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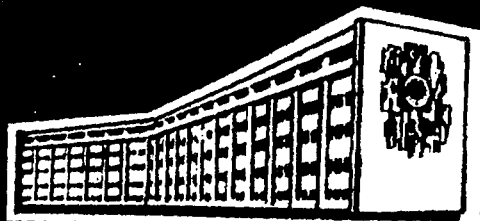
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INTERLAYER INTERACTIONS IN
ABSORPTION AND REFLECTION
SPECTRA OF BISMUTH HSC CRYSTALS



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Межслоевые взаимодействия в спектрах поглощения и отражения висмутовых кристаллов ВТСП

В работе теоретически анализируются особенности оптических спектров отражения и поглощения ВТСП кристаллов на основе висмута и таллия. Предложенный подход учитывает сложный характер структуры кристаллов, возможность локализации возбуждений в изолированных слоях и последующего обмена возбуждениями, вследствие межслоевого взаимодействия, между купратами (CuO) и квази вырожденными с ними висмутовыми слоями (BiO/ZrO). Получены выражения для энергий возбуждений и интенсивностей соответствующих переходов. Показано, что только часть возбуждений, общее число которых определяется количеством слоев в элементарной ячейке, будет проявляться в оптических спектрах поглощения и отражения.

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Interlayer Interactions in Absorption and Reflection Spectra of Bismuth HTSC Crystals

The HTSC reflection and absorption optic spectra peculiarities are analysed in the paper on the basis of bismuth and thallium. The approach suggested takes into account the complex character of crystals structure, possible localization of excitations in the isolated layers and further excitations exchange due to the interlayer interaction between cuprate (CuO) and quasi-degenerate bismuth layers (BiO/ZrO). The expressions for the excitation and intensity energies of the corresponding transitions are obtained. It is shown that only part of excitations whose number is determined by the number of layers in the unit cell will be manifest in optical reflection and absorption spectra. The experimental results on spectral dependence of crystal reflection coefficients are analysed.

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1. INTRODUCTION

The bismuth HTSC compounds synthesized and described recently (see, e.g., Refs. [1-3]) are now intensively studied.

The general chemical formula characterizing these substances has the form $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4}$ ($n = 1, 2$ and 3) and contains no rare-earth elements in its composition. These crystals have two-dimensional square lattice of copper atoms (Cu) bonded by oxygen atoms as in the case of $(\text{La}, \text{Sr})_2\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$ but also have the space structure caused mainly by Bi_2O_2 layers.

Considerable efforts are focussed on studying the optical properties of these compounds. In particular, a specific interest is attracted to the fact how the Bi_2O_2 layers effect essentially the physical properties, optical ones included, of $\text{Bi}_2\text{O}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4}$ crystals.

The purpose of this paper is to study theoretically certain optical peculiarities inherent to these crystals proceeding from their structural properties. Therefore, we describe in brief the experimental fact established so far for these compounds and caused by the electron transitions (until we do not deal with peculiarities of their phonon spectra).

The most complete data on optical properties of these substances are given in Refs. [4, 5]. According to the data of the above-cited papers there hold the following facts.

a - Measurement of conduction monocrystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ reveals anomalous anisotropy in the conductivity between the directions in parallel ρ_{\parallel} and perpendicularly ρ_{\perp} to the C-axis

$\rho_{\parallel}/\rho_{\perp} \sim 10^5$. This points out to the fact that the conduction CuO_2 layers are well isolated along the direction of C-axis by Bi_2O_2 layers. The band calculation results reveal then that Bi_2O_2 layers provide holes for conducting CuO_2 layers

b - The $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ specimens become superconductors at $T_c \sim 80$ K. The $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ specimens are metal but they do not manifest superconductivity higher than 42 K.

c - The measuring data on the reflection [4] reveal that spectra of both compounds $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ are rather close by their properties (Fig. 1a) but differ much from the analogous spectra in crystals $(\text{Ca}, \text{Sr})\text{CuO}_2$ (Fig. 1b) and also from the spectra of $(\text{La}, \text{Sr})_2\text{CuO}_4$ compounds that have become classic ones, Fig. 1c (Figs. 1a, b illustrate how the planes are disposed in the

unit cell of Bi-containing crystals).

d - The most distinguished peculiarity of Bi-containing cuprate crystals is the presence of wide, intensive enough peak in the range of 4-5 eV. This maximum is not quite determined because of complex structure of crystals of Bi-containing cuprates, although, in principle, there are two possibilities to explain this maximum:

- the first one is associated with the optical transition in Bi_2O_2 and/or SrO layers between Bi 6p and d2p;

- the second - with the transition between Cu 3d and O 2p in CuO_2 layer. In principle, the overlap of these transitions is possible.

e - It is characteristic that the other cuprates, without Bi_2O_2 layers, also manifest sometimes the peculiarity in the region 4 eV, but the maximum observed is narrower (Fig.1b) and unitary. The peak is ascribed to the internal excitation of CuO_2 layer.

The main conclusion of Ref.[4], based on the data of the other authors and ours consists in the following.

The fact that peculiarity of $E \sim 4$ eV in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ crystals consists of several peaks and forms relatively intensive and wider reflection band than in $(\text{Ca,Sr})\text{CuO}_2$ (Figs.1a,b) implies that excitations in CuO_2 layer same as in $\text{Bi}_2\text{O}_2/\text{SrO}$ layers give, probably, the total contribution to this wide reflection peak. Generally speaking, the $\text{Bi}_2\text{O}_2/\text{SrO}$ layers can also be isolating and have no direct relation to superconductivity.

2. MODEL OF CRYSTAL

According to experimental data we'll assume the conducting planes and nonconduction $\text{Bi}_2\text{O}_2/\text{SrO}$ layers to be isolated and the excitations, effected in them, to be preserved well enough in zero approximation. Thus, such a crystal can be regarded as unidimensional molecular crystal where the above planes interacting between themselves play the role of molecules. One can use here the approach well known in the theory of molecular crystals [6-8] having noted one important point: not all layers, involved in the unit cell, are identical. Frequencies of the transitions in them are somewhat different, so that this situation is more close to

the case of combined Fermi-Davydov resonance [8] where the frequencies in a single molecule can be different.

3. EXCITATION ENERGIES AND INTENSITIES OF TRANSITIONS

Let the number of layers in the unit cell of crystal be G then the wave functions of the ground Ψ^0 and excited Ψ states, and also the Hamiltonian operator of such layered crystal can be written as:

$$\Psi^0 = \prod_{n=1}^{GN} \Psi_{n\alpha}^0; \quad (1)$$

$$\Psi = \sum_{n=1}^{GN} C_{n\alpha} \Psi_{n\alpha} \prod_{m\beta (\neq n\alpha)} \Psi_{m\beta}^0; \quad (1a)$$

$$H = \sum_{n=1}^{GN} H_{n\alpha} + \frac{1}{2} \sum_{n\alpha \neq m\beta}^{GN} V_{n\alpha, m\beta} \quad (2)$$

$$H_{n\alpha} \Psi_{n\alpha} = E_{n\alpha} \Psi_{n\alpha}; \quad H_{n\alpha} \Psi_{n\alpha}^0 = E_{n\alpha}^0 \Psi_{n\alpha}^0. \quad (3)$$

Here N enumerates the number of unit "layered" cells in the main region of crystal cycle; $\Psi_{n\alpha}^0$ and $\Psi_{n\alpha}$ are the wave functions of an isolated layer in the ground and excited states, respectively; $H_{n\alpha}$ is the Hamiltonian of an isolated layer;

$V_{n\alpha, m\beta}$ is the interaction operator of $n\alpha$ and $m\beta$ layers; $C_{n\alpha}$ are the probability amplitudes of the fact that the excitation obeying the normalization condition, arises in the layer.

The excitation energy of the crystal, containing G molecules in the unit cell, is determined by the standard expression which after the Fourier-transformation takes the form

$$\Delta H = \sum_{\kappa\alpha} \bar{a}_{\kappa\alpha} a_{\kappa\alpha} (\Delta \varepsilon_{\alpha} + \mathcal{D}_{\alpha}) + \sum_{\kappa\alpha\beta} \bar{a}_{\kappa\alpha} a_{\kappa\beta} M_{\alpha\beta}(\kappa); \quad (1)$$

$\Delta \varepsilon_{n\alpha} = \Delta \varepsilon_{\alpha}$; $\mathcal{D}_{n\alpha} = \mathcal{D}_{\alpha}$; $M_{\alpha\beta}(\kappa) = \sum_m H_{n\alpha, m\beta} e^{i\kappa(m-n)}$.
Here $\vec{\kappa}$ is one-dimensional wave vector directed perpendicularly to the layers, i.e., along the crystal axis.

The operator of the interaction between crystal and electromagnetic field is written as

$$H_{int} = - \sum_{\alpha\beta} E_{\alpha\beta} d_{\alpha\beta} \Rightarrow \sum_{\alpha\beta} (\vec{E}_0 e^{i(Qm - \omega t)} + \text{c.c.}); \quad (5)$$

Intensity of transitions per single "layered" unit cell is determined by $I_0 \sim \frac{1}{N} \langle \Psi^* | H | \Psi_0 \rangle^2$ which, with account, taken of (1), (1a) equals

$$I_0 \sim |\vec{E}_0 \sum_{\kappa d} \vec{d}_d \vec{a}_{\kappa d} \delta(Q-\kappa)|^2 = |\sum_d a_{Qd} \vec{a}_d|^2; \quad (6)$$

$$\sum_d |a_{Qd}|^2 = 1; \quad a_{\kappa d} = a_{Qd} \delta(Q-\kappa). \quad (7a)$$

Using Eqs. (4) and (7) and varying them in \vec{a}_{Qd} we get the system of equations determining the excitation spectrum of the layered crystal, i.e. the energy band position and their intensities are determined by the expressions (6) and (7)

$$\sum_{\beta} [(E - \Delta \epsilon_d - D_d) \delta_{d\beta} - M_{d\beta}(Q)] a_{\beta Q} = 0. \quad (8)$$

It was stated in the Introduction that there exists the family of Bi-containing crystals. In particular, the studies of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ exhibit similar singularities in the reflection spectra (doublet structure in the energy range 4-5 eV), although crystals have rather different unit cells.

We classify them as:

- a - four-layered, $\text{Bi}_2\text{Sr}_2\text{CaCuO}_6$, that have two CuO_2 layers and two Bi_2O_2 layers in the unit cell and
- b - three-layered, $\text{Bi}_2\text{Sr}_2\text{CuO}_6$, that have one cuprate CuO_2 layer and two $\text{Bi}_2\text{O}_2/\text{SrO}$.

We consider each of the above-mentioned models taking into account the following rules similar for both models: the interaction of the layer neighbours only is take into account, with regard to the longwave length approximation the wave vector dependence in the matrix elements is omitted assuming $M_{d\beta}(Q) = M_{d\beta}$.

4. $\text{Bi}_2\text{Sr}_{2-x}\text{LaCu}_2\text{O}_{8+x}$ CRYSTALS (FOUR-LAYERED CASE)

a) Excitation energies

The model unit cell corresponding to this crystal is shown in Fig. 1a. The spectrum bands position is found from the determinant of the system (8). With account taken of the unit cell symmetry and the equality

$$\begin{aligned} \Delta \varepsilon_1 - \mathcal{D}_1 - M_{11} &= \Delta \varepsilon_4 - \mathcal{D}_4 - M_{44} = \varepsilon_1 \\ \Delta \varepsilon_2 - \mathcal{D}_2 - M_{22} &= \Delta \varepsilon_3 - \mathcal{D}_3 - M_{33} = \varepsilon_2 \\ M_{12} = M_{21} = M_{34} = M_{43} &= a, \quad M_{13} = M_{31} = M_{24} = M_{42} = 0 \\ M_{14} = M_{41} &= b; \quad M_{23} = M_{32} = d, \end{aligned}$$

the determinant Δ_4 can be written as

$$\Delta_4 = [(E - \varepsilon_1)(E - \varepsilon_2) - (a^2 - bd)]^2 - [d(E - \varepsilon_1) + b(E - \varepsilon_2)]^2 = 0. \quad (9)$$

Having done the replacements

$$\begin{cases} \varepsilon_1 - \varepsilon_2 = 2\delta \\ \varepsilon_1 + \varepsilon_2 = 2E_0 \end{cases} \Rightarrow \begin{cases} \varepsilon_1 = E_0 + \delta \\ \varepsilon_2 = E_0 - \delta \end{cases}; \quad \varepsilon = E - E_0. \quad (10)$$

we get

$$\varepsilon^2 + \varepsilon(b+d) + \delta(b-d) - \delta^2 - a^2 + bd = 0, \quad (11a)$$

$$\varepsilon_{1,2} = \frac{1}{2} \{ -(b+d) \pm \sqrt{(2\delta - (b+d))^2 + 4a^2} \};$$

$$\varepsilon^2 - \varepsilon(b+d) - \delta(b-d) - \delta^2 - a^2 + bd = 0, \quad (11b)$$

$$\varepsilon_{3,4} = \frac{1}{2} \{ (b+d) \pm \sqrt{(2\delta + (b+d))^2 + 4a^2} \};$$

b) Bands Intensity

The intensity of the corresponding transitions are determined by Eq.(6) in which the coefficients $a_{qd} = a_q$ should be found from the system (8). We express the coefficients a_d through a_1 :

$$a_2 = \frac{d_2}{d_1} a_1, \quad a_3 = \frac{d_3}{d_1} a_1, \quad a_4 = \frac{d_4}{d_1} a_1, \quad (12)$$

and, using the normalization condition (7) we get for a_1

$$a_1 = \frac{\pm |d_1|}{\sqrt{\sum_{i=1}^4 d_i^2}} \quad (13)$$

where

$$d_1 = b(E - \varepsilon_2)^2 + d(a^2 - bd);$$

$$d_2 = a[b(E - \varepsilon_2) + d(E - \varepsilon_1)]; \quad (14)$$

$$d_3 = a[(E - \varepsilon_1)(E - \varepsilon_2) - (a^2 - bd)];$$

$$d_4 = [(E - \varepsilon_2)^2(E - \varepsilon_1) - a^2(E - \varepsilon_2) - d_2(E - \varepsilon_1)];$$

We now easily calculate the sum $\sum_{d=1}^3 \vec{d}_d a_d$ in Eq.(6). It should then be borne in mind that 1 and 4 layers a_d as well as the 2 and 3 ones are identical, therefore, $\vec{d}_1 = \vec{d}_4$, $\vec{d}_2 = \vec{d}_3$. Then

$$I_0 \propto \left| \vec{E}_0 \sum_{d=1}^3 \vec{d}_d a_d \right|^2 = \frac{\left\{ \vec{E} [\vec{d}_1 (E - \epsilon_1 - d) + \vec{d}_2 a] [(E - \epsilon_2 + d)(E - \epsilon_1 - d - a^2)] \right\}^2}{\sum_{d=1}^3 d_i^2} \quad (15)$$

The given expression should be calculated at the energy values (i.e. $\epsilon_{1,4}$) determined by the roots of equations (11a,b).

We emphasize one important fact. It is clear that the numerator of Eq.(15) vanishes at certain energy values due to the presence of the second co-factor in square brackets. The relations between dipole moments of different layers \vec{d}_1 and \vec{d}_2 may be arbitrary. The energy values at which zero arises in the numerator are the following

$$\epsilon_{\pm} = \frac{1}{2} \left\{ -(\delta + d) \pm \sqrt{[2\delta - (\delta - d)]^2 + 4a^2} \right\} \quad (16)$$

They correspond exactly to the values of the roots of eqs.(11a). This means that in the crystal absorption spectrum the bands discussed will not be observed since their intensity is zero.

When the interaction between different layers dominates ($a \gg |b|$, $|a| \gg |d|$), and $\delta = 0$ for the roots $\epsilon_{3,4}$ of the allowed bands we have $\epsilon_{3,4} \approx \pm a$. The first operand of Eq.(15) that determines bands polarization gets transformed as

$$[\vec{d}_1 (E - \epsilon_2 - d) + \vec{d}_2 a] \Rightarrow a [\mp \vec{d}_1 + \vec{d}_2]$$

This result corresponds exactly to the effect of Davydov's bands polarization for crystals having layers with different orientation of dipole moments.

5. $\text{Bi}_2\text{Sr}_2\text{CaO}_{6+x}$ CRYSTAL (THREE-LAYERED CASE)

a) Excitation Energies

The model unit cell is shown in Fig.1a. The bands position is again determined from the system of equations (8) whose determinant for three-layered crystal with account of the unit cell symmetry and the equality

$$\Delta \epsilon_1 + D_1 + M_{11} = \Delta \epsilon_3 + D_3 + M_{33} = \epsilon_1$$

$$\Delta \epsilon_2 + D_2 + M_{22} = \epsilon_2$$

$$M_{12} = M_{21} = M_{23} = M_{32} = a, \quad M_{13} = M_{31} = d$$

is reduced to

$$\Delta_3 = (E - \epsilon_2 + d) [(E - \epsilon_2)(E - \epsilon_1) - 2a^2] = 0. \quad (17)$$

The roots of the equation (18) are equal to

$$\epsilon_1 = \delta - d$$

$$\epsilon_{2,3} = \frac{1}{2} [d \pm \sqrt{(2\delta + d)^2 + 8a^2}]. \quad (18)$$

b) Band Intensities

Using the known scheme we find from the system of equations (8)

$$a_2 = \frac{d_2}{d_1} a_1, \quad a_3 = \frac{d_3}{d_1} a_1 \quad (19)$$

$$a_1 = \frac{\pm |d_1|}{\sqrt{\sum_{i=1}^3 d_i^2}} \quad (20)$$

where

$$d_1 = [d(E - \epsilon_2) + a^2],$$

$$d_2 = a[E - \epsilon_2 + d], \quad (21)$$

$$d_3 = [(E - \epsilon_1)(E - \epsilon_2) - a^2].$$

Substitution of equations gives the following expression for intensity three-layered crystal transitions

$$I_0 = \frac{\{\bar{E}_0 [d_1 \vec{d}_1 (E - \epsilon_2) + a \vec{d}_2] (E - \epsilon_2 + d)\}^2}{\sum_{i=1}^3 d_i^2} \quad (22)$$

It is readily seen comparing it with (19) that with the value of one of the determinant Δ_3 roots, namely $E = \epsilon_1 = \delta - d$, the numerator vanishes again.

Thus, for both crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ instead of quartet and triplet, respectively, only doublets of bands will be observed in the absorption spectrum in the frequency range of

resonance transitions of CuO_2 layers quasi-degenerate with transitions in $\text{Bi}_2\text{O}_2/\text{SrO}$ layers.

A detailed analysis, taking into account the effect of different parameters on the spectra, will be done below. Here we note the following:

- a) The results obtained can readily be applied to explain the optic spectra and other crystals having no more than 4 layers. In particular, HTSC crystals on the basis of Tl (thallium) for which the transitions only in cuprate CuO_2 layers are actual. For this it is sufficient to take $|\vec{d}_1| = |\vec{d}_2|$ and set the angle θ between the dipole moment vectors of different layers to be zero.
- b) Besides that, if in the layered crystals the phonon excitations are well localized in the layers the results obtained can be used also to explain the rules in optical phonon spectra.

6. INFLUENCE OF CRYSTAL PARAMETERS ON INTENSITY DISTRIBUTION IN ABSORPTION BANDS (DISCUSSION OF RESULTS)

The expression for the absorption bands position (11a,b) and (18) and their intensities (15) and (22) are dependent in a complicated manner on the crystal parameters. But parameters δ and d give only shift in energies ϵ_1 and ϵ_2 , thus, only the interaction parameter of $\text{Bi}_2\text{O}_2\text{-CuO}_2$ layers will be important. The Figs. 3 and 4 show some of such dependences. The circles (white and black) show in energy scale the positions of the transition bands, and vertical lines from these points to the intersection with enveloping one (points A,B) give the values for the corresponding intensities. It is clear that the intensities ratio can be different and depends on mutual orientation of dipole moments of the transitions (angle θ). The figure 3 characterizes the 4-layered structure, Fig. 4 - the 3-layered one. For the two values of roots (ϵ_1 and ϵ_2) the intensity of the transitions is zero, whereas in Fig. 3 this is effected for one root only $|\epsilon_1|$. For both crystals, although they have different structure the band doublets only are observed.

More complete information on optical properties of crystals can be obtained from theoretical calculations of spectral dependences of its optical characteristics.

It was noted in part I of the paper that experimental results

on optical studies of BI-containing cuprate HTSC crystals were connected mainly with the reflection spectra $R(\omega)$. Thus, theoretical results obtained there, make it possible to determine the number of bands manifest in the spectrum, their relative intensities, but do not allow one to compare directly the calculation results with experiment.

In this part of the paper we investigate spectral characteristics of absorption and refraction coefficients $\mathcal{K}(\omega)$ and $n(\omega)$, respectively. The frequency dependence of the reflection coefficient $R(\omega)$ is then compared to experiment. This makes it possible not only to verify the reasonableness of the model suggested, but also to determine some parameters of the crystal such as interlayer interaction constants, transition oscillator force, unperturbed excitation energies, etc.

7. DIELECTRIC FUNCTION OF CRYSTAL

To study the spectral distribution within the transitions it is convenient to go over to the second quantization representation. According to the known procedure (see, e.g., [6,7]) the excitation energy operator of the crystal conserves, in Heitler-London approximation, the previous form [4], where the values $a_{\kappa\alpha}^+, a_{\kappa\alpha}$ can now be regarded as Bose-operators.

The dielectric function is expressed through the dipole moment operators [6,8]

$$\epsilon_{xx}(\kappa\omega) = \epsilon_{0xx} - \frac{4\pi}{V} \sum_{\alpha\beta} d_{\alpha x} d_{\beta x} \ll (a_{\kappa\alpha} + a_{\kappa\alpha}^+) \epsilon; (a_{-\kappa\beta} + a_{-\kappa\beta}^+) \gg_{\omega}. \quad (23)$$

Here $V = \ell \cdot S$ is the unit cell volume, ℓ is the dimension along the axis, S - the layer area.

In the layer axis $/x, y/$ the crystal lattice is square, thus, $\epsilon_{\alpha x} \Rightarrow \epsilon_{\alpha}$.

To find the spectral dependence $\epsilon_{xx}(\omega, \kappa)$ it is necessary to get the Fourier components in the relation (30).

8. EQUATIONS FOR GREEN'S FUNCTIONS

We assume the Hamiltonian operator (4) determining the properties of the system to be supplemented with the operator H_{pa} generating the excitation damping and take into account its influ-

ence in the finite formulae introducing the damping constants γ_i .

Out of Green's functions, incoming into (23), only those with resonances in the energy range $E \sim \epsilon_1, \epsilon_2$ prove to be of interest, namely,

$$G_{\alpha\beta}(\kappa, \omega) = \langle\langle a_{\kappa\alpha}(t), a_{\kappa\beta}^+(0) \rangle\rangle_{\omega}. \quad (24)$$

The equation for the Green's functions $|t_k=1|$ (24) with account taken of the Fourier-transformation is reduced to:

$$\sum_{\gamma} \Delta_{\alpha\gamma}(\kappa, \omega) G_{\gamma\beta}(\kappa, \omega) = \delta_{\alpha\beta} \quad (25)$$

where

$$\Delta_{\alpha\gamma}(\kappa, \omega) = (\omega - \tilde{\omega}_{\alpha}) \delta_{\alpha\gamma} - M_{\alpha\gamma}(\kappa); \quad \tilde{\omega}_{\alpha} = \Delta\epsilon_{\alpha} + \omega_{\alpha} - i\gamma_{\alpha}. \quad (26)$$

Using the algebra rules the solution to the system (25) is written as

$$G_{\gamma\beta}(\kappa\omega) = \frac{i}{\Delta(\kappa\omega)} \sum_{\alpha} \delta_{\alpha\beta} A_{\alpha\gamma} = \frac{A_{\beta\gamma}(\kappa\omega)}{\Delta(\kappa\omega)}, \quad (27)$$

where

$$\Delta(\kappa\omega) = \det \|\Delta_{\alpha\gamma}(\kappa\omega)\| \quad (28)$$

$A_{\alpha\gamma}(\kappa\omega)$ is an algebraic addition to the element $\Delta_{\alpha\beta}(\kappa\omega)$ of the determinant $\Delta(\kappa\omega)$ of the system. Thus, the sum $\sum_{\alpha} \delta_{\alpha\beta}$ in the relation (27) gets transformed to simple enough equation

$$\sum_{\alpha\beta} d_{\alpha\alpha} d_{\beta\beta} G_{\alpha\beta}(\kappa\omega) = \sum_{\alpha\beta} d_{\alpha\alpha} d_{\beta\beta} \frac{A_{\beta\alpha}(\kappa\omega)}{\Delta(\kappa\omega)}. \quad (29)$$

It is necessary now to calculate the values $A_{\alpha\beta}(\kappa\omega)$ using Eqs. (26), (28) and to find the sum (29) determining the contribution to the dielectric function of the crystal (23) of resonance states. The calculation will be done for each of the above-concerned models of crystals (four- and three-layered ones).

9. $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ CRYSTAL (FOUR-LAYERED CASE)

The determinant of the system $\Delta(\kappa\omega)$ coincides with Eq. (9), (we are again limited by the longwavelength approximation $\kappa \rightarrow 0$).

It must be borne in mind that $\varepsilon_1 \Rightarrow \tilde{\varepsilon}_1 = \varepsilon_1 - i\gamma_1$, $\varepsilon_2 \Rightarrow \tilde{\varepsilon}_2 = \varepsilon_2 - i\gamma_2$. Taking this into account we rewrite the determinant (9) in a somewhat transformed way

$$\Delta_4(\omega) = [(\omega - \tilde{\varepsilon}_1 + b)(\omega - \tilde{\varepsilon}_2 + d) - a^2][(\omega - \tilde{\varepsilon}_2 - d)(\omega - \tilde{\varepsilon}_1 - b) - a^2] \quad (30)$$

Algebraic additions $A_{\beta\alpha}(k\omega)$ can be obtained from the relations (26), (28) and taking into account that $d_{1x} = d_{4x}$, $d_{2x} = d_{3x}$ the Eq.(29), after some cumbersome, though not complicated transformations, will be reduced to

$$\sum_{\alpha\beta} d_{\alpha x} d_{\beta x} G_{\alpha\beta}(k\omega) = \frac{2 \{ [d_{1x}^2(\omega - \tilde{\varepsilon}_2 - d) + 2d_{1x}d_{2x}a + d_{2x}^2(\omega - \tilde{\varepsilon}_1 - b)] \}}{[(\omega - \tilde{\varepsilon}_2 - d)(\omega - \tilde{\varepsilon}_1 - b) - a^2]} \quad (31)$$

It is clear from Eq.(31) that in the frequency range $\omega \sim \tilde{\varepsilon}_1, \tilde{\varepsilon}_2$ there are only two poles rather than four ones as it follows from the determinant of the system (30). Thus, just the poles (11c) where, by part I, the transition intensities are nonzero, are conserved.

We note the following: if we neglect the interaction between layers of different nature, i.e. CuO_2 and $\text{Bi}_2\text{O}_2/\text{SrO}$ assuming

$a = 0$, the sum (31) is represented as the set of two oscillators

$$\sum_{\alpha\beta} \Rightarrow 2 \left\{ \frac{d_{1x}^2}{\omega - \tilde{\varepsilon}_1 - b} + \frac{d_{2x}^2}{\omega - \tilde{\varepsilon}_2 - d} \right\}, \quad (32)$$

where the coefficient 2 takes into account the presence of two identical cuprate and bismuth layers. The shift in energy by the value b for $\tilde{\varepsilon}_1$ ($\text{Bi}_2\text{O}_2/\text{SrO}$ layer) and by the value d for $\tilde{\varepsilon}_2$ (CuO_2 layer) is due to the interaction between the layers of the same type. Therefore, the role of these parameters is non-important. The parameter a is determinant.

Taking into account Eq.(31) the dielectric function of the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystal can be written as

$$\varepsilon_{xx}(\omega) = \varepsilon_{\text{ox}x} - \frac{4\pi d_{2x}^2 \{ \beta^2(\omega - \tilde{\varepsilon}_2 - d) + 2\beta a + (\omega - \tilde{\varepsilon}_1 - b) \}}{\nu [(\omega - \tilde{\varepsilon}_2 - d)(\omega - \tilde{\varepsilon}_1 - b) - a^2]}, \quad (33)$$

where $\beta = d_{1x}/d_{2x}$ is the value that indicates in relative units the ratio between the components of dipole moments of the transitions (oscillator forces) in $\text{Bi}_2\text{O}_2/\text{SrO}$ and CuO_2 layers, respectively.

only assume that $\vec{E} \parallel \vec{x} \parallel \vec{d}_{2x}$ then it is not necessary that $\vec{d}_{21} \parallel \vec{d}_{22}$.

To study the reflection spectra $R(\omega)$ it is necessary to find the real and imaginary parts of dielectric permeability $\epsilon(\omega)$. Since the lattices of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ crystals in the layer plane are square one can omit the indices x, x at $\epsilon_{c,xx}$. The expression (33) can then be represented as

$$\epsilon(\omega) = \epsilon_0 - \frac{4\pi \cdot 2 d_{2x}^2}{V \epsilon_0} \frac{(\bar{\epsilon}_1 - i\bar{\epsilon}_2)}{c^2 + q^2} \quad (34)$$

If we take explicitly into account the damping of excitations $\bar{\epsilon}_1 = \epsilon_1 - i\gamma_1, \bar{\epsilon}_2 = \epsilon_2 - i\gamma_2$ after simple transformations, introducing the values

$$x = \frac{\epsilon - \delta - \beta}{\epsilon_0}; \quad y = \frac{\omega + \delta - d}{\epsilon_0}, \quad a = \frac{\alpha}{\epsilon_0} \quad (35)$$

$$c = [xy - \gamma_1\gamma_2 - a^2], \quad q = [y\gamma_2 + x\gamma_1] \quad (36)$$

(here $\delta = \omega - E_0$, ϵ_0 and δ are determined by (10)) we get

$$\bar{\epsilon}_1 = \left\{ \gamma_2 \left[(\beta a + x)^2 + \gamma_1 \left[(a + \beta y)^2 + \beta^2 \gamma_2^2 \right] \right] \right\} \quad (37)$$

It is clear that imaginary part of the dielectric function responsible for the energy absorption by the system, is positive. The real part is sign-varying and equals

$$\bar{\epsilon}_2 = \left\{ x \left[\beta^2 (\gamma_1^2 + \gamma_2^2) - a^2 \right] + y \left[(x^2 + \gamma_1^2) - a^2 \beta^2 \right] + 2\beta a [xy - \gamma_1\gamma_2 - a^2] \right\} \quad (38)$$

after substitution of (37), (38) into (34) with account taken of the equality

$$\epsilon(\omega) = \epsilon_0 + 4\pi(x' + x'')$$

we obtain

$$x' = \frac{2 d_{2x}^2}{\epsilon_0 V} \frac{\bar{\epsilon}_1}{c^2 + q^2} \quad (39a)$$

$$x'' = \frac{2 d_{2x}^2}{\epsilon_0 V} \frac{\bar{\epsilon}_2}{c^2 + q^2} \quad (39b)$$

10. $\text{Bi}_2\text{Te}_2\text{O}_6$ CRYSTALS (THREE-LAYERED CRYSTALS)

The determinant characterizing the excitation spectrum of this crystal coincides with expression (17)

$$\Delta_3(\omega) = (\omega - \epsilon_1 + d) [(\omega - \bar{\epsilon}_2)(\omega - \bar{\epsilon}_1 - d) - 2a^2], \quad (40)$$

where, as in the previous case, one should change $\epsilon_2 \Rightarrow \epsilon_2 - i\gamma_2$. To find the dielectric permeability $\epsilon_{xx}(\omega)$ we calculate the sum (39). After simple algebraic transformations we get

$$\sum_{\alpha\beta} d_{1x} d_{2x} \frac{A_{\alpha\beta}(k\omega)}{\Delta(k\omega)} = \frac{[2d_{1x}^2(\omega - \bar{\epsilon}_2) + 4ad_{1x}d_{2x} + d_{2x}^2(\omega - \epsilon_1 - d)]}{[(\omega - \bar{\epsilon}_2)(\omega - \bar{\epsilon}_1 - d) - 2a^2]} \quad (41)$$

The cofactor corresponding to one of the determinant roots (17) gets reduced, so that the root $\omega = \bar{\epsilon}_1 - d \Rightarrow \delta - d$ for which the intensity of the corresponding transition was zero (22), disappears. When there is no interaction between the layers of different nature ($a=0$) the expression (41) is approximated by two independent oscillators:

$$\sum_{\alpha\beta} = \left\{ \frac{2d_{1x}^2}{\omega - \bar{\epsilon}_2} + \frac{d_{2x}^2}{\omega - \bar{\epsilon}_1} \right\}. \quad (42)$$

Let's pay attention to the following important circumstance. The results (32) and (42) reveal that the approximation $\epsilon(\omega)$ for an arbitrary system by the sum of independent oscillators is possible only under extremely weak interaction in the system. In the general case such an expression should involve also the interaction between the oscillators, but the approximation is determined by the specific model.

The dielectric function with account taken of (41) is written as

$$\epsilon(\omega) = \epsilon_0 - \frac{4\pi d_{2x}^2 [2\beta^2(\omega - \bar{\epsilon}_2) + 4a\beta + (\omega - \bar{\epsilon}_1 - d)]}{V [(\omega - \bar{\epsilon}_2)(\omega - \bar{\epsilon}_1 - d) - 2a^2]}. \quad (43)$$

We single out the real and imaginary parts of $\epsilon(\omega)$. The dielectric permeability is written in the standard form

$$\epsilon(\omega) = \epsilon_0 + 4\pi(X' + X'') = \epsilon_0 - \frac{4\pi d_{2x}^2 (\phi_1 - i\phi_2)}{V E_0 (c^2 + q^2)}, \quad (44)$$

where we introduce the notations

$$\Phi_1 = \left\{ \gamma_1 [(x_1^2 + \gamma_1^2) - 4\beta^2 a^2] + 2x_1 [\beta^2 (\gamma_1^2 + \gamma_2^2) - a^2] + 4\beta a [x_1 \gamma_1 - \gamma_1 \gamma_2 - 2a^2] \right\} \quad (45)$$

$$\Phi_2 = \left\{ 2\gamma_1 [(a + \beta \gamma_1)^2 + \beta^2 \gamma_2^2] + \gamma_2 [(x_1 + 2\beta a)^2 + \gamma_1^2] \right\} \quad (46)$$

Then

$$x_1 = \frac{\varepsilon - \delta \cdot d}{\varepsilon_0} ; \quad \gamma_1 = \frac{\varepsilon + \delta}{\varepsilon_0} , \quad (\varepsilon = \omega - E_0), \quad (47)$$

$$c = [x_1 \gamma_1 - \gamma_1 \gamma_2 - 2a^2], \quad q_0 = [\gamma_1 \gamma_1 + x_1 \gamma_2]. \quad (48)$$

Comparing (44), (45) and (46) we find

$$X' = - \frac{d_{zx}^2}{\sqrt{\varepsilon_0}} \frac{\Phi_1}{c^2 + q^2} ; \quad (49a)$$

$$X'' = \frac{d_{zx}^2}{\sqrt{\varepsilon_0}} \frac{\Phi_2}{c^2 + q^2} \quad (49b)$$

The value that characterizes losses is again positive. The magnitude $F = \frac{d_{zx}^2}{\sqrt{\varepsilon_0}}$ to be called the transition oscillator force, arises for both crystals.

11. REFLECTION COEFFICIENT, DISCUSSION OF RESULTS

The reflection coefficient is determined by the following equation [9]

$$R(\omega) = \left| \frac{n(\omega) - 1}{n(\omega) + 1} \right|^2 = \frac{(n-1)^2 + \kappa^2}{(n+1)^2 + \kappa^2} , \quad (50)$$

where the complex refraction index $n(\omega) = n + i\kappa$ is connected with the dielectric permeability function

$$\varepsilon(\omega) = \varepsilon' + i\varepsilon'' = n^2(\omega) = (n + i\kappa)^2 \quad (51)$$

We then easily get from (44) and (51)

$$\varepsilon' = \varepsilon_0 + 4\pi X' = n^2 - \kappa^2 \quad (52a)$$

$$\varepsilon'' = 4\pi X'' = 2n\kappa \quad (52b)$$

The two last expressions make it possible to find the refraction index n and the absorption coefficient κ

$$n = \sqrt{\frac{\sqrt{(\epsilon')^2 + (\epsilon'')^2} + \epsilon'}{2}}; \quad (53a)$$

$$k = \sqrt{\frac{\sqrt{(\epsilon')^2 + (\epsilon'')^2} - \epsilon'}{2}}; \quad (53b)$$

Having substituted into (53a), (53b) the relations (52a), (52b) we determine the dependences taking into account the specific expressions for the X' -real and X'' -imaginary parts of susceptibilities obtained, correspondingly, for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$. Below we give a series of theoretical curves for the four- and three-layered crystals. As is shown by the calculation data, the influence of the parameters a, β (b, d are set to be equal to zero) on the shape of the reflection spectrum, is appreciable, however, the low-frequency edge of both crystals could not be made so sharply varying as in experiment at any variations of the parameter, because of disregard of plasma excitations. The latter can easily be taken into account having added the term

ϵ_0 to $-\frac{\omega_p^2}{\omega^2}$. Then we get

$$\epsilon_0 \Rightarrow \tilde{\epsilon}_0 = \epsilon_0 - \frac{(\hbar\omega_p)^2}{E_0^2(1+\epsilon)^2} = \epsilon_0 - \frac{\Delta}{(1+\epsilon)^2};$$

Here $\Delta = \frac{(\hbar\omega_p)^2}{E_0^2}$, ω_p is plasma frequency, E_0 and ϵ are determined by the expressions (10).

The influence of the parameter Δ , i.e. plasma frequency ω_p is well shown in Fig.4 where the values of the parameters a, β are chosen so, that the peak of two-hump reflection maximum of the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystal observed in experiment (Fig.1a) is described perfectly well. If $\Delta=0$ (Fig.4 curve 1) the reflection $R(\omega)$ in the low-frequency range gets constant value determined by $\tilde{\epsilon}_0$. If Δ is small ($\Delta=0.05$ curve 2) the reflection curve bends down slowly, so that there arises certain hump, but, having attained the smallest value, goes sharply up. When the parameter Δ is large enough ($\Delta = 0.25$, curve 3), what corresponds to $\hbar\omega_p \sim 2$ eV, the reflection curve falls abruptly with next sharp raising up to the value $R \approx 1$; i.e. in a complete analogy with experimental curve (Fig.1a). Thus, the main details of the reflection curve are described by theoretical dependence. This becomes clear from comparison of experimental results

with calculations. Fig. 4 for $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ crystal and Fig. 5 for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystal. Two additional dependences (curves 5 and 3, respectively) are also shown there: the real and imaginary (curves 4 and 2) parts of susceptibility for both types of crystals. For X' the doublets of weakly allowed bands are distinctly manifest, and X'' has somewhat different forms at almost coincident reflection spectra $R(\omega)$. The interlayer interaction effect (of $\text{Bi}_2\text{O/Sr}_2\text{O}$ and CuO_2 layers) is illustrated in Fig. 7.

The difference in its influence for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{CaCuO}_8$ crystal is obvious. In particular, if $a \rightarrow 0$ only one maximum appears in $\text{Bi}_2\text{Sr}_2\text{CaCuO}_6$ crystal. Since the analytic values $\epsilon'(\omega)$ and $\epsilon''(\omega)$ are found, in our calculation the latter may be the basis for checking the correctness of applying the Kramers-Kronig method widely used to determine the other optical characteristics of crystal $n(\omega)$, $\chi(\omega)$ etc. according to the reflection spectra.

It follows from Fig. 1a that for both crystals the reflection spectrum is a curve monotonously decreasing with increasing energy E , where the weakening maxima coincident actually in the position for both crystals, get arisen. It is seen also that the second maxima within the range $E \sim 9-10$ eV are similar in shape to the first ones ($E \sim 4-5$ eV) what is especially clear for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystal. This similarity indicates that the above maxima represent high-energy iterations of the first doublet ($E \sim 4-5$ eV). Although they can shift slightly to the low-energy side by analogy with the shift of high-energy states in the oscillator problem. To verify this assumption the multiquanta iterations $E = n E_0$ ($n = 1, 2, \dots$) of the 1-st doublet $E_0 \approx 4.5$ eV are shown with arrows in Fig. 1a. The position of arrows, as is readily seen, follows after the position of maxima in the reflection spectra of both crystals.

The maximum within the range $E \sim 30$ eV is somewhat specific, since the state $\sim 6 E_0$ turns out to be close to a new intense excitation $E_v \sim 30$ eV. As a result of their interaction there appears a well pronounced gap in the reflection spectrum. The increasing intensity of the reflection in the quantum transition region $5 E_0$ also testifies in favour of such assumption.

Comparing the doublets E_0 and $2 E_0$ (Fig. 1a) we see that maxima ratio has changed. From comparison with the calculation data (Figs. 7a, b, 8a, b, curves 1-3) we may conclude that this is

associated with decreasing constant of the interaction between the layers of different origin (constant "a").

Taking into account the comments to Bi-crystals we can consider the situation in Fig.1b for (Ca,Sr) CuO₂ crystal. A sharp maximum $E_a \sim 1.7$ eV and more uniform $E_b \sim 4-5$ eV are clearly observed, so that in the energy range $E \sim 5$ eV there is observed the threshold. If we indicate with arrow the energy range

$3E_a$ we get the position of this threshold. The spectrum has also several arrows, pointing out to multiquanta iterations of the two first transitions ($n E_a, m E_b$). It is clear that in this case there holds the reasonable correlation.

The analysis performed shows that layers in these compounds prove to be important structural units and optical properties of the crystal are determined by the properties of separate layers.

The approach based on the molecular crystals correlates with Prieman conclusions [10] about possible important role of excitons arising in the layers separating CuO₂ layers for HTCC crystals.

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FIGURE CAPTIONS

- Fig. 1. Experimental reflection spectra $R(\omega)$ for the crystals:
 a - $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{CuO}_6$; b - $(\text{Ca}, \text{Sz})\text{CuO}_2$.
- Fig. 2. Position of absorption bands ($\mathcal{E}_1 \pm \mathcal{E}_4$) and their intensities (vertical segments) for certain values of the sets of four-layered crystal parameters: $a = 0,05$; $b = 0$; $d = 0$; $\beta = 0,2$; $\delta = 0$; $E_0 = 4,5$ eV. $1-\theta = 60^\circ$; $2-\theta = 180^\circ$.
- Fig. 3. Position of absorption bands ($\mathcal{E}_1 \pm \mathcal{E}_3$) and distribution of three-layered crystal intensities: $a = 0,05$, $d = 0$, $\beta = 0,2$, $E_0 = 4,5$ eV; $1 - \theta = 60^\circ$, $2 - \theta = 180^\circ$.
- Fig. 4. Change in the form of the reflection spectrum $R(\mathcal{E})$ of the four-layered crystal in the plasma frequency function (parameter Δ):
 curve - 1, $\Delta = 0$;
 curve - 2, $\Delta = 0,05$;
 curve - 3, $\Delta = 0,25$;
 curve - 4, imaginary parts of susceptibility in the region of resonance transitions (Δ -independent). Insertion shows comparison of experiment with calculation (χ' and χ''). Parameters: $a = 0,05$; $b=d=0$; $\delta = 0,1$; $\beta = -0,4$; $F = 0,035$; $E_0 = 4,5$ eV; $\mathcal{E}_0 \approx 3,5$; $\Delta = 0,25$);
 curve - 5 real $\chi'(\mathcal{E})$.
- Fig. 5. The form of the reflection spectrum $R(\mathcal{E})$ of three-layered crystal (curve 1). (Parameters: $a = 0,05$; $d = 0$; $\delta = 0,1$; $\beta = 0,4$; $F = 0,035$; $E_0 = 4,5$ eV, $\mathcal{E}_0 \approx 3,5$; $\Delta = 0,25$). Insertion - comparison of experiment with calculation.
- Fig. 6. The spectral dependence of the reflection coefficient $R(\omega)$ in the region of mixed $\text{CuO}_2 - \text{Bi}_2\text{O}_2/\text{SrO}$ transition for the four-layered crystal under varying interaction

parameter of the layers of different origin: $a \equiv \frac{a}{E_0}$

a - $a = 0$;

b - $a = 0,05$;

c - $a = 0,15$.

Fig.7. Change in the reflection spectrum $R(\omega)$ for the three-layered crystal under varying parameter a :

a - $a = 0$; b - $a = 0,05$; c - $a = 0,15$.

The rest parameters in Figs.6,7 are same as in Figs.5 and 4.

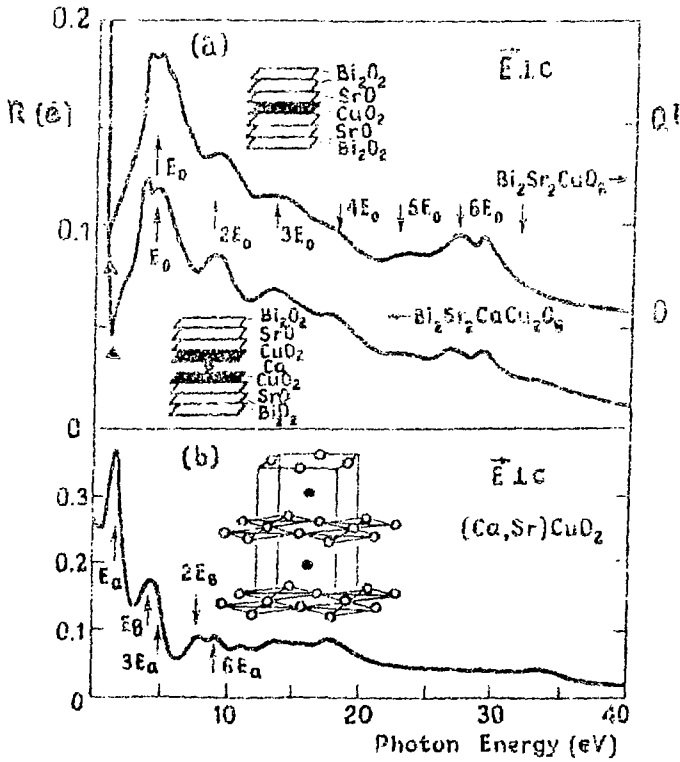


Fig. 1

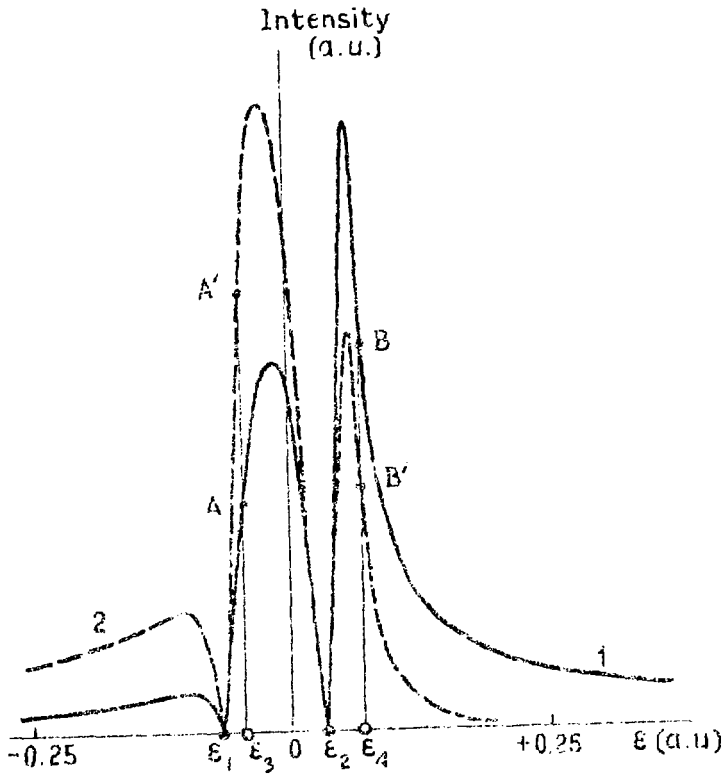


Fig. 7

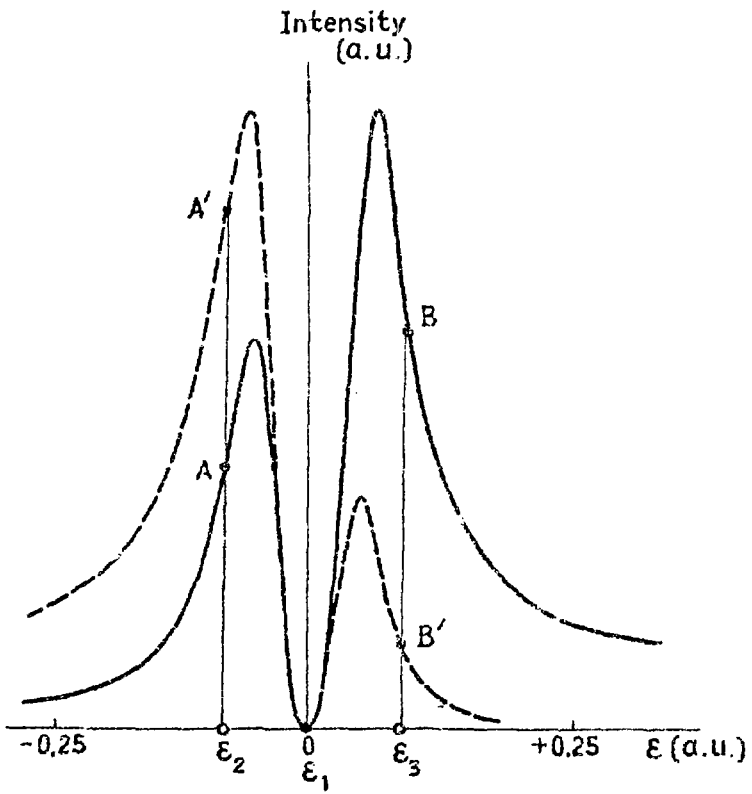


Fig. 3

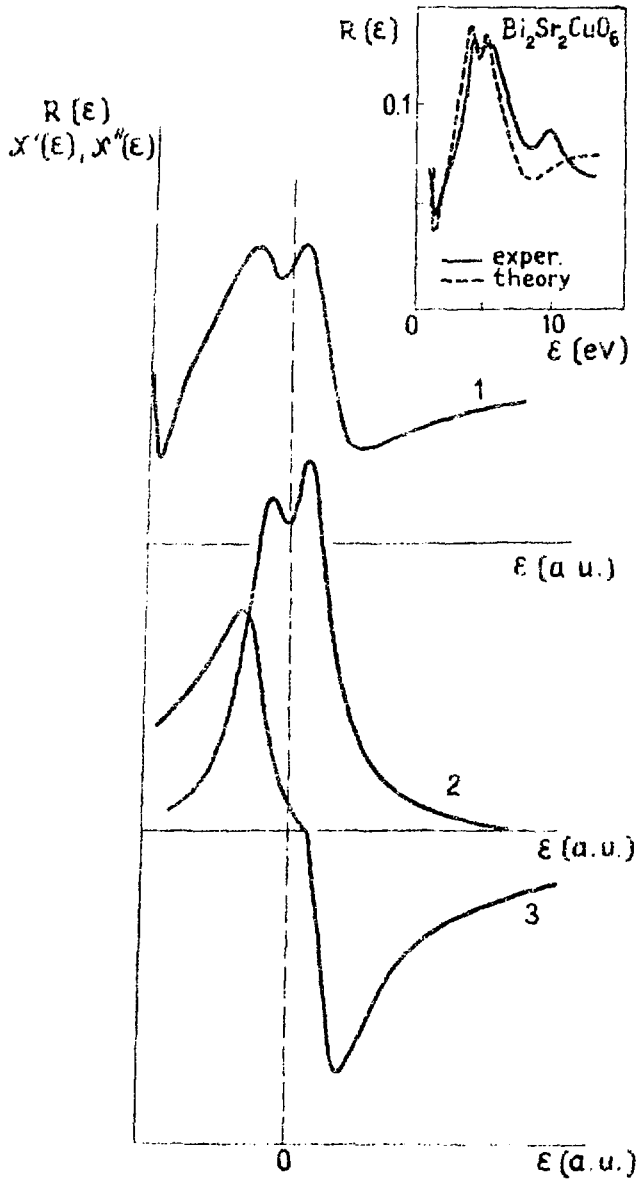


FIG. 4

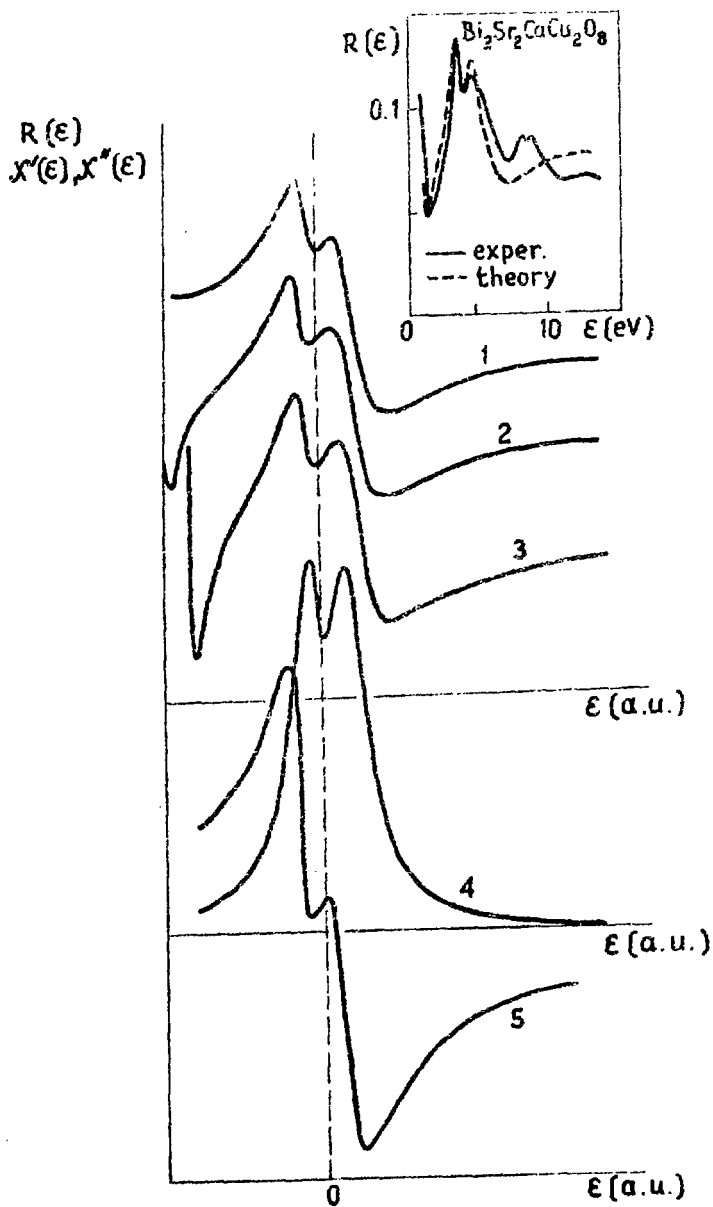


Fig.5

$R(\epsilon)$
 $X''(\epsilon), X'(\epsilon)$

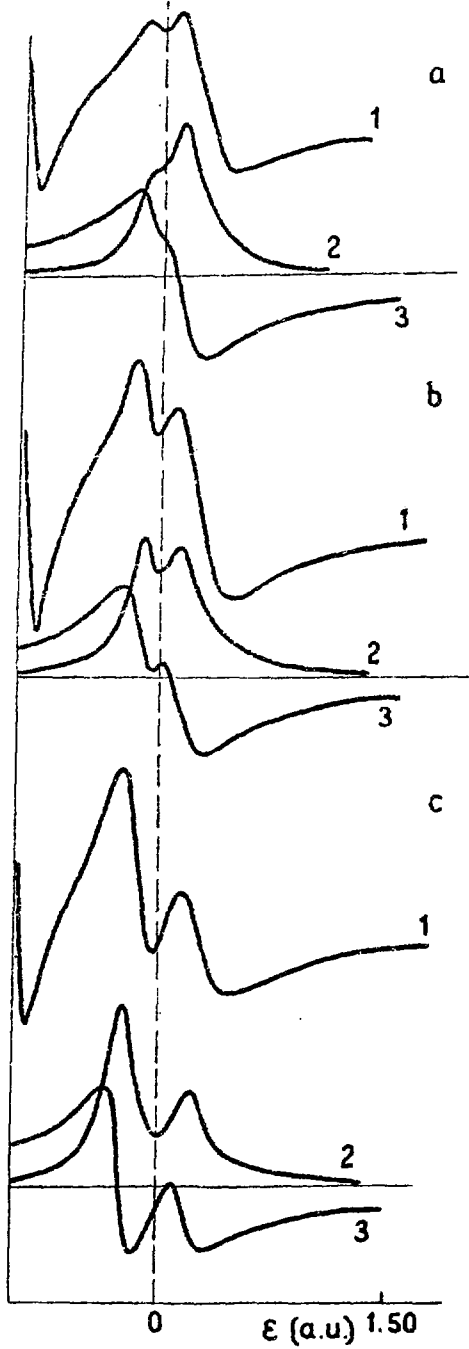


Fig.6

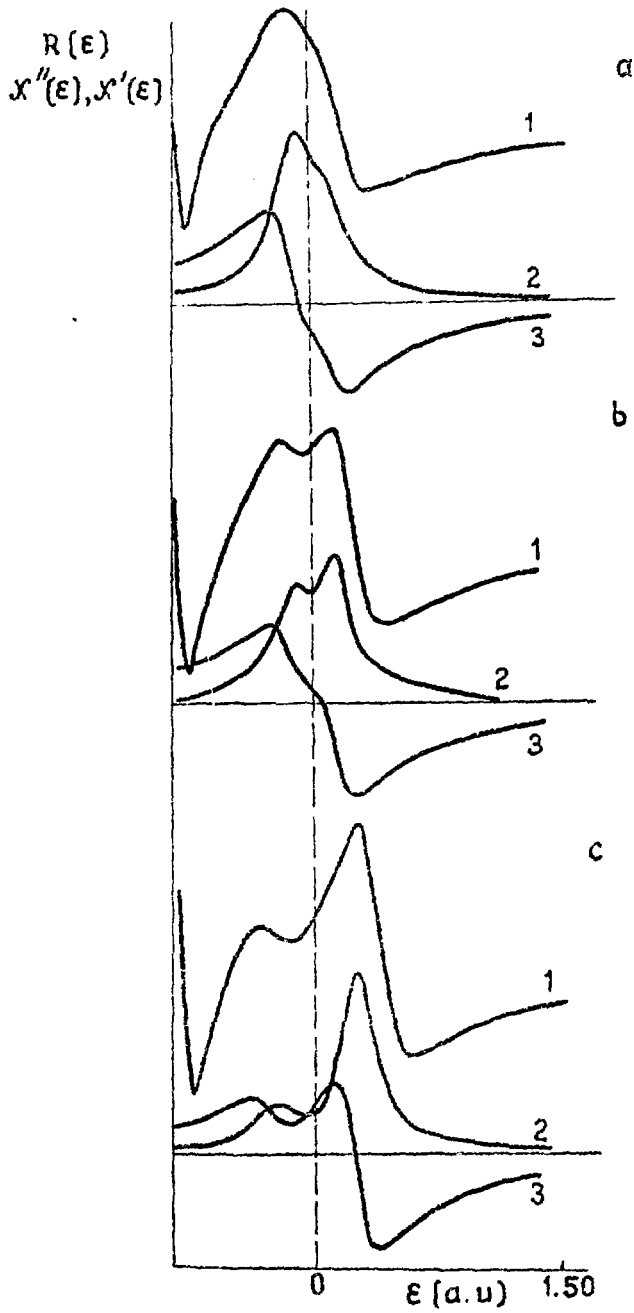


Fig.7

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