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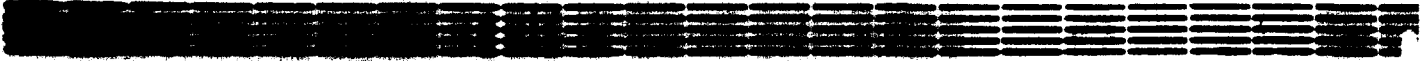
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ЕРЕВАНСКИЙ ФИЗИЧЕСКИЙ ИНСТИТУТ
YEREVAN PHYSICS INSTITUTE



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NONDIAGONAL COMPONENT OF LINEAR PHOTOVOLTAIC TENSOR
IN DOPPED LiNbO_3 AND LiTaO_3 CRYSTALS

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1. Introduction

In noncentrosymmetric crystals, at their homogeneous illumination, there arises a steady-state current [1]. According to the phenomenological theory [2], the expression for the photovoltaic current density has the form:

$$j_i = \beta_{ine} E_n E_e^* + i\gamma_{ie} [EE^*]_e, \quad (1)$$

where E_n , E_e are the components of the electric field of light wave, β_{ine} and γ_{ie} are the components of the symmetric third-rank tensor and the second-rank pseudotensor, respectively. The first term in this expression connects the photovoltaic current density to the field of linearly polarized wave, and is called "linear photovoltaic current". The second term is nonvanishing for elliptically polarized wave only, and is called "circular photovoltaic current".

In $\text{LiTaO}_3\text{:Fe}$, $\text{LiNbO}_3\text{:Fe}$ and $\text{LiNbO}_3\text{:Cu}$ crystals, which belong to $3m$ point group, the third-rank tensor, β_{ine} , has four independent components: β_{33} , β_{31} , β_{22} and β_{15}^s . In this

work we have presented results of the measurement of all the four independent components of the linear photovoltaic effect in doped LiTaO_3 and LiNbO_3 crystals. The measurements were done using the technique suggested in [4] and described in [5,6] .

The task was as follows: to clarify the influence of different admixtures in LiNbO_3 and LiTaO_3 crystals on the nature of the currents determined by the ρ_{15}^s component. By measuring the angular dependence of the current determined by this component in the $\text{LiNbO}_3\text{:Cu}$ crystal, there was found that the shape of the curve did not coincide with that theoretically predicted. In this connection here we present results of a series of experiments which were performed with the aim to find out the reason of discrepancy with theory. On the basis of obtained results we have made an assumption about the relation of this phenomenon to breaking of crystal uniaxiality.

2. Experiment

The experimental arrangement for photovoltaic current measurements is shown in Fig.1. A 1 kW xenon lamp (L) was used as a source of light. The light was monochromated by means of a CP -26 spectrometer (M), then polarized by a polarizer (P), and by means of a lens system (l) was focused on the crystal. The crystal was fastened on a measuring head (Fig.2) which could be turned independently in two mutually perpendicular planes with the help of a remote control. The head with the sample, in turn was placed in a shielded vessel. The contacts were made by vacuum deposition of silver. The photovoltaic current was measured by an electrometer (E1). The samples were oriented with respect

to crystallographic axes up to 10 angular minutes by means of X-ray diffraction. All the results obtained were corrected with regard to the absorption and non-uniformity of the crystal illumination according to the technique proposed in Ref. [3]. The method proposed earlier in [4] was used in the present work to measure the currents determined by the β_{15}^s component. The essence of the method is the following: if the XOY face of the crystal is exposed to the light wave incident at a certain angle in the XOZ plane and polarized in the same plane (Fig.3), then only the extraordinary wave will propagate in the crystal, with nonzero components of its electric field vector along both X and Z axes. The photovoltaic current along the X axis will contain a spatially uniform component which is defined by the expression:

$$j_x = \beta_{15}^s I^{(t)} \sin 2\alpha, \quad (2)$$

where

$$I^{(t)} = I(1 - R_{\parallel})$$

$$R_{\parallel} = \frac{\operatorname{tg}^2(\alpha^0 - \alpha)}{\operatorname{tg}^2(\alpha^0 + \alpha)}$$

$$\alpha = \sin^{-1}[(\sin \alpha_0)/n_e]$$

and I is the intensity of incident light, $I^{(t)}$ is the intensity of light in the sample, α^0 is the angle of incidence, α is the angle between the light propagation direction and the Z axis in the sample, R is the Fresnel reflectivity of the sample face. To measure the angular dependence of photovoltaic current densi-

ty, j_x , the measuring head was rotated in the XOZ plane. The measurement accuracy of the ratio of photovoltaic current to intensity was estimated no better than $\pm 13\%$.

3. Experimental Results and Discussion

3.1. Measurement of the β_{15}^S Photovoltaic Tensor Component in $\text{LiTaO}_3:\text{Fe}$ Crystal.

The experimental investigation was carried out on $\text{LiTaO}_3:\text{Fe}$ samples (0.038 wt %) which had a parallelepiped form with sizes $X:Y:Z = 2.75:4.8:4.25 \text{ mm}^3$. Three of the nine polarization dependences of photovoltaic current density for photon energy of exciting light 2.6 eV are presented in Fig.4. These dependences agree perfectly well with the theoretical predictions following from equation (1), which allows to calculate the values of β_{31} , β_{33} and β_{22} .

The angular dependence of photovoltaic current density along the X axis is shown in Fig.4b. It also fits well Eq.(2). When turning the sample around the X axis by 180° , the current determined by the β_{15}^S component changes its sign, this also being in agreement with the proposed model. The values of β_{15}^S were calculated from the slopes of angular dependence of j_x , according to Eq.(2).

The spectra of photovoltaic tensor components are shown in Fig.5. When comparing the results obtained, one may notice that all the components grow with increasing photon energy of incident light. A characteristic feature of β_{33} , β_{22} and β_{15}^S spectra is the presence of a shoulder in the energy range from 2.1 eV to 2.7 eV. To make possible a more detailed analysis,

the constants β_{33} , β_{22} and β_{15}^S were normalized to β_{31} , as the spectrum of β_{31} contains no sharp features. It can be seen from Fig.6 that the ratio β_{33}/β_{31} depends on the quantum energy least of all (is independent of the quantum energy with an accuracy up to 35%). From Fig.6 one also can see an obvious correlation between the spectral dependences for β_{22}/β_{31} and β_{15}^S/β_{31} . The curves qualitatively recur in shape. The maxima and minima occur for both curves at nearly the same photon energies. Such a correlation allows to assume that the linear photovoltaic currents propagating in the direction normal to \vec{C} axis in $\text{LiTaO}_3:\text{Fe}$ arise due to the photoexcitation of carriers from the same admixture or defect centers.

3.2. Measurement of β_{15}^S Photovoltaic Tensor Component in $\text{LiNbO}_3:\text{Fe}$ Crystals.

The measurements were carried out on $\text{Li NbO}_3:\text{Fe}$ samples (0.03 wt %) which had a parallelepiped form with sizes $X:Y:Z = 4.2:5.2:1.7 \text{ mm}^3$. The polarization dependences in these crystals as well as in $\text{LiTaO}_3:\text{Fe}$ crystals agree perfectly well with the theoretically predicted dependences. The angular dependence of photovoltaic current along the X axis also fits Eq.(2) perfectly well. And like in the case with $\text{LiTaO}_3:\text{Fe}$ crystals, according to the cited model, when turning the sample around the Z axis by 180° , the current determined by β_{15}^S component changes its sign.

The spectral dependences of linear photovoltaic tensor components are shown in Fig.7. Comparing the results, one can notice that the spectra of β_{15}^S correlate with those of β_{33} , β_{31} and β_{22} , all the components grow with increasing photon

energy, and at energies higher than 3 eV there is observed a cut-off in all the curves. Issuing from the aforesaid, one can conclude that the nature of the centers determining the photovoltaic currents, which is connected with all the four components, is the same.

3.3. Measurement of β_{15}^S Photovoltaic Tensor Component in $\text{LiNbO}_3:\text{Cu}$.

The measurements were carried out on $\text{LiNbO}_3:\text{Cu}$ samples (0.1 and 0.03 wt %) which had a parallelepiped form with sizes $X:Y:Z = 2.1:4.6:(1.1 - 4.3) \text{ mm}^3$. The polarization dependences of the photovoltaic currents which are determined by β_{22} , β_{31} and β_{33} components in this crystal agree with theoretical predictions. The spectral dependences of these components, which are shown in Fig.8, also are in agreement with the known data [3].

However the angular dependence of photovoltaic current j_x , which is determined by β_{15}^S component, differs from the dependence described by expression (2) as well as from the results which were obtained using $\text{LiTaO}_3:\text{Fe}$ and $\text{LiNbO}_3:\text{Fe}$ crystals [5,6]. This dependence is shown in Fig.9. When the angle of inclination α is changed from -10° to $+10^\circ$, the curve is growing, but near 0° it begins to oscillate. The frequency of oscillations depends on photon energy very faintly. When the energy of photons increases from 2 eV to 4 eV, the frequency of oscillations decreases by a factor of 1.5. The amplitude of oscillations depends linearly on the light intensity. When turning the sample around the Z axis by 180° , the current j_x changes its sign, and the oscillations on the angular dependence preserve too. To check up these results, we made samples with different dimen-

sions from different parts of the single crystal, and also made samples from the single crystal with Cu concentration 0.3 wt %. The external view of obtained angular dependences of j_x did not change. But in the case with samples with higher concentration of admixture the frequency of oscillations increased.

The investigation of conoscopic figures has shown that in the regions along X-cuts the samples have a considerable biaxiality, while in the middle part the crystal is strictly uniaxial (Fig.10). To remove the residual stress which is possible during crystals processing and which perhaps is the cause of deflection from uniaxiality, the samples were annealed during 20 minutes in a H_2 atmosphere at $700^\circ C$. After annealing almost the whole volume of the samples have got uniaxiality. The angular dependences j_x changed after annealing, and these dependences in some samples fitted Eq.(2). The results of measurements have shown that in the crystal $LiNbO_3:Cu$ (0.1 wt %) the component $\beta_{15}^S = 6.9 \cdot 10^{-9} V^{-1}$ for photon energy of exciting light 2.6 eV.

Thus we have made an assumption that the cited phenomenon is the result of crystal uniaxiality breaking. The nature of this phenomenon can be assessed more precisely with the use of additional experiments.

In conclusion the author would like to express his gratitude to Dr S.Odoullov and Dr V.Obuhovsky for the discussion of the results, and to Dr I.Kiseleva and Dr K.Belabaev for growing and preparation of crystals.

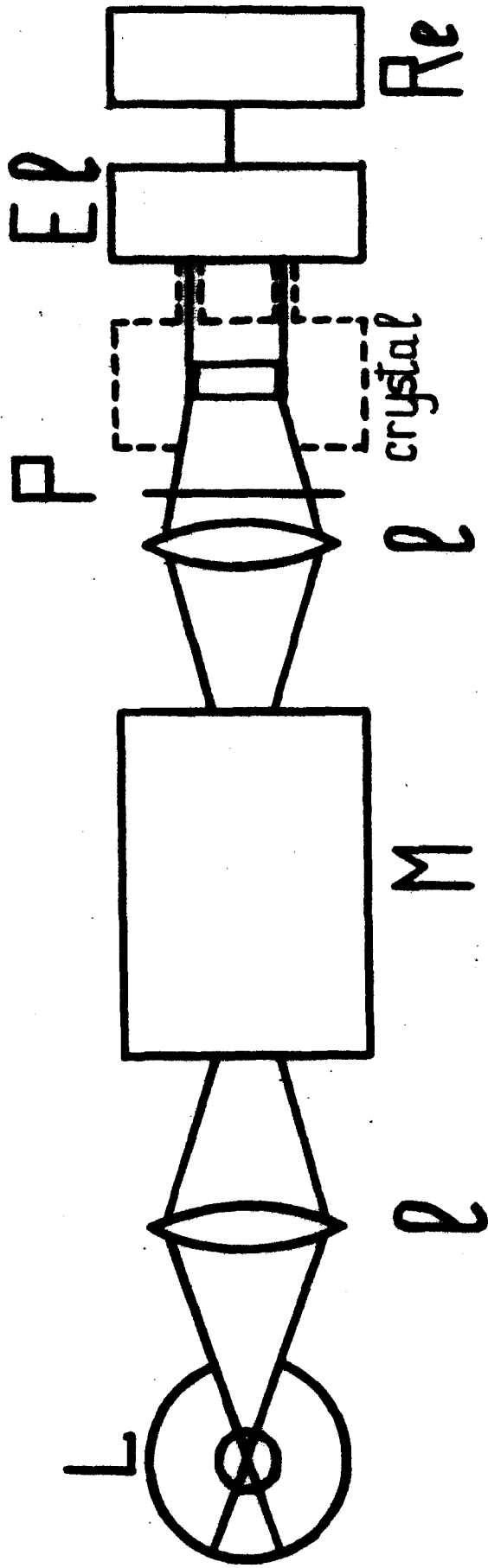


FIG. 1

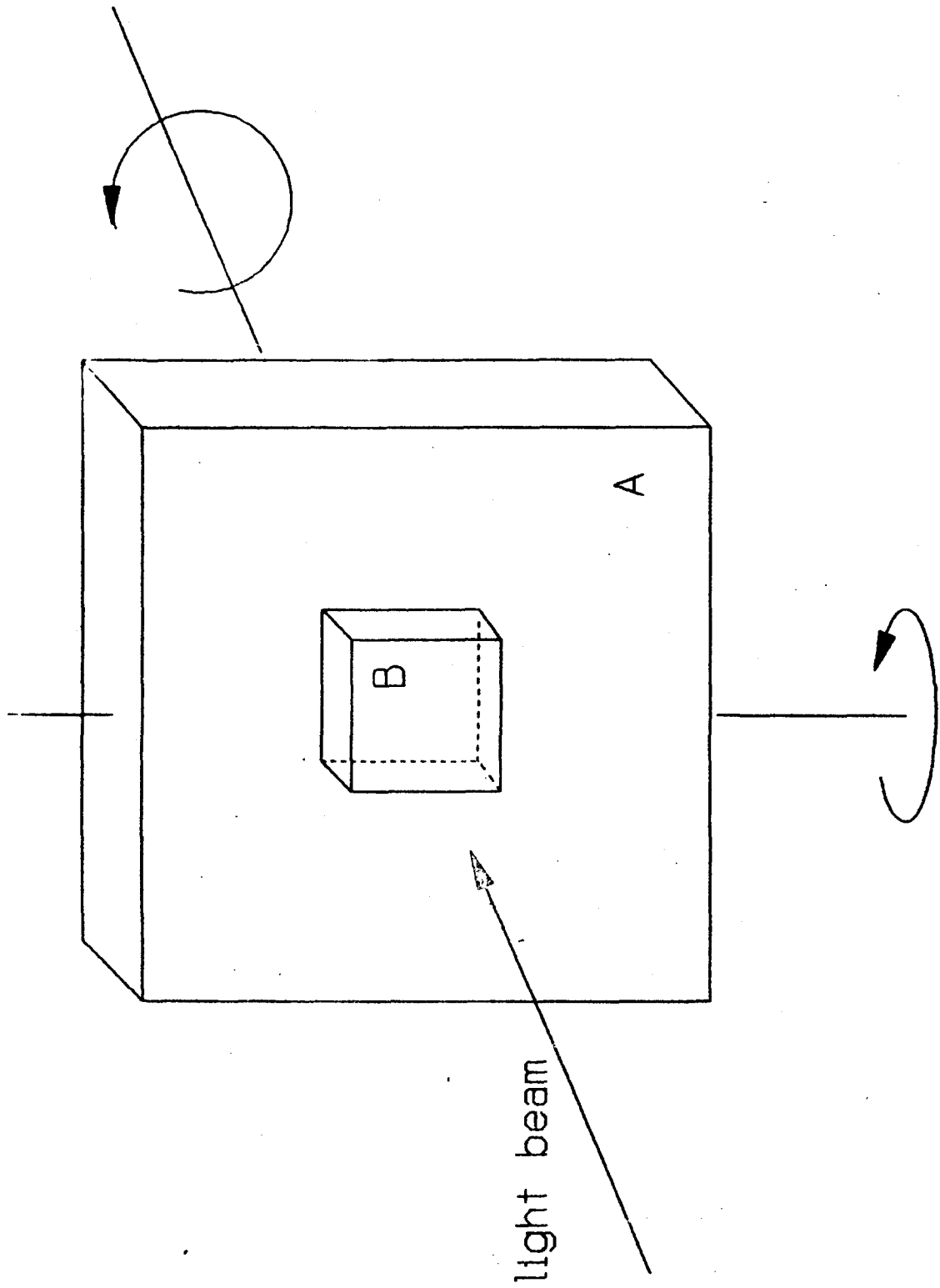


FIG. 2

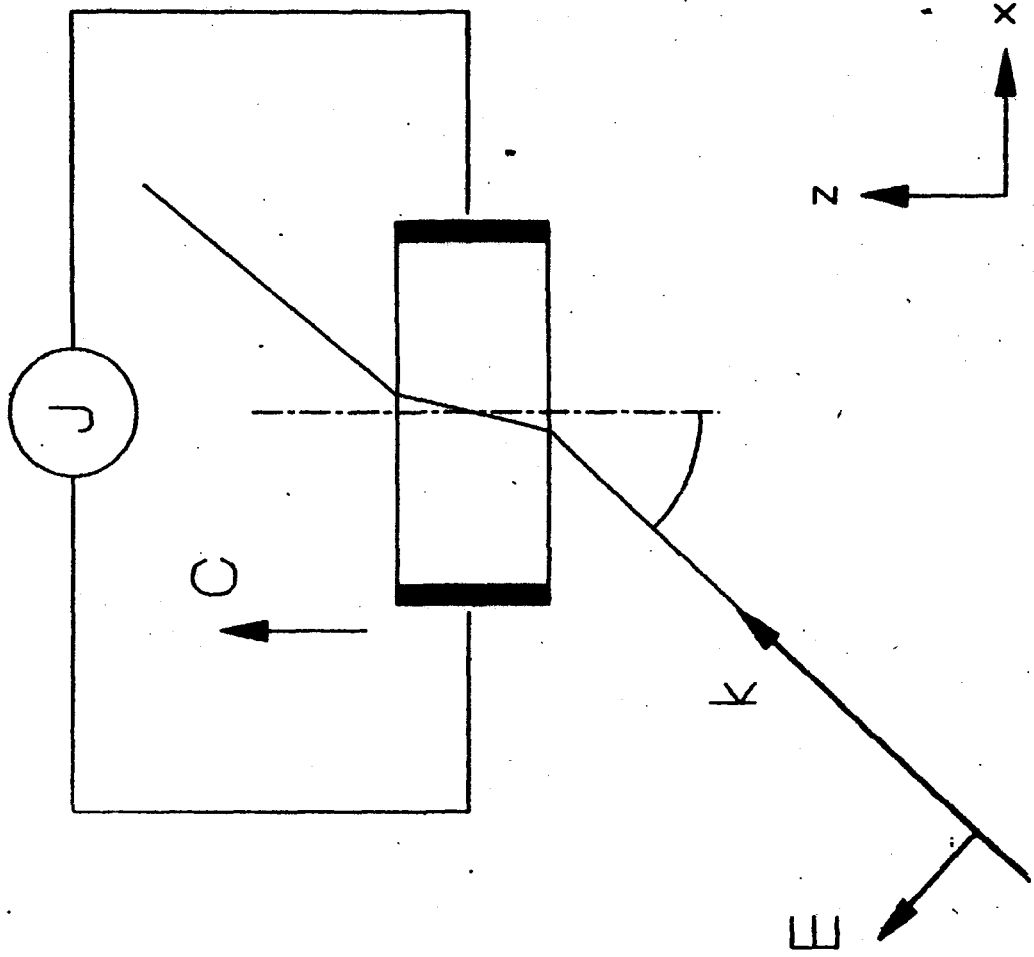


FIG. 3

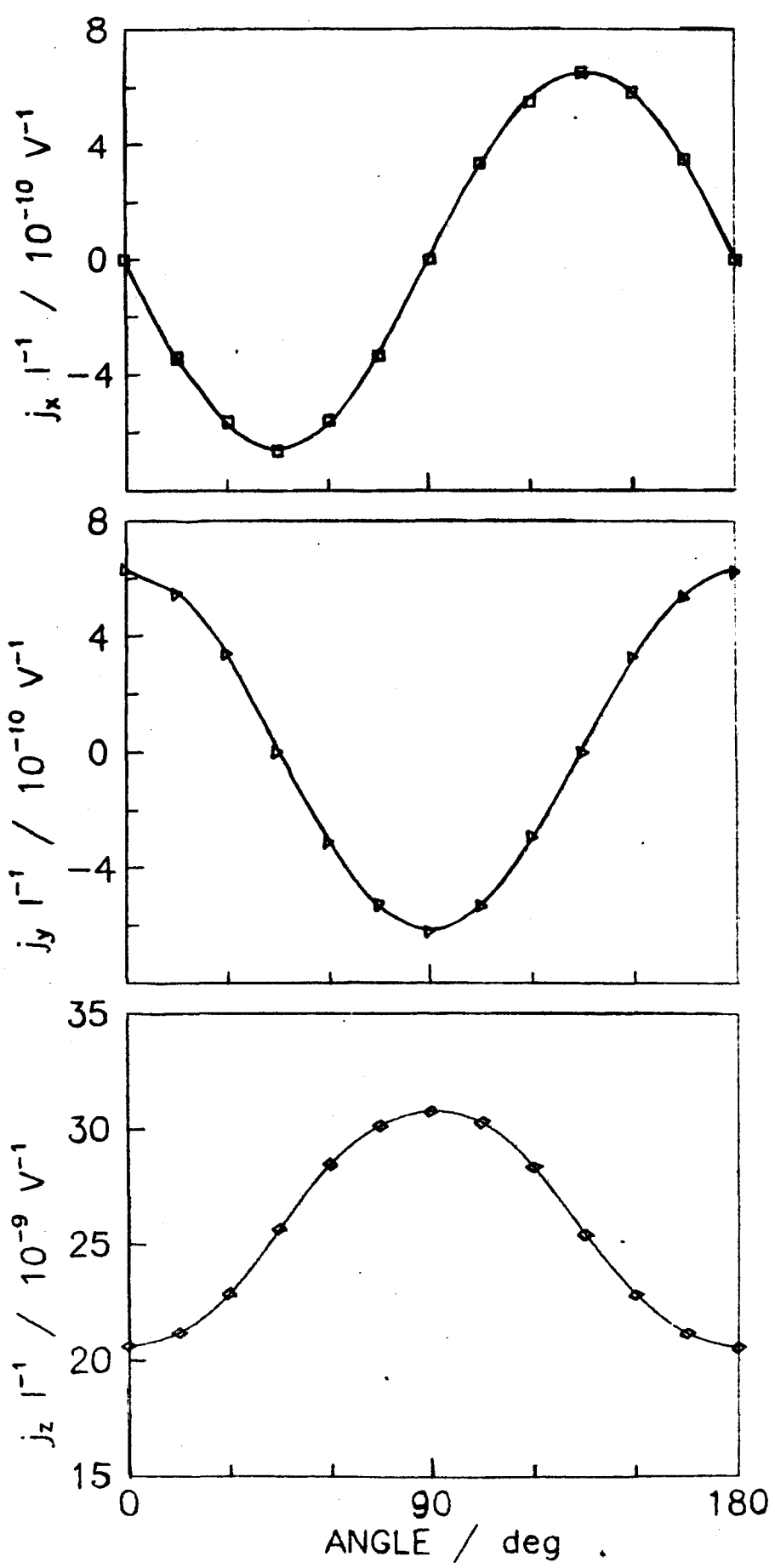


Fig. 4a

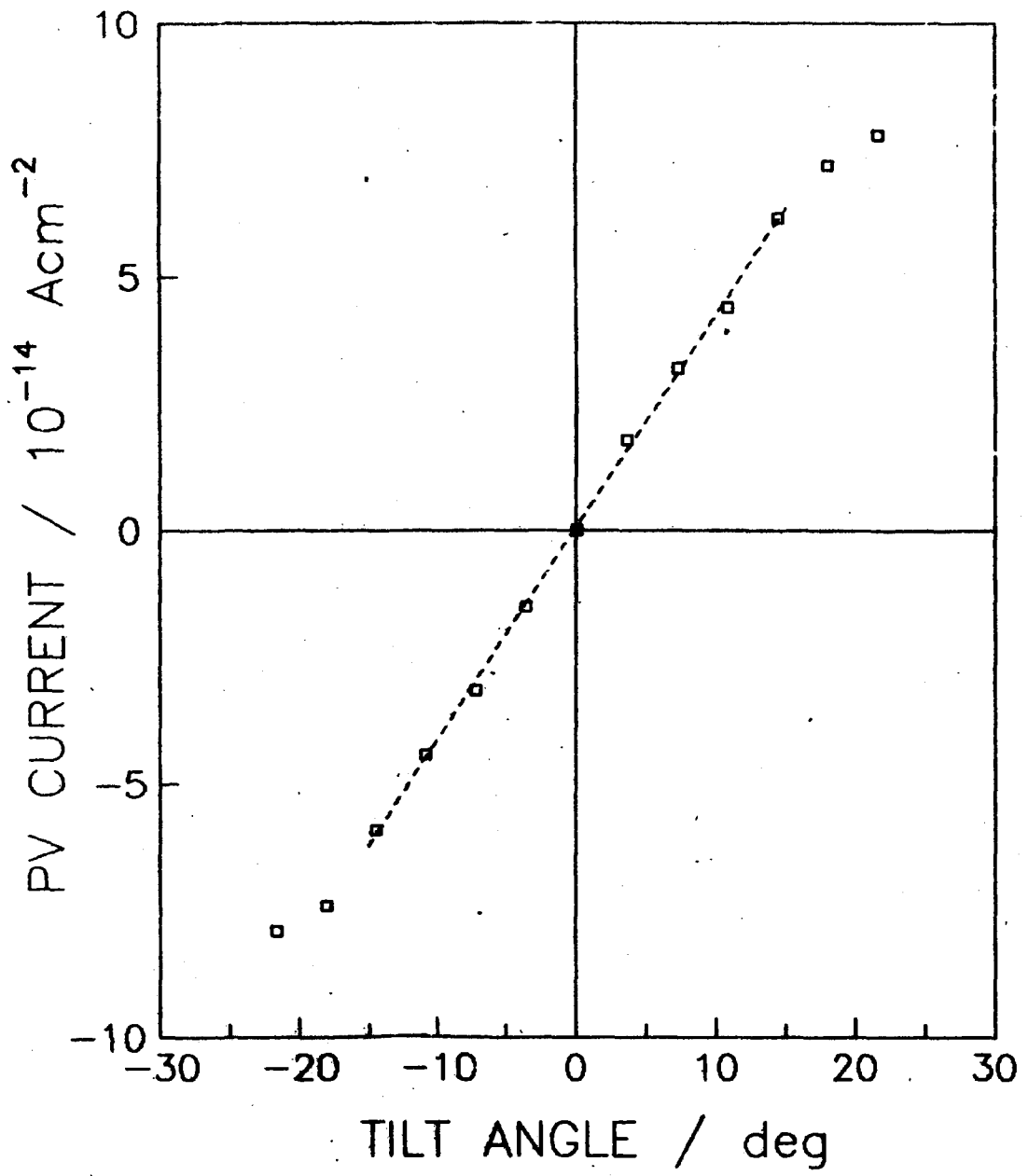


Fig.4b

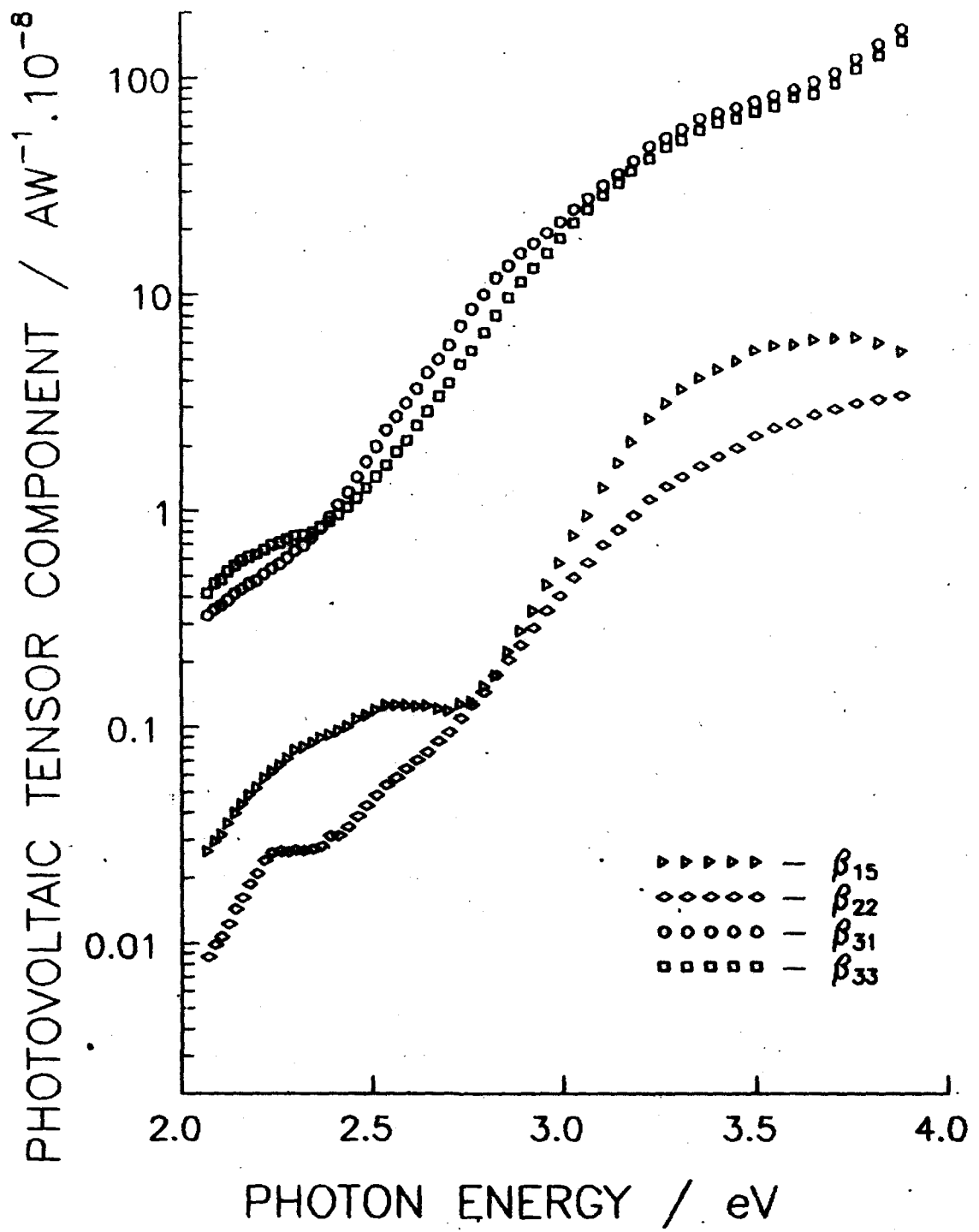


Fig.5

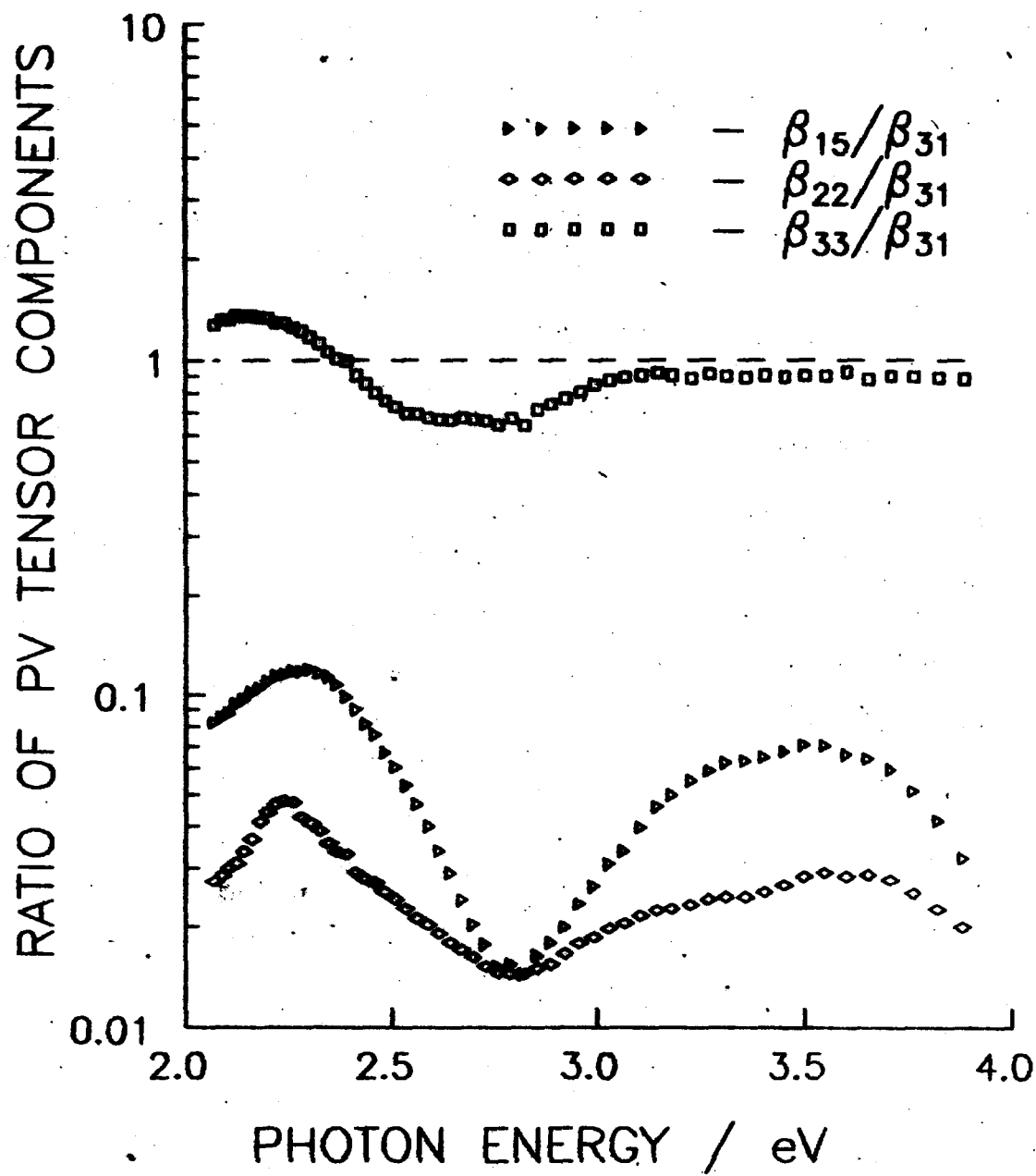


Fig.6

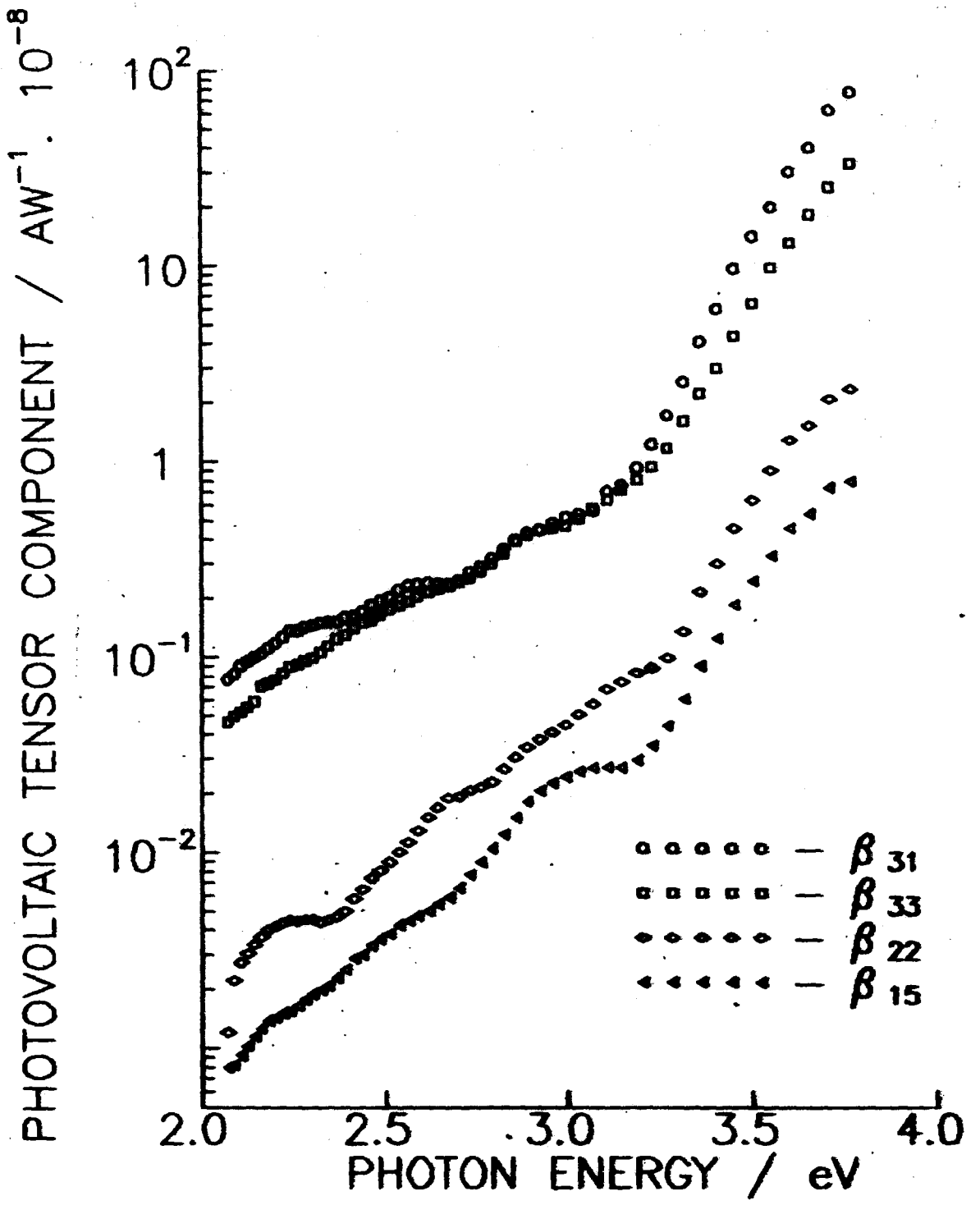


Fig. 7

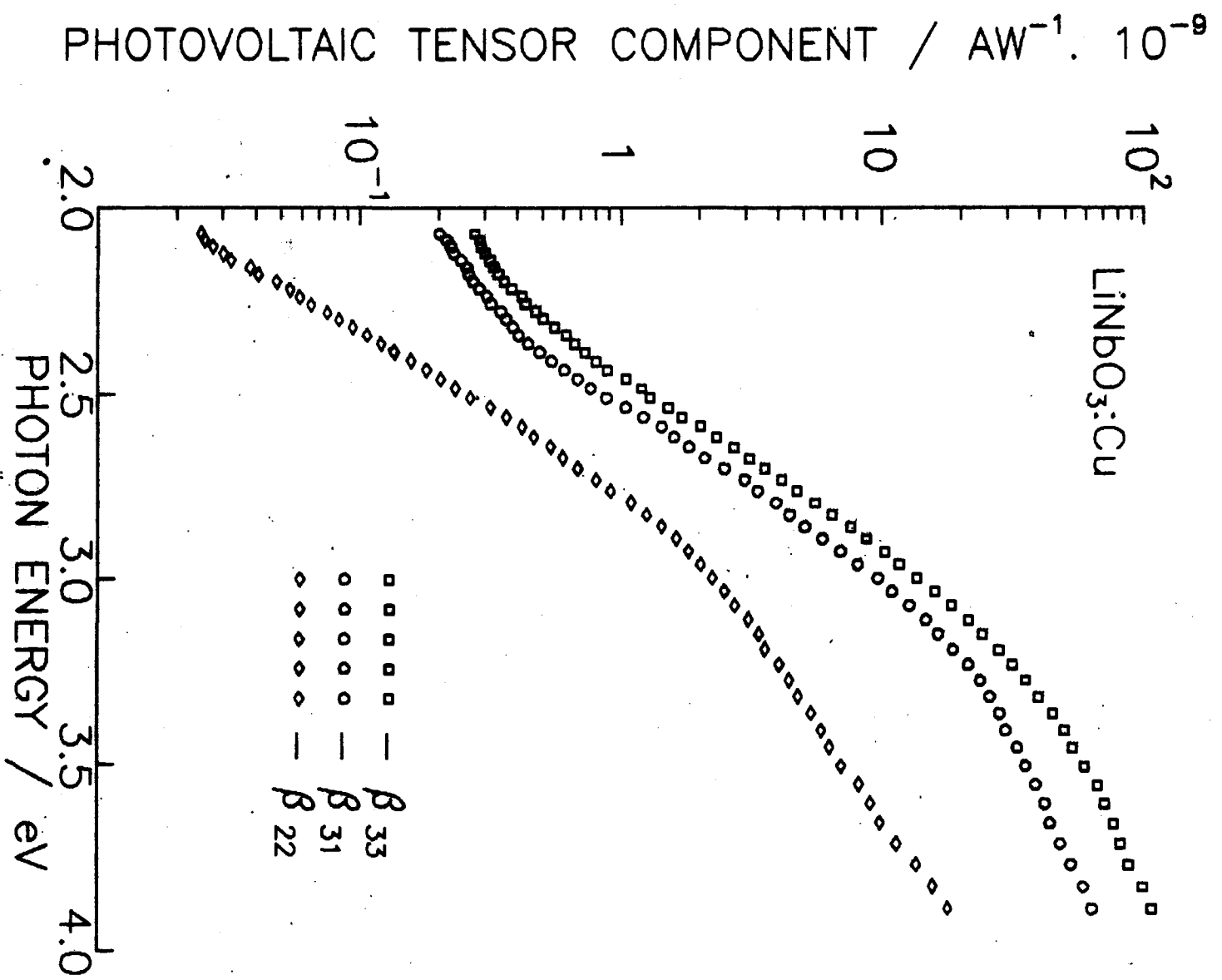


Fig. 8

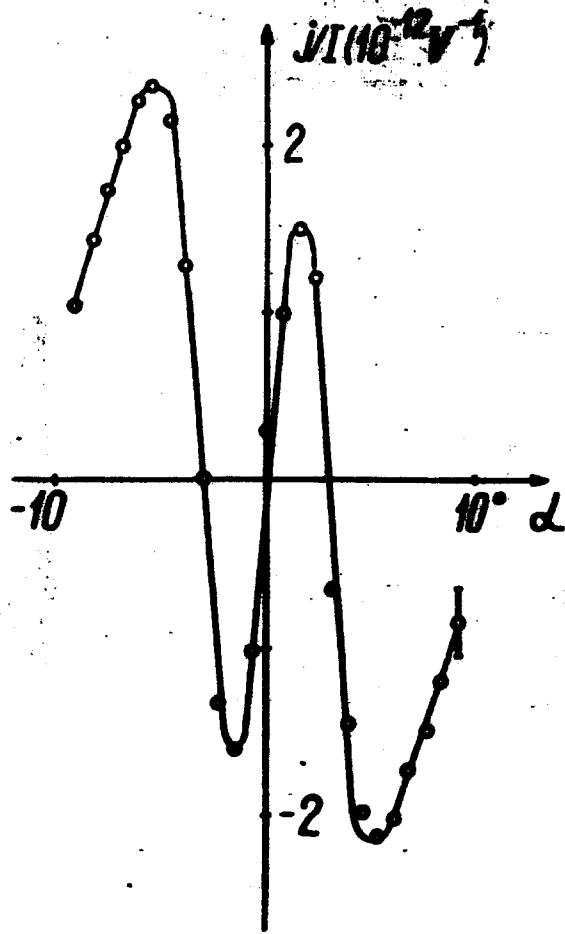


Fig.9

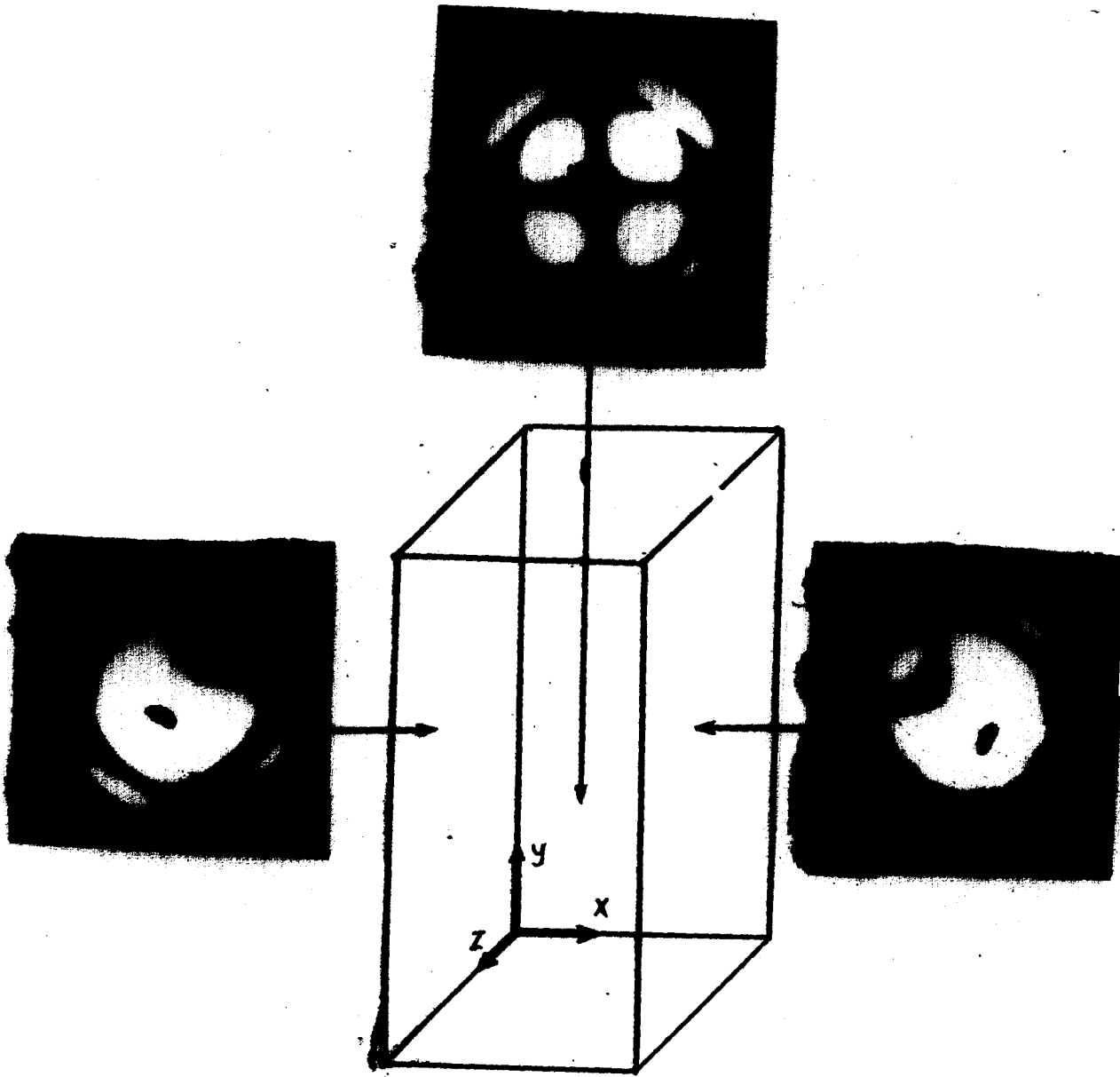


Fig. 10

Figure Captions

- Fig.1. The scheme of experimental arrangement for photovoltaic current measurements.
- Fig.2. Schematic representation of measuring head: Measuring head (A) with crystal (B) inside can be turned around two axes as shown by arrows.
- Fig.3. Experimental geometry for β_{15}^5 measurement.
- Fig.4. (a) - Photovoltaic current density versus polarization angle at 2.6 eV. Solid lines represent the best fit to calculated dependences.
$$j_x/I = -\beta_{22} \sin 2\theta \quad ,$$
$$j_y/I = \beta_{22} \cos 2\theta \quad \text{and} \quad j_z/I = \beta_{31} + (\beta_{33} - \beta_{31}) \cos^2 \theta .$$
In each curve θ is the angle between polarization vector and crystallographic axis. (b) - Angular dependence of photovoltaic current density j_x for photon energy 2.6 eV in $\text{LiTaO}_3:\text{Fe}$ (0.038 wt %).
- Fig.5. Spectra of photovoltaic tensor components for $\text{LiTaO}_3:\text{Fe}$ (0.038 wt %).
- Fig.6. Spectra of photovoltaic tensor component ratios in $\text{LiTaO}_3:\text{Fe}$ (0.038 wt %).
- Fig.7. Spectra of photovoltaic tensor components for $\text{LiNbO}_3:\text{Fe}$ (0.03 wt %).
- Fig.8. Spectra of β_{31} , β_{33} and β_{22} photovoltaic tensor components for $\text{LiNbO}_3:\text{Cu}$ (0.1 wt %).
- Fig.9. Angular dependence of photovoltaic current density j_x for photon energy 2.6 eV in $\text{LiNbO}_3:\text{Cu}$ (0.1 wt %).
- Fig.10. Conoscopic figures observed in $\text{LiNbO}_3:\text{Cu}$ prior to annealing.

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С.И. КАРАБЕЖЯН, В.В. ОБУХОВСКИЙ
НЕДИАГОНАЛЬНАЯ КОМПОНЕНТА ТЕНЗИОНА
ДЛИННОГО ФОТОВОЛЬТАИЧЕСКОГО ЭЛЕМЕНТА
В ПРИСВИСЫХ КРИСТАЛЛАХ LiNbO_3 И KLiO_3

(на английском языке, перевод Асрян З.П.)

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