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Three-channel wavelength modulation spectrometer*

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In this paper a three-channel wavelength modulation spectrometer is described. This spectrometer records simultaneously either the reflectivity and derivative reflectivity spectra or reflectivity and logarithmic derivative reflectivity spectra. The applied feedback loops permit to minimize errors caused by the differentiation of the optical system spectral characteristic. Errors caused by the dark current of the photomultiplier and the light noise are reduced by applying an additional third channel.

1. Introduction

In the recent years the derivative spectroscopy as a method of investigation of the band structure of solids has been developed. There are various techniques of obtaining the derivative spectra. Their advantages and disadvantages have been widely discussed elsewhere [1, 2]. To obtain the derivative spectra of reflectivity of solids the following methods are most often applied: electroreflectance, piezoreflectance, thermoreflectance and wavelength modulation. In all these methods, except the latter one, the modulation of the light beam is obtained by the application of a periodic perturbation directly to the sample. This perturbation alters the band structure or some other properties of the sample. The derivative spectrum obtained in this way depends strongly on the sample response to the perturbation. To interpret appropriately the obtained spectrum it is necessary to know the variation of the band structure as a function of the applied perturbation. However, the knowledge of such properties of solids is rather limited and therefore the derivative reflectivity spectra, obtained with the use of so-called internal modulation techniques, are difficult to interpret. Despite these difficulties, the internal modulation techniques are often employed. This is due to the fact that the derivative spectra may be obtained easier by employing the internal modulation, than by using the wavelength modulation, which is the external one. In other words, we do not introduce any changes into physical parameters of the sample but we modulate the incident light. This method does not destroy the state of the sample and the obtained spectrum is simply the first or the second derivative of the conventional spectrum. Wavelength modulation may be used to study the ef-

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fects associated with the changes of temperature, stress, magnetic or electric fields. However, unlike the other modulation techniques, the wavelength modulation method requires special and accurate construction of the optical system in order to eliminate a huge background in the derivative spectrum. This background results from the differentiation of the spectral characteristic of various optical components in the system and may be comparable or even larger that the sample signal. The separation of the sample spectrum from the background is a serious problem.

In wavelength modulation spectrometer described in this paper, the background can be minimized by the use of two feedback loops. Errors caused by the dark current and the light noise are reduced in the third channel applied in our spectrometer. The permanent reduction of the dark current and the light noise signals is especially important, when the supply of the photomultiplier is being controlled during the measurement procedure.

To obtain wavelength modulation of light, various methods have been proposed [3–8]. In our spectrometer we use a vibrating flat mirror inserted outside the monochromator.

In the next Sections of our paper a description of the optical and electronic system is given, the principles of operation of our spectrometer are discussed and some examples of recorded spectra presented.

2. Optical system

The wavelength modulation spectrometer has been built on the basis of the grating monochromator GDM-1000. The diagram of the optical part of our spectrometer is shown in Fig. 1. The entrance part of this system is analogous to that suggested by Zucca and Shen [8], except that they used a torsional tuning fork to make the mirror vibrate, while we have employed a loudspeaker specially constructed for this purpose. We took advantage of the fact that at the frequency of 1620 Hz the loudspeaker vibrating system gets into resonances. The external slit E performs the function of the entrance slit of the monochromator. The light emerging from the slit E is focussed by the spherical mirror Z_1 in the entrance slit plane of the monochromator. The mirror M (modulator) vibrating

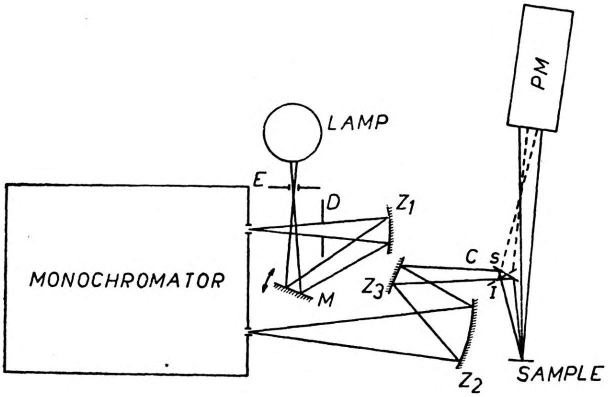

Fig. 1. Scheme of the optical system of the wavelength modulation spectrometer

with the frequency of 1620 Hz is inserted between E and Z_1. Thus, in the plane of the entrance slit of the monochromator we obtain a vibrating image of the slit E, so that the exit slit gives the wavelength-modulated light with the frequency equal to that of the modulator vibrations and with the modulation depth dependent on the amplitude of the modulator vibrations. Due to the use of the grating monochromator, the modulation depth is kept constant over the whole measuring range.

The monochromatic light beam with the modulated wavelength emerging from the exit slit of the monochromator is directed onto the beam chopper C by means of the mirrors Z_2 and Z_3. The beam chopper splits the measurement cycle into: sample part, reference part and sampling of the dark current, and the light noise signals. There are various ways by which the spectrometer channels may be constructed and the signals employed [8–14]. In our arrangement beam splitting is performed by the mirror C, which may be put into two positions: S and I. In position S (the sample channel) the light beam is focussed on the sample and the light reflected falls on the surface of the photodetector PM. In position I (the reference channel) the light beam is sent directly from the chopper to the photodetector. The period time during which the mirror C changes its position (and no light falls on the photodetector) is used to measure the signal of the dark current and light noise (the third channel). Due to the geometry of this system the images on the photodetector in the S and I positions are somewhat different in size. It should be emphasised that we have tested two ideas of the beam splitter construction. The first one was suggested by ZUCCA and SHEN [8], HART [10] and MORITANI et al. [11]. Here, an alternately reflecting or transmitting chopper was used. The latter, used in our arrangement, is similar to that proposed by THEIS and BUSSE [12]. In the first arrangement light beams passing through the reference and sample channels are reflected from different mirrors. This is a major disadvantage of this idea. Such a splitting technique requires the chopper and mirrors of very good quality. However, due to different rates of the ageing mirrors, the asymmetry of the channels is observed. These problems do not occur in the second arrangement, as the light beam reflected from the chopper mirror into the reference and sample channels does not encounter any additional mirrors after splitting. This method reveals, however, one disadvantage. The sizes of images on the photodetector are somewhat different. For the photomultiplier M12 FQ51, employed by us, error caused by the use of different mirrors appears to be greater than that caused by different sizes of images on the photocathode of the photomultiplier. Therefore, we applied the second method of beam splitting.

In our beam splitter the changes of the chopper mirror position are produced by torsional vibrations of the specially constructed mechanical system. The vibration frequency may be regulated in the range of 2–6 Hz. With our beam splitter the ratio of the measurement time in the sample or reference channel to the switching time may be regulated in the range of 3 : 1 to 8 : 1. Our beam splitter seems therefore to have better properties than the one proposed by Theis and Busse.

The moving diaphragm D [8] located in front of the monochromator is an important component of the optical system. Its task is to minimize the alternating signal generated as a result of the differentiation of the optical system characteristic (the modulation background). The signal generated by the additional modulation of the light beam on a sawtooth diaphragm *D* should have the same amplitude and opposite phase with respect to the modulation background. The minimization of the modulation background is especially important when sharply structured light sources are used.

3. Operation principles

On the one hand, the output signal in the reference channel without the modulation can be expressed as

$$U_I(\lambda) = I(\lambda)\, R_z(\lambda)\, Y(\lambda) + V_T, \tag{1}$$

where λ is the wavelength of the light, $I(\lambda)$ – the intensity of the light beam emerging from the exit slit of the monochromator, $R_z(\lambda)$ – the overall reflectivity of the mirrors in the optical system between the monochromator and the photomultiplier, $Y(\lambda)$ – the coefficient for converting light intensity into the final output electric signal, V_T – the signal proportional to the dark current and the light noise.

On the other hand, the output signal in the sample channel is given by

$$U_s(\lambda) = I(\lambda)\, R_z(\lambda)\, R(\lambda)\, Y(\lambda) + V_T, \tag{2}$$

where $R(\lambda)$ is the reflectivity of the sample.

If the wavelength modulation of light is given by

$$\lambda = \lambda_0 + \Delta\lambda \sin\omega t,$$

where $\Delta\lambda$ is the modulation depth, ω is the wavelength modulation frequency, and if we denote

$$U(\lambda) = I(\lambda)\, R_z(\lambda)\, Y(\lambda),$$

then the output signals from the photomultiplier in reference and sample channels can be expressed as

$$W_I = U(\lambda_0) + \left.\frac{dU}{d\lambda}\right|_{\lambda_0} \Delta\lambda \sin\omega t + V_T + A(\lambda_0)\sin(\omega t + \pi), \tag{3}$$

and

$$W_s = R(\lambda_0)\, U(\lambda_0) + U(\lambda_0) \left.\frac{dR}{d\lambda}\right|_{\lambda_0} \Delta\lambda \sin\omega t + R(\lambda_0) \left.\frac{dU}{d\lambda}\right|_{\lambda_0} \Delta\lambda \sin\omega t + V_T$$
$$+ R(\lambda_0)\, A(\lambda_0)\sin(\omega t + \pi), \tag{4}$$

respectively, where $A(\lambda)$ is the amplitude of the correction signal used to minimize the modulation background.

If we add the signal V_K to the signals W_I and W_s we obtain the signals V_I and V_s given by

$$\begin{aligned} V_I &= W_I + V_K, \\ V_s &= W_s + V_K. \end{aligned} \tag{5}$$

The signal V_K is generated in the third channel of the electronic system and fulfils the relation

$$V_K = -V_T. \tag{6}$$

If the electronic system ensures the fulfilment of the following relations:

$$\begin{aligned} U(\lambda_0) &= \text{const} = H, \\ V_s - R(\lambda_0)V_I &= U_{\text{out}1} \end{aligned} \tag{7}$$

the output signal is given by

$$U_{\text{out}\,1} = \alpha H \frac{dR}{d\lambda} \Delta\lambda, \tag{8}$$

where α and H are the constant coefficients determined in the process of calibration of the apparatus. If

$$R(\lambda_0)U(\lambda_0) = \text{const} = H$$

and (9)

$$V_s - R(\lambda_0)V_I = U_{\text{out}\,2}$$

we obtain the output signal given by

$$U_{\text{out}\,2} = \alpha H \frac{1}{R(\lambda_0)} \frac{dR}{d\lambda}. \tag{10}$$

Relations (8) and (10) give the quantities $\frac{dR}{d\lambda}$ and $\frac{1}{R}\frac{dR}{d\lambda}$, respectively.

4. Electronic system

The block diagram of the electronic system is shown in Fig. 2. The output signal from the photomultiplier *PM* is split into: sample channel, reference channel and correction (third) channel. In the sample and reference channels the respective signals V_s and V_I are sent to the reducer of the switching noise *RSN*. After the reduction of the switching noise the signals V_s and V_I are sent from the reducer *RSN* to the lock-in nanovoltmeter and measured with the use of the reference signal V_{ref} generated in the wavelength modulator *M*. The signal from the lock-in nanovoltmeter is recorded by the X-t recorder. In this way we record the derivative reflectivity spectra. However, it is also possible to record the conventional spectrum of the reflectivity using for this purpose the signal obtained in the block R_0. The block *SHV* which controls the high voltage power supply ensures the fulfilment of the relations $U(\lambda) = \text{const}$, and $U(\lambda)R(\lambda) = \text{const}$. In the third channel, equipped with the memory of the V_T signal, the correction signal V_K is obtained. In order to minimize the modulation background the *RMB* block has been constructed.

Its task is to generate signals which control the position of the diaphragm D in the light beam. The unit S ensures the signalling of faulty work of the electronic system in the case when the permissible values of V_s, V_I or V_T signals are exceeded.

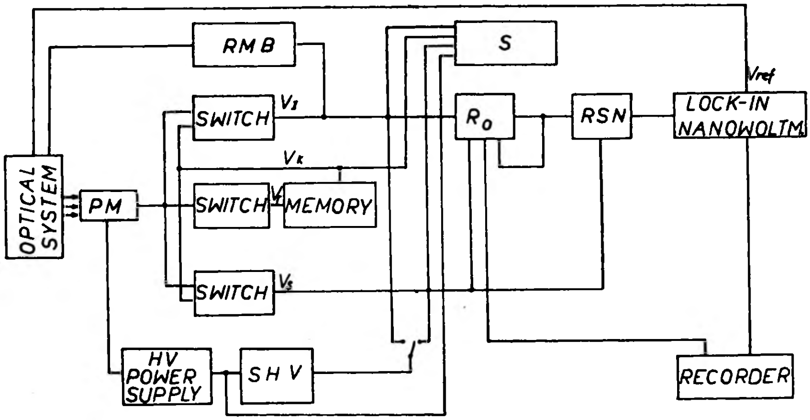

Fig. 2. Block diagram of the electronic system used in the wavelength modulation spectrometer

5. Examples of application

The experiments have been carried out in order to confirm the capabilities of our spectrometer. Conventional and derivative spectra of reflectivity in interference filter obtained by means of our spectrometer are shown in Fig. 3. The superiority of the derivative spectrum is out of question. The details which are hardly seen in the conventional spectrum (points A, B, in Fig. 3) are distinctly marked in the derivative spectrum. In the filter being investigated the essential changes in reflectivity occurred in the narrow interval of wavelength. So far, the wavelength modulation technique has been generally used to in-

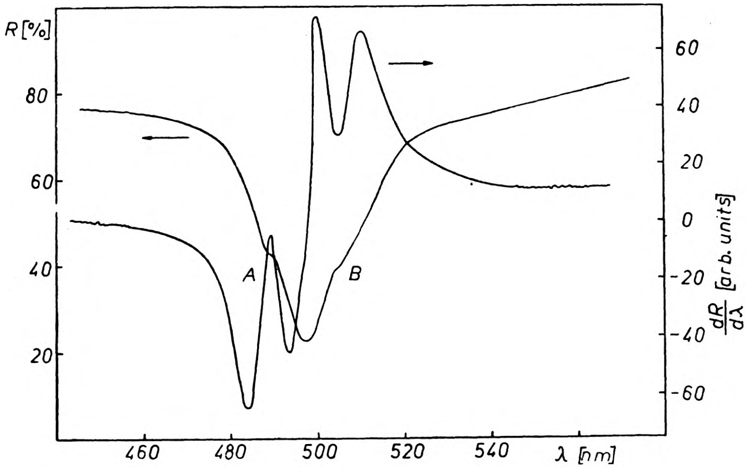

Fig. 3. Conventional and derivative reflectivity spectra of interference filter used to test the spectrometer

vestigate the solids with a sharp structure, such as semiconductors, especially at low temperatures. Our spectrometer has been tested for recording the reflectivity spectra of the solids the structure of which is not sharp. To obtain the derivative reflectivity spectra of these solids the internal modulation techniques have been most often used.

Figure 4 shows the derivative reflectivity spectrum $\frac{1}{R}\frac{dR}{d\lambda}$ of electrochemically deposited gold, recorded in the visible region. The measurement was done at room temperature. Figure 5 shows the reflectivity spectra of three copper samples of different age. Copper was evaporated on glass in vacuum. The samples prepared in this way were aged in air. The measurements were also done at room temperature. As seen in Fig. 5, the ageing of the probe results not only in the decrease of reflectivity over the whole spectral range, but also in the change of the shape of the curve $R(\lambda)$. For aged samples smaller changes of reflectivity $R(\lambda)$ are observed. These changes are better pronounced in the derivative spectra presented in Fig. 6.

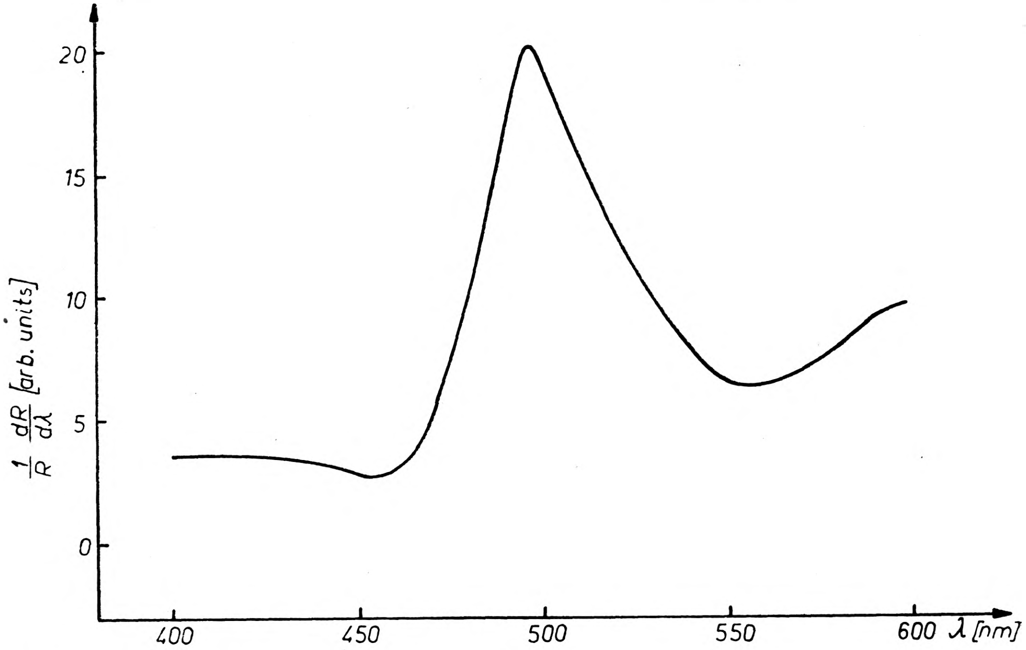

Fig. 4. Logarithmic derivative reflectivity spectrum of electrochemically deposited gold

5. Conclusions

In this paper we have presented three-channel wavelength modulation spectrometer which possesses the following essential features:

1. Errors caused by the dark current and light noise are reduced. This is achieved by the utilization of the switching time chopper mirror and due to the additional third-

channel equipped with memory of the dark current and light noise. To our knowledge this is an unparalleled way of permanent reduction of the errors caused by the dark current and the light noise.

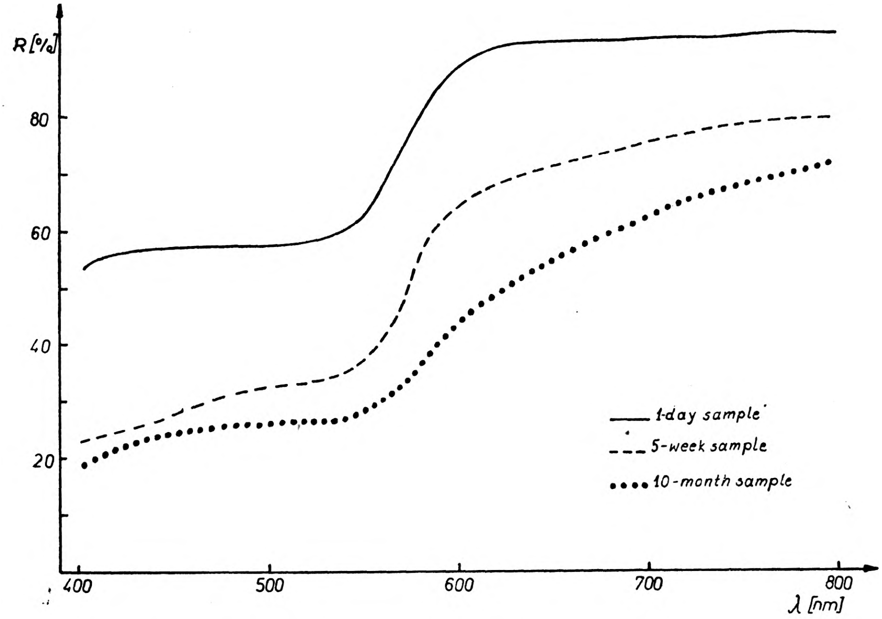

Fig. 5. Reflectivity spectra of the copper samples of different ages

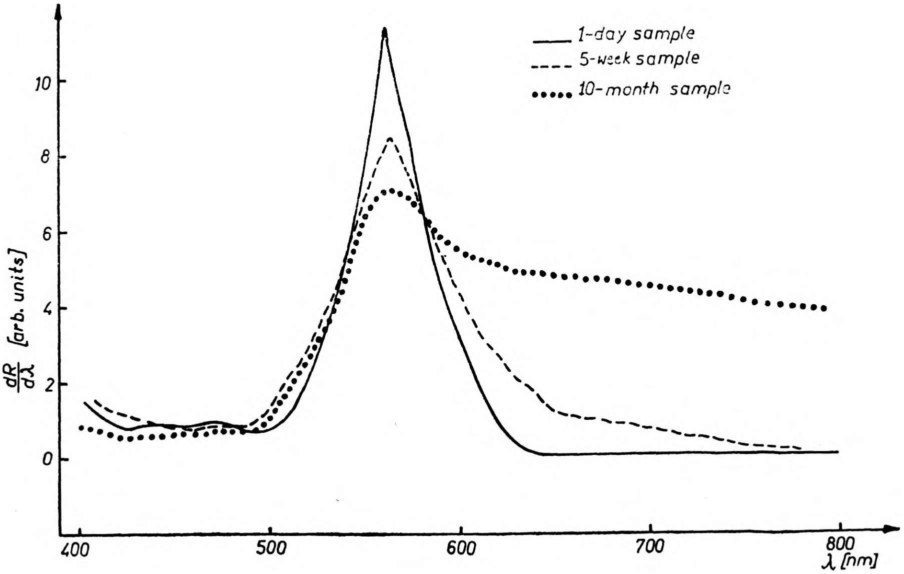

Fig. 6. Derivative reflectivity spectra of the copper samples of different ages

2. High frequency of modulation is applied. To our knowledge the employed frequency of 1620 Hz is now the highest frequency of wavelength modulation ever used. High frequency of wavelength modulation (also used by Zucca and Shen [8]) permits to achieve more advantageous signal to noise ratio than in the case of low frequencies.
3. Large modulation background is minimized.
4. Modulation depth is constant over the whole measuring range.

This spectrometer permits to record the derivative reflectivity spectra of solids, which do not have a sharp structure. Even if the signal is very weak, due to little changes of reflectivity and small modulation depth, it can be separated from noise.

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Трёхканальный спектрометр с модуляцией длины волны

В настоящей работе описан трёхканальный спектрометр с модуляцией длины волны. Он одновременно регистрирует спектры коэффициента отражения и производной коэффициента отражения или спектра коэффициента отражения и логарифмической производной коэффициента отражения. Применение в этом спектрометре петли обратных связей позволило минимизировать ошибки, вызванные дифференцированием спектральной характеристики оптической системы. Ошибки, вызванные темновым током фотоумножителя и сетевым шумом уменьшаются посредством применения дополнительного, третьего канала.