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HOLOGRAPHIC SPECTROSCOPY

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THE groundwork for the method of Fourier spectroscopy was laid already by the works of Michelson. But the greatest progress in this method was reached after the publication of the papers of Fellgett[1], Jacquinot[2], Strong[3], and J. and P. Connes[4,5], when it became possible to effect Fourier transformations with the aid of the electronic digital computer. However, the use of expensive computers, and also the presence of a large time interval between the registration of the spectrum and its reconstruction, have prevented extensive use of the Fourier-spectroscopy method. To simplify it, specialized analog computers were constructed.

At the same time, the method of photographic registration of the interference pattern is also used. The interference pattern is projected in the plane of the photographic plate and is photographed. The obtained image is then subjected to microphotometric analysis and subsequent numerical reduction[11].

In[12,13], a spectrum analyzer making it possible to obtain the spectrum directly without recording the interference pattern was proposed. In addition, the effect of the displacement of the interferometer mirrors was eliminated.

The old methods of reconstructing the spectrum gave way to holographic spectroscopy[6,14]. The gist of the method consists in the following. During the first stage one records the spectral hologram, and in the second stage the investigated spectrum is reconstructed. This method makes it possible to register the spectrum relatively simply and requires the use of electronic computers for its reconstruction.

1. PRINCIPLE OF HOLOGRAPHIC SPECTROSCOPY

The idea of this method[10] serves as the basis of a holographic spectroscope, in which the hologram plays simultaneously the role of the dispersive and focusing elements.

The hologram H is registered in the following manner (Fig. 1a). The object is a very narrow slit F_1 parallel to H; the reference wave comes from a point source S_0 located in the symmetry plane P of the slit F_1.

A laser emitting a monochromatic light of wavelength λ illuminates F_1 and S_0 simultaneously. After photographic processing, the hologram H occupies its original position relative to S_0, and F_1 is removed. If the light coming from S_0 now includes several wavelengths, then the hologram reconstructs as many different images F_1' of the slit F_1, one for each wavelength λ'.

Let us examine the process of localization of the image of the slit for each wavelength. We place the axis Ox $\perp$ H, and the Oy axis on the intersection of the planes P and H (see Fig. 1a). We take the points S_0, S_1 and S_1', which lie in the symmetry plane P. Each of these points is determined by its polar coordinates (Fig. 1b) OS = R $> 0, -\pi/2 < \theta < \pi/2$. The waves emitted by these points and arriving at the point M(0, y) of the Oy axis can be written in the form

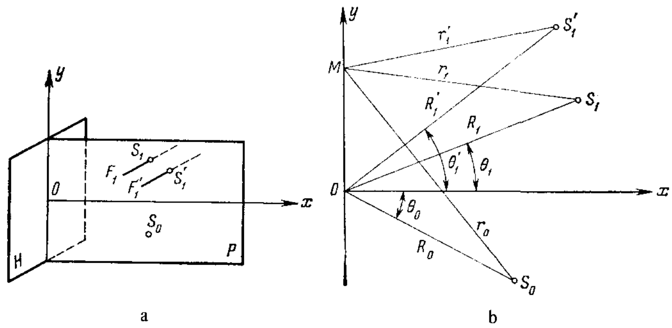

FIG. 1. Hologram registration scheme [10]. H—hologram; F_1—slit parallel to H; S_0—point source from which the reference wave comes; P—symmetry plane; F_1'—image of slit F_1 for the wavelength λ'; S_0, S_1, and S_1'—points lying in the symmetry plane P, OS = R > 0, $-\pi/2 < \theta < \pi/2$, M—observation point.

$$S = a \exp [ik(r - R)],$$

where k = $2\pi/\lambda$ and r = MS.

Since $y/R \ll 1$, r − R can be expanded in a series

$$r - R \approx -y \sin\theta + \frac{y^2}{2R} \cos^2\theta. \quad (1)$$

Comparing the phases of the two waves and putting λ/λ' = u, we obtain from (1)

$$\sin\theta_1' = u(\sin\theta_1 - \sin\theta_0) + \sin\theta_0, \quad (2)$$

$$\frac{\cos^2\theta_1'}{R_1'} = u\left(\frac{\cos^2\theta_1}{R_1} - \frac{\cos^2\theta_0}{R_0}\right) + \frac{\cos^2\theta_0}{R}. \quad (3)$$

These relations determine completely the position of the point S_1' corresponding to a radiation λ'. From formula (2) we can obtain an equation for the dispersion:

$$\frac{d\theta_1'}{d\lambda'} = \frac{\sin\theta_1 - \sin\theta_0}{\lambda\cos\theta_1'}. \quad (4)$$

The dispersion $d\theta_1'/d\lambda'$ does not depend on R_0, R_1, or R_1', and is the higher the larger $|\theta_1 - \theta_0|$. If it is desirable to obtain the spectrum almost perpendicular to the Ox axis, it is necessary to choose $\theta_1 = 0$ for $\lambda = 0.50\ \mu$. Then

$$\frac{d\theta_1'}{d\lambda'} = -\frac{\sin\theta_0}{\lambda}.$$

For $\theta_0 = -45°$ and $\lambda = 0.50\ \mu$ we have $d\theta_1'/d\lambda' = 1.4 \times 10^{-4}$ rad/Å or approximately 0.5 min/Å. Under favorable conditions, such an instrument makes it possible to separate two lines at a distance on the order of 2 Å.

During the process of construction of the spectroscope, the pointlike source S_0 was replaced, from light-

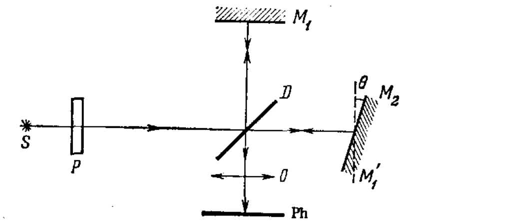

FIG. 2. Diagram of Michelson interferometer. S—source; P—diffuse scatterer; M_1, M_2—mirrors; D—beam splitter, O—output lens, Ph—hologram; θ—angle between M_2 and the image M_1' of mirror M_1.

strength considerations, by a slit source parallel to F_1. A hologram of very small dimensions (25 mm^2) and an adjustable slit source were mounted on the ends of a tube 25 cm long. By placing a strongly stopped-down photographic camera behind the hologram, it was possible to resolve the yellow doublet of sodium (5890 and 5896 Å). The resolving power of the spectroscope[10] is estimated at approximately 1 Å. It is important that the light strength of the holographic spectroscope can be increased by using bleached holograms.

An undesirable factor in the use of this method is the excessive difference between the intensities of the two interfering beams, which reduces the light intensity. This shortcoming can be overcome by the method described in the next section, and also by other methods of obtaining coherent point sources.

2. HOLOGRAPHIC SPECTROSCOPY USING INTERFEROMETERS

If the screen with two openings is replaced by a Michelson interferometer adjusted in such a way that both images of the source at the exit overlap fully, and then one of the mirrors is inclined until fringes of equal thickness appear, then in this case we again realize the method of holographic spectroscopy.

Such a method has an advantage over the preceding one since the use of beam splitters with special coating makes it possible to greatly increase the fraction of the effective light flux by equalizing the intensities of both beams.

Such an assumption is valid for an ideal beam splitter, when the amplitudes of the reflected and refracted rays are equal.

In[6], a Michelson interferometer was used for this purpose* (Fig. 2). The mirrors in the interferometer were placed at an angle other than a right angle to the optical axis, in such a way that the wave fronts reflected from each of the mirrors were at a small angle θ. The photographic plate registered a system of equal-thickness fringes, produced in the air gap between one of the mirrors and the image of the other†. The interferometer wedge gave 30 fringes/mm. The light source was a mercury lamp and a diffuse scatterer was used to illuminate the surface of the mirror uniformly.

We present the derivation of the equation of the system of fringes in the plane of the photographic plate. If E(x, y, z) is the electric vector of a plane wave inclined at an angle $\theta/2$ to the z axis of the interferometer, then the field in the (x, y) plane, which is perpendicular to the z axis, is described as follows:

$$E(x, y) = E_0 \exp\left[2\pi i\nu x \sin\frac{\theta}{2} + \omega t\right]. \quad (5)$$

In this expression $\nu = 1/\lambda$ cm^{-1} and $\omega = 2\pi c\nu$, where c is the velocity of light. We assume now that a second plane wave of the same wavelength λ, incident at an angle $(-\theta/2)$ to the z axis, produces in the same plane a field

$$E(x, y) = E_1 \exp\left[2\pi i\nu x \sin\left(-\frac{\theta}{2}\right) + \omega t + \varphi\right], \quad (6)$$

where φ is the phase difference. A photographic plate placed in the (x, y) plane registers the intensity of the interference pattern:

$$[I(x, y)]_\lambda = \frac{1}{2}E_0^2 + \frac{1}{2}E_1^2 + E_1E_0\cos\left[2\pi\nu x\sin\frac{\theta}{2} - \varphi\right]. \quad (7)$$

We assume that the interferometer is adjusted in such a way that $\varphi = 0$, and also $E_0^2 = E_1^2 = I(\nu)$, where $I(\nu)$ is the intensity of the source with wave number ν. We shall assume that θ is a small angle. Then expression (7) takes the form

$$[I(x, y)]_\lambda = I(\nu)\,[1 + \cos 2\pi\nu\theta x]. \quad (8)$$

Because of the incoherence between the spectral lines of the source, the hologram obtained from a source with a complex spectrum will have the following form:

$$I(x) = \int_0^\infty I(\nu)\,[1 + \cos 2\pi\nu\theta x]\,d\nu. \quad (9)$$

It is known that the amplitude H(x) of the radiation passing through the hologram with intensity I(x) is

$$H(x) \approx [I(x)]^{-\gamma/2}, \quad (10)$$

where γ is the contrast coefficient of the photographic plate.

In the case when the spectrum is a set of n narrow spectral lines separated by sufficiently large distances, $I(\nu)$ can be represented in the form

$$I(\nu) = I_1\delta(\nu - \nu_1) + I_2\delta(\nu - \nu_2) + \ldots + I_n\delta(\nu - \nu_n).$$

Then substitution of this expression in (9) and (10) yields

$$H(x) = \left[\sum_{i=1}^{n} I_i\,(1 + \cos 2\pi\nu_i\theta x)\right]^{-\gamma/2}. \quad (11)$$

If $\gamma < 2$, then approximately

$$H(x) \approx \sum_{i=1}^{n} I_i - \frac{\gamma}{2}\sum_{i=1}^{n} I_i\cos 2\pi\nu_i\theta x. \quad (12)$$

From this we find that the variable components observed as a result of analysis of the structure of the reconstructed hologram H(x) are proportional to the initial intensities I_i and to the hologram contrast coefficient γ, while the spatial frequencies are $N_i = \nu_i\theta = \theta/\lambda_i$.

In[6] the spectrum was obtained by illuminating the holographic Fourier interference pattern with a laser beam of wavelength 6328 Å. Five mercury lines were separated in the wavelength band from 4047 to 6234 Å. The resolution was 50 Å.

*It is possible to use for analogous purposes optical systems of interferometers with compensated optical paths, for example with a triangular ray path [7].

†When an objective is used to project the interference pattern, it is necessary to take into account the bandwidth of the spatial frequencies of such an objective.

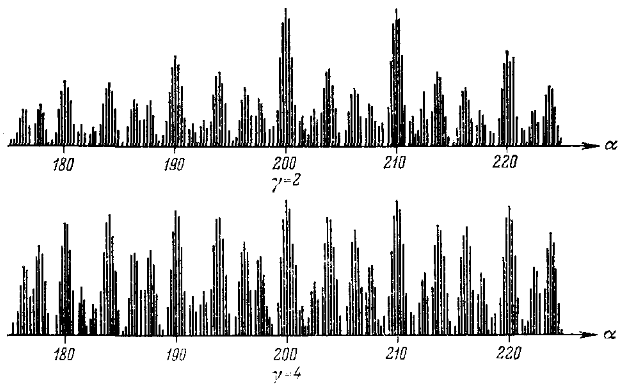

FIG. 3. Dependence of the relative intensity of the fundamental and combination frequencies on the contrast coefficient γ of the photographic plate. The calculation is based on the formula

$$G(\Delta t_k) = \left\{\sum_{n}^{n_1} E_n (1+\cos\omega_n \Delta t_k)\right\}^{-\gamma/2}, \quad k = 0, 1, 2, \ldots, N,$$

In the particular case for $E_n = 1$ and $\gamma = 2$ and 4. The parameter $\alpha = \omega/(\Delta\omega)_0$, where $(\Delta\omega)_0$ is the resolution limit.

3. COMBINATION FREQUENCIES

When the holographic method was used to investigate the spectra of mercury and cadmium lamps[7] it was noted that false images appeared in the reconstructed hologram. The author of[8] interpreted this phenomenon as the appearance of "ghosts" with combination spatial frequencies. It is obvious that upon subsequent photometry these "ghosts" can greatly distort the spectrum and lower the resolution. We shall therefore dwell in greater detail on the nature of the "ghosts."

We write down expression (10), called the hologram equation, for a finite number of terms of the Fourier series:

$$H(x) \approx [I(x)]^{-\gamma/2} = K\left\{\sum_{i=1}^{n} I_i(\nu)\,[1+\cos 2\pi\nu_i\delta]\right\}^{-\gamma/2}, \tag{13}$$

where δ is the path difference.

We expand (13) in series and retain the first four terms:

$$K\left\{\sum_{i=1}^{n} I_i(\nu)\,[1+\cos 2\pi\nu_i\delta]\right\}^{-\gamma/2} =$$
$$= K\left\{\sum_{i=1}^{n} I_i(\nu) - \frac{\gamma}{2}\sum_{i=1}^{n} I_i(\nu)\cos 2\pi\nu_i\delta + \frac{1}{2}\frac{\gamma}{2}\left(\frac{\gamma}{2}+1\right)\sum_{i=1}^{n}\sum_{k=1}^{n} I_i(\nu) I_k(\nu)\right.$$
$$\times \cos 2\pi\nu_i\delta\cos 2\pi\nu_k\delta - \frac{1}{6}\frac{\gamma}{2}\left(\frac{\gamma}{2}+1\right)\left(\frac{\gamma}{2}+2\right)\sum_{i=1}^{n}\sum_{k=1}^{n}\sum_{l=1}^{n} I_i(\nu) I_k(\nu) I_l(\nu)$$
$$\left.\times \cos 2\pi\nu_i\delta\cos 2\pi\nu_k\delta\cos 2\pi\nu_l\delta\right\} + \ldots,$$

$$H(\delta) \approx K\left\{\sum_{i=1}^{n} I_i(\nu) - \frac{\gamma}{2}\sum_{i=1}^{n} I_i(\nu)\cos 2\pi\nu_i\delta +\right.$$
$$+\frac{\gamma}{8}\left(\frac{\gamma}{2}+1\right)\sum_{i=1}^{n}\sum_{k=1}^{n} I_i(\nu) I_k(\nu)\,[\cos 2\pi\delta(\nu_i+\nu_k) + \cos 2\pi\delta(\nu_i-\nu_k)] -$$
$$-\frac{\gamma}{48}\left(\frac{\gamma}{2}+1\right)\left(\frac{\gamma}{2}+2\right)\sum_{i=1}^{n}\sum_{k=1}^{n}\sum_{l=1}^{n} I_i(\nu) I_k(\nu) I_l(\nu)\,[\cos 2\pi\delta(\nu_i+\nu_k+\nu_l)$$
$$\left.+\cos 2\pi\delta(\nu_i+\nu_k-\nu_l) + \cos 2\pi\delta(\nu_k+\nu_l-\nu_i) + \cos 2\pi\delta(\nu_l+\nu_i-\nu_k)]\right\} + \ldots \tag{14}$$

In this expression the first term corresponds to the dc component; the second term describes the intensity of the frequencies of the spectrum, and the third term corresponds to the intensity of the "ghosts."

Assume that we investigate a very weak emission in which the intensity of some one frequency ν_k is much stronger than all the remaining frequencies ν_i. Then the intensity of the "ghosts" corresponding to the sum $(\nu_k + \nu_i)$ and the difference $(\nu_k - \nu_i)$ of this frequency and of all the remaining frequencies of the spectrum will exceed the intensities not only of the remaining "ghosts," but also of the fundamental frequencies. In particular, it is possible to use a laser to produce such a powerful radiation at the frequency ν_k.

It is much more convenient to use for the analysis of this phenomenon simulation of the process of obtaining spectra with the aid of a computer. Thus, in particular, if we have two spectral lines with equal intensities and frequencies $\omega_1 = 200$ and $\omega_2 = 210$, then at $\gamma = 2$ and 4 (Fig. 3) we can predict the intensities of all the component frequencies.

The foregoing results have shown that in the method of holographic spectroscopy it is necessary to pay special attention to the choice of photographic materials and the method of processing them. On the other hand, particular interest attaches to making good use of the nonlinear process of obtaining holograms with $\gamma \geq 1$ and accordingly to registration of the spectra at the combination spatial frequencies. This effect is analogous to the phenomenon of Raman scattering in matter.

In similar fashion, it is also possible to use for spectroscopic purposes the effect of the nonlinearity of the refractive index of certain crystals to obtain frequency multiplication by illumination with light from a laser and the investigated source. This phenomenon has already been used earlier to convert laser radiation to other frequencies[15].

4. CONCLUSION

For practical utilization of the method in question, it is, as usual, necessary to find the form of the apparatus function. Such a function was derived in[16]. It was found to coincide in form with the function obtained in ordinary Fourier spectroscopy[14]. From this it is possible to determine beforehand the angle between the wave fronts, *starting from the requirement that the resolving power* R, the cross section of the beam L, and the angle between the interfering fronts α should satisfy the relation

$$R = \frac{2L\alpha}{\lambda}. \tag{15}$$

In this case when the interferometer is illuminated with light of wavelength λ, the number of equal-thickness fringes located on both sides of the "zeroth" fringe is equal to R. The permissible angular dimensions of the source for a given R should satisfy the relation

$$\Omega \leq \frac{2\pi}{\nu\delta_m} = \frac{4\pi}{R},$$

where δ_m is the maximum path difference between the interfering fronts, equal to $\delta_m = (L/2)\cos\alpha$.

The method can be useful for the registration of rapidly occurring processes and sources of weak glow in the spectral region where photographic materials can be used.

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