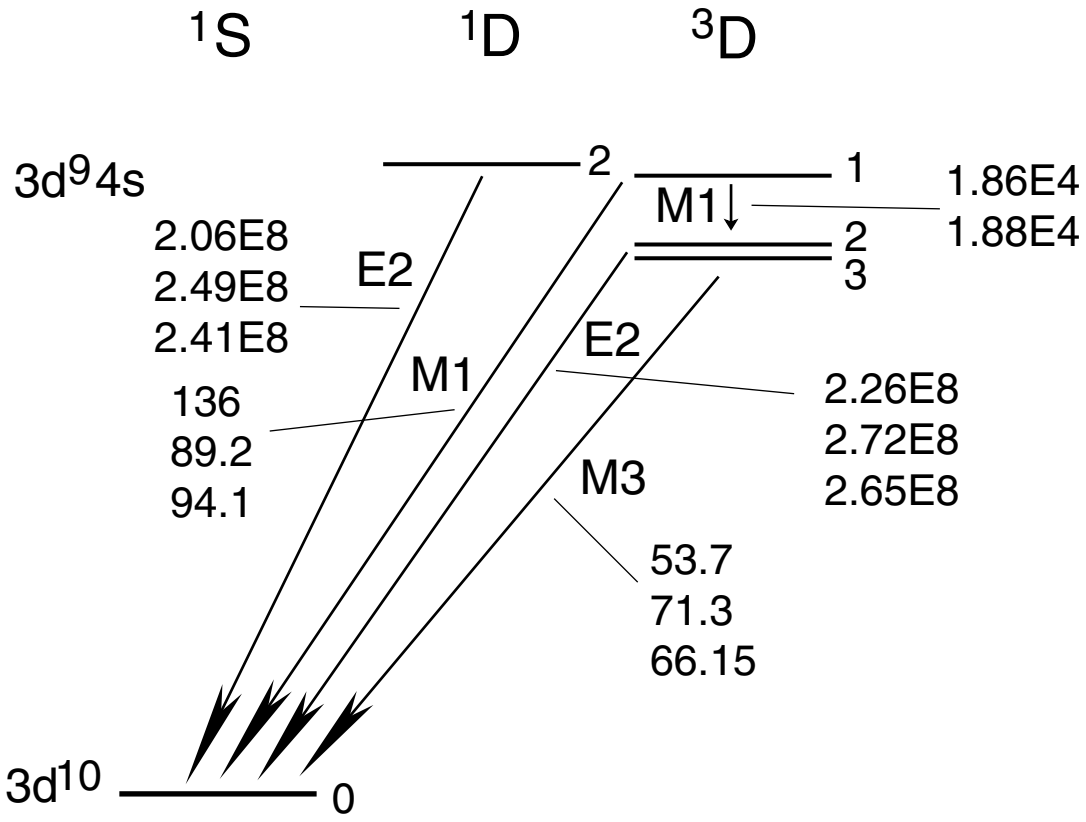

FIG. 1: Lowest levels and principal decays in Ni-like ions. The J values of the levels are noted at the right end of the level bars. The numbers associated with the principal decays are the calculated transition probabilities A_{ki} in the format aEb meaning a $\cdot 10^b\,s^{-1}$. The first of each entry is from calculations by Safronova *et al.* [26, 27], the second from our own GRASP code calulations (see [19]), the third from the GRASPVU code calculations by Yao *et al* [17], where available. All of these numbers are for an isotope without hyperfine structure.

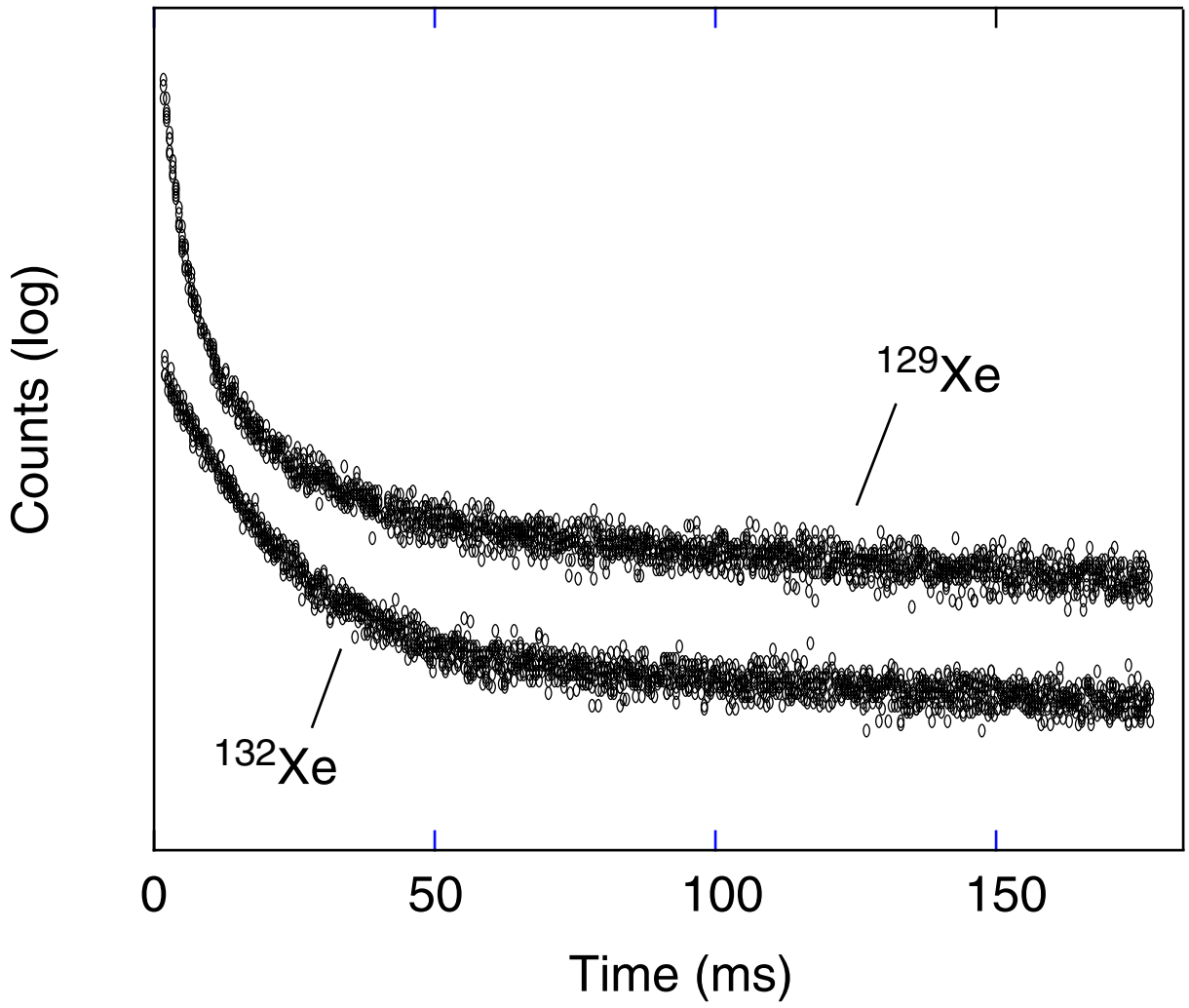

FIG. 2: Decay curves obtained with a proportional counter detector, when injecting isotopically pure ^{129}Xe (upper data set) or ^{132}Xe (lower data set) into SuperEBIT. Both data sets have initial amplitudes of more than 1000 counts; they have been vertically displaced to show their shapes without overlap. Evidently the decay curve of an isotope without hyperfine structure is less complex, consisting of a single-exponential radiative decay and a tail that represents electron capture by charge exchange with neutral residual gas. The curve for ^{129}Xe has two major radiative decay components; isotopes of larger nuclear spin are expected to show even more components.