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E6-86-232

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LASER SPECTROMETER PARAMETERS

Submitted to "Journal of Physics E:
Scientific Instruments"

1986

Recently optical methods for nuclear moments via hyperfine structure determination, based on dye laser spectrometers, have been developed ^{/1,2/}. An atomic beam laser spectrometer has been built in Dubna and described elsewhere ^{/3,4/}. The parameters of such spectrometers are in general defined and experimentally determined ^{/1,2/}. Nevertheless, in the course of the laser group experimental work the development of theoretical methods for their determination from experiment and for their prediction to achieve optimal development of the set up appeared necessary. As a first step a theory of atomic beam collimation was developed ^{/5/}. As a second step this work is proposed, containing a precise definition of the parameters to be dealt with, possibilities for their determination, prediction and improvement. The methods developed here have been applied to the spectrometer optimization ^{/3,4/}.

1. Atomic beam and fluorescence light parameters

The type of set up we have in mind is described in refs. ^{/3,4/}. To determine its parameters (section 2), firstly the distribution of the atomic flux out of the oven over space and velocity is necessary. To rely on experiment only about such a distribution in a multidimensional space, dependent on several parameters connected with sample plus oven plus collimator, laser and detector properties, is rather hopeless. Therefore a theoretical approach has been developed and experimentally checked ^{/5/}. It yields finally the Doppler line width at 1/2 height $\Delta\nu$, and the following integral quantities: χ [atom] is the local number of atoms (in the interaction region of atomic beam with laser ray and fluorescence light, in our case 6 mm^3); φ [atom/s], the local flux of atoms (per second, intersecting the interaction region cross section, 6 mm^2); Ψ [atom/s], the total flux of atoms (per second, leaving the oven collimator); general notation θ of the set (χ, φ, Ψ) .

Secondly, information on the excitation and registration equipment is necessary, including laser line width, reaction of the photon counting set up to effect and background (section 3) and on the

interaction process between atomic beam, laser ray and emitted resonance fluorescence light (section 4). We shall see in section 4 that some estimates are possible, helping the laser spectrometer optimization. However such estimates are rather approximate, and therefore experimental information is necessary. We are going to summarize it in the quantity N_e [count/s]: the effect, e.g. the number of counts per second in the peak of the hyperfine structure over background (section 4), and also in the background N_b [count/s]. In the last case experimental information is unavoidable, but it can be helped by some model dependencies (section 5). However a possible method for background improvement and its limitations will be discussed too (section 6). Both N_e and N_b are accepted to be corrected for dead time and deviations from single photon counting, as discussed in section 3.

2. Resolution, efficiency and sensitivity

The resolution defined by $\Delta\nu_r$ is determined via the theoretically obtained Doppler width $\Delta\nu = \Delta\nu_d$ and the experimentally known laser line width $\Delta\nu_l$ by some type of summation. E.g. approximating both distributions by Gauss shapes (or by Lorentz shapes respectively), the resolution will be the reciprocal value of:

$$\Delta\nu_r = \sqrt{\Delta\nu_l^2 + \Delta\nu_d^2} \quad (\Delta\nu_r = \Delta\nu_l + \Delta\nu_d). \quad (1)$$

We can define the reciprocal efficiency set of parameters $\theta_e = (\chi_e, \psi_e, \gamma_e)$ by theoretical θ and experimental N_e quantities as follows:

$$\theta_e = \theta/N_e. \quad (2)$$

It means that χ_e [atom*s/count] represents the local number of atoms producing an effect of 1 count per second in the peak of the hyperfine structure; ψ_e [atom/count], the local flux of atoms producing the same effect; γ_e [atom/count], the total flux of atoms producing the same effect.

To introduce the sensitivity, we use moreover t_c [s]: the measuring time per channel. Also a constant a is the minimal ratio of effect over statistical error of effect plus background, for which the effect is accepted to be observable (we assume as usually $a = 3$). Let us denote the minimal observable effect by N_o [count/s]. Then:

$$N_o t_c = a \sqrt{(N_o + N_b) t_c}. \quad (3)$$

Solving (3) with respect to N_o and defining the reciprocal sensitivity set of parameters $\theta_s = (\chi_s, \psi_s, \gamma_s)$ as the corresponding minimal observable quantities, we find:

$$\theta_s = \theta_e N_o = a \left(\frac{a}{2} + \sqrt{\frac{a^2}{4} + N_b t_c} \right) \theta / (N_e t_c) \quad (4)$$

χ_s [atom] represents the minimal observable local number of atoms; ψ_s [atom/s], the minimal observable local flux of atoms; γ_s [atom/s], the minimal observable total flux of atoms.

We see immediately two limiting cases of formula (4). In the first one of background low enough $a^2 \gg N_b t_c$, the sensitivity is limited by statistics:

$$\theta_s = a^2 \theta_e / t_c = a^2 \theta / (N_e t_c). \quad (5)$$

And in the second case of background high enough $a^2 \ll N_b t_c$, the sensitivity is limited by background:

$$\theta_s = a \sqrt{N_b / t_c} \theta_e = a \sqrt{N_b / t_c} \theta / N_e. \quad (6)$$

3. Single photon counting

In this section the optimal choice and regime of the photon counting system will be discussed. Methods for data correction when deviations from these conditions appear will be proposed.

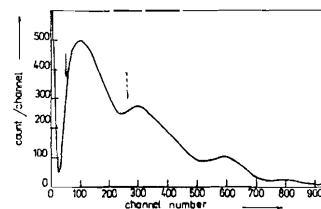


Fig. 1. Electron pulse amplitude distribution of the chosen photomultiplier FEU 79 at high counting rate. Discrimination level for single photon counting is indicated by a vertical solid arrow; for double photon counting, by a vertical dashed arrow.

We start with fig. 1, taken from ref. ^{13/}. As is shown there, the choice of the photomultiplier should be such that the one-, two-, and so on photon peaks should be well distinguishable. Single photon counting would be achieved when the discrimination level is somewhat below the one-photon peak maximum.

To understand why this is preferable, and what type of corrections

should be introduced if it is violated, let us remind the well known Poisson statistical distribution of many-fold photon bursts ^{1/6/}. We denote by $N = N_g + N_b$ [s^{-1}] the real effect plus background rate; by N_x , the measured x-fold burst rate; N_s , the measured saturation rate (N_s^{-1} [s]; the overall dead time of the whole photon counting system). Then:

$$N_x = \frac{N}{1 + N/N_s} \frac{(N/N_s)^{x-1}}{(x-1)!} e^{-N/N_s} \quad (7)$$

Let us also denote by N_x^∞ the measured x- and more-fold burst rate. Then evidently:

$$N_x^\infty = \sum_{\xi} N_\xi = \frac{N}{1 + N/N_s} \left[1 - \sum_{\xi=0}^{x-1} \frac{(N/N_s)^\xi}{\Gamma(\xi)} e^{-N/N_s} \right] \quad (8)$$

Notice that for $x = 1$ the expression in brackets equals 1, and (8) gives the usual dead time correction of the measured N_1^∞ to real rate N . For $N \ll N_s$, $N_x^\infty \sim (N)^x$, so that the relation of the single photon counting rate N_1^∞ to the real rate N remains linear. However for $x \geq 2$ this linearity is violated, and for decreasing real rate N , the measured one N_x^∞ decreases much faster, with the x-th power of N . Therefore the ratio of statistical error to effect, for small effect N , will increase with increasing x . This means also that the two-, three- and so on photon peaks, compared to the one-photon peak of the electron pulse amplitude distribution in fig. 1, will progressively disappear with decreasing photon intensity N .

The situation described has been checked experimentally. For this purpose the real counting rate N has been obtained: 1) for a discrimination level corresponding to $x = 1$, deriving N from the experimental counting rate N_1^∞ by:

$$N_1^\infty = \frac{N}{1 + N/N_s} \quad (9)$$

2) for a discrimination level corresponding to $x = 2$, deriving N from the experimental counting rate N_2^∞ by:

$$N_2^\infty = \frac{N(1 - e^{-N/N_s})}{1 + N/N_s} \quad (10)$$

The results for N have been compared and have turned out to be approximately the same.

4. Effect enhancement

To enhance the effect N_e , at a given total flux of atoms Ψ , means to increase the total efficiency ψ_e^{-1} (2) and to increase the total sensitivity ψ_s^{-1} (4). Let us therefore, first of all, discuss the total reciprocal efficiency ψ_e (2) and its various components (factors) $\psi_e^{(k)}$, related to different processes in the laser spectrometer. We have summarized their values obtained in the last version of the set up, with their eventual best values limits, in table 1.

Table 1

Reciprocal efficiency components $\psi_e^{(k)}$ for an odd (or doubly odd) and a (doubly) even nucleus (both of the "standard" type described in text of section 4)

k	Component		Nucleus	$\psi_e^{(k)}$		
				Now	Limit	
1	collimation			5×10^2	1×10^2	
2	atomic registration	photon production	excitation	1×10^0	1×10^0	
3			transition pro-	odd	8×10^{-1}	8×10^{-1}
3		duction	even	1×10^{-2}	1×10^{-2}	
4		component pro-	odd	6×10^0	6×10^0	
4		duction	even	1×10^0	1×10^0	
5	atomic registration	photon detection	light collection	1.4×10^1	2×10^0	
6			photomultiplier counting	1.5×10^1	5×10^0	
7			filter transmission	4×10^0	1×10^0	
Total $\psi_e = \prod_{k=1}^7 \psi_e^{(k)}$				odd	2×10^6	5×10^3
				even	4×10^3	1×10^1

The collimation reciprocal efficiency $\psi_e^{(1)} = \psi_e / \psi_e = \Psi / \Psi$ is already near to the limit: an improvement of only 5 times is expected in realistic cases, as explained in ref. ^{1/5/}.

The registration reciprocal efficiency ψ_e decomposes into 2 groups of components. The photon production group consists of 3 components: firstly, the excitation $\psi_e^{(2)}$, being the ratio of the Doppler broadened to the real (natural plus laser) line width. In our case it is near to 2, and is very difficult to be improved to 1

if complete hyperfine structure component resolution is preserved. However we have corrected it to 1 to compare with the real case of ref. /4/. In fact, in that case we have an average overlapping of about 2 line components, enhancing the efficiency by the same factor. Secondly, the transition production $\psi_e^{(3)}$ is the number of transitions of one atom during its 4×10^{-6} s long flight through the 1 mm thick laser ray. In the case of a doubly even nucleus with a "standard" excited level radiation lifetime 4×10^{-8} s, about 10^2 transitions per atom could be expected for saturation at high enough laser power. However, in the case of an odd or doubly odd nucleus with several ground state sublevels, optical pumping to them would soon make the sublevel under resonance empty, so that roughly 1.25 transitions per atom could take place for saturation at a high enough laser power. Thirdly, the component production $\psi_e^{(4)}$ is the number of ground state sublevels, and thus $\psi_e^{(4)-1}$ will be the ratio of sublevel to total level populations. As an example we have taken the $^{151,153}_{63}\text{Eu}$ case, with a nuclear ground level spin 5/2 and an atomic ground level spin 7/2, i.e. with a number of ground state sublevels 6, accepted here as "standard". Both last components cannot be improved by changing the set up. They depend on the investigated transition, atom and nucleus only.

The photon detection group consists of 3 components again. Firstly, the light collection $\psi_e^{(5)}$ is the ratio of 4π to the collected light solid angle. Secondly, the photomultiplier counting $\psi_e^{(6)}$ depends on its properties and is usually not better than 10, only by extra measures to become 5. So, generally, a 20 times improvement could be the limit result. Thirdly, the interference filter transmission $\psi_e^{(7)}$ 4 times reduction can be avoided, in order to get another 4 times improvement, only in cases when oven background is less than laser background. Otherwise it is necessary to use it in order to improve the effect to oven background ratio, in our case about 100 times /4/.

Let us note at the end that the total reciprocal efficiency of our present table 1, first column (now), odd nucleus: $\psi_e = 2 \times 10^6$, is in agreement with the experimental one of ref. /4/, table 2, second row: $\psi_e = 1.8 \times 10^6$. The limit of improvement is about 10^2 times lower or better, due to rows 1, 5, 6, without the doubtful possibility of other 4 times due to row 7.

5. Background reduction

The background N_b components $N_b^{(k)}$, whose reduction will increase the sensitivity ψ_s^{-1} (4, 6), are mainly an experimental problem. Therefore they have been studied by our group, and the results published in ref. /4/.

However, there are some approximate model dependencies which facilitate the background determination (especially the oven background components: $N_b^{(3')}$ with interference filter or $N_b^{(3'')}$ without it) for any sample temperature T and laser power I . They, together with our results $N_b^{(k)}$ of ref. /4/ at standard \bar{T} and \bar{I} , are shown in table 2. For the ^{63}Eu I line 5765.20 \AA , $\lambda = 5768 \text{ \AA}$, $\nu = c/\lambda$ is the interference filter maximum transmission ($\nu = 1/4$)

Table 2

Background components $N_b^{(k)}$ with their dependence on sample temperature T and laser power I ($\nu; \nu_1, \nu_2$: see text section 5; h : Plank and k : Boltzmann constant)

k	Component	$N_b^{(k)}$ at $\bar{T} = 1350 \text{ K}$ $\bar{I} = 100 \text{ mW}$	$N_b^{(k)}/\bar{N}_b^{(k)}$ dependence on T, I
1	photomultiplier noise	40	const.
2	laser background	2500	I/\bar{I}
3'	with interference filter	500	$(e^{\frac{h\nu}{kT}} - 1) / (e^{\frac{h\nu}{kT}} - 1)$
3''	without interference filter	90000	$\int_{\nu_1}^{\nu_2} \frac{\nu^3 d\nu}{e^{\frac{h\nu}{kT}} - 1} / \int_{\nu_1}^{\nu_2} \frac{\nu^3 d\nu}{e^{\frac{h\nu}{kT}} - 1}$
Total $N_b = \sum_{k=1}^3 N_b^{(k)}$			

frequency, $\Delta\lambda = 75 \text{ \AA}$: transmission bandwidth, $\Delta\nu = c\Delta\lambda/\lambda^2$; ν_1, ν_2 are the empirical limits of photomultiplier average sensitivity.

Empirical oven background reduction /4/ with interference filter, as compared to the case without it, good description by:

$$N_b^{(3')}/N_b^{(3'')} = \eta \frac{\nu^3 \Delta\nu}{e^{\frac{h\nu}{kT}} - 1} / \int_{\nu_1}^{\nu_2} \frac{\nu^3 d\nu}{e^{\frac{h\nu}{kT}} - 1} \quad (11)$$

is achieved with: $\lambda_1 = 6750 \text{ \AA}$, $\lambda_2 = 4000 \text{ \AA}$. The oven background reduction and the oven background to effect ratio reduction with interference filter is as follows: at $T = 900 \text{ K}$ 400 and 100 times, at $T = 1350 \text{ K}$ 180 and 45 times.

6. Photon burst technique

This is a basic possibility of effect to background ratio enhancement /6, 7/. A simplified modification of this technique has been proposed by Yu.P. Gangrsky: to use x - or more-fold bursts by simply raising the discrimination level to the minimum below the x -photon peak of fig. 1, $x = 2, 3, \dots$

Suppose n photons are emitted per atom ($n \gg 1$ only in even-even nuclei: section 4) during its flight through the laser ray. The probability to obtain a x -fold coincidence is:

$$P_x = \binom{n}{x} \omega^x (1-\omega)^{n-x}, \quad (12)$$

where ω is the photon detection efficiency. For $n\omega$ fixed and $n \rightarrow \infty$ this can be approximated by a Poisson distribution again /6/:

$$P_x = \frac{(n\omega)^x}{x!} e^{-n\omega}. \quad (13)$$

The probability to obtain a x - or more -fold coincidence is then

$$P_x^n = \sum_{\xi=x}^n P_\xi = 1 - \sum_{\xi=0}^{x-1} P_\xi. \quad (14)$$

We look at the $x = 1$:

$$P_1^n = 1 - (1-\omega)^n \quad (15)$$

and the $x = 2$ case:

$$P_2^n = 1 - (1-\omega)^{n-1} [1 + (n-1)\omega] \quad (16)$$

by using (12). We note firstly that for small ω $P_1^n \approx P_1 \approx n\omega$, which shows an improvement of the overall efficiency $n_{\text{even}}/n_{\text{odd}} \approx 80$ times. Secondly, for $\omega \ll 1$, for the coincidence case, the ratio of (16) to (15) is:

$$c = P_2^n / P_1^n \approx P_2 / P_1 \approx (n-1)\omega/2 \quad (17)$$

c is actually the coincidence effect increase when going from $x = 1$ to $x = 2$. Let us compare it with the random case ratio of (10) to (9) r , applied to effect or background $N = N_{e,b}$ $r_{e,b} \ll N \ll N_s$:

$$r = N_2^\infty / N_1^\infty \approx N / N_s \quad (18)$$

r_b is actually the background increase when going from $x = 1$ to $x = 2$. So $(c + r_b) / \sqrt{r_b}$ is the enhancement of the ratio of effect to statistical error of background, and thus the improvement of sensitivity θ_s^{-1} (6) when doing the same, i.e., going from single ($x = 1$) to double ($x = 2$) counting mode ($c \gg r_b$ for small N_s).

To see how it looks with our data /4/, we present it in table 3 for photons emitted per atom from an odd nucleus $n = 1.25$ and (doubly) even nucleus $n = 10^2$ (both of the "standard" type described in section 4), photon detection efficiency now $\omega = 1.25 \times 10^{-3}$ and in the limit of improvement $\omega = 10^{-1}$ (see table 1), background normal $N_b = 10^3 \text{ s}^{-1}$ and strongly reduced $N_b = 25 \text{ s}^{-1}$, saturation rate $N_s = 10^6 \text{ s}^{-1}$.

Table 3

Sensitivity θ_s^{-1} (6) improvement $c/\sqrt{r_b}$ from (17, 18) when going from single to double counting mode for different nucleus, efficiency and background: see text section 6

k	Nucleus	Efficiency	Background	$c/\sqrt{r_b}$
1	odd	now	normal	4.9×10^{-3}
2	odd	now	reduced	3.1×10^{-2}
3	odd	limit	normal	4.0×10^{-1}
4	odd	limit	reduced	2.5×10^0
5	even	now	normal	2.0×10^0
6	even	now	reduced	1.2×10^1
7	even	limit	normal	1.6×10^2
8	even	limit	reduced	9.9×10^2

Looking at table 3 we can say that for odd or doubly odd nuclei, in the cases $k = 1, 2, 3$ the sensitivity becomes worse, and only in the case $k = 4$, i.e., if we improve both efficiency and background to the limit, there is a non-essential improvement. Thus this modification is of no use for odd nuclei. For (doubly) even nuclei there is a non-essential improvement for $k = 5$ if efficien-

cy and background remain as they are, but the improvement becomes one order of magnitude for $k = 6$, i.e., with improved background, two orders of magnitude for $k = 7$, i.e., with improved efficiency, and three orders of magnitude for $k = 8$, i.e., with improved both efficiency and background. Thus such a modification might become useful for doubly even nuclei. In general it requires an essentially increased measuring time t_c , but at the same time this leads to an improvement of the single photon counting mode sensitivity θ_s^{-1} by (6), which acts in addition to the ratio of the double to single mode sensitivity $c/\sqrt{F_b}$, derived by (17, 18) and shown in table 3.

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Received by Publishing Department
on April 14, 1986.

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Наджаков Е.Г.

E6-86-232

Параметры лазерного спектрометра

Обсуждены основные параметры лазерного спектрометра с атомным пучком: разрешение, эффективность и чувствительность. Предложены формулы для их вывода на основе эксперимента. Показан оптимальный выбор и режим системы счета фотонов. Найден метод коррекции данных при отклонениях от этих условий. Указаны возможности предсказания и улучшения эффекта и фона. Критически представлена техника фотонных вспышек, в сравнении со счетом одиночных фотонов, и рассмотрен вопрос, при каких условиях она могла бы улучшить чувствительность.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1986

Nadjakov E.G.

E6-86-232

Laser Spectrometer Parameters

The basic atomic beam laser spectrometer parameters: resolution, efficiency and sensitivity are discussed. Formulae for their derivation from experiment are proposed. The optimal choice and regime of the photon counting system is indicated. A method is found for data correction when deviations from these conditions appear. The possibilities for effect and background prediction and improvement are pointed out. The photon burst technique is critically presented, compared to the single photon counting, and the question under what conditions it could improve the sensitivity is considered.

The investigation has been performed at the Laboratory of the Nuclear Reactions, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1986