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Yu.P.Gangrsky, D.V.Karaivanov, K.P.Marinova*,
B.N.Markov, L.M.Melnikova, G.V.Mishinsky,
S.G.Zemlyanoi, V.I.Zhemenik

HYPERFINE SPLITTING AND ISOTOPE SHIFT OF ^{22}Na, ^{23}Na BY HIGH RESOLUTION LASER SPECTROSCOPY AT THE ATOMIC D_2-LINE

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*Faculty of Physics, University of Sofia, Bulgaria

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1 Introduction

In earlier experiments with high resolution on-line laser spectroscopy the hyperfine (hf) structure and the isotope shift (IS) of the D_2 line of $^{21-31}$Na relative to ^{23}Na have been 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 have also been investigated [3] and the quadrupole coupling constants B of the $3p\ ^2P_{3/2}$ excited state have been deduced. Thus, additional information on the nuclear deformation has been 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 optical 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 the parameters of experiment (laser frequency scan, frequency stabilization etc.) were computer controlled.

^{22}Na ($T_{1/2} = 2.602$ years) has been produced by the reaction $^{19}\mathrm{F}(\alpha, n)$ in a target of AlF_3 . The sodium atoms have been extracted 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 of 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 more 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 quantity down to 10^{13} atoms. In this way all the experimental conditions have been optimized. In particularly, an experimental FWHM of the detected resonance lines of the order of 20 - 23 MHz had been established due to a reasonable limiting of 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 to about 1 μW. This was done to reduce the power broadening which under such condition gives a maximal FWHM of 12 MHz [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 ground $3s\ ^2S_{1/2}$ state. In our measurements of the ^{22}Na hf splitting each group has been 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 has been used to choose correctly the start 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 thus, for each of them more as 10 spectra were recorded with a sufficiently good signal to noise ratio.

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.

The hf splitting of the $3p\ ^2P_{3/2}$ level for ^{23}Na has been also measured in the course of our test experiments. This has been 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 Fig.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 procedure of the spectra decomposition included two fixed parameters: the number of the hf splitting components and the FWHM tested by studing well separated resonance peak ($F' = 2 \rightarrow F = 3$ for ^{23}Na). As can be seen from Figs. 1 to 3 the ratios of the fitted line amplitudes correspond in general to the theoretical expected ones.

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. The centroid of each peak was measured with respect to the two nearest-neighbour etalon peaks. 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 implied. 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 not taken into account.

The experimental values of the hf splitting spacings 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 critical 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 are obtained by quantum -beat spectroscopy [5,13]. The recently data of [9] deduced by laser-induced resonance are also shown, as well as the theoretical 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 determinations.

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

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

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. Such a result agrees with the fact, that in the mass region of the light nuclei the nuclear hf anomaly is negligibly small [17,18], 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 [19]. 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. [20]) 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, in most cases the following is assumed:
(i) d^2V/dz^2 is proposed 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 stable isotope and on an optical configuration which is best suited for a reliable estimation or an *ab initio* calculation of d^2V/dz^2;
(iii) Q_{ref} can 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.

An early method to obtain d^2V/dz^2 were the calculations of $< r^{-3} >$ values in magnetic dipole hyperfine coupling and to introduce Sternheimer corrections to account for quadrupole shielding [21]. This was the way in which the spectroscopic quadrupole moments for the sodium isotope chain $^{21,25-29}$Na [3] have been deduced. With the Hartree-Slater calculations of [21] for ^{23}Na, providing the best agreement with measured magnetic moments, the relation

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

has been obtained and used to calculate Q_s. Table 3 presents the thus obtained $Q_s(23)$ with different $B(23,^2P_{3/2})$ values. As can be seen, $Q_s(23)$ values are with more than 10% larger than the muonic $Q_s(23) = 0.1006(20)$ b of [22]. This exceeds 6-7 times the quoted errors. The last are only the statistical experimental uncertainties of the B factors to which one had to add an uncertainty of about 5% as estimate in [21].

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

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

Due to the improved value of $B(23)$ the present $Q_s(23)$, as well as this from [5] (see Table 3), is about 2% smaller than the value reported in [23]. Nevertheless, a disagreement of about 6% with the muonic Q_s still remains

and it is larger than the quoted errors. It should be noted, that in difference to [5] the error bars of Q_s have been obtained using the conventional statistical formula for the error of a product of two independently obtained quantities. 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 [23,26] and, therefore, it will be avoided here.

Table 3 also summarises the spectroscopic quadrupole moments of ^{22}Na extracting in different ways. Note, that under the assumption (i) the relations (4) and (5) hold for each sodium isotope. All the data of $Q_s(22)$ have been obtained with the only known value of $B(22.^2P_{3/2})$ of the present work. Due to larger experimental uncertainty of the electric quadrupole constant for ^{22}Na than for ^{23}Na, different methods result in $Q_s(22)$ values scattered in the limits of the quoted errors. This situation will be obviously more pronounced for the radioactive isotopes measured in [3] because of the much more larger experimental uncertainties of B-factors than in the present work. For this reason, in evaluating the data for the radioactive Na isotopes one properly chosen method and/or reference Q_s can be used. In what follows eq.(5) is accepted, obtained by the most reliable MCHF calculations [23] (with an uncertainty less than 1%), which gives $Q_s(22) = 0.185(11)$ b.

The sign of the $Q_s(22)$, 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 $Q_0 = +0.444(26)$ b.

From the literature two quite different values for the intrinsic quadrupole moment of ^{22}Na are known: $Q_0 = 0.07(2)$ b [27] which is much smaller and $Q_0 = 0.50(1)$ b which in opposite is larger than the value reported in the present work.

In the first case [27] a level-crossing experiment has been performed and Q_0 has been extracted from the spectroscopic quadrupole moment using eq.(6). The result may indicate that eq.(6) holds no more in the case of very low Q_s values. At least, this has been argued by the authors [27]: one has also to have in mind the relative low experimental precision.

In the second case [19,28] Q_0 is deduced from measurements of the rotational levels lifetimes (here the weighted mean value from the data in [19] is given) and 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 [19.28] are also sensitive to zero point vibrations; ii) [19.28] contain a model dependence in the basic assumption about the nuclear shape assumed as rotational ellipsoid; iii) in evaluating of the quadrupole moment the admixture of bands has been neglected in [19]. 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 [19].

Shell-model calculations (see Ref. [19]) show remarkably close agreement with the experiment predicting a relatively large quadrupole deformation of ^{22}Na: the theoretical Q_0 values lie between those of [19] 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 β_2 are given in Fig.4. These are the β_2 values obtained by recalculation of the data from Ref. [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 β_2 for the neighbouring nuclei $_{10}$Ne and $_{12}$Mg. It is seen, that $\beta_2(22)$ follows very well the general trend of β_2 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 [3] for ^{22}Na and from [31] for ^{23}Na. Further, with our experimental values of the frequency intervals between the hf splitting 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 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 in two such lines. In this way, if one eliminates the wavelength dependence of the transition IS (expressed by the normal mass shift) J–dependence of the IS can be revealed. The thus obtained IS for the $3p\ ^2P_{3/2}$ level reffered to the $3p\ ^2P_{1/2}$ level is: $\Delta T^{23,22} = -1.34(74)$ MHz.

The situation is different in comparison to that for lithium and potassium where the IS in D_2 line has been found from 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 enhancement of the IS experimental accuracy for sodium, especially in the case of D_1 line, is required to make some reliable conclusions.

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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.
22Na	7.31(4)	4.71(28)	this work
	7.38(6)	3.76(45)	[4]
23Na	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]

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 equation	Q (23), b	Q (22), b	Ref. for the method	B-value
hfs + HS cal.	(4)	0.1145(25)*		[21]	[13]
		0.1126(12)*			[5]
		0.1126(22)*	0.195(12)*		this work
hfs + MCHF cal.	(5)	0.1089(25)		[23]	[13]
		0.1070(15)			[5]
		0.1070(23)	0.185(11)		this work
muonic value		0.1006(20)		[22]	
	(3)		0.174(11)		this work
nuclear theory		0.1024		[24]	
	(3)		0.177(11)*		this work
nuclear theory		0.1034		[25]	
	(3)		0.179(11)*		this work

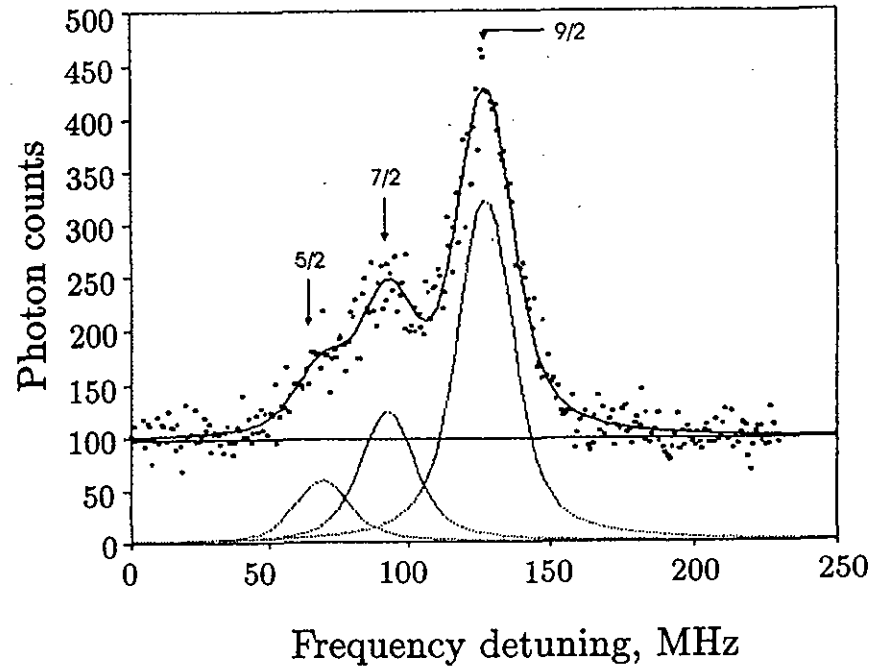

Fig.1. Example of a typical flag-pattern of D_2 line in the transition $F' = 7/2 \rightarrow F = 9/2, 7/2, 5/2$ for ^{22}Na. F' and F are the total quantum numbers of the lower and upper levels of the transition, respectively. A deconvolution of the observed peaks as well as the overall fitted spectrum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

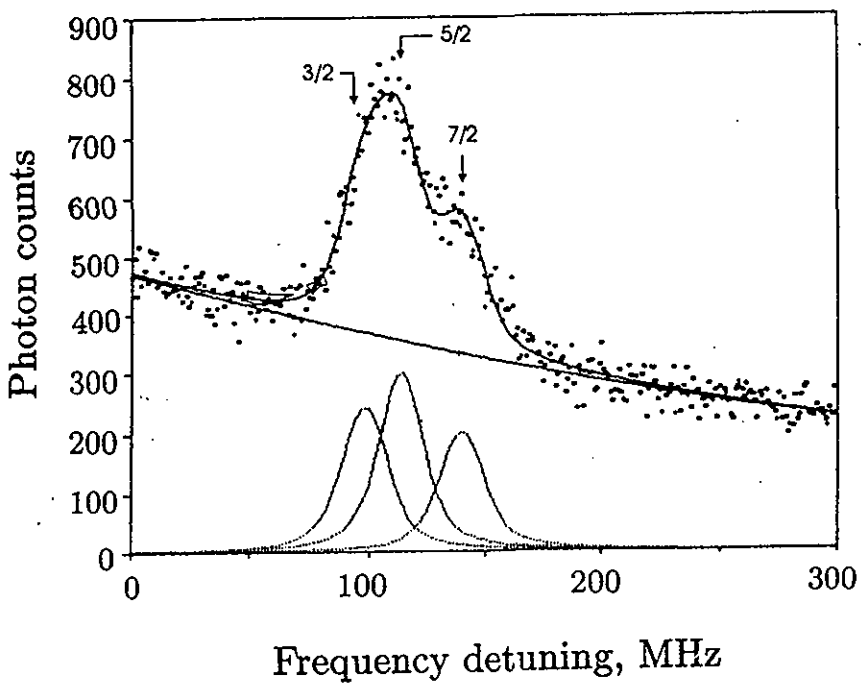

Fig.2. Example of a typical D_2 line single run in the transition $F' = 5/2 \rightarrow F = 7/2, 5/2, 3/2$ levels of ^{22}Na. The rest as in Fig.1.

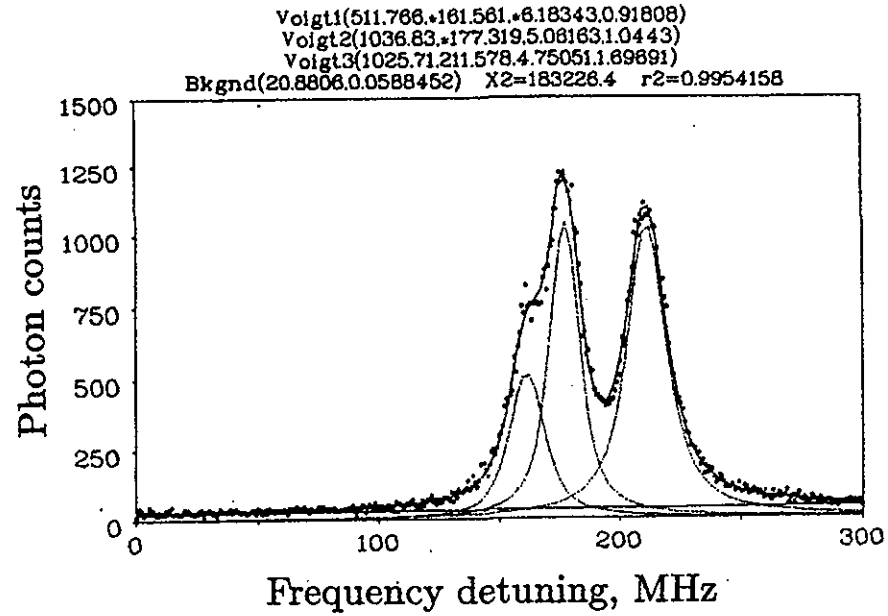

Fig.3. Example of a deconvolution of the observed resonance peaks in D_2 sodium line for ^{23}Na in the transition $F' = 1 \rightarrow F = 2, 1, 0$. In this case the FWHM of the resonance lines was 18 MHz.

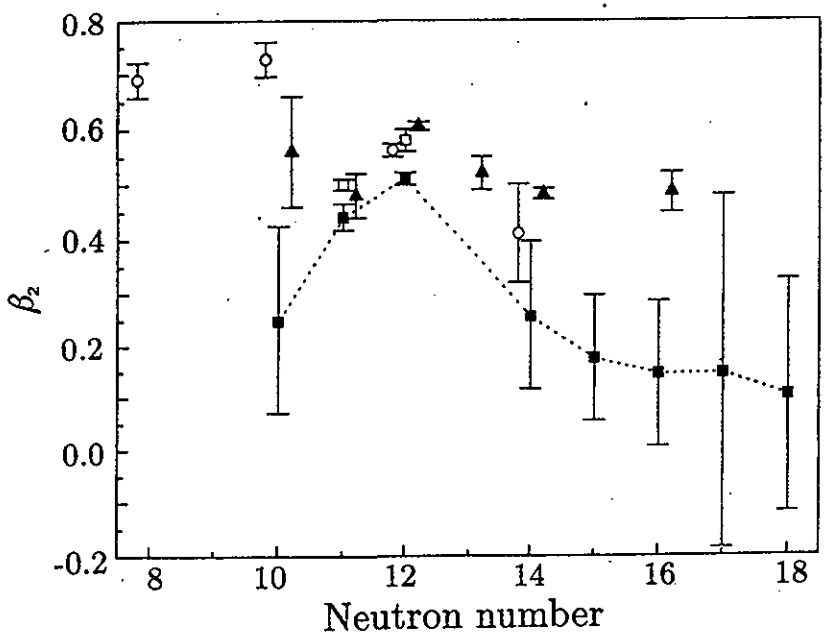

Fig.4. Experimental nuclear quadrupole deformation of $_{11}$Na (full squares) and of the neighbouring $_{10}$Ne (full circles) and $_{12}$Mg (open triangles) isotopes. The data for the even-even Ne and Mg isotopes have been deduced from $B(E2)$ measurements [29], for the odd-even isotopes - using the rotational levels lifetimes given in [30]. The data points for Na connected with dotted line have been obtained by laser-spectroscopic measurements. The open squares present the values of β_2 for ^{22}Na and ^{23}Na obtained by nonoptical methods from [19] and [30] respectively.

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