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Hyperfine splitting and isotope shift in the atomic D_2 line of ^{22,23}Na and the quadrupole moment of ^{22}Na

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Abstract. The hyperfine structure of the D_2 optical line in ^{22}Na and ^{23}Na has been investigated using high resolution laser spectroscopy of a well-collimated atomic beam. The hyperfine splitting constants A and B for the excited $3p\ ^2P_{3/2}$ level for both investigated sodium isotopes have been obtained. They are as follows: $A(22) = 7.31(4)$ MHz, $B(22) = 4.71(28)$ MHz, $A(23) = 18.572(24)$ MHz, $B(23) = 2.723(55)$ MHz. With this data, using the high precision MCHF calculations for the electric field gradient at the nucleus, the electric quadrupole moment of ^{22}Na has been deduced: $Q_s(22) = +0.185(11)$ b. The sign of $Q_s(22)$, determined for the first time, indicates a prolate nuclear deformation. A precise value of the isotope shift ^{22,23}Na in the D_2 line has also been obtained.

PACS. 21.10.Ky Electromagnetic moments – 27.30.+t $20 \leq A \leq 38$ – 31.30.Gs Hyperfine interactions and isotope effects

1 Introduction

In earlier experiments with high resolution on-line laser spectroscopy the hyperfine (hf) structure and the isotope shift (IS) of the D_1 line of $^{21-31}$Na relative to ^{23}Na were studied [1,2]. Nuclear properties (spins I, magnetic moments μ_I and changes in the mean square charge radius) were deduced from these experiments. The D_2 line hf structures of $^{21,25-29}$Na were also investigated [3] and the quadrupole coupling constants B of the $3p\ ^2P_{3/2}$ excited state were deduced. Thus, additional information on the nuclear deformation was gained. However, in this systematic study over a long sodium isotope chain there is a lack of information about the electric quadrupole constant B, and respectively, about the nuclear quadrupole moments. Up to now, no data on quadrupole moments of ^{22}Na, ^{24}Na, as well as of neutron-rich ^{30}Na, ^{31}Na are available. This is obviously due to two reasons: a) the hf splitting of the upper $3p\ ^2P_{3/2}$ state of the D_2 optical transition for the above mentioned isotopes is rather small and it needs extremely high resolution of the used apparatus; b) the low production rate of the neutron-rich isotopes allowed to make conclusions only about the IS [3].

Preliminary results on the electric quadrupole moment of ^{22}Na, obtained by off-line laser specroscopy, have been published, to the best of our knowledge, for the first time in our previous work [4]. The present paper is devoted to a more precise study of the hf structure of ^{22}Na at the D_2 line. It has the following purposes:

i) to obtain high accuracy values of magnetic dipole, $A(22)$, and electric quadrupole, $B(22)$, coupling constants of the excited $3p\ ^2P_{3/2}$ state;
ii) to extract a reliable value of the electric quadrupole moment $Q_s(22)$ and thus extend our knowledge on the nuclear deformation in the region of light deformed nuclei.

Some additional results on the D_2 line have been obtained and are reported as well:

iii) new values of the hf splitting constants $A(23)$ and $B(23)$ of the $3p\ ^2P_{3/2}$ excited state with a precision of the same order of magnitude as the most accurate known data of [5];
iv) IS of ^{22}Na relative to ^{23}Na, which may give information about the IS of the $3p\ ^2P_{3/2}$ level relative to the $3p$ $^2P_{1/2}$ level.

2 Experimental details

In the present work, an off-line experimental method has been used, based on the detection of the laser-excited fluorescence in a well-collimated atomic beam. The experimental setup has been described in detail previously [6,7], and partially in [8]. A cw dye laser with a system of active frequency stabilization (Spectra Physics 380D) pumped with an Ar ion laser was operated using dye of R6G. To reduce the Doppler broadening, both the atomic and the laser beams were collimated, and the laser beam crossed the atomic beam perpendicularly. The laser-induced resonance fluorescence was detected with a photomultiplier, operating in a single photon counting mode. The spectra were recorded by a multichannel analyzer synchronized with the laser frequency tuning. A frequency calibration was provided by recording the transmission signal of the laser light trough a temperature stabilized Burleigh CFT-500 confocal etalon with a free spectral range of 150 MHz. The data acquisition system as well as all parameters of the experiment (laser frequency scan, frequency stabilization etc.) were computer controlled.

^{22}Na ($T_{1/2}$ = 2.602 years) was produced by the reaction ^{19}F(α, n) in a target of AlF$_3$. The sodium atoms were extracted chemically from the target in the form of NaCl. NaCl water solution containing $2 \cdot 10^{13}$ atoms of ^{22}Na was dried on a tantalum foil. The heating of the sample in a Ta crucible to a temperature of about 600 - 700°C resulted in thermal dissotiation of the chloride and provided a stable atomic beam of ^{22}Na for about an hour. Care has been taken to minimize the abundance of ^{23}Na by annealing the tantalum foil and crucible in a vacuum at $T \sim 1800$°C for a several hours. Due to contamination of the target and of the used chemical reagents with the stable sodium isotope, the final ratio of ^{23}Na to ^{22}Na in the sample was about 150:1.

It is well known that in a Doppler limited spectroscopic method the resolution is strongly connected to the efficiency of the system. In order to perform reliable measurements with the low abundant radioactive ^{22}Na great care has been taken to find an acceptable compromise between resolution and sensitivity. Numerous tests of the apparatus have been done with samples containing only the stable ^{23}Na in quantities down to 10^{13} atoms. In this way all the experimental conditions were optimized. In particular, an experimental FWHM of the detected resonance lines of about 20 - 23 MHz was established by reasonably limiting the residual Doppler broadening. It should be noted that all spectra were measured using a series of neutral density filters to attenuate the laser power density to about 100 μW$/sm^2$. This was done to reduce the power broadening which under such condition gives a maximal FWHM of about 12 MHz (see e.g. [9]) and compares the natural linewidth of 9.7 MHz [10].

The large difference in the hf splittings of the lower and upper levels of the investigated D_2 sodium transition results in a typical flag-pattern spectrum: two narrow triplet groups defined by the small hf splitting of the excited $3p$ $^2P_{3/2}$ state are quite spaced because of the large splitting ($\sim$ 1220 MHz) of the $3s$ $^2S_{1/2}$ ground state. In our measurements of the ^{22}Na hf splitting each group was recorded separately but together with the low energy flag-pattern group of the stable sodium isotope to obtain information on the IS. A reference atomic beam of ^{23}Na was used to choose correctly the starting frequency of the laser scan. The laser frequency was tuned over a region of about 800 MHz and one scan took typicaly 200 - 250 s. For high resolution recording a typical frequency interval represented by each channel was 0.8 - 1 MHz. Two samples of ^{22}Na were used, one for each hf splitting group, and for each of them more than 10 spectra were recorded with a sufficiently good signal to noise ratio.

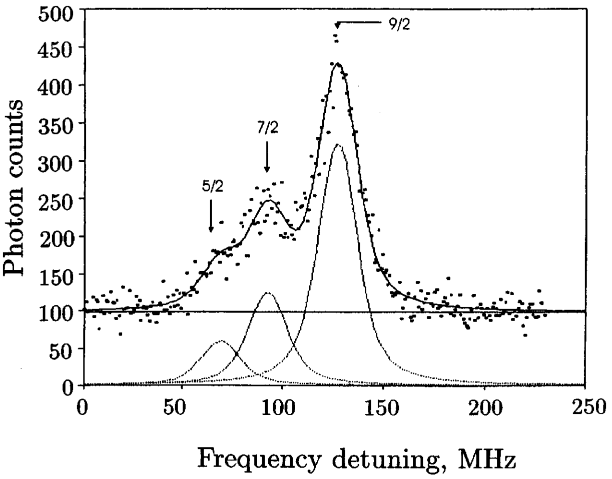

Fig. 1. Example of a typical flag-pattern of D_2 line in the transition $F' = 7/2 \rightarrow F = 9/2, 7/2, 5/2$ for ^{22}Na. F' and F are the total quantum numbers of the lower and upper levels of the transition, respectively. A deconvolution of the observed peaks as well as the overall fitted spectrum are also shown

Spectra of the laser resonance fluorescence from the radioactive ^{22}Na, detected by the photomultiplier in a single scan, are shown in Fig.1 and Fig.2. The peaks in Fig.1 arise from the excitation of the $3s$ $^2S_{1/2}$ $F' = 7/2$ level to the $3p$ $^2P_{3/2}$ $F = 9/2, 7/2, 5/2$ levels. In Fig.2, the peaks result from exciting the $F' = 5/2$ ground level to the $3p$ $^2P_{3/2}$ $F = 7/2, 5/2, 3/2$ levels. The high abundance of the stable sodium isotope in the investigated samples led to a significant enhancement of the background, due to the Lorentzian wings of the spectral lines. Their influence is more pronounced in the case of Fig.2 because of the smaller distance of this peak group to the most intensive ^{23}Na peak.

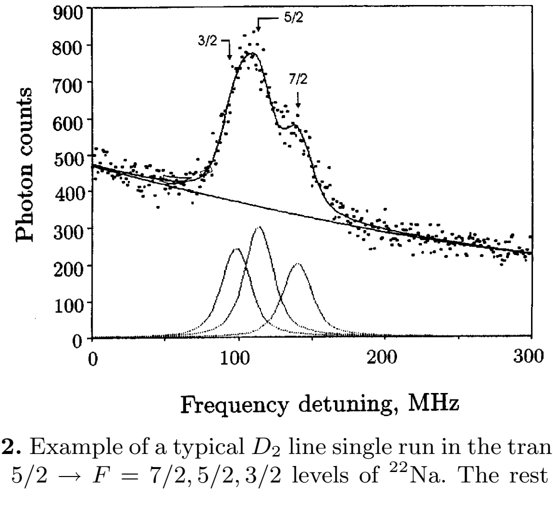

Fig. 2. Example of a typical D_2 line single run in the transition $F' = 5/2 \rightarrow F = 7/2, 5/2, 3/2$ levels of ^{22}Na. The rest as in Fig.1

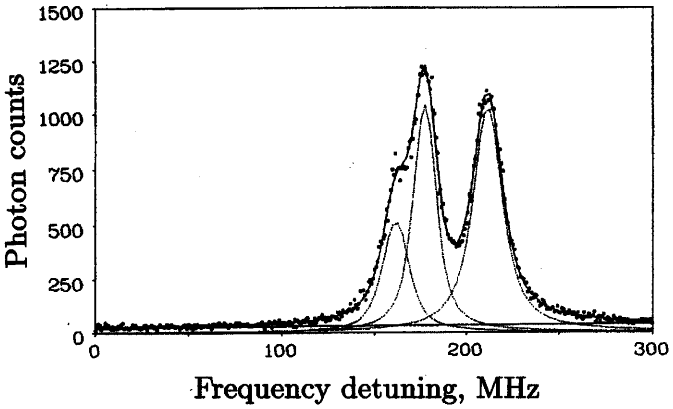

Fig. 3. Example of a deconvolution of the observed resonance peaks in D_2 sodium line for ^{23}Na in the transition $F' = 1 \rightarrow F = 2, 1, 0$. In this case the FWHM of the resonance lines was 18 MHz

The hf splitting of the $3p\ ^2P_{3/2}$ level for ^{23}Na was also measured in the course of our test experiments. This was done with a higher resolution corresponding to FWHM of about 18 MHz. Fig.3 gives an example of the hf splitting pattern arising by the excitation from the $F' = 1$ hf component of the ground level to the $F = 2, 1, 0$ hf components of the $3p\ ^2P_{3/2}$ level. This peak group is about two times narrower, and therefore, worse resolved than the pattern arising from the $F' = 2$ ground level hf component.

3 Data analysis and results

As can be seen from Figs. 1 to 3, the observed transitions were only partially resolved with the present collimation of the atomic beam. For this reason it was necessary to make deconvolution of the unresolved resonance peaks. The centroid of each peak was established by fitting the data with Voigt profiles to properly describe the convolution of the Lorentzian shape of the natural linewidth with the Gaussian distribution caused by the residual Doppler broadening using the well- known program PEAKFIT. The centroid of each peak was measured with respect to the two nearest-neighbour etalon peaks.

The procedure of decomposing the spectra included two fixed parameters: the known number of the hf splitting components and the values of the FWHM (see the previous section) corresponding to different collimations of the atomic beam. They were determined experimentally by the well separated resonance peak ($F' = 2 \rightarrow F = 3$ for ^{23}Na). The two width parameters in the Voigt profile of this peak, corresponding to the observed FWHM, were used in the fit of all other spectra measured with the same resolution. It was established that such conditions led to the best fit of the spectra: with normalized χ^2-value close to 1 and smallest errors for the peak position values. As can be seen from Figs. 1 to 3, also the ratios of the fitted line amplitudes correspond in general to the theoretically expected ones.

The fitting procedure gave statistical errors composed of both the uncertainty in the peak position determination and the uncertainty originating from the frequency calibration. These errors were typically in the range 0.9 - 0.03 MHz. It was found, however, that the spread between different runs was larger than the statistical errors involved. These occasional errors could, e.g., be caused by changes in the evaporation rate, the laser intensity or scan frequency linearity. The error estimates given in this paper correspond to one standard deviation of the spread between different runs. Thus, the errors given include both known and unknown occasional errors as well as statistical errors. The systematic error due to the uncertainty of about 0.01% of the free spectral range of the frequency calibration interferometer are negligible.

The experimental values of the hf splittings for the excited $3p\ ^2P_{3/2}$ state of ^{22}Na and ^{23}Na are presented in Table 1. From these values the magnetic dipole, A, and electric quadrupole, B, constants of the $3p\ ^2P_{3/2}$ state are deduced and given in Table 2. The method used was a fit of the hf splitting intervals to the well known two-parameter first-order hf splitting formula [11]. It can be written in the form

$$\Delta W(F, F-1) = A \cdot F + B \cdot \frac{3F[F^2 + 1/2 - I(I+1) - J(J+1)]}{2I(2I-1)J(2J-1)}, \qquad (1)$$

if applied for the energy intervals $\Delta W(F, F-1)$ between two hf splitting components F and $F-1$ of a given atomic level J. Table 2 summarizes also the results from different source references. In the present work the precision of the experimental A and B values for ^{22}Na has been improved compared to our previous paper [4] due to a) the optimization of the experiment which led to a better signal to noise counting ratio, and b) a more than two times higher number of the recorded spectra for each hf splitting group.

As about ^{23}Na, it should be emphasize that extensive studies of the $3p\ ^2P_{3/2}$ hf splitting have been carried out. They are reviewed and critically discussed in many papers, e.g. [5,12]. Table 2 lists the two results of $A(23)$ and $B(23)$ having the smallest quoted uncertainties found in the literature: they were obtained by quantum-beat spectroscopy [5,13]. The recent data of [9] deduced by laser-induced resonance are also shown, as well as the theoretically predicted $A(23)$ value [14]. References to other measurements can be found in the article by Yei et al. [5]. As can be seen from Table 2 (see also Table II of [5]), our results on A and B for ^{23}Na are in very good agreement with the most precise data of [5] and considerably more accurate than any other previous determination.

Using i) our value for $A(22,^2P_{3/2})$ and $A(23,^2P_{3/2})$ from Table 2; ii) the magnetic dipole coupling constants for the ground $^2S_{1/2}$ state from [15,16], and iii) the gyromagnetic ratios $g_I = \mu_I/I$ of both Na isotopes measured by paramagnetic resonance (see [17] and the references therein) we obtain that the following relations hold

$$A(22,^2P_{3/2})/A(23,^2P_{3/2}) = A(22,^2S_{1/2})/A(23,^2S_{1/2}) = g_I(22)/g_I(23),$$

Table 1. Experimental, ΔW_{exp}, and calculated, ΔW_{cal}, values of the energy separation between the excited state hyperfine levels F and $F-1$ for ^{22}Na and ^{23}Na in D_2 line. Here $\delta = \Delta W_{cal} - \Delta W_{exp}$. All energies are in MHz

^{22}Na				^{23}Na			
F	ΔW_{exp}	ΔW_{cal}	δ	F	ΔW_{exp}	ΔW_{cal}	δ
4.5	36.62(35)	36.44	-0.18	3	58.44(10)	58.439	-0.001
3.5	23.79(19)	23.95	+0.16	2	34.42(7)	34.421	+0.001
2.5	15.07(30)	14.75	-0.32	1	15.86(17)	15.849	-0.011

Table 2. Hf splitting constants A and B for the $3p\ ^2P_{3/2}$ level of ^{22}Na and ^{23}Na. For comparison the values of the hf splitting constants obtained from other authors are presented, too

isotope	A, MHz	B, MHz	Ref.
^{22}Na	7.31(4)	4.71(28)	this work
	7.38(6)	3.76(45)	[4]
^{23}Na	18.572(24)	2.723(55)	this work
	18.534(15)	2.724(30)	[5]
	18.64(6)	2.77(6)	[13]
	18.62(21)	2.11(52)	[9]
	18.53		theory [14]

where the values of the first, second and third ratio are respectively 0.3936(22), 0.393706(11) and 0.3937(7). The agreement between the absolute values is excellent, althought the quoted uncertainties of our data are not the best one could wish. The result agrees with the expectation, that in the mass region of the light nuclei the nuclear hf anomaly is negligibly small [18,19], and thus, it is in a support of the reliability of our hf splitting measurements.

4 Discussion

4.1 Quadrupole moment of ^{22}Na

The ^{22}Na isotope belongs to the region of well-deformed light nuclei. A large quadrupole deformation in this region has been deduced from the large matrix elements of the $E2-$intraband transitions [20]. The value of the hf constant B deduced in the present work offers an alternative and model-independent way to obtain an additional information about the nuclear deformation.

A review of the present status of our knowledge on quadrupole moments Q (see e.g. [17,21]) shows that the many Q moments of radioactive nuclei have been determined by optical methods as these methods are very sensitive and fast. From the B factors of the hf splitting and electric field gradient d^2V/dz^2 at the nucleus the value for Q can be derived:

$$Q = -B/(d^2V/dz^2). \tag{2}$$

Since the calculation of d^2V/dz^2 is difficult, the following assumptions are made:

(i) d^2V/dz^2 is isotope independent and thus, for two isotopes A and A'

$$Q(A)/Q(A') = B(A)/B(A'); \tag{3}$$

(ii) a reference Q moment, Q_{ref}, is known from optical measurements on a stable isotope for an atomic configuration which is best suited for a reliable estimation or an *ab initio* calculation of d^2V/dz^2;
(iii) Q_{ref} can alternatively be taken from muonic measurements of stable isotopes, as the wave function of the single muon, and thus, d^2V/dz^2 is almost undisturbed by other electrons.

A conventional method to obtain d^2V/dz^2 is to calculate $< r^{-3} >$ values from the hyperfine coupling constants with Sternheimer corrections accounting for quadrupole shielding [22]. This is the way in which the spectroscopic quadrupole moments for the sodium isotope chain $^{21,25-29}$Na [3] were deduced. With the Hartree-Slater calculations of [22] for ^{23}Na, providing the best agreement with measured magnetic moments, the relation

$$Q_s = 0.04134 \cdot B(^2P_{3/2}) \tag{4}$$

has been obtained and used to calculate Q_s. Table 3 presents the $Q_s(23)$ thus obtained with different $B(23,^2P_{3/2})$ values. As can be seen, $Q_s(23)$ values are by more than 10% larger than the muonic $Q_s(23) = 0.1006(20)$ b of [23]. This exceeds 6-7 times the quoted errors. These are only the statistical experimental uncertainties of the B factors to which one has to add an uncertainty of about 5% as estimated in [22].

Recently d^2V/dz^2 for ^{23}Na was obtained alternatively from high-quality *ab initio* calculations of atomic (or molecular) system without any correction factors (see [24] and the references therein). With the final value of the electric field gradient at the nucleus -0.1083(9) a.u. [24] one obtains:

$$Q_s = 0.0393(3) \cdot B(^2P_{3/2}) \tag{5}$$

(in (4) and (5) Q_s is in b and $B(^2P_{3/2})$ in MHz). Due to the improved value of $B(23)$ the present $Q_s(23)$, as well as the one from [5] (see Table 3), is about 2% smaller than the value reported in [24]. Nevertheless, a disagreement of about 6% with the muonic Q_s still remains which is larger than the quoted errors. Disagreements between optical and muonic Q moments have been observed also in other nuclides. A detailed discussion of the possible causes can be found, e.g. in [24,27].

Table 3. The quadrupole moments of ^{22}Na compared to values from the literature and of ^{23}Na. Here HS denotes Hartree-Slater; MCHF - multiconfiguration Hartree-Fock; * - that the quoted errors include only the experimental uncertainties of the electric quadrupole hf constant

Method	used	$Q_s(23)$, b	$Q_s(22)$, b	References for the:	
	equation			method	$B-$value
hfs + HS cal.	(4)	0.1145(25)*		[22]	[13]
		0.1126(12)*			[5]
		0.1126(22)*	0.195(12)*		this work
hfs + MCHF cal.	(5)	0.1089(25)		[24]	[13]
		0.1070(15)			[5]
		0.1070(23)	0.185(11)		this work
muonic value		0.1006(20)		[23]	
	(3)		0.174(11)		this work
nuclear theory		0.1024		[25]	
	(3)		0.177(11)*		this work
nuclear theory		0.1034		[26]	
	(3)		0.179(11)*		this work

Table 3 also summarises the spectroscopic quadrupole moments of ^{22}Na extracted in different ways. All the data of $Q_s(22)$ were obtained with the only known value of $B(22,{}^2P_{3/2})$ of the present work. Due to the larger experimental uncertainty of the electric quadrupole constant for ^{22}Na compared to ^{23}Na, different methods result in $Q_s(22)$ values scattered within the limits of the quoted errors. In what follows (5) is accepted, obtained by the most reliable MCHF calculations [24] (with an uncertainty less than 1%), which gives $Q_s(22) = 0.185(11)$ b.

The sign of $Q_s(22)$, determined for the first time, indicates a prolate nuclear deformation. An evaluation of the deformation parameter beta can be derived from the intrinsic quadrupole moment Q_0 which is related to the spectroscopic quadrupole moment Q_s via the well-known projection formula

$$Q_s = Q_0[I(2I-1)/(I+1)(2I+3)]. \tag{6}$$

For ^{22}Na with $I = 3$ the intrinsic quadrupole moment is $Q_0 = +0.444(26)$ b.

From the literature a value for the intrinsic quadrupole moment of ^{22}Na is known: $Q_0 = 0.50(1)$ b [20,28] which is deduced from measurements of the rotational level lifetimes (here the weighted mean value from the data in [20] is given). The larger $Q_0(22)$ value can be due to the following reasons: i) our quadrupole moment takes into account only static deformation, while the results of [20,28] are also sensitive to zero point vibrations; ii) [20,28] contain a model dependence in the basic assumption about the nuclear shape assumed as rotational ellipsoid (as a matter of fact, all Q_0 values and in particular formula (6) are model dependent as well); and, iii) in evaluating of the quadrupole moment the admixture of bands has been neglected in [20]. Having all this in mind, one could say that Q_0 value deduced in the present work is in a reasonable agreement with the results of [20].

Shell-model calculations (see [20]) show a remarkably close agreement with the experiment predicting a relatively large quadrupole deformation of ^{22}Na: the theoretical Q_0 values lie between those of [20] and of the present work. The collective effects which are described macroscopically by a deformation of the nucleus, are reproduced in the shell-model calculations by the coherent effect of the six valence nucleons - 3 protons and 3 neutrons.

The deformation parameter $\beta_2 = 0.441(24)$ has been calculated using the relation

$$Q_0 = \frac{3}{\sqrt{5\pi}} Z R_0^2 \beta_2 [1 + \frac{1}{8}\sqrt{\frac{5}{\pi}}\beta_2],$$

where $R_0^2 = 0.0144 A^{2/3}$ b. The experimental results for $\mid \beta_2 \mid$ in the Na region are given in Fig.4. These are the $\mid \beta_2 \mid$ values obtained by recalculation of the data from [3] plus our values of β_2 for ^{22}Na and ^{23}Na. For comparison Fig.4 shows also the $N-$dependence of the deformation parameter $\mid \beta_2 \mid$ for the neighbouring nuclei $_{10}$Ne and $_{12}$Mg. It is seen, that $\beta_2(22)$ follows very well the general trend of $\mid \beta_2 \mid$ versus N dependence for the investigated sodium isotopes.

4.2 Isotope shift ^{22,23}Na

Using the already known hf splitting constants the determination of the center of gravity of the D_2 line is straightforward. The following values of the hf splitting constants have been used: for $^2P_{3/2}$ level from the present work (Table 2), for $^2S_{1/2}$ level from [16] for ^{22}Na and from [31] for ^{23}Na. Further, with our experimental values of the frequency intervals between the hf structure components of ^{22}Na and ^{23}Na the isotope shift of both isotopes in D_2 line have been deduced:

$$\delta\nu^{23,22}(D_2) = \nu(23) - \nu(22) = 757.72(24)\ \ MHz.$$

The most precise value of the IS between both isotopes in the D_1 line is $\delta\nu^{23,22}(D_1) = 758.5(7)$ MHz [32]. Both D_1 and D_2 transitions are from a common level - the $^2S_{1/2}$ ground level in the optical spectra of NaI. The influence of the common level is cancelled when comparing the IS

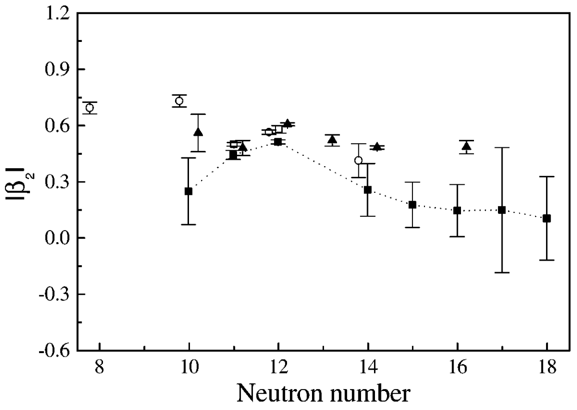

Fig. 4. Experimental deformation parameter $| \beta_2 |$ of $_{11}$Na (full squares) and of the neighbouring $_{10}$Ne (open circles) and $_{12}$Mg (full triangles) isotopes. The data for the even-even Ne and Mg isotopes were deduced from $B(E2)$ measurements [29], for the odd-even isotopes - using the rotational level lifetimes given in [30]. Since the laser-spectroscopic data points for Na except ^{22}Na and ^{23}Na are with huge error bars, these points are connected with dotted line to guide the eye and to suggest the general trend of the $| \beta_2 |$ behaviour in the studied N-region. The open squares present the values of β_2 for ^{22}Na and ^{23}Na obtained by nonoptical methods from [20] and [30] respectively

in two such lines. The thus obtained IS for the $3p\ ^2P_{3/2}$ level relative to the $3p\ ^2P_{1/2}$ level is: $\Delta T^{23,22} = -0.78(74)$ MHz.

The situation is different in comparison to that for lithium and potassium where the IS in D_2 line has been found by many authors to be larger than the IS in D_1 line, e.g for Li one obtains $\Delta T^{7,6} = 1.8(2)$ MHz [33]. Obviously an improvement of the IS experimental accuracy for sodium, especially for the D_1 line, is required to draw reliable conclusions.

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