Laser Spectroscopy Measurements of $^{72-96}\text{Kr}$ Spins, Moments and Charge Radii

M. Keim, E. Arnold[†], W. Borchers[‡], U. Georg, A. Klein, R. Neugart
Institut für Physik, Universität Mainz, D-55099 Mainz, Germany

L. Vermeeren

Instituut voor Kern- en Stralingsfysika, K.U. Leuven,
B-3001 Leuven, Belgium

R. E. Silverans

Laboratorium voor Vaste-Stoffysika en Magnetisme, K.U. Leuven,
B-3001 Leuven, Belgium

P. Lievens[¶] and the ISOLDE Collaboration

PPE Division, CERN, CH-1211 Geneva 23, Switzerland

Abstract

The spins, moments and radii of krypton isotopes have been investigated by collinear fast-beam laser spectroscopy in combination with ultra-sensitive collisional ionization detection. The sequence of isotopes under study ranges from the neutron deficient $N = Z = 36$ isotope ^{72}Kr to the neutron rich ^{96}Kr ($N = 60$). The mean square charge radii in the neighbourhood of the $N = 50$ neutron shell closure exhibit a pronounced shell effect which has recently been explained in the framework of relativistic mean field theory. The results for the neutron deficient nuclei are related to the shape coexistence of strongly prolate and near-spherical states which is known from nuclear spectroscopy. Here, an inversion of the odd-even staggering is observed below the neutron number $N = 45$. The neutron rich transitional nuclei are influenced by the $N = 56$ subshell closure. In contrast to the $N = 60$ isotones ^{97}Rb , ^{98}Sr and ^{100}Zr , the new isotope ^{96}Kr is not strongly deformed.

Keywords: NUCLEAR STRUCTURE $^{72,74-96,79m,81m,83m,85m}\text{Kr}$; measured isotope shifts, hfs; deduced hyperfine constants, spins I , magnetic dipole moments μ , electric quadrupole moments Q_s , mean square charge radii $\langle r^2 \rangle$; Collinear fast-beam laser spectroscopy, collisional ionization detection.

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[†] Present address: Heraeus Noblelight GmbH, Hanau

[‡] Present address: Agfa Gevaert AG, Leverkusen

[¶] Present address: Laboratorium voor Vaste-Stoffysika en Magnetisme, K.U. Leuven, B-3001 Leuven, Belgium.

1 Introduction

The elements in the region around the $Z = 40$ shell closure have been the subject of detailed experimental and theoretical investigations [1]. The ground state properties of these nuclei depend very sensitively on small changes in the proton and neutron number. This has been shown by extended nuclear spectroscopy studies of the level schemes and lifetimes and by laser spectroscopy studies of nuclear ground state properties. Two regions of strong ground state deformation with deformation maxima $\beta_2 \approx 0.4$ were established for $Z \approx 38$, $N \approx 38$ and $Z \approx 38$, $N \approx 60$ [2, 3]. The rapid transition from a spherical shape around the $N = 50$ shell closure to such a strong deformation reflects the reinforcement of several shell gaps in the single particle spectrum (see [4, 5] and references therein). Laser spectroscopy experiments on rubidium [6, 7] and strontium [8, 9, 10, 11, 12] yielded the spins, moments and changes in mean square charge radii $\delta\langle r^2 \rangle$ for sequences of isotopes covering the neutron shell closure and both regions of deformation. These results provide a rather coherent description of the changes in nuclear structure as a function of the neutron number. We have carried out similar measurements on the krypton isotopes ranging from the self-mirror nucleus ^{72}Kr to the $N = 60$ isotope ^{96}Kr . Previously, optical spectroscopy data only covered the range of isotopes between $A = 78$ and $A = 90$, where the nuclear shape is essentially spherical [13, 14, 15, 16, 17]. The new results for $^{72,74-96}\text{Kr}$ extend considerably the experimental information about the electromagnetic ground state properties, particularly in the deformation region around $N = 40$ and for the neutron rich isotopes up to $N = 60$.

2 Experiment

The experiment was performed at the ISOLDE 2 on-line isotope separator using the technique of collinear fast-beam laser spectroscopy [18]. The hyperfine structures and isotope shifts of the ground states of $^{72,74-96}\text{Kr}$ and the isomers $^{79m,81m,83m,85m}\text{Kr}$ were measured in the transition $5s[3/2]_2 \rightarrow 5p[3/2]_2$ of the atomic spectrum of krypton at 760 nm. In addition to the standard fluorescence detection, the much more sensitive collisional ionization scheme [19, 20] was used for the isotopes ($^{72,74,75}\text{Kr}$ and $^{93-96}\text{Kr}$) produced with low yields between 10^7 and 10^3 atoms per second. Two reaction mechanisms were employed for the production of the krypton isotopes at the 600 MeV proton beam of the CERN Synchro-Cyclotron. The neutron deficient isotopes were produced by spallation in a niobium powder target and the neutron rich ones by fission of uranium in a UC_2 target. A cooled transfer line connects the target to a plasma ion source from which the ions are extracted and electrostatically accelerated to 60 keV. This cold transmission line suppresses strongly the contamination of the noble gas beams by less volatile isobars, which is essential for the application of the sensitive collisional ionization detection. The experimental setup is shown schematically in Figure 1. The mass-separated ion beam is superimposed collinearly on a cw laser beam and neutralized by charge exchange in a caesium vapour cell containing about $2 \cdot 10^{14}$ atoms per cm^2 . With an ionization potential of 3.9 eV for caesium, the metastable $5s[3/2]_2$ level in atomic krypton at a binding energy of 4.1 eV is populated quasi-resonantly in the charge transfer reaction. The atoms in this state are excited by the laser light to the $5p[3/2]_2$ state which decays either back to $5s[3/2]_2$ or via $5p[3/2]_1$ to the $4p^6\ ^1S_0$ ground state. In the standard optical detection

scheme this excitation is detected by counting the fluorescence photons which are focussed on the entrance surface of a light guide and transmitted to a photomultiplier [18]. The much more sensitive collisional ionization detection [19, 20, 21] takes advantage of the state dependence of stripping cross-sections in an atomic collision. For the loosely bound high-lying metastable state this cross-section is considerably larger than for the ground state. At resonance the metastable $5s[3/2]_2$ state is depopulated by optical pumping to the ground state. Downstream of the optical interaction region the beam of krypton atoms is transmitted through a gas target at low pressure, where the atoms are partly ionized in collisions with molecules. The gas enters the target chamber through a needle valve, and it is differentially pumped by two turbomolecular pumps. The ions created here are electrostatically separated from the remaining neutral atoms and focussed onto the cathode of a secondary electron multiplier. Since the cross-section of the stripping reaction is low for atoms in the ground state, the optical resonance causes a drop in the ion count rate. There are two main reasons for the enormous gain in sensitivity achieved by using this technique instead of the optical detection. (i) The counting efficiency for ions is nearly 100%, while for photons it is below 1% because of the low quantum efficiency of the photomultiplier and the solid angle of the collection optics. (ii) the constant background due to stray laser light is avoided. However, an additional background may be caused by isobars which are as well neutralized in the caesium cell and reionized in the gas target. As mentioned before, isobaric contaminations are largely suppressed by the cold transfer line between the production target and the ion source. Systematic studies of the ionization cross-sections led to the choice of a Cl_2 target with a density of $5 \cdot 10^{14}/\text{cm}^2$. The ionization efficiency for the total beam was 15% and a flop out signal in the ion count rate up to about 30% could be achieved for even-even isotopes which have no hyperfine structure. With an assumed 30% population of the metastable $5s[3/2]_2$ state, this corresponds to an ionization efficiency of 30% for the atoms in the metastable and 10% for those in the ground state. Taking into account the beam line transmission losses (70%) and the neutralization efficiency in the caesium vapour (70%), one ends up at a detection limit of about 100 atoms/s. Beam intensities in excess of this lower limit give a signal-to-noise ratio ≥ 5 within in a measuring time of 30 seconds per channel. Examples of experimental spectra are shown in Figure 2 for the hyperfine structures of ^{95}Kr ($I = 1/2$) and ^{93}Kr ($I = 1/2$) measured by ion counting, and for ^{89}Kr ($I = 3/2$) measured by fluorescence photon counting. In these odd-A isotopes the signal intensity is distributed over up to 13 hyperfine components. The output beam currents of the separator were about $6 \cdot 10^8$ atoms/s for ^{89}Kr (measuring time 2 min), $6 \cdot 10^6$ atoms/s for ^{93}Kr (measuring time 2 min) and $2 \cdot 10^4$ atoms/s for ^{95}Kr (measuring time 1 h). The relatively long measuring time for ^{95}Kr is due to the strong isobaric contamination of the $A = 95$ beam, probably consisting of stable ^{95}Mo from the ion source or of doubly ionized radioactive mercury.

3 Results and evaluation of I , μ_I , Q_s and $\delta\langle r^2 \rangle$

3.1 Hyperfine structure

For the odd-A isotopes, the nuclear spins I and the magnetic dipole and electric quadrupole interaction constants A and B of both states, $5s[3/2]_2$ and $5p[3/2]_2$ are compiled in Tables 1 and 2. The results for ^{81}Kr , ^{83}Kr and ^{85}Kr agree well with those of earlier

experiments [14, 17, 22, 23]. The spins and the A - and B -factors were obtained from the experimental hyperfine structure spectra using a least square fitting procedure. For low-spin nuclei ($I \leq 2$) already the number of hyperfine structure components determines the spin directly. In the high-spin cases ($I = 5/2, 7/2, 9/2$) it is found that the hyperfine structure intervals and the relative amplitudes are consistent with only one of the possible spin assumptions. Values for the magnetic moments are obtained with reference to the NMR measurement performed by Brinkman [24] for the stable isotope ^{83}Kr . The diamagnetic correction is taken from the calculations of Feiock and Johnson [25]. With the value of Faust and Chow Chiu [22] for the A -factor of the $5s[3/2]_2$ state, the magnetic moments of all other krypton isotopes are obtained by the relation

$$\frac{\mu_I^x I^{83}}{\mu_I^{83} I^x} (1 - \Delta^{x,83}) = \frac{A^x}{A^{83}} \quad . \quad (1)$$

The small hyperfine structure anomaly $\Delta^{x,83}$ is mainly caused by the Bohr-Weisskopf effect [26]. It is estimated according to the empirical rule found by Moskowitz and Lombardi [27] for a series of mercury isotopes, taking into account the Z -dependence established by Bohr and Weisskopf [26]. The corrections to the magnetic moments reach from less than $10^{-4}\mu_N$ to $2 \cdot 10^{-3}\mu_N$, depending on the assumptions made for the ls -coupling of the unpaired nucleon. Since this effect cannot be calculated reliably, the estimated values have been added to the statistical error caused by the A -factor measurement and quoted for the magnetic moments of Table 1. The spectroscopic quadrupole moments Q_s are calculated from the B -factors of the $5s[3/2]_2$ level. The electric field gradient is derived from the magnetic hyperfine structure constant $a_{p3/2} = -204.1(4)$ MHz of the $p_{3/2}$ hole according to the Breit-Wills analysis for the $4p^5 5s$ configuration by Trickl et al. [28]. As relativistic corrections we use those calculated by Schwartz [29]. Since no value for the Sternheimer shielding factor has been published, we follow the analysis of the corresponding states in xenon [20] where a small contribution was estimated and the correction was neglected. This yields the calibration factor $B(5s[3/2]_2)/Q_s = -1775$ MHz/barn. The deviation of our value for the quadrupole moment of ^{83}Kr from those of Kuiper [23] and Faust and Chow Chiu [22] is due to the more complete Breit-Wills analysis of the A -factors [28] used to calibrate the electric hyperfine field gradient. An alternative calibration deduces the field gradient fine structure splitting of the $4p^5 5s$ configuration. It results in a 10% larger quadrupole moment. Our preference for the calibration using the Breit-Wills analysis of the A -factors is based on the situation in xenon [20], where these quadrupole moments are confirmed by the result of a muonic hyperfine structure measurement. The quadrupole moments of Table 2 are given with a conventionally estimated error of 10% covering the uncertainties of the calibration as well as the expected small Sternheimer shielding correction [30]. Ratios of quadrupole moments correspond to the ratios of the B -factors and have much smaller (only statistical) errors. In Tables 1 and 2, the nuclear moments (including errors) quoted from earlier experiments on ^{81}Kr , ^{83}Kr and ^{85}Kr are taken from the original publications. Discrepancies to our results are exclusively due to the evaluation procedures, whereas the A - and B -factors are all in very good agreement.

3.2 Isotope shift

Isotope shifts in the transition $5s[3/2]_2 \rightarrow 5p[3/2]_2$ were measured for the even-A and the odd-A isotopes. They are listed in Table 3, where the errors in parentheses are purely statistical, the ones in square brackets are the linear sum of these and the systematic errors which arise mainly from the determination of the Doppler shifts from the acceleration voltages and ion source potentials¹. The isotope shifts can be decomposed into the field shift $\delta\nu_f^{A,A'}$ and two terms accounting for the finite mass of the nucleus, the normal and the specific mass shifts $\delta\nu_n^{A,A'}$ and $\delta\nu_s^{A,A'}$:

$$\begin{aligned}\delta\nu^{A,A'} &= \delta\nu_f^{A,A'} + \delta\nu_n^{A,A'} + \delta\nu_s^{A,A'} \\ &= \delta\nu_f^{A,A'} + \frac{m_{A'} - m_A}{m_{A'} m_A} (M_n + M_s) \quad ,\end{aligned}\quad (2)$$

where the usual convention for the sign is $\delta\nu^{A,A'} = \nu^{A'} - \nu^A$. The normal mass shift constant is given by $M_n = \nu m_e$ with the electron mass m_e . The specific mass shift constant M_s takes into account the correlation of the electron motion. The field shift is related to the change in electron density at the site of the nucleus occurring in the optical transition, $\Delta|\Psi(0)|^2$, and a nuclear parameter $\lambda^{A,A'}$ which can be expanded in a series of radial moments $\delta\langle r^{2k} \rangle^{A,A'}$,

$$\delta\nu_f^{A,A'} = E f(Z) \lambda^{A,A'} = \pi a_0^3 \frac{\Delta|\Psi(0)|^2}{Z} f(Z) \sum_k \frac{c_k}{c_1} \delta\langle r^{2k} \rangle^{A,A'} \quad . \quad (3)$$

The function $f(Z) = C_{unif}/\lambda_{unif}$ is sensitive to the course of the Dirac wave function at the extended nucleus. It is derived from the parameter $C_{unif} = \delta\nu_{unif}/E$ calculated by Blundell et al. [31] for a uniformly charged sphere of radius $R = R_0 A^{1/3}$ ($R_0 = 1.2$ fm). In the medium mass region the moments higher than $\langle r^2 \rangle$ contribute only about 2% to λ [10]. They are roughly taken into account by the normalisation of $f(Z)$ to $\delta\langle r^2 \rangle_{unif}$ instead of λ_{unif} , i. e. $f'(Z) = C_{unif}/\delta\langle r^2 \rangle_{unif}$, with $\langle r^2 \rangle_{unif} = 3/5 R_0^2 A^{2/3}$. The electronic normalization factor E is calculated using two different semiempirical approaches based on the magnetic hyperfine structure and the Goudsmit-Fermi-Segrè (GFS) formula [32], respectively. In both cases the change in the electron density is derived from the absolute value of the $5s$ electron density, $\Delta|\Psi(0)|^2 = \beta |\Psi(0)|_{5s}^2$. For the screening factor β we adopt the value $\beta = 1.1$ [15] which is based on the systematics of comparable transitions [33]. In the GFS approach the electronic factor E is given by

$$E = \frac{\beta}{(n - \sigma)^3} \left(1 - \frac{d\sigma}{dn}\right) \quad . \quad (4)$$

The quantum defect $\sigma = 3.175$ is obtained from the term energy, and the differential change $\frac{d\sigma}{dn}$ is derived using the relation [32]

$$\frac{d\sigma}{dn} = \frac{\frac{d\sigma}{dT}}{\frac{d\sigma}{dT} - \frac{n - \sigma}{2T}} \quad (5)$$

¹For differences of the isotope shifts listed in Table 3 it is a good approximation to calculate the errors by taking the quadratic sum of the statistical errors plus the differences of the systematic errors. The same is true for the mean square charge radii $\delta\langle r^2 \rangle$.

and a second order polynomial fit of σ as a function of the term energies T of the $ns[3/2]_2$ levels ($n = 5, \dots, 9$). With $(1 - \frac{d\sigma}{dn}) = 1.123$ this gives $E(GFS) = 0.203$. For the alternative evaluation of E from the magnetic hyperfine structure we use the single-electron interaction constant $a_{5s} = -363.7(1.3)$ MHz calculated by Trickl et al. [28] for the stable isotope ^{83}Kr . This was derived in the Breit-Wills analysis of the experimental A -factors of the $^{1,3}P_1$ and 3P_2 levels. The probability density at the nucleus becomes

$$|\Psi(0)|_{5s}^2 = \frac{3 a_{5s}}{8\pi a_0^3 R_\infty \alpha^2 \frac{m_e}{m_p} g_I F_r (1 - \delta)(1 - \varepsilon)} \quad (6)$$

Adopting $F_r(1 - \delta) = 1.132$ from Blundell and Palmer [34] and neglecting the very small correction ε for the Bohr-Weisskopf effect [26], we find $E(hfs) = 0.179$. The discrepancy of 13% between the results of the two procedures can be attributed to the model dependent uncertainties in the Breit-Wills analysis as well as to simplifications of the GFS formula. For the further calculation of $\delta\langle r^2 \rangle$ we thus use the mean value $E = 0.19(2)$ which yields the total electronic isotope shift constant $F = E f(Z) = -608(61)$ MHz/fm² for the isotope pair $^{84,86}\text{Kr}$. As usual the systematic error of this calibration is assumed to be 10% so that in our case both calculated values of E are compatible with the adopted mean value.

In contrast to the situation in heavy elements, the mass shift contributes considerably to the isotope shift in the medium mass region. Therefore, the evaluation of $\delta\langle r^2 \rangle$ requires a good estimate of the specific mass shift. The specific mass shift constant M_s is determined in a King plot analysis of the optical isotope shift against the muonic field shift data which are available for the six stable krypton isotopes [35]. The muonic values for $\delta\langle r^2 \rangle^{A,A'}$ were derived model dependently from the transition energies assuming a Fermi II distribution with constant diffuseness parameter [35]. A King plot slope fixed to the semi-empirical electronic factor gives the specific mass shift $\delta\nu_s = -0.22(9)\delta\nu_n$. Here, the error of the specific mass shift could be reduced significantly compared to former evaluations [13, 15] using preliminary muonic data. The result is in agreement with the empirical estimate by Heilig and Steudel [36], $\delta\nu_s \approx 0.3(0.9)\delta\nu_n$. The changes in the radii $\delta\langle r^2 \rangle$ obtained by this analysis are listed in Table 3. The errors quoted in parentheses correspond to the statistical errors of the isotope shifts. Systematic errors have been added linearly: The errors in square brackets include the voltage calibration errors given already for the isotope shifts, and those in braces contain the additional errors arising from the uncertainties in the electronic factor and specific mass shift. Figure 3 shows the $\delta\langle r^2 \rangle$ values together with the latter error intervals. It is obvious from this figure that all calibration errors only affect the absolute values of $\delta\langle r^2 \rangle$, but not the structure of the curve (see also Footnote ¹). Included in Table 3 are the results by Schuessler et al. [15] and by Cannon [17] for the stable and three radioactive isotopes. The values of Gerhardt et al. [13] for $\delta\langle r^2 \rangle$ were evaluated from the isotope shifts in a different electronic transition. Discrepancies are mainly due to the different specific mass shift estimates, while the isotope shifts, except for ^{90}Kr , are in good agreement. As systematic errors are not considered in the publication by Schuessler et al. [15], it is difficult to judge this discrepancy in the results for ^{90}Kr .

4 Discussion

4.1 Mean square charge radii

The variation of the radii along the chains of isotopes in the neighbouring elements krypton and strontium [10] is shown in Figure 4 with respect to the neutron magic nuclei ^{86}Kr and ^{88}Sr . For both of these curves, the evolution of $\langle r^2 \rangle$ is quite similar, except at the neutron number $N = 60$. The non-appearance of a sharp onset of strong deformation at $N = 60$ is the most remarkable deviation of krypton from the behaviour of the neighbouring chains of isotopes. An abrupt change in nuclear shape between $N = 59$ and 60 has been observed for all other elements in the $Z \approx 40$ region and is qualitatively reproduced by shell correction calculations [3, 4, 37]. It is most pronounced for rubidium, strontium and zirconium and diminishes gradually with increasing proton number. This effect is attributed to the interaction of the prolate neutron shell gap at $N = 60$ and the $1g_{9/2}$ shell occupied by the protons [5]. In a prolate deformed nuclear potential the downsloping low- Ω orbitals of the $g_{9/2}$ shell become lower in energy than the high- Ω orbitals of the pf shell. A local minimum in the potential energy surface at $\beta_2 \simeq 0.4$ develops for $N > 58$, when the protons occupy the lowered Nilsson levels of the $g_{9/2}$ configuration. This effect is reinforced by the interaction with the neutrons occupying the spin-orbit partner $g_{7/2}$ and the intruder state from the $h_{11/2}$ shell [38]. For $N = 60$, with a sufficient number of valence neutrons available for the proton-neutron interaction, this deformed state becomes ground state. The non-appearance of strong deformation in ^{96}Kr can thus be explained by the lack of protons moving to the downsloping $g_{9/2}$ orbitals and stabilizing the deformation. A further quantitative discussion of the radii will be based on the predictions of the finite range droplet model [37] including quadrupole deformation. In Figures 3 and 4, these are represented by nearly straight isodeformation lines for deformation parameters β_2 between 0 and 0.4. The droplet model mean square charge radius of the nucleus is expressed by an effective sharp radius R_Z of the proton distribution and an expansion of the nuclear shape in terms of Legendre polynomials (see eq. (10)). For even-even nuclei the quadrupole deformation reflected in mean square charge radii may be derived from the reduced transition strength $B(E2)$ between the 0^+ ground state and the first excited 2^+ state. The $B(E2)$ value is related to the intrinsic quadrupole moment Q_0 by

$$Q_0 = \sqrt{\frac{16\pi}{5} \frac{B(E2)}{e^2}} \quad . \quad (7)$$

In the spirit of Kumar [39] the intrinsic quantity Q_0 defined by equation (7) represents a root mean square value containing nonzero contributions from quadrupole vibrations also for nuclei that are not permanently deformed. In the droplet model [40] the dependence of Q_0 on the quadrupole deformation parameter α_2 is given by

$$Q_0 = \frac{6}{5}ZR_Z^2(\alpha_2 + \frac{4}{7}\alpha_2^2 - \frac{1}{7}\alpha_2^3 - \frac{94}{231}\alpha_2^4) + \frac{48}{175}C'ZR_Z^2(\alpha_2 + \frac{6}{7}\alpha_2^2 - \frac{4}{5}\alpha_2^3 - \frac{1984}{1155}\alpha_2^4) \quad , \quad (8)$$

with C' the Coulomb redistribution correction. The parameter α_2 is related to the more common β_2 by $\alpha_2 = \sqrt{5/(4\pi)}\beta_2$. Values for $B(E2)$ based on lifetime measurements have been compiled by Raman et al. [41]. In krypton these data are available only for the even isotopes $^{74-86}\text{Kr}$. A rough estimate of the remaining $B(E2)$ values can be obtained from the energies of the first 2^+ level using the empirical relation given by Grodzins [42],

$$B(E2) = (12 \pm 4) \frac{Z^2}{A} \frac{1}{E_{2^+}} \quad (keV \ e^2 \ barn^2) \quad . \quad (9)$$

We follow this estimate for the isotopes $^{72,88,90,92}\text{Kr}$, and for ^{94}Kr where the 2^+ energy has been reported recently [43]. Altogether, the deformation from $B(E2)$ values and the droplet model description of the radii,

$$\langle r^2 \rangle = \frac{3}{5}R_Z^2(1 + \alpha_2^2 + \frac{10}{21}\alpha_2^3 - \frac{27}{35}\alpha_2^4) + \frac{12}{175}C'R_Z^2(1 + \frac{14}{5}\alpha_2^2 + \frac{28}{15}\alpha_2^3 - \frac{29}{5}\alpha_2^4) + 3b^2 \quad (10)$$

result in a prediction of the $\delta\langle r^2 \rangle$ curve which is also shown in Figure 3. The diffuseness parameter b is assumed to be constant. For the position of the reference point with respect to the isodeformation lines we have adopted $\beta_2 = 0.145$ (accounting for zero-point motion) from the measured $B(E2)$ value [41] of ^{86}Kr . On the neutron rich side $\langle r^2 \rangle$ increases only slightly more than predicted for constant deformation by the droplet model. Thus the radii indicate only little deviation from a spherical nuclear shape. The droplet model radii obtained with deformations of $\beta_2 \approx 0.2$ from the energies of the 2^+ levels (compared to $\beta_2 \approx 0.15$ for ^{86}Kr at $N = 50$) predict this trend qualitatively. It is explained by the stabilizing influence of the spherical shell gap in the single particle spectrum at $N = 56$ [4] which is in competition with several shell gaps at large prolate and oblate deformation ($Z = 34, 36, 38$, $N = 58$). On the other hand, lifetime measurements for the first 2^+ levels in the equivalent strontium isotopes up to $N = 56$ give exceptionally low $B(E2)$ values [44] of about half the predictions of the Grodzins rule. Presumably the situation in krypton is similar, which means that the increase of the radii contains additional contributions. Mach et al. [44] suggested to ascribe these to octupole vibrations. However, deviations of experimental radii from droplet model predictions in the neighbourhood of shell closures are a generally observed phenomenon. Their origin is probably more complex, related to the fact that the droplet model approach (including only one shape parameter) is not a good approximation for near-spherical nuclei. The same arguments should apply to the isotopes just below the $N = 50$ shell closure. Here, the radii increase steadily from the spherically shaped ^{86}Kr through the transitional region to the strongly deformed nuclei around $N = 40$. The very sharp kink at $N = 50$ and the increase of the radii with decreasing neutron number for $N < 50$ is a general feature also observed for the isotones in rubidium [6, 7] and strontium [10, 45].

Although the droplet model calculations in this region are based on directly measured $B(E2)$ values, they fail to reproduce the described trends except for the strongly deformed isotopes around $N = 40$. The discrepancy reflects the fact that a shallow minimum of the $B(E2)$ values cannot explain a distinct slope change of the $\delta\langle r^2 \rangle$ curve. Independent of experimental deformation values, Myers and Rozmej [46] investigated the influence of collective zero-point quadrupole motion on the radii and ended up with similar conclusions. The problem also persists in recent calculations of deformations and radii using the shell correction method [47]. Another suggestion raised in connection with earlier measurements of krypton [13] and strontium [45] radii below $N = 50$ emphasizes the effect of the skin thickness. Changing the diffuseness parameter of the droplet model by only a few percent would be sufficient to reproduce the experimental $\delta\langle r^2 \rangle$ [45]. Information about the skin thickness is obtained from elastic electron scattering experiments [48]. The analysis of the data is usually based on a spherical charge distribution so that the effects related to quadrupole deformation are contained in the variation of the surface thickness. For molybdenum and zirconium which are located in the region of the $N = 50$ shell closure, a separation of both effects has been performed by taking into account the β_2 values derived from $B(E2)$ values [45]. This leads to a remaining change of the thickness parameter of the order of magnitude required to explain the changes in the charge radii. Although the importance of the skin thickness aspect is supported by a macroscopic picture obtained from Hartree-Fock plus BCS calculations on strontium [10], the principal difficulty of a geometrical model remains in the separation of surface diffuseness and the possible modes of (dynamic) deformation. After many vain attempts to describe the shell effects in mean square charge radii by self-consistent Hartree-Fock calculations [2, 10], there has recently been a surprising breakthrough owing to the application of relativistic mean field (RMF) theory. First calculations of this type were presented by Sharma et al. [49] for the example of lead. It turned out that the spin-orbit interaction which is added phenomenologically in non-relativistic calculations plays an important role for changes in the spatial arrangement of nuclear orbitals. The shell effect, i.e. the kink in the development of $\delta\langle r^2 \rangle$ at magic neutron numbers, naturally emerges from these calculations. Recently, this approach has been employed to investigate the radii and deformations of Kr, Sr and Zr nuclei in the $N = 50$ region. The results are presented in an accompanying paper by Lalazissis and Sharma [50], and Figure 3 includes the charge radii calculated for the krypton isotopes. Even though triaxiality is observed in the transitional region (see, e.g., ref. [2]), the trends of the radii are already well described by the RMF theory based on a simple axially symmetric potential. Forcing the nuclei into an axially symmetric shape may be responsible for a few unexpected sign changes of the theoretical deformation values, but these will not show up in the radii which are mainly sensitive to β_2^2 . The isotopes in the region around $N = 40$ are strongly deformed as expected from nuclear spectroscopy [51, 52] and from shell correction calculations [4]. These deformations have been attributed to the influence of the shell gaps in the single particle spectrum at $Z = N = 38, 40$ [4]. Both, the maximum of $\langle r^2 \rangle$ reached for $N = 39$ (^{75}Kr) and the decrease towards lower masses ($^{72,74}\text{Kr}$) is reproduced qualitatively by the droplet model including deformation derived from the $B(E2)$ values. Here the deformation reaches saturation at $\beta_2 \approx 0.4$. For ^{72}Kr , the high 2_1^+ energy would suggest a sudden decrease of the deformation to $\beta_2 = 0.26$ which is not observed in the radii. This inconsistency might be related to the effect of shape coexistence known for the even nuclei in the $N = Z = 38$ region $^{76,74}\text{Kr}$. Here, a strongly prolate 0_1^+ ground state coexists with a low-lying 0_2^+ state of spherical or slightly

oblate shape [51]. The mixing of the bands built on these states increases the energy of the first 2^+ state with respect to the 0^+ ground state, thus yielding too low deformation estimates from the Grodzins rule. Nazarewicz et al. [4] predicted an oblate ($\beta \approx -0.35$) ground state of ^{72}Kr ($N = Z = 36$) coexisting with a strongly deformed prolate band. It is directly seen from Figure 3 that the mean square charge radius confirms the large deformation, but it can not distinguish between prolate or oblate shape. An analysis of the low-lying level structure by Dejbakhsh et al. [53] is at least consistent with the assumption of a reversal of the prolate and oblate 0^+ levels in ^{72}Kr compared to the heavier isotopes. Another remarkable observation in the region of light krypton isotopes is the inversion of the odd-even staggering below the neutron number $N = 45$. This means that in contrast to the usual behaviour the radii of the odd isotopes are larger than the mean values of their even neighbours. The same effect has already been noticed in the Sr isotones [9]. An explanation could be found in the influence of the odd neutron on the soft even-even core, stabilizing a well defined strongly prolate shape of the ground state. A detailed discussion of this subject will be presented in a separate publication [54].

4.2 Spins and moments

The spins and moments of all krypton isotopes investigated are listed in Tables 1 and 2 and shown in Figure 5. Previously the spins had been known – from nuclear spectroscopy or hyperfine structure measurements – for the ground states and isomers in the sequence $77 \leq A \leq 87$. Ambiguous spin assignments were made for ^{75}Kr [55] and ^{89}Kr [56], and no values have been existing in the literature for $^{91,93,95}\text{Kr}$. The spins together with the magnetic moments reveal near spherical shell model states built on the $1g_{9/2}$, $2p_{1/2}$, $2d_{5/2}$ and $3s_{1/2}$ neutron configurations (in some cases coupled to $I = j - 1$) as well as strongly deformed Nilsson states.

The $I = 9/2$ ground states of $^{83,85}\text{Kr}$ can be described by rather pure $g_{9/2}$ configurations. Their magnetic moments are about half the Schmidt value of $-1.91\mu_N$. The absolute values increase towards the $N = 50$ shell closure, because the quenching due to the contribution of core polarization is reduced. The change in the spectroscopic quadrupole moments corresponds to the trend of the single-particle values. The $N = 45$ isotope ^{81}Kr and the isomer ^{79m}Kr have the spin $I = 7/2$. As in ^{83}Sr this corresponds to an anomalously coupled $(g_{9/2}^3)_{7/2}$ configuration [8]. This assumption is supported by the nuclear moments. The g -factors should be nearly independent of the coupling of identical nucleons. This is in accordance with the experimental values for the $I = 7/2$ states, $g_I(^{81}\text{Kr}) = -0.2594(6)$ and $g_I(^{79m}\text{Kr}) = -0.2247(6)$, compared to those of the ordinarily coupled $I = 9/2$ states, $g_I(^{83}\text{Kr}) = -0.2157109(7)$ and $g_I(^{85}\text{Kr}) = -0.2234(4)$. A dressed three quasiparticle calculation [57] also yields $g_I = -0.22$ for the excited $(g_{9/2}^3)_{7/2}$ state in ^{83}Kr , compared to the experimental value of $g_I = -0.268(1)$. The electric quadrupole moment of ^{81}Kr , $Q_s(^{81}\text{Kr}) = 0.64(7) \text{ barn}$, is somewhat larger than the values for ^{83}Kr and ^{85}Kr . This is not unexpected as it is known that the anomalous coupling to the $I = 7/2$ state results in an enhanced collectivity [58]. The spectroscopic quadrupole moments in the transitional region depend sensitively on the coupling between the single-particle and collective motion. Therefore, it is difficult to interpret the relatively small quadrupole moment of $0.40(4) \text{ barn}$ for ^{79m}Kr without detailed nuclear model calculations. Concerning the radii there is at least a pronounced difference between ^{79m}Kr for which the odd-even staggering is normal (i.e. the radius is smaller than the mean of the adjacent

even isotopes) and ^{81}Kr for which the staggering is inverted. In terms of deformation this corresponds to the trends of the quadrupole moments. The group of $I = 1/2$ nuclei consisting of the ground state of ^{79}Kr and the isomers $^{81m,83m,85m}\text{Kr}$ are assigned to the $p_{1/2}$ shell. All these magnetic moments are very close to the single particle value of $\mu_{s.p.} = 0.638\mu_N$, and they increase only slightly while approaching the shell closure. This is a characteristic property of the $p_{1/2}$ states which are nearly unaffected by configuration mixing and mesonic contributions [59]. From a calculation by Nazarewicz et al. [4] the $1/2^-$ ground state of ^{79}Kr is predicted to be dominated by the $[301\ 1/2]$ Nilsson orbital (corresponding to $p_{1/2}$), either prolate with $\beta_2 = 0.25$ or oblate with $\beta_2 = -0.14$. The magnetic moment is not sensitive enough to deformation to decide upon the proper choice between these two possibilities. However, from the radii the assignment of the stronger deformed prolate state should be favoured. Above the $N = 50$ shell closure up to the $N = 56$ subshell closure the nuclei belong clearly to the $d_{5/2}$ configuration. The $I = 5/2$ ground states of ^{87}Kr and ^{91}Kr are assigned to a $d_{5/2}$ single neutron (particle or hole) state. The magnetic moment of ^{87}Kr has been calculated by Nag and Pal [60]. They include core polarization and an exchange magnetic moment correction to the single particle moment. The theoretical result of $\mu_I(^{87}\text{Kr}) = -1.1207\mu_N$ is in fair agreement with the experimental value of $-1.023(2)\mu_N$. For ^{91}Kr , the magnetic moment is $-0.583(2)\mu_N$. Such a decrease in the absolute value with the number of neutrons in the open $d_{5/2}$ shell is partly attributed to configuration mixing with 1^+ states created by excitations from the $d_{5/2}$ to the $d_{3/2}$ shell. The spectroscopic quadrupole moment of the $d_{5/2}$ isotopes changes sign from negative to positive as the $d_{5/2}$ shell is filling up. This effect is reproduced by the single particle formula

$$Q_{s.p.} = \frac{2j-1}{2j+2} \langle r^2 \rangle \frac{2j+1-2n}{2j-1} \quad (11)$$

with the occupation number n . Even quantitatively the ratio $\frac{Q_s(^{87}\text{Kr})}{Q_s(^{91}\text{Kr})} = -1.00(3)$ agrees with the single-particle approach yielding a ratio of -1 . As usual the experimental quadrupole moments are a factor of 2 larger than those predicted by eq. (11). The ground state spin of ^{89}Kr has been determined to be $3/2$, while former experiments gave no firm spin assignment [56]. In contrast to the other $N = 45$ isotones, the $(d_{5/2}^3)_{3/2}$ cluster state becomes lower in energy than the $d_{5/2}$ single particle configuration, but the difference in energy of these two states is only 29 keV [56]. The magnetic moment of the $3/2^+$ ground state is relatively small. The g -factor should be similar to those of the single $d_{5/2}$ neutron states. The experiment yields $g_I(^{89}\text{Kr}) = -0.220(2)\mu_N$ which is about 30% of the single particle value $g_{s.p.} = -0.765\mu_N$ as in ^{91}Kr . The electric quadrupole moment is close to zero, in accordance with the shell-model estimate for the $(d_{5/2}^3)_{3/2}$ configuration [61]. The $N = 57$ and $N = 59$ isotopes, ^{93}Kr and ^{95}Kr , have the spin $I = 1/2$. This is expected for near-spherical ground states, because the macroscopic-microscopic model [3] as well as Hartree-Fock plus BCS calculations [10] yield the $s_{1/2}$ level lowest in energy above the $N = 56$ subshell closure. The assumption of a near-spherical shape is also supported by the slowly increasing collective contribution to the radii. The assignment of an $s_{1/2}$ configuration is not in contradiction to the very small magnetic moments (about 20% of the single particle value of $-1.91\mu_N$) which can be explained by quenching due to configuration mixing. The $g_{9/2}$ and $d_{5/2}$ neutron shells as well as the $p_{3/2}$ proton shell are completely occupied, while the corresponding spin-orbit partners with $j = l - 1/2$ are empty. Thus a large number of particles is available for M1-excitations of the core.

In $^{93,95}\text{Kr}$ the quenching is even stronger than in the Sr isotones [10], which might be caused by the additional degrees of freedom due to the holes in the $f_{5/2}$ proton shell. The very neutron deficient isotopes $^{75,77}\text{Kr}$ turn out to be the only well deformed odd nuclei investigated in this experiment. Their ground states of $I = 5/2$ can be associated with the $[422\ 5/2]$ Nilsson orbital originating from the $g_{9/2}$ shell. According to the calculation of Nazarewicz et al. [4] these states have deformations of $\beta_2 = 0.38$ (^{75}Kr) and $\beta_2 = 0.34$ (^{77}Kr). For ^{77}Kr the $3/2^-$ state corresponding to $[301\ 3/2]$ from the $f_{5/2}$ configuration (observed at 65 keV) is expected to become the ground state. Assuming strong coupling the intrinsic quadrupole moments $Q_0 = 3.1(4)$ barn (corresponding to $\beta_2 = 0.43(5)$) for ^{75}Kr and $Q_0 = 2.6(3)$ barn (corresponding to $\beta_2 = 0.37(4)$) for ^{77}Kr are derived from Q_s . This indicates a larger deformation for ^{75}Kr , which is also reflected in the evolution of the rms radii. The large inverted odd-even staggering of this nucleus indicates a deformation-stabilizing effect of the unpaired neutron that has been discussed in connection with nuclear spectroscopy work (see references in [55]). The quadrupole moment of ^{77}Sr [12] is about 30 % larger than for the isotone ^{75}Kr . This indicates a deformation difference which is also suggested by the results of theoretical calculations [4], though to a much smaller extent. A similar difference for $N=41$ between ^{79}Sr and ^{77}Kr seems to be responsible for the $[422\ 5/2]$ ground state in ^{77}Kr instead of the theoretically expected $[301\ 3/2]$ (ref. [4]) that is found experimentally in ^{79}Sr . On the other hand, the comparison of spectroscopic quadrupole moments of different elements is subject to the independent calibration errors assumed to be nearly 10 % each. The magnetic moments of both these krypton isotopes are almost identical and about 60 % larger than for the $N = 39$ isotone ^{77}Sr . Particle-rotor calculations performed for ^{77}Sr [12] reproduce the krypton moments much better than the strontium one. This is probably fortuitous, because a small magnetic moment, in which the single-particle and collective contributions nearly cancel out, is very sensitive to small admixtures to the $[422\ 5/2]$ main Nilsson wave function.

5 Conclusion

The hyperfine structure and isotope shift measurements on 24 krypton isotopes in the ground state and 4 isomers extend far beyond the range of previous investigations covering only the stable and a few radioactive isotopes close to stability. The access to isotopes far from stability has become possible due to the very sensitive spectroscopic technique that combines the advantages of collinear laser fast-beam spectroscopy with the efficient detection of ions [19]. The results comprise directly measured spins, magnetic dipole and electric quadrupole moments and changes in the mean square charge radii for nearly all known isotopes. Furthermore, ^{96}Kr with the neutron number $N = 60$ has been observed for the first time. Together with the earlier laser spectroscopy work on strontium ($^{77-100}\text{Sr}$) and rubidium ($^{76-98}\text{Rb}$) the experiment provides a large basis of nuclear ground state data covering the regions of particular interest around $Z \simeq N \simeq 38$ and $N \simeq 60$. The spins and moments of the odd isotopes $^{79-95}\text{Kr}$ reveal near spherical shell model states built on the $2p_{1/2}$, $1g_{9/2}$, $2d_{5/2}$ and $3s_{1/2}$ configurations. The well deformed ground states of ^{75}Kr and ^{77}Kr are assigned to a rather pure $[422\ 5/2]$ Nilsson configuration with prolate deformation of about $\beta_2 \simeq 0.35$. The deformed shape is stabilized by the unpaired neutron, which is reflected in slightly larger radii of the odd isotopes compared to their even neighbours [54]. For the self-mirror nucleus ^{72}Kr , the prediction of an oblate ground state can be neither

confirmed nor disproved. In any case, a possible oblate deformation would be as large as the the obviously prolate one of the neighbouring isotopes. Apart from these details, the main features observed in the radii include the pronounced slope change at $N = 50$ and the non-appearance of a strongly deformed ground state at $N = 60$. An abrupt change of shape at $N = 60$ occurs for all other elements around $Z = 40$ and should be expected from theoretical calculations [2, 3] also for krypton. The absence of this effect is attributed to a reduced proton-neutron interaction for $Z = 36$ in comparison to the isotones with $Z \geq 38$. Commonly, the radii along a sequence of isotopes are (fairly well) described by the droplet model including quadrupole deformation from empirical data. While this approach is quite satisfactory for strongly deformed nuclei in comparison with the spherical ones of closed neutron shells, it fails to reproduce the neighbourhood of the shell closures, where (dynamic) deformation represented by the $B(E2; 2^+ \rightarrow 0^+)$ values passes through a shallow minimum, whereas the kink in the radii as a function of the neutron number would require a pronounced V-shaped behaviour. In other mass regions it has been proposed to explain this so-called shell effect by (i) the combined effect of many multipole orders in the zero-point motion of the nuclear charge distribution [62] or (ii) the admixture of the giant quadrupole resonance into the ground state [63]. Also the shell effect in the radii. Recently, it has been shown by Sharma et al. [49] that a relativistic mean field approach properly describes the development of the radii of Pb isotopes across the $N = 126$ shell. Such calculations were also performed for krypton and strontium [50], reproducing the main features of the radii around $N = 50$ which are the subject of our present discussion. An obvious difficulty, however, persists in the unclear relationship between the different aspects of the microscopic and collective descriptions.

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FIGURE 1: Schematic view of the experimental setup. FIGURE 2: Experimental spectra

of the hyperfine structures of ^{95}Kr and ^{93}Kr measured by ion counting and of ^{89}Kr measured by conventional optical detection. The frequency resolution is 7.7 MHz/channel for ^{93}Kr and ^{95}Kr , and 3.7 MHz/channel for ^{89}Kr . FIGURE 3: Differences of mean charge

radii for the krypton isotopes relative to ^{86}Kr . The uncertainty due to the electronic factor E and the specific mass shift constant M_s is indicated by the two enveloping lines. Isodeformation lines and predictions from experimental β_2 values are taken from the finite range droplet model. The large error bars for $^{72,88,90,92,94}\text{Kr}$ are due to the rough estimate of the quadrupole deformation from the energy of the first 2^+ state. Open squares represent the results of RMF calculations [50]. FIGURE 4: Radii of the krypton isotopes in

comparison with those of strontium. The strontium values are taken from [10, 11, 12]. FIGURE 5: Magnetic dipole moments and electric quadrupole moments of the krypton

isotopes in comparison with those of the isotones in strontium. The symbols for moments belonging to the same nuclear state are connected by lines.

Table 1: A -factors and magnetic dipole moments of the odd- A krypton isotopes. The A -factors are given in MHz. The magnetic moments are obtained with reference to the value for ^{83}Kr given by Brinkman [24] including the diamagnetic correction from ref. [25]. The errors of the magnetic moments consist of the statistical errors given for the A -factors plus the estimated uncertainty accounting for hyperfine structure anomalies (see text). Literature values of the hyperfine structure constants are given for comparison.

A	I	$A(5s[3/2]_2)$	$A(5p[3/2]_2)$	$\mu_I[\mu_N]$	ref.
75	5/2	-240.2(1.2)	-106.8(1.2)	-0.531(4)	
77	5/2	-263.6(0.9)	-117.5(0.9)	-0.583(3)	
79	1/2	1212.2(1.1)	539.9(1.1)	0.536(2)	
79m	7/2	-254.1(0.4)	-113.1(0.4)	-0.786(2)	
81	7/2	-293.4(0.5) -294.02(27)	-130.5(0.4) —	-0.908(2) -0.909(4)	[17]
81m	1/2	1324.7(2.4)	589.1(2.4)	0.586(2)	
83	9/2	-243.8(0.3) -243.970(4) -243.9693(2) -243.87(5)	-108.3(0.3) — — -108.49(12)	-0.970699(3) — — —	[23] [22] [17]
83m	1/2	1335.8(2.8)	592.9(1.5)	0.591(2)	
85	9/2	-252.7(0.3) -252.81(8) -253.5(4)	-112.5(0.4) -112.46(14) —	-1.005(2) -1.0055(4) -1.000(2)	[17] [14]
85m	1/2	1432.2(1.6)	637.3(1.6)	0.633(2)	
87	5/2	-462.8(0.4)	-205.9(0.4)	-1.023(2)	
89	3/2	-248.4(0.4)	-110.7(0.4)	-0.330(3)	
91	5/2	-263.7(0.5)	-117.6(0.5)	-0.583(2)	
93	1/2	-934.1(1.0)	-415.9(1.0)	-0.413(2)	
95	1/2	-927.4(2.1)	-412.5(1.7)	-0.410(3)	

Table 2: B -factors and electric quadrupole moments of the odd-A krypton isotopes with $I > 1/2$. The B -factors are given in MHz. The additional error of 10% applied to the quadrupole moments accounts for the uncertainties in the evaluation of the electric field gradient and should include the missing Sternheimer shielding correction.

A	I	$B(5s[3/2]_2)$	$B(5p[3/2]_2)$	Q_s [barn]	ref.
75	5/2	-1995(10)	-384(10)	1.12(12)	
77	5/2	-1663(8)	-321(7)	0.94(10)	
79m	7/2	-710(7)	-141(7)	0.40(4)	
81	7/2	-1128(4) -1117.2(48)	-216(5)	0.64(7) 0.629(13)	[17]
83	9/2	-454(4) -452(12) -452.170(4) -453.1(7)	-86(5) — — -85.7(16)	0.26(3) 0.251(5) 0.270(13) —	[23] [22] [17]
85	9/2	-774(5) -775.8(12) -775.6(27)	-148(6) -154.0(23) —	0.44(5) 0.433(8) 0.629(13)	[17] [14]
87	5/2	528(3)	101(3)	-0.30(3)	
89	3/2	-291.4(1.2)	-56.6(1.4)	0.16(2)	
91	5/2	-531(4)	-101(4)	0.30(3)	

Table 3: Isotope shifts $\delta\nu^{86,A}$ and differences in the mean square charge radii $\delta\langle r^2\rangle^{86,A}$. Results of Gerhardt et al. [13], Schuessler et al. [15] and Cannon [16, 17] on isotopes close to stability are given for comparison. Errors in parentheses are purely statistical. The errors quoted in square brackets include systematic errors from the voltage calibration of the isotope shift measurement, and those quoted in braces represent total errors of $\delta\langle r^2\rangle$ including the uncertainties of the evaluation from the isotope shifts.

A	$\delta\nu^{86,A}[\text{MHz}]$	$\delta\langle r^2\rangle^{86,A}[\text{fm}^2]$	ref.
72	-278(11)[36]	-0.168(18)[58]{165}	
74	-335(3)[24]	0.030(5)[40]{109}	
75	-421(5)[24]	0.221(7)[39]{79}	
76	-352(3)[21]	0.156(4)[34]{75}	
77	-356(3)[19]	0.209(5)[31]{60}	
78	-305(2)[16] -304.3(4.0) -306.8(1.9) —	0.172(3)[26]{52} 0.122(7) — 0.126	[15] [16] [13]
79	-276(3)[15]	0.168(4)[24]{47}	
79m	-250(3)[15]	0.126(4)[24]{49}	
80	-216(4)[14] -221.6(4.0) -221.3(1.7) —	0.114(7)[24]{44} 0.088(7) — 0.087	[15] [16] [13]
81	-181(2)[11]	0.099(4)[18]{34}	
81m	-170(5)[14]	0.080(8)[22]{41}	
82	-139(2)[9] -141.6(4.0) -142.7(1.6) —	0.071(3)[14]{28} 0.053(7) — 0.054	[15] [16] [13]
83	-89.5(1.8)[6.9] -90.2(6.0) -94.1(1.4)	0.031(3)[11]{24} 0.015(10) —	[15] [17]
83m	-93(4)[9]	0.037(6)[14]{26}	
84	-72.4(5)[3.8] -73.3(4.0) -73.4(1.1) —	0.042(1)[6]{12} 0.033(7) — 0.033	[15] [16] [13]
85	-29(3)[5] -33.5(3.3)	0.009(4)[7]{11} —	[17, 16]
85m	-6(3)[5]	-0.028(5)[8]{17}	
86	0	0	
87	-53.3(1.7)[3.4]	0.125(3)[6]{26}	
88	-126(3)[6] -132.5(8.0)	0.282(4)[10]{53} 0.304(13)	[15]
89	-164(3)[8]	0.379(4)[12]{71}	
90	-213(6)[13] -244.0(8.0)	0.495(10)[21]{99} 0.570(13)	[15]
91	-254(4)[12]	0.597(6)[20]{114}	
92	-327(3)[13]	0.751(5)[22]{139}	
93	-344(3)[15]	0.811(4)[23]{151}	
94	-432(3)[17]	0.989(4)[26]{180}	
95	-447(2)[18]	1.045(3)[29]{194}	
96	-533(6)[24]	1.217(10)[40]{229}	