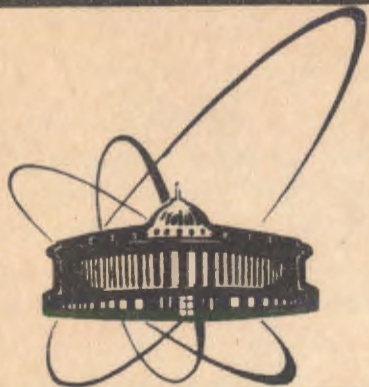


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TUNABLE IR SYNCHROTRON LIGHT SOURCE
OF HIGH LUMINOSITY FOR SPECTROSCOPIC
INVESTIGATIONS OF HTSC MATERIALS

1991

Middle and far range infrared spectroscopy has achieved good progress using Fourier interferometers and "black body" radiation sources of temperature $T \sim 1000 \dots 1500$ K and radiation power $P = 10^{-8}$ W in the region $\Delta\lambda/\lambda = 0.1$. Further increase in the efficiency of spectroscopic investigations depends on development of highly intensive radiation sources for the range $\lambda \geq 100 \mu\text{m}$.

These sources can be cyclic electron accelerators (synchrotron radiation sources (SR)), and sources of variable magnetic structure (free-electron lasers (FEL)) [1]. The difference between a synchrotron radiation source and a "black body" source used for infrared spectroscopy in the far IR range is that spectral density of the former decreases as λ^{-1} while for the latter it is λ^{-3} [2]. The SRS radiation power was measured in the long wave region with the Brookhaven NSLS source [1]. The results showed that in the spectral range $\lambda > 100 \mu\text{m}$ the power ratio of the synchrotron radiation to the black body radiation is $P_{\text{SL}}/P_{\text{b.b.}} = 10$ [3]. In specialised storage systems the radiation is mainly in the soft X-ray range and only a small part of it is in the infrared range. The SR spectrum is characterised with a parameter $\lambda_c = 4.2R/\gamma^3$, where R is the electron ring radius, γ is the relativistic factor. For the NSLS storage ring one has $R = 2.2$ m, $\lambda_c = 59$ Å [3]. To estimate the radiation power in the far IR range one uses the asymptotic formula ($\lambda > \lambda_c$) [1]

$$N_\lambda \approx 5.27 \cdot I_e (\text{A}) \cdot E (\text{Gev}) \cdot \left(\frac{\lambda_c}{\lambda} \right)^{1/3} \cdot \frac{\Delta\lambda}{\lambda}; \quad (1)$$

where I_e (A) is the electron ring current in amperes, E is the energy in GeV. Higher SR power in the infrared range is possible if one has 1) a larger horizontal aperture of the optical channel, 2) a higher current accumulated in the ring.

The available maximum values of these parameters at the NSLS [3] are

$$\begin{aligned} \epsilon_x &= 90 \text{ mrad}, I_e = 750 \text{ mA}, \\ \epsilon_z &= 90 \text{ mrad}. \end{aligned}$$

The optimum source (with high pulsed luminosity of SR for the infrared range) is a storage ring with a high current.

Substituting $I_e = \frac{N_e e c}{2\pi R}$, $\lambda_c = R/E^3$ in formula (1) we get

$$N(\lambda) = c \cdot \frac{\Delta\lambda}{\lambda} \cdot \frac{N_e}{R^{2/3}}. \quad (2)$$

Formula (2) shows that the best radiator for the IR range is an electron ring with the minimal radius R and the maximal number of electrons. As shown in ref.[4], a pulsed electron accumulator is best suited for SR generation in a submillimetre range. The parameters of the electron ring for the pulsed accumulator are:

$$R = 3-4 \text{ cm}, E_e = 20-25 \text{ MeV}, N_e = 5 \cdot 10^{12}, I_e = 1200-1300 \text{ A}. [5]$$

With these parameters, the radiation power gain in the long-wave range, which depends on the electron current ratio, is ~ 2000 .

In ref.[4] they propose to use a pulsed electron accelerator for submillimetre spectroscopy of high- T_c superconductors, namely for energy gap measurements by film transmission spectra. Ref.[6] was the first to present the results of film transmission studies with $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ on a MgO support in a synchrotron radiation beam. A fast scanning Michelson interferometer was used for the spectral range $r = (50-500) \text{ cm}^{-1}$. The measurement interval for an entire spectrum was $\sim 3 \text{ min}$, the instrumental resolution was $\Delta r = 1 \text{ cm}^{-1}$, $(\lambda/\Delta\lambda) = 100$. These experimental results show a high efficiency of the synchrotron radiation in submillimetre spectroscopy.

Before using a pulsed electron accumulator as a source for submillimetre spectroscopy it is necessary to measure the absolute power in a long wave range $\lambda \geq 100 \text{ }\mu\text{m}$, where interference effects are possible for the radiation

wavelength is comparable with the linear dimensions of an electron density grain.

The absolute power was also measured at $T = 4.2 \text{ K}$ by an InSb detector with a set of narrow-band interference filters. The radiation power measured for the vertical acceptance $\theta_z = 30 \text{ mrad}$ and the wavelength $\lambda = 300 \text{ }\mu\text{m}$ was $\Delta\lambda/\lambda = 0.1$ $P(\lambda) = 1.5 \cdot 10^{-5} \text{ W}$. It is $5 \cdot 10^{-4} \text{ W}/\mu\text{m}$ in terms of the total power of radiation from the ring, the expected value of N_e is $2 \cdot 10^{12}$, $P(\lambda) = 6 \cdot 10^{-4} \text{ W}/\mu\text{m}$, $\lambda = 300 \text{ }\mu\text{m}$. The linear relation of the radiation power and the number of electrons is observed for any number of electrons in its variation range.

To increase the acceptance of the optical channel we developed a multimirror line with spherical mirrors. The experiments showed that it transferred about 5% of the total synchrotron radiation power to the length $L = 200 \text{ cm}$. Test bed investigations at the transport length $L = 20 - 30 \text{ cm}$ yielded the axial acceptance values about $\theta_\phi \approx 1000 \text{ mrad}$. The measurements in this optical system showed that the vertical acceptance θ_z allowed all radiation to be collected; the value of the acceptance is $\theta_z = 120 \text{ mrad}$ for $\lambda = 300 \text{ }\mu\text{m}$.

With a multimirror optical channel of length $L = 30 \text{ cm}$ the pulsed SR power is expected to be $P = 6 \cdot 10^{-3} \text{ W}$ on the sample $P(\lambda) = (2-3) \cdot 10^{-4} \text{ W}/\mu\text{m}$ in the band $\Delta\lambda/\lambda = 0.1$ at $\lambda = 300 \text{ }\mu\text{m}$. The time duration of the radiation depends on the modes of electron ring formation with the (R, Z) trajectory and is $\tau = (10^{-4} - 10^{-2}) \text{ s}$; the repetition frequency is $f = 1 \text{ Hz}$.

The above results allow a conclusion that a pulsed electron accumulator with the parameters $N_e = 5 \cdot 10^{12}$, $R = 3 \text{ cm}$, $E = 20-25 \text{ MeV}$ makes it possible to shape submillimetre radiation beams in the range of $\lambda > 100 \text{ }\mu\text{m}$ with a record pulsed power $P = (5-6) \cdot 10^{-3} \text{ W}$ in the band $\Delta\lambda/\lambda = 0.1$.

Spectroscopic investigations with this pulsed SR beam involve an important task of creating a pulsed spectrometer. For this purpose we are now upgrading an infrared monochromator with diffraction gratings in order to extend the spectrum range to $700 \text{ }\mu\text{m}$. The detector is an

optic-acoustic receiver (OAR) with a mylar window and a receiver based on Ge:Ga admixture conductance at helium temperature. If a diffraction monochromator is used, the transmission spectrum taking time, e.g. from 50 - 300 μ s, with $\Delta\lambda/\lambda = 10^{-2}$ will correspond to 10^4 pulses, or about 3 hours at the frequency $f = 1$ Hz, while for $\Delta\lambda/\lambda = 10^{-1}$ $t = 30$ min.

To study HTSC materials for the effects that manifest themselves as smooth dependence of transmission or reflection on the wave-length, one can employ the fact that during formation of the electron ring in the pulsed accumulator the short-wave boundary of the SR spectrum moves towards shorter waves, i.e. time evolution of the spectrum takes place. A typical scale of time evolution for a typical compression mode depends on the variation of the ring parameters $R = R(t)$, $E = E(t)$ and, consequently, $\lambda_c = \lambda_c(t)$ (see Fig.1).

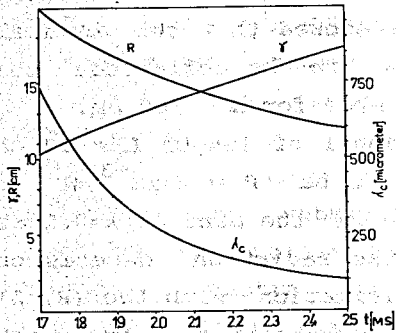


Fig.1. Variations of the electron ring radius, electron energy, and critical wavelength of the SR spectrum in the chosen time range of the compression cycle.

The prompt SR power spectrum of an electron ring $P(\lambda)$ can be described by the following formula [7]:

$$P(\lambda) = \frac{3^{5/2}}{16\pi^2} \cdot \left(\frac{e^2 c}{R^3}\right) \cdot \gamma^7 \cdot \left(\frac{\lambda_c}{\lambda}\right)^3 \cdot \int_0^{\infty} K_{5/3}(x) \cdot dx ; \quad (3)$$

where R is ring radius; γ is the gamma factor ($\gamma = E_e/mc^2$); e , m , E are the charge, mass and energy of electron; λ_c is the critical wavelength defined as $\lambda_c = 4\pi R/\gamma^3$; $K_{5/3}(x)$ is the Macdonald function of a real argument. The distribution maximum is on the wavelength $\lambda_m = 0.42\lambda_c$.

For practical calculations we isolated a universal power function from expression (1):

$$f(y) = y^3 \cdot \int_y^{\infty} K_{5/3}(\alpha) \cdot d\alpha ; \quad y = \lambda_c/\lambda . \quad (4)$$

Calculations of the SR power spectra at different time moments of the electron ring compression cycle are shown in Fig.2.

If the SR flow is measured by a detector with the time constant much smaller than the chosen time interval and with the spectral sensitivity $D_0(\lambda)$, the time function of the detector current can be represented in the following form:

$$I_0(t) = \int_0^{\infty} P[t, \lambda_c(t), \lambda] \cdot D_0(\lambda) \cdot d\lambda . \quad (5)$$

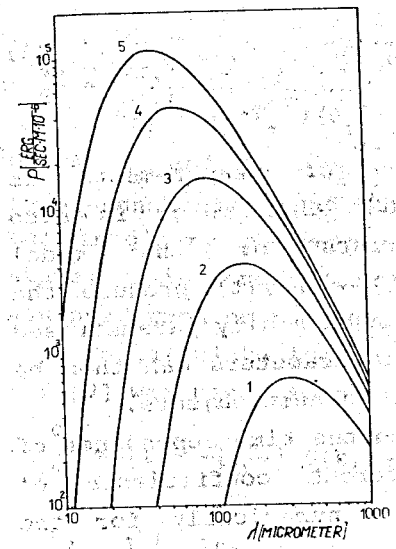


Fig.2. Prompt SR power spectra at different moments of time: (1)- $t = 1.7$ ms, (2)- $t = 1.9$ ms, (3)- $t = 2.1$ ms, (4)- $t = 2.3$ ms, (5)- $t = 2.5$ ms.

Practically, one can integrate from the lower limit $\lambda_1 = K\lambda_c$, where K is a constant of very small value (e.g. for $K = 0.05$ $P(\lambda_1, \lambda_c) = 10^{-6}P(\lambda_c, \lambda_m)$), to the upper limit λ_2 , where the detector sensitivity is negligible ($D(\lambda_2) \ll D(\lambda_{max})$).

Placing the object of interest in front of the detector, we find that the penetration factor of the object $T(\lambda)$ is connected with the time function of the detector signal $I_0(t)$ through an integral equation:

$$I_s(t) = \int_{\lambda} P[t, \lambda_c(t), \lambda] \cdot D_s(\lambda) \cdot T(\lambda) \cdot d\lambda ; \quad (6)$$

which can easily be reduced to an integral Voltair equation of the first kind.

Yet, these measurements are difficult to perform because the total radiation power in the ring compression mode is a rapidly increasing time function. Thus, special equipment with a large dynamic range ($\sim 10^5$) is necessary, and the reconstruction accuracy of the sought-for function $T(\lambda)$ becomes lower. The situation changes for the better if two channels are used simultaneously for SR flow measurement and the ratio of the detector signals $I_s(t)/I_o(t)$ is registered:

$$\frac{I_s(t)}{I_o(t)} = \frac{\int_{\lambda_1}^{\lambda_2} f(y) \cdot D_s(\lambda) \cdot T(\lambda) \cdot d\lambda}{\int_{\lambda_1}^{\lambda_2} f(y) \cdot D_o(\lambda) \cdot d\lambda} \quad (7)$$

In this case the dynamic range of measurements is significantly narrower (down to 10^2) and, which is also important, the experimental errors or the model representation of the functions $R(t)$ and $\gamma(t)$ produce the minimal effect. The function $\lambda_c(t)$ can be easily parametrised in the chosen time interval, and the parameters can then be fitted by the results of measurements on test objects.

The direct problem of calculating the time dependence of the signal $I_s(t)/I_o(t)$ with different coefficients of transmission curves $T(\lambda)$ was solved numerically for the above-mentioned measuring technique. A typical spectral sensitivity curve of a Ge:Ga detector for infrared radiation, common for both channels, was used. The calculations were performed in the time interval 1.7-2.5 ms, which corresponds to the scanning of the 10-200 μm wavelength range by the short-wave boundary of SR.

In Fig.3 there are different transmission coefficients

used in the calculations. Curves 1 and 2 are shown for illustration, they are the step function and the Gaussian distribution. Curves 3, 4, 5 correspond to the transmission coefficients of superconducting films calculated by formula (8):

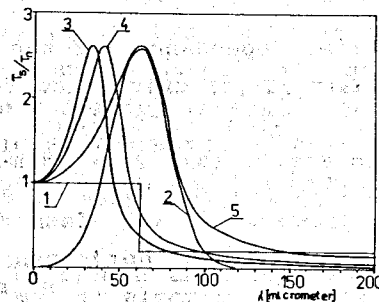


Fig.3. Transmission coefficients used for calculation of the time dependence of the signal. (1) is the step function, (2) is the Gaussian distribution, (3-5) are the transmission coefficients of the superconducting film calculated according to [8,9]. (3) $-2\epsilon_0 = 28$ meV, (4) $-2\epsilon_0 = 23$ meV, (5) $-2\epsilon_0 = 15$ meV.

$$\frac{T_s}{T_N} = \frac{1}{[T_N^{1/2} + (1-T_N^{1/2})(\sigma_1/\sigma_N)]^2 + [(1-T_N^{1/2})(\sigma_2/\sigma_N)]^2} \quad (8)$$

where T_S and T_N are the transmission coefficients for a super-conducting and normal state; σ_1 , σ_2 are the real and the imaginary parts of conductance in the superconducting state; σ_N is conductance in the normal state. The calculation was performed on the assumption that $T_N = C = 0.02$ [6].

To calculate the ratios σ_1 and σ_2 a theory developed in ref.[9] was used. According to it,

$$\frac{\sigma_1}{\sigma_N} = \left(1 + \frac{2\epsilon_0}{h\omega}\right) \cdot E(k) - 2 \cdot \left(\frac{2\epsilon_0}{h\omega}\right) \cdot K(k) ; \quad h\omega > 2\epsilon ; \quad (9)$$

$$\frac{\sigma_2}{\sigma_N} = \frac{1}{2} \cdot \left\{ \left(\frac{2\epsilon_0}{h\omega} - 1\right) \cdot E(k') + \left(\frac{2\epsilon_0}{h\omega} - 1\right) \cdot K(k') \right\} ;$$

where the arguments of the elliptical integrals E and K are:

$$k = \left| \frac{2\epsilon_0 - h\omega}{2\epsilon_0 + h\omega} \right| ; \quad k' = (1 - k^2)^{1/2} ;$$

$2\epsilon_0$ is the width of the energy gap, ω is the light frequency.

Curves 3, 4, 5 in Fig.3 are the penetration coefficients of a superconducting film at different energy gap widths, $2\epsilon_0 = 28, 23$ and 15 MeV respectively. Fig.4 shows the functions

$I_s(t)/I_o(t)$ calculated for the penetration coefficients mentioned.

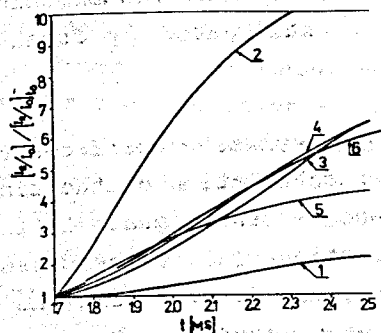


Fig. 4. Time dependence of the signal $I_s(t)/I_o(t)$ calculated for the transmission coefficients shown in Fig. 3. (3): $2\epsilon_0 = 28$ meV, (4): $2\epsilon_0 = 23$ meV, (5): $2\epsilon_0 = 15$ meV.

In Fig. 5 there are time derivative functions $d/dt(I_s/I_o)$ calculated for the time evolution in the time interval $t = (1.7-2.5)$ ms. It is seen that each value of the energy gap corresponds to a certain value of the time maximum for the function $d(I_s/I_o)/dt$.

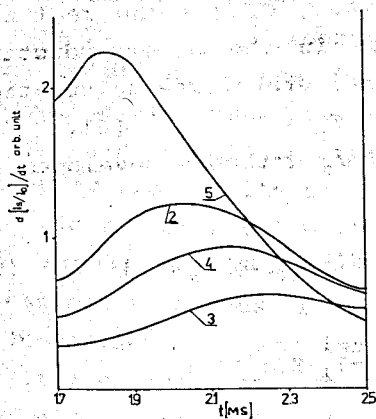
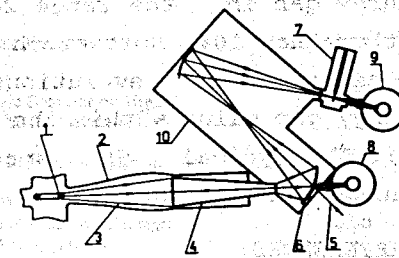


Fig. 5. Time dependence of the derivative $d(I_s/I_o)/dt$ for values of the energy gap. (3): $2\epsilon_0 = 28$ meV, (4): $2\epsilon_0 = 23$ meV, (5): $2\epsilon_0 = 15$ meV.

Fig. 6 schematically shows the experimental installation and the optical channel for extraction of synchrotron radiation. Since spectroscopic investigations of HTSC materials go on in parallel with investigations of charged states of ions in an electron ring, a brass conical light

guide with the acceptance $\theta_\varphi = \theta_z = 130$ mrad was used for extraction of synchrotron radiation. The transmission coefficient was estimated to be $\sim 80\%$. A conical light guide allows the maximum radiation power to be transferred to the sample in a cryostat with an adjustable temperature range from 5 to 300 K. To measure transmission coefficients of HTSC materials, two beams are shaped by mirrors. One passes through the sample, the other is a reference beam to measure the radiation intensity on the sample.

Fig. 6. Schematic of the installation and the optical channel for measurement of penetration spectra HTSC films. 1: electron ring, 2: light guide cone, 5: laser beam, 6: spherical mirrors, 7: cryostat with sample, 8: detector for IR radiation.



According to this method, the beams of a copper vapour laser ($\lambda = 0.51 \mu\text{m}$, $P = 30$ kW) and a nitrogen laser ($\lambda = 0.337 \mu\text{m}$, $P = 3$ kW) are sent through the monitoring channel to study evolution of a long-wave transmission spectrum in the destructive mode of superconductivity. For classical measurement of transmission spectra a modified monochromator with separable echelettes up to $\lambda = 600 \mu\text{m}$ is placed behind the mirror channel. The radiation is detected by an optic-acoustic receiver and a photodetector based on admixture photoconductance Ge:Ga.

Let us estimate the minimum R and γ values at which radiation is reliably registered when a two-beam method is used to measure the transmission coefficient. With the electron ring compression time $t = 2.0$ ms, $R = 15$ cm, $\gamma = 15$, the maximum spectral radiation power, $\lambda_M = 0.42\lambda_c = 96 \mu\text{m}$ we obtain $P_{\text{max}} = 2.7 \cdot 10^{-16}$ W/ μm . If we have the number of electrons $N_e = 5 \cdot 10^{12}$, the optical channel acceptance $\theta_z = \theta_\varphi = 130$ mrad, the transmission coefficient of a YBaCo

film on a MgO support for the wavelength $\lambda = 100 \mu\text{m}$ $T = 10^{-2}$, then the radiation power on the detector will be $P_D = P(\lambda_M) \cdot G \cdot \Delta\lambda \cdot N_e \cdot T = 4 \cdot 10^{-6}$ W. For the Ge:Ga admixture photo-receiver the minimal registered power is $P = 10^{-10}$. Thus, the synchrotron radiation with $R = 15$ cm, $\gamma = 14$ is reliably registered. It corresponds to the long-wave boundary of the scanning beginning $\lambda = 100 \mu\text{m}$. The calculations show that two-channel registration of the radiation passed through a HTSC film and monitored by the time evolution I_s/I_0 and $d/dt(I_s/I_0)$ allows one to find the value of the energy gap in the range $2\epsilon_0 = (15-28)$ meV with an accuracy better than 10%. Software has been developed for the analysis of experimental evolutions I_s/I_0 in order to determine the energy gap value within the B-C-S theoretical calculations (9). The typical SR spectrum scanning time is ~1 ms in tuning from $100 \mu\text{m}$ to $10 \mu\text{m}$.

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