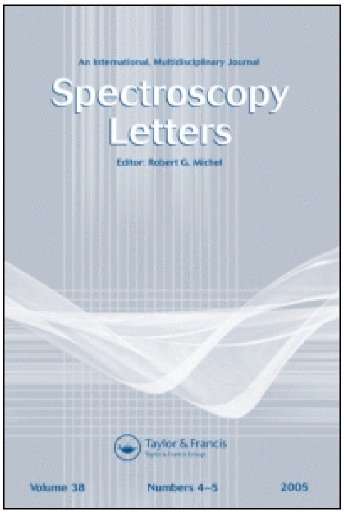

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M. A. Rebolledo[a]; J. A. Martínez-Betorz[a]
[a] Departmento de Física Aplicada, Facultad de Ciencias, Universidad de Zaragoza, Zaragoza, Spain

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QUANTUM BEAT DETECTION BY PHOTON STATISTICS TECHNIQUES

M.A. Rebolledo and J.A. Martínez-Betorz

Departamento de Física Aplicada, Facultad de Ciencias,

Universidad de Zaragoza, 50009 Zaragoza, Spain.

ABSTRACT

In this paper we study the usefulness of photon statistics techniques in quantum beat spectroscopy when the intensity of the detected light beam is very small. It is concluded that accurate results can be obtained if the Fourier transform of the time-interval probability is measured.

INTRODUCTION

Quantum beat spectroscopy[1)] is a high resolution technique where two very close energy levels are coherently excited by a pulsed laser. If the fluorescence from the excited levels to a lower energy level is detected a beating intensity is obtained (quantum beats) whose beating frequency provides us the difference of the energies of the two excited levels. Quantum beats can also be detected by phosphorescence[2], superfluorescence[3] or transmission[4] phenomena.

When high excited levels are measured by quantum beats a very small intensity can be obtained that can be masked by noise. Bearing in mind that very noisy signals can be analyzed succesfully by measuring their intensity autocorrelation functions[5] we think this technique could be useful in quantum beat detection. On the other hand small intensity periodic signals can be very accurately analyzed[6-9] by means of the Fourier transform of the time-interval probability, obtained from a repeatedly measurement of the time-interval between two consecutive photopulses when using a photon counting system to detect the light beam. Therefore this technique could also be useful in quantum beat spectroscopy.

The aim of this paper is to study the usefulness of these techniques for small values of the intensity.

METHOD

If a quantum beat signal is measured where only a decay constant, Γ, appears the detected intensity can be expressed[1] as

$$I(t) = I_0[1+V\cos(2\pi\nu_0 t+\delta)]\exp(-\Gamma t) \tag{1}$$

ν_0 being the quantum beat frequency. For noisy signals the coherent excitation of the energy levels is periodically repeated and a periodic repetition of the quantum beat signal is obtained. If a synchronous average procedure is used noise is reduced. Nevertheless as I_0 decreases noise increases and a poor noise reduction is obtained by this method.

In this paper we intend to analyze very noisy quantum beat signals by means of the normalized intensity autocorrelation function or by means of the Fourier transform of the time-interval probability.

The normalized intensity autocorrelation function is defined[5] as

$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle^2} \tag{2}$$

If the time-intervals, θ, between consecutive photopulses are repeatedly measured the Fourier transform of the time-interval probability is defined as

$$Q_F(\nu) = \int_0^\infty W(\theta)\cos(2\pi\nu\theta)d\theta \tag{3}$$

$W(\theta)$ being the time-interval probability.

In order to know the shape of $g^{(2)}(\tau)$ and $Q_F(\nu)$ we used an experimental setup to measure these quantities. A signal generator was fed into a LED to produce a light beam with an intensity given by

$$I(t) = I_0\{1+V\cos[2\pi\nu_0(t-nP)+\delta]\}\exp[-\Gamma(t-nP)]$$
$$nP \leqslant t < (n+1)P \quad (n = 0,1,2,\ldots) \tag{4}$$

that is a periodic repetition of the quantum beat signal (Eq. (1)) with a period P. A photon counting system controlled by a computer was used to measure $g^{(2)}(\tau)$ and $Q_F(\nu)$.

Fig. 1 shows the results obtained for $g^{(2)}(\tau)$ when ν_0 = 36380 Hz, V ≈ 0.3, Γ ≈ 12500 Hz and P = 678 µS. Since $g^{(2)}(\tau)$ do not changes when I_0 does (Eq. (2)) we used a high value for the mean number, $\bar{n}_p$, of photopulses detected in a period P ($\bar{n}_p$ = 100) to reduce noise in the graph of $g^{(2)}(\tau)$. It can be observed that $g^{(2)}(\tau)$ oscillates with a frequency approximatelly equal to ν_0.

Fig. 2 shows the results obtained for $Q_F(\nu)$ for the same signal used to measure $g^{(2)}(\tau)$. Since $Q_F(\nu)$ changes when I_0 does and we want to apply this technique for small intensities we attenuated the light beam to obtain $\bar{n}_p$ = 1. In this case the value of ν_0 was ν_0 = 36930 Hz because of the signal generator instabilities did not allow us to use the same frequency used to measure $g^{(2)}(\tau)$. It can be observed that the maximum of the taller peak occurs for $\nu \approx \nu_0$. The two other maxima occur for $\nu \approx \nu_0 \pm 1/P$.

Taking the above observations in mind we can propose a very simple method to obtain ν_0. When measuring $g^{(2)}(\tau)$ we can determine ν_0 by obtaining the frequency of its oscillations. This can be done by computing the Fourier transform

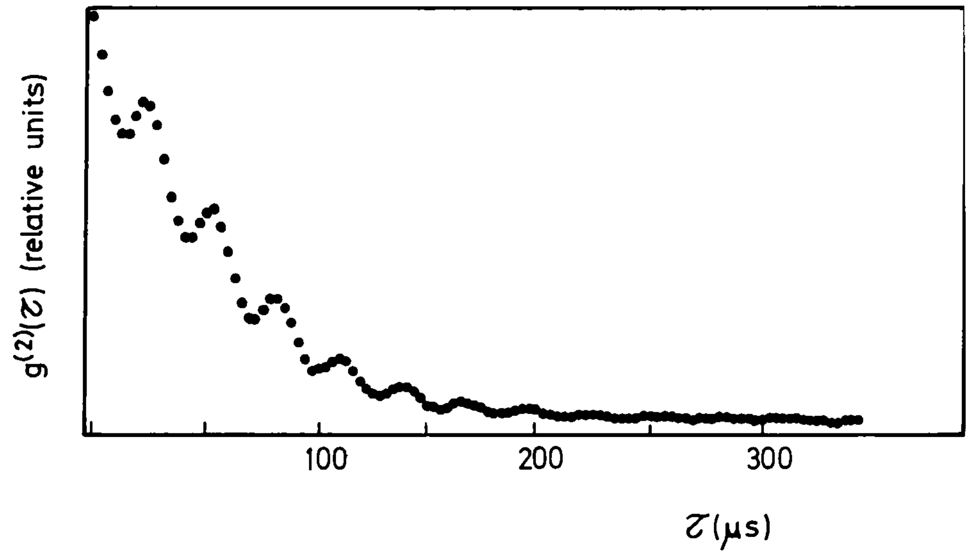

Fig. 1.- Experimental values of $g^{(2)}(\tau)$ for ν_0 = 36380 Hz.

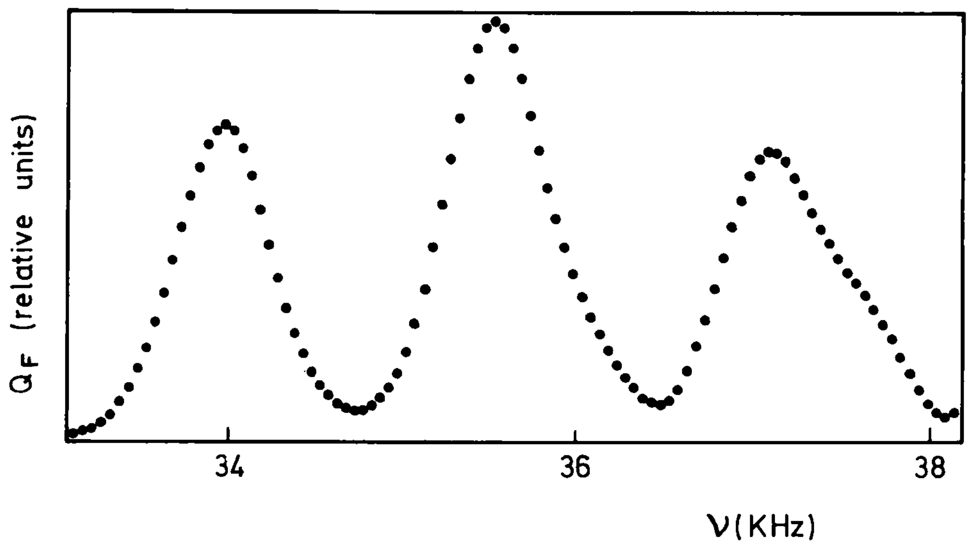

Fig. 2.- Experimental values of $Q_F(\nu)$ for ν_0 = 36930 Hz.

of $g^{(2)}(\tau)$ and obtaining the value of ν that corresponds to its maximum. When measuring $Q_F(\nu)$ we can determine ν_* by obtaining the value of ν that corresponds to its central maximum.

Since the signals obtained in our experimental setup were not stable because of the instabilities in the function generator, we studied the errors involved in the determination of the beat frequency, ν_*, by using a computer simulation method to obtain values of $g^{(2)}(\tau)$ and $Q_F(\nu)$. This computer simulation method was used and experimentally checked in previous papers[8,9].

RESULTS AND CONCLUSIONS

The errors were studied for two values of the number, N_0, of oscillations of the quantum beat signal in a period P. For each one of them three values of the mean number, $\bar{n}_p$, of photopulses in a period were used that corresponded to three small values of the mean number, $\bar{n}_0$, of photopulses in an oscillation. The values of $\bar{n}_0$ and N_Γ/N_0 (N_Γ = number of oscillations in a decay time) remained unchanged when N_0 was varied. With these values of N_0, N_Γ, $\bar{n}_p$, $\bar{n}_0$ and two values for V we obtained 12 cases that are listed in table I.

For all cases ten series of simulated values of $g^{(2)}(\tau)$ (for 101 values of τ) and $Q_F(\nu)$ (for 101 values of ν) were obtained from 10^4 simulated values of the time-interval θ. The values of ν_* were obtained as explained above. From the

Table I.- Cases for which errors in ν_0 were obtained for ν_0 = 10 KHz and δ = 0 (Eq. (4)).

Case	N_0	N_r	$\bar{n}_p$	$\bar{n}_0$	V
1	9	3	1.50	0.167	0.5
2	9	3	1.50	0.167	0.3
3	9	3	0.50	0.056	0.5
4	9	3	0.50	0.056	0.3
5	9	3	0.15	0.017	0.5
6	9	3	0.15	0.017	0.3
7	24	8	4.00	0.167	0.5
8	24	8	4.00	0.167	0.3
9	24	8	1.33	0.056	0.5
10	24	8	1.33	0.056	0.3
11	24	8	0.40	0.017	0.5
12	24	8	0.40	0.017	0.3

ten different values obtained for ν_0 we calculated the systematic error

$$e_s = \left| \frac{10^4 - \langle \nu_0 \rangle}{10^4} \right| \tag{5}$$

and the statistics error σ (the relative mean standard deviation). Table II shows the results. It can be observed that in general systematic errors are larger than statistics errors. To avoid this difficulty the method to obtain ν_0

Table II.- Values of the systematic (e_S) and statistics (σ) errors in ν_0 for the cases in table I.

	$g^{(2)}$		Q_F	
Case	e_S(%)	σ(%)	e_S(%)	σ(%)
1	3.32	0.466	1.83	0.256
2	3.83	0.791	2.01	0.744
3	2.82	0.959	1.52	0.133
4	2.15	2.01	1.56	0.265
5	2.71	1.03	1.45	0.018
6	2.48	3.23	1.50	0.127
7	0.98	0.177	0.736	0.227
8	1.39	0.546	0.729	0.688
9	1.01	0.326	0.458	0.058
10	1.40	0.840	0.414	0.213
11	1.30	4.63	0.400	0.029
12	1.05	5.30	0.436	0.135

must be improved by obtaining analytical expresions for $g^{(2)}(\tau)$ and $Q_F(\nu)$ and studying them.

With respect to statistics errors it can be observed that when obtaining ν_0 from $g^{(2)}(\tau)$ they increase as the intensity of the light beam decreases. Instead when obtaining ν_0 from $Q_F(\nu)$ the errors decrease as the intensity does. Therefore it is obvious that the measurement of $Q_F(\nu)$ is a more appropriate technique than intensity correlation technique for very small intensity quantum beat signals. From the above results we can conclude that the measurement of $Q_F(\nu)$ can be an accurate technique in very small intensity quantum beat experiments, if analytical expresions for $Q_F(\nu)$ are obtained in order to avoid systematic errors.

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