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INTERFEROMETRIC MEASUREMENTS OF SPECTRAL HOLES IN THE VISIBLE

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Abstract. A simple interferometric method like Fourier spectroscopy is suggested for spectral measurements in the visible. The method is based on the measurement of the envelope of interferometric oscillations and allows to find the symmetrical part of the spectrum. The method is applicable to the investigation of the spectra of external radiation as well as of absorption and secondary emission. Photochemically burned multi-frequency spectral holes (step $\simeq 1.65$ cm^{-1}) in impurity-doped polystyrene were studied by using a Michelson-type interferometer and a cw picosecond dye laser.

Key words: spectral hole burning, Fourier spectroscopy, interferometry.

1. INTRODUCTION

Interferometric methods, e. g. the method of Fourier spectroscopy, are widely used for spectral measurements of infrared radiation, allowing one to achieve a very high signal-to-noise ratio. In the visible these methods are seldom used due to serious difficulties in practical applications. The difficulties arise from the fact that the mean frequencies ω_0 of spectral lines exceed by many orders their widths δ (usually $\omega_0/\delta \geqslant 10^3$). Therefore the Fourier transform of the spectrum measured by these methods shows from thousands up to millions (or even more) oscillations. To find the spectrum all the oscillations should be resolved and the position (phase) of every oscillation fixed with high accuracy. Naturally, there may be cases in which the spectra in the visible are very broad ($\delta \sim \omega_0$). In such cases the interference methods by subfemtosecond pulses with a white-light continuum spectrum appear to be rather efficient [1–5], e. g. for the measurement of absorption spectra. However, in ordinary cases, $\omega_0/\delta \gg 1$, the mentioned difficulties remain also when ultrashort pulses are used.

When in the case $\omega_0/\delta \gg 1$ a usual Fourier spectrometer is used, which contains a moving mirror (controlling the difference between the optical passes of the interferometer shoulders; see e. g. [6]), then the mirror should be moved at a very small velocity over a long distance, otherwise the oscillations will be averaged to zero and the signal will be lost. The performing of such kind of measurements with the required stability and accuracy is rather difficult. The main difficulty is connected with the absolute measurement of the time t, or (which is the same here) the phase of oscillations: it should be performed with the accuracy essentially exceeding the mean period of oscillations $t_0 = 2\pi/\omega_0$. In contrast to that,

the measurements of the amplitude of oscillations may be performed with much smaller accuracy. This possibility comes from the fact that in the actual case, $\delta/\omega_0 \ll 1$, the amplitudes are well characterized by their envelope (so-called visibility function, see e. g. [7]), because they change considerably slower in the course of time, $t \sim \delta^{-1} \gg t_0$.

Here the question arises what kind of information about the spectrum can be obtained if only simple measurements of the envelope of amplitudes are performed. The answer is: one can find the sum $I_c(\omega) = I(\omega) + I(\omega - 2\Omega)$, where $\Omega = \omega - \omega_0$ is the difference between the frequency ω of the spectrum and its central frequency ω_0. In the case of symmetrical spectra $I_c(\omega)$ and $I(\omega)$ coincide [7]. It is important to underline that these kinds of measurements in the visible can be used not only for spectral investigations of an external radiation but also for the study of absorption and excitation spectra. Feasibility of the interferometric method proposed below is demonstrated by studying the multi-frequency spectral holes burned into the inhomogeneous absorption band of persistent spectral hole-burning media.

2. INTERFEROMETRIC METHOD

The method of Fourier spectroscopy is based on the dependence of the radiation passed through a Michelson-type interferometer on the optical delay time t between the interferometer shoulders

$$I(\omega; t) = \frac{1}{2} I(\omega)\,(1 + \cos \omega t). \tag{1}$$

Here $I(\omega)$ and $I(\omega; t)$ are the spectra of the radiations entering and coming out of the interferometer, respectively; $t = lc^{-1}$, l being the difference between the optical passes of the two shoulders of the interferometer. In the standard method of Fourier spectroscopy the radiation under investigation is passed through the interferometer. The total intensity of the passed radiation,

$$J(t) = \int_{-\infty}^{\infty} d\omega I(\omega; t) = \frac{1}{2}\,(J_F(0) + J_F(t)), \tag{2}$$

is studied as a function of t. Here

$$J_F(t) = \int_{-\infty}^{\infty} d\omega\, I(\omega) \cos \omega t \tag{3}$$

is the cosine Fourier transform of the spectrum. The physical nature of the t-dependence of the intensity of the passed-through radiation obviously lies in the interference phenomenon. For $\omega_0 \gg \delta$ this dependence is of an oscillatory type with a very slow change of the amplitude and the period. This can be clearly seen from the following representation of the Fourier transform, $J_F(t)$ [7]:

$$J_F(t) = A(t) \cos\,(\omega_0 t + \varphi(t)), \tag{4}$$

where

$$A(t) = (J_c^2\,(t) + J_s^2\,(t))^{1/2} \tag{5}$$

is the amplitude (visibility),

$$\varphi(t) = \arccos(J_c(t)/A(t)) \tag{6}$$

is the phase (period) correction,

$$J_c(t) = \int_{-\infty}^{\infty} d\Omega\, I(\omega_0+\Omega)\cos\Omega t \tag{7}$$

and

$$J_s(t) = \int_{-\infty}^{\infty} d\Omega\, I(\omega_0+\Omega)\sin\Omega t \tag{8}$$

are the cosine and sine Fourier transforms of the shifted spectrum. Both $A(t)$ and $\varphi(t)$ depend slowly on t in comparison with the quickly oscillating term $\cos\omega_0 t$. To find the spectrum $I(\omega)$ both the amplitude $A(t)$ and the phase $\omega_0 t+\varphi$ of the Fourier transform should be measured. If the phase correction $\varphi(t)$ is neglected (not measured), then $A(t)\approx J_c(t)$ and the Fourier transform of the envelope of the amplitudes gives

$$\frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, e^{-i(\omega_0-\omega)t}A(t) \approx \frac{1}{2}[I(\omega)+I(\omega-2\Omega)]. \tag{9}$$

Consequently, the measurement of the envelope of the amplitudes of oscillations allows one to find the even sum of the spectrum $I(\omega)$ and of the shifted spectrum $I(\omega-2\Omega)$ (with regard to the central frequency ω_0).

The same setup can be used also for the measurements of absorption and excitation spectra. Here the radiation, which is used for the measurement of the absorption or excitation of secondary emission, is first directed to the interferometer and after passing through it, to the sample. The total intensity of the light passed through the sample (in the case of transmittancy-absorption measurements) or the intensity of the secondary emission under investigation is recorded for different delay times t. Because of the slow change of the amplitudes of oscillations in relation to t, only few oscillations in the vicinity of the chosen discrete points, $t_n = 2\pi cn/\delta N$ (situated far from each other in comparison with the period t_0), should be recorded ($n=0, \pm1, \pm2, \ldots, \pm N$; N is determined by spectral resolution).

Let us consider, e. g., the measurement of the transmittancy spectrum $K(\omega)$ of a sample; the absorption spectrum can be found from the dependence of $K(\omega)$ on the thickness of the sample. The intensity of the radiation passed through the sample is

$$W(t) = \int d\omega\, K(\omega)I(\omega; t) = \frac{1}{2}(v(0)+v(t)), \tag{10}$$

where

$$v(t) = \int d\omega\, \bar{K}(\omega)\cos\omega t \tag{11}$$

is the cosine Fourier transform of the product $\bar{K}(\omega) = K(\omega)I(\omega)$ of the transmittancy and the initial spectra. If $I(\omega)$ is broad in comparison with $K(\omega)$, then $v(t)$ coincides with the Fourier transform of the transmittancy spectrum $K(\omega)$.

In a total accordance with the above-presented discussion $v(t)$ oscillates with the optical period $t_0 = 2\pi c/\omega_0$, while it takes a much longer time $t \sim 2\pi c/\delta \gg t_0$ for remarkable changes in the amplitude $a(t)$ of oscillations (and their period) to occur. If only the envelope of the amplitudes $a(t)$ (i. e. the visibility function) is measured, then, by performing the Fourier transform

$$K_c(\omega) = \frac{1}{2\pi} \int dt \; e^{-i(\omega_0 - \omega)t} a(t), \tag{12}$$

one can find the even combination of the spectrum $\bar{K}(\omega)$ and the shifted spectrum $\bar{K}(\omega - 2\Omega)$.

3. EXPERIMENTAL

Here we demonstrate experimentally the proposed interferometric method for the investigation of photochemically burned spectral holes. As a sample a polystyrene slab was used, which had been doped with two dyes: octaethylporphine and protoporphine (both 10^{-3}—10^{-4} mol/l). The inhomogeneously broadened 0—0 impurity absorption band at 1.8 K was 200 cm^{-1} fwhm and its maximum occurred at 619 nm. The optical density of the slab was approximately $D = 3.0$, the thickness $d \simeq 4$ mm and the working aperture in a liquid helium cryostat, $\Phi \simeq 10$ mm.

A picosecond cw Rhodamine 6G laser was used as the light source for both the burning of holes in the spectra and the detection of these spectra by the interferometric method. The duration, spectral width and repetition rate of the pulses were $\simeq 4$ ps, 4—5 cm^{-1} and 82 MHz, respectively.

The same Michelson interferometer was used for spectral hole burning and for spectral measurement. The input laser beam was divided by the interferometer into two parts, which were directed collinearly onto the sample. The optical path of the first beam could be changed with regard to the second one by a variable delay line with a step of 10 nm.

Two laser pulses with a time separation $t = 20$ ps were used for multifrequency hole burning in the transparency spectra of the sample. Coherent superposition of temporally not overlapping pulses is provided by the long phase coherence time, $T_2 \simeq 1.6$ ns [8] of the excited molecules. The temporal separation is Fourier transformed into the spectral structure of the burned hole. In particular, the width of the hole envelope is equal to the spectral width of the pulse. The transparency spectrum with burned holes in these cases is symmetric: $I(\omega) \approx I(\omega - 2\Omega)$ and in the weak burning case $I(\omega)$ is given by a simple formula, $K(\omega)J(\omega;\ t_1)$, where t_1 is the delay time of the interferometer shoulder (see [9] where this type of hole burning was also performed and studied). After hole burning the same sample was used for transmittancy measurements. To avoid additional hole burning during the measurement, the intensity of the light pulses was strongly ($\simeq 500$ times) reduced in comparison with the hole burning intensity.

Two different spectrum measurements by the interferometric method were performed. Spectral holes were burned in the spectrum with low (9 mJ/cm^2) and medium (42 mJ/cm^2) exposure levels with an exposure time of about 140 s. After the hole burning the same sample was used for transmittancy measurements. Several fine scannings of the delay line were performed during the delay time $t = 0$—50 ps. The light passed through the sample was recorded by a photomultiplier coupled to a photon counting system.

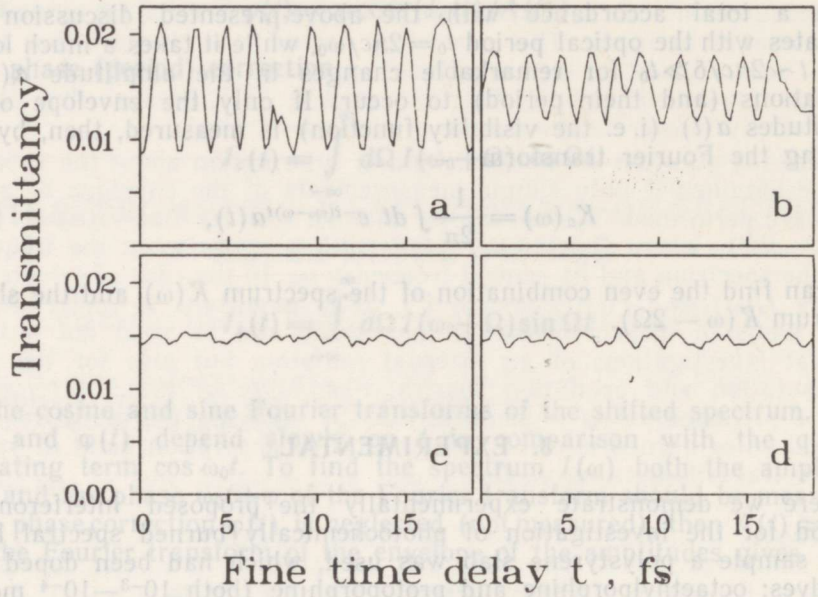

Fig. 1. Interferometric oscillations of the radiation passed through the sample at delay time values (*a*) $t_1=20$ ps, (*b*) $t_2=23$ ps, (*c*) $t_3=26$ ps, (*d*) $t_4=40$ ps. The sample was previously irradiated by two laser pulses with time separation 20 ps and exposure dose 42 mJ/cm².

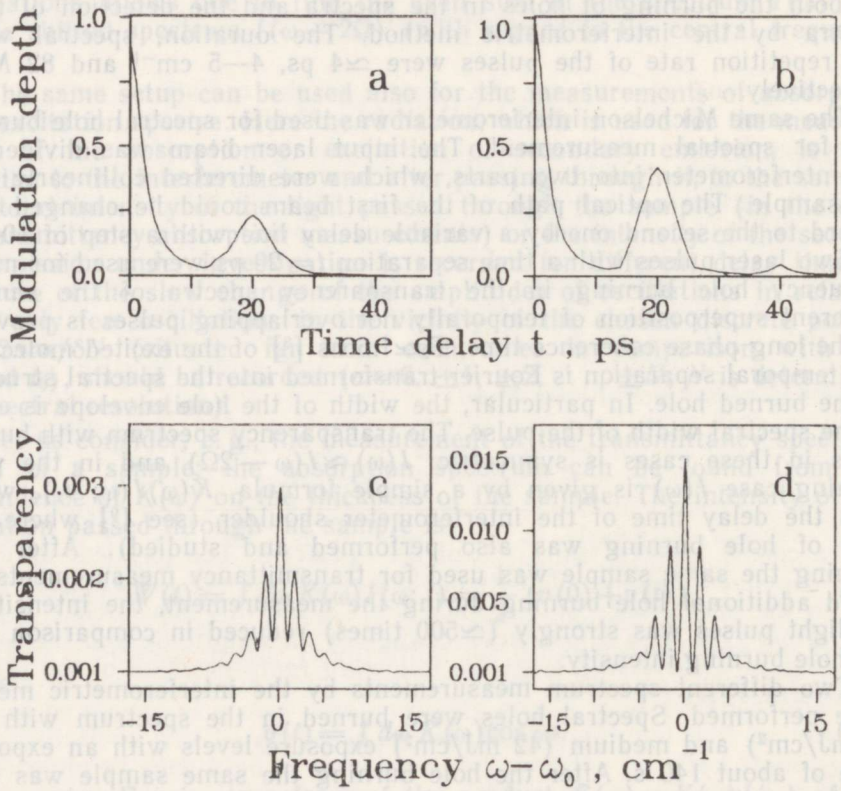

Fig. 2. The envelopes of the Fourier spectra in samples after the hole-burning dose (*a*) 9 mJ/cm² and (*b*) 42 mJ/cm², and the corresponding transparency spectra (*c*) and (*d*) calculated by the cosine Fourier transform.

Fig. 1 shows the measured oscillations at the delay time values $t_1=20$ ps, $t_2=23$ ps, $t_3=27$ ps and $t_4=40$ ps. In Figs. 2a, 2b the envelopes of the oscillation amplitudes are presented. In Figs. 2c, 2d the results of the cosine Fourier transforms of the measured envelopes of the amplitudes of oscillations are presented. The obtained spectra agree qualitatively well with the expected analogous spectra measured in [10] by the usual method. They both have a fine structure with a step of 1.65 cm^{-1}. The spectrum in Fig. 2d shows deeper minima, which corresponds to a stronger hole burning, performed by $\simeq 5$ times higher exposure dose (in comparison with the spectrum in Fig. 2c).

4. CONCLUDING REMARKS

In this work a simple interferometric method for spectral measurements in the visible is proposed. In the case of symmetrical spectra it is enough to record the envelope of interferometric amplitudes. The possibility of studying the fine structure of hole-burning spectra by using a Michelson-type interferometer and a cw picosecond dye laser is shown experimentally.

The proposed method has some advantages in comparison with the usual method of spectral investigations by spectrometers. Firstly, the equipment for these measurements is quite simple and cheap. Secondly, the signal-to-noise ratio can be higher, since a) all radiation is registered here, not only its single spectral component, b) a large aperture can be achieved in the measurements. Therefore this method can be useful for the investigation of weak secondary emission spectra in the case of laser monitoring of external objects (e. g. impurities of atmosphere).

Finally, we point out that if two-photon registration of radiation is possible (e. g. in the case of excitation by ultrashort pulses), then measurement can be extremely simplified. In this case the usual setup with a moving mirror can be used. There is no need to resolve interferometric oscillations: as the recording signal is $\sim I^2(t)$ (instead of $I(t)$), it is not averaged to zero even when the single interference oscillations are not resolved.

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REFERENCES

1. Tokunaga, E., Terasaki, A., Kobayashi, T. Opt. Lett., 1992, **17**, 16, 1131—1133.
2. Halbout, J.-M., Tang, C. L. Appl. Phys. Lett., 1982, **40**, 9, 765—767.
3. Cotter, D., Ironside, C. N., Ainslie, B. J., Girdlestone, H. P. Opt. Lett., 1989, **14**, 6, 317—319.
4. Finlayson, N., Banyai, W. C., Seaton, C. T., Stegeman, G. I. J. Opt. Soc. Am. B, 1989, **6**, 4, 675—684.
5. LaGasse, M. J., Anderson, K. K., Haus, H. A., Fujimoto, J. G. Appl. Phys. Lett., 1989, **54**, 21, 2068—2070.
6. Demtröder, W. Laser Spectroscopy. Springer, Berlin—Heidelberg—New York, 1982.
7. Chamberlain, J. The Principles of Interferometric Spectroscopy. John Wiley & Sons, Chichester—New York—Brisbane—Toronto, 1979.
8. Kaarli, R., Rätsep, M. Laser Physics, 1992, **2**, 4, 517—524.
9. Sõnajalg, H., Gorokhovskii, A., Kaarli, R., Palm, V., Rätsep, M., Saari, P. Opt. Comm., 1989, **71**, 6, 377—380.
10. Kikas, J., Kaarli, R., Rebane, A. Proc. Acad. Sci. ESSR. Phys. Math., 1984, **33**, 1, 124—127.

SPEKTRAALSÄLKUDEGA NEELDUMISSPEKTRI MÕÕTMINE INTERFEROMEETRILISEL MEETODIL

Vladimir HIŽNJAKOV, Margus RÄTSEP

On välja töötatud Fourier' spektroskoopiale sarnanev interferomeetriline meetod spektraalseteks mõõtmisteks spektri nähtavas piirkonnas. Meetodit on võimalik rakendada nii väliskiirguse kui ka neeldumis- ja sekundaarkiirguse spektraalseks uurimiseks. Sümmeetriliste spektrite korral on võimalik piirduda interferomeetriliste ostsillatsioonide mähisjoone mõõtmisega. Michelsoni tüüpi interferomeetrit ja kvaasipidevat pikosekundilist värvilaserit kasutades on uuritud lisandimolekuli neeldumisspektrisse spektraalsälkamise meetodil põletatud aukude peenstruktuuri lahutusega $\simeq$1,65 cm^{-1}.

ИЗМЕРЕНИЕ СПЕКТРАЛЬНЫХ ПРОВАЛОВ ИНТЕРФЕРЕНЦИОННЫМ МЕТОДОМ

Владимир ХИЖНЯКОВ, Маргус РЯТСЕП

Предложен простой интерференционный метод изучения оптических спектров в видимом диапазоне, являющийся упрощенным вариантом спектроскопии Фурье. Метод основан на измерении огибающей интерферометрических осцилляций и позволяет определить симметручную часть спектра. Метод может быть использован для изучения спектра внешней радиации, а также спектров поглощения и вторичного свечения. С помощью интерферометра Майкельсона и пикосекундного лазера на красителе измерен спектр многочастотных спектральных провалов (с шагом $\simeq$1,65 cm^{-1}), предварительно выжженных в примесной полосе поглощения полистирола.