Spontaneous electroabsorption in cadmium telluride and gallium arsenide

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(Submitted June 8, 1973)
Zh. Eksp. Teor. Fiz. **65**, 2007–2015 (November 1973)

Electroabsorption of plane polarized light near the band edge of CdTe and GaAs is investigated. The results of an harmonic analysis of light absorption in samples located in a strong alternating electric field are interpreted under the assumption that optical transitions occur in crystal regions with strong stationary anisotropic fields. From the aggregate of the experimental data it can be concluded that the fields are directed along a certain polarity of the ⟨111⟩ crystallographic axes.

The structure of the fundamental absorption edge in semiconductors depends on many factors: on the quantity and form of the impurities, on the temperature and the character of the phonon spectrum and so on. A major role in the formation of tails is played in particular by level-forming fluctuations of the impurity potential;[1-3] however, as has been shown in[4-6], the role of microfields in the formation of absorption-edge structure is not limited solely to the formation of levels in a forbidden band, since the fluctuating impurity potential can be perceived by the carriers as an external electric field, and the absorption of light in this field as the spontaneous Keldysh-Franz effect. It is natural that microfields arise not only in impurity fluctuations, but also in p-n junctions, in dislocations, in regions of residual stresses, at the surface and so forth.

Our aim in the present paper is to study the spontaneous electro-absorption[7-9] due to anisotropic microfields, which arise for one reason or another in cubic crystals without a center of inversion such as GaAs and CdTe.

EXPERIMENTAL METHOD

Electroabsorption in GaAs and CdTe was studied with the aid of a differential method described previously.[10] Monochromatic plane-polarized light was passed through the sample. The sample was either placed in a capacitor or ohmic contacts were been placed on it. A strong electric field $E = E_1 \cos \Omega t$ was applied to it perpendicular to the direction of propagation of the light ($\Omega = 500$ Hz).

The crystallographic planes and axes of polished single crystals of GaAs (resistivity at $300°K \rho = 10^8$ Ω-cm) and CdTe ($\rho = 10^4$ Ω-cm) were determined by x-ray structural analysis. The oriented samples were cut either in the shape of plane-parallel bars with dimensions $2 \times 2 \times 1$ mm (the 2×2 mm plane was polished) or in the shape of a cylinder with diameter 5 mm and height 1–2 mm.

The light was normally incident on the 2×2 mm plane of the bar and normal to the base of the cylinder. Rotating the cylinder made possible smooth variation of the direction of the external field relative to the crystallographic axes. Light transmitted through the investigated sample fell on a photodetector (FÉU-28). Two signals were taken from the load resistance of the photodetector—an alternating signal proportional to the change in the light intensity ΔI and arising as a result of the periodic variation of the absorption coefficient of the crystal under the action of the external sinusoidal

field, and a constant signal proportional to the intensity I of the light transmitted through the sample. Harmonic analysis of the periodic signal ΔI was carried out with the help of a U2-6 narrow-band amplifier. After amplification of both signals by means of a divider, the first signal was then divided by the second. As a result, the ratio $\Delta I/I$, obtained on a recorder with excellent accuracy, allowed us to establish the value of the change in the light absorption coefficient:

$$\Delta\alpha(E) = \alpha(E) - \lim_{E \to 0} \alpha(E). \tag{1}$$

EXPERIMENTAL RESULTS

The experimental investigations of the electroabsorption of GaAs (at temperatures of 300 and $80°K$) and CdTe (at $T = 80°K$) gave the following basic results.

1. Under the action of the external electric field $E_1 \cos \Omega t$, a complicated light modulation takes place in the studied samples, due to a shift in the absorption band edge.[7-9] The harmonic analysis of the signal showed that the light was modulated with the frequencies Ω, 2Ω, 3Ω and so forth.

2. For fixed values of the energy of the light quantum energy $\hbar\omega$, the changes in the light absorption coefficient $\Delta\alpha_1$ (at the frequency Ω) and $\Delta\alpha_2$ (at the frequency 2Ω) depend differently on the value of E_1. As is seen from Fig. 1a, the dependence $\Delta\alpha_1(E)$ for GaAs contains a linear segment. At the same time, as is seen from Figs. 1b, $\ln \Omega\alpha_2$ depends linearly on $\ln E_1$ with the slope 2, i.e., $\Delta\alpha_2 \sim E_1^2$. In[7] similar dependences were cited for CdTe.
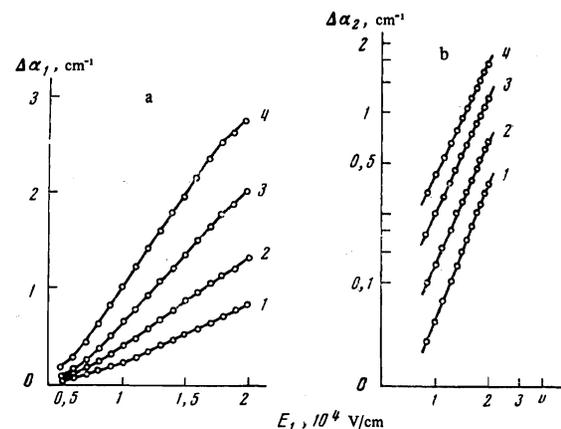
FIG. 1. Dependence of $\Delta\alpha_1$ (a) and $\Delta\alpha_2$ (b) on E_1 in GaAs for fixed values of $\hbar\omega$: 1 — $\hbar\omega = 1.368$ eV; 2 — $\hbar\omega = 1.372$ eV; 3 — $\hbar\omega = 1.376$ eV; 4 — $\hbar\omega = 1.380$ eV.
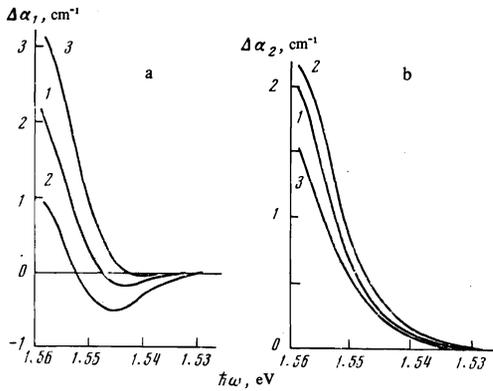
FIG. 2. Dependence of $\Delta\alpha_1$ (a) and $\Delta\alpha_2$ (b) on $\hbar\omega$ in CdTe at $E_1 = 10^4$ V/cm; $1 - E_c = 0$, $2 - E_c = 10^3$ V/cm, $3 - E_c = 10^3$ V/cm.

3. If, in addition to the external electric field of the form $E_1 \cos \Omega t$, a constant bias E_c acts on the crystal, then the ratio of the quantities $\Delta\alpha_1/\Delta\alpha_2$ and $\Delta\alpha_3/\Delta\alpha_2$ changes.

Figure 2a shows curves of $\Delta\alpha_1$ vs. $\hbar\omega$ for two opposite directions of E_c in CdTe; it is seen that the sign and the value of the change $\Delta\alpha_1$ depend on the direction and magnitude of E_c. The analogous changes in $\Delta\alpha_2$ have the opposite sign (Fig. 2b).

4. Measurements carried out on samples of cylindrical shape showed that the value of the ratio $\Delta\alpha_1/\Delta\alpha_2$ depends on the orientation of the external electric field relative to the crystallographic axes of the crystal. Here the character of the change in the quantity depends on the orientation of the plane of polarization of the plane-polarized light relative to the direction of the external field.

In the case in which the base of the cylinder lies in the (110) plane and the light polarization vector $e \parallel E_1$, a dependence of $\Delta\alpha_1$ on φ is obtained, where φ is the angle between the [110] axis and the direction of E_1 (Fig. 3a). As is seen from the figure, the amplitude of the fundamental absorption harmonic is maximal in the $\langle 111 \rangle$ directions and the phases of the signal are opposite for opposite directions of the same axis.

The analogous dependence for light with $e \perp E_1$ has a somewhat different shape (Fig. 3b).

Simultaneously measured values of the second harmonic showed that $\Delta\alpha_2$ is almost independent of φ.

DISCUSSION OF RESULTS

The Keldysh-Franz effect has been studied in[11-13] for straight bands of a semiconductor. In particular, expressions have been obtained for the electroabsorption for all critical points of the spectrum. In the paper by Franz,[14] the case of a diffuse edge has been investigated. It was found that in all cases of semiconductors with vertical bands (the point M_0 according to the van Hoff classification), which included GaAs and CdTe the value of $\Delta\alpha(E)$ is a function of $|E|$. Therefore, if an external field $E \cos \Omega t$ is applied to samples of GaAs or CdTe, then $\Delta\alpha(|E|)$ ought to contain only even harmonics in Ω. The odd harmonics of $\Delta\alpha$ ought to appear in the presence of a constant external bias E_c.[15]

The presence of odd harmonics in the absence of an external bias allows us to assume that the role of the constant component of the field is taken by microfields
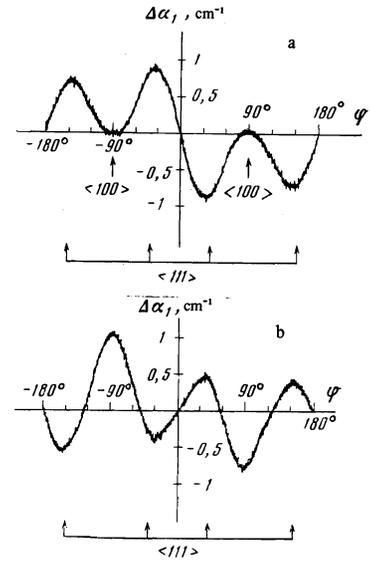
FIG. 3. Dependence of $\Delta\alpha_1$ of plane-polarized light on the orientation of E_1 relative to the crystallographic axes in the (110) plane, measured on a sample of GaAs of cylindrical shape: $\hbar\alpha = 1.376$ eV; $E_1 \approx 10^4$ V/cm; a – $e \parallel E_1$; b – $e \perp E_1$.

that arise for one reason or another in the crystal. However, for the generation of odd harmonics of $\Delta\alpha(E)$ the fact of the existence of an internal field is alone insufficient.

We assume that there corresponds a certain volume ΔV_{E_0} to each value of the intensity of the microfield E_0 in the crystal. Then $\Delta\alpha$ can be expressed in the form of a sum:

$$\Delta\alpha(E_1) = \frac{1}{V} \sum_{E_0} \Delta V_{E_0} \Delta\alpha(|E_0 + E_1 \cos \Omega t|), \qquad (2)$$

and the expression for the amplitude of the first harmonic will have the form

$$\Delta\alpha_1(E_1) = \frac{1}{\pi V} \sum_{E_0} \Delta V_{E_0} \int_0^{2\pi} \alpha(|E_0 + E_1 \cos \Omega t|) \cos \Omega t \, d\Omega t$$

$$= \frac{1}{\pi V} \sum_{E_0, \vartheta > \pi/2} (\Delta V_{E_0} - \Delta V_{-E_0}) \int_0^{2\pi} \alpha(E) \cos \Omega t \, d\Omega t \qquad (3)$$

Here V is the volume of the sample and ϑ is the angle between E_0 and E_1.

It is clear that if $\Delta V_{E_0} = \Delta V_{-E_0}$, then $\Delta\alpha_1 = 0$. Thus, the fact that we need asymmetry of the microfield for the presence of odd harmonics, i.e.,

$$\Delta V_{E_0} \neq \Delta V_{-E_0}. \qquad (4)$$

is a direct consequence of (3).

When the macropotential is a constant quantity over the entire sample, the condition (4) indicates a different rate of change of the micropotential for opposite walls of the potential well. It is quite evident that the expression (3) is valid only when the conditions of quasi-classicity are satisfied.

If we assume that

$$E_1/E_0 \ll 1, \qquad (5)$$

then the expression for the amplitude of the first harmonic is obtained by simple differentiation (2) with respect to the alternating field.

For the case in which we can neglect the fact of the

diffuseness of the absorption-band edge, the calculations of Aspnes[13] are valid. According to[13],

$$\Delta\alpha(E) = R\theta^{1/2}\{Ai'^2(\eta) - \eta Ai^2(\eta)\} = R\sqrt{\frac{\Delta\hbar\omega}{\hbar}}F(\eta),\qquad(6)$$

where

$$\eta = \frac{\Delta\hbar\omega}{\hbar\theta},\qquad \theta^3 = \frac{e^2E^2}{2\mu\hbar},\qquad \Delta\hbar\omega = E_g - \hbar\omega,$$

$Ai(\eta)$ is the Airy function, μ the reduced mass of the carriers, R some constant quantity.

Setting $\Delta V_{E_0} = V$, we obtain the following expressions, depending on the setup of the problem: for the case $E_0 = $ const

$$\Delta\alpha_i(\eta, E_0, E_i) = \frac{1}{3}R\left(\frac{e^2}{2\mu\hbar}\right)^{1/6}(Ai'^2 + \eta Ai^2)\frac{E_i}{E_0^{2/3}}\cos\vartheta;\qquad(7a)$$

for the case $\Delta\hbar\omega = $ const

$$\Delta\alpha_i(\eta, \Delta\hbar\omega, E_i) = \frac{1}{3}R\left(\frac{e^2\hbar}{2\mu}\right)^{1/2}\eta(Ai'^2 + \eta Ai^2)\frac{E_i}{\Delta\hbar\omega}\cos\vartheta.\qquad(7b)$$

The difference between the functions $F(\eta)$ and $\eta(Ai'^2 + \eta Ai^2) \equiv \Phi(\eta)$ is important in the limit $\eta \to 0$:

as $\eta \to 0$, $F(\eta) \to F_{Max}$, $\Phi(\eta) \sim \eta \to 0$,

if $\eta \gg 1$ then $\Phi(\eta) \sim \eta^{1/4}\exp(-4/3\eta^{3/2})$. (8)

The function $\Phi(\eta)$ is easily tabulated[16] and its graph is shown in Fig. 4. The presence of a sharply expressed maximum on the $\Phi(\eta)$ curve allows us to affirm that under otherwise equivalent conditions the largest contribution to the first harmonic of the absorption will be made by that region of the microfield ($E_{eff}, \Delta E_0$) which corresponds to the halfwidth $\Delta\eta$ of the function $\Phi(\eta)$:

$$E_{eff} = \sqrt{\frac{2\mu}{e^2\hbar^2}}(\Delta\hbar\omega)^{1/2}\eta_{ext}^{-3/2}.\qquad(9)$$

Limitation of the absorption at the first harmonic at large values of E_0 improves to some degree the conditions for validity of the representation of the absorption in the form (3). Limitation of the absorption for small values of E_0 (the limit $\eta \gg 1$) cuts off the region of the microfield in which the condition (5) is definitely known not to be satisfied.

For a definite $\Delta\hbar\omega_i$ the absorption in the interval ($E_{eff}, \Delta E_0$) determines a certain excess volume:

$$\Delta V(\Delta\hbar\omega, \bar\vartheta) = \frac{1}{\cos\bar\vartheta}\sum_{\Delta E_0, \vartheta < \pi/2}(\Delta V_{E_0} - \Delta V_{-E_0})\cos\vartheta.\qquad(10)$$

Here $\bar\vartheta$ is the angle between E_1 and the axis of maximum asymmetry.

Thus, for the case of absorption at the first harmonic we get the expression

$$\Delta\alpha_i(\Delta\hbar\omega, E_i) \sim \frac{1}{V\Delta\hbar\omega}\Phi(\eta_{ext})\Delta V(\Delta\hbar\omega, \bar\vartheta)E_i\cos\bar\vartheta.\qquad(11)$$

If we take into account only one type of carrier in the valence band,[17-19] then, in accord with (11), the value of $\Delta\alpha_1$ should depend linearly on E_1 for a constant $\hbar\omega$. The slope of this linear segment determines the excess volume $\Delta V(\Delta\hbar\omega, \bar\vartheta)$. In the presence of a definite model
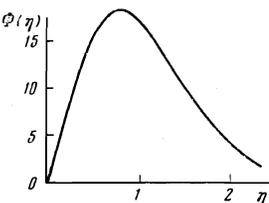
FIG. 4. Dependence of the weighting function Φ on η.

of generation of the microfield, the dependence $\Delta V(\Delta\hbar\omega, \bar\vartheta)$ would allow us to determine the character of the micropotential.

Measurements of the dependence of $\Delta\alpha_1$ on E_1 (Fig. 1a) show that $\Delta\alpha_1(\Delta\hbar\omega, E_1)$ in fact contains segments that are linear in E_1, and the slopes fall off with increase in $\Delta\hbar\omega$. However, one important difference is the fact that the linear segments do not extrapolate to the origin of the coordinates.

At least two different explanations of this fact are possible. One point of view is based on the assumption of screening of the external field by the carriers. Since the signals that we observed at the fundamental originate only from those regions of the microfield into which the external alternating field penetrates, then the intensity E_1 in these regions decreases to the value E_1^*, i.e., it is assumed that the field $E_1 - E_1^*$ removes the screening in the region of the microfield $E_{eff}(\Delta\hbar\omega)$. Then extrapolation of the linear segments as $\Delta\alpha_1 \to 0$ cuts off those values of $E_1 - E_1^*$ for which the screening of the field $E_{eff}(\Delta\hbar\omega)$ is removed, and since larger E_0, screen more strongly, $E_1 - E_1^*$ increases with an increase in $\Delta\hbar\omega$ according to (9),

The other possible explanation is based on the representation of the microfield as a nonsymmetric potential well with a large number of levels. If the width of the well is d, then the quantity dE_1 determines the excess of one edge of the well over the other, or that portion of the microfield which becomes accessible for carriers not localized in the well. On reversal of the external field, different regions ΔV will be released because of the nonsymmetric character of the well; here $\Delta V_{E_0} \neq \Delta V_{-E_0}$.

Study of the absorption at the second harmonic has shown that $\Delta\alpha_2 \sim E_1^2$ (Fig. 1b), i.e., in the real case, when the edge of the absorption band is diffuse, the procedure of expansions similar to (5)—(7) is valid right up to terms of second order in E_1.

As follows from what has been said above, the odd harmonic of the absorption originates from a certain excess volume of the microfield $\Delta V(\Delta\hbar\omega, \bar\vartheta, E_1)$. If we apply a constant bias E_C to the sample in the direction ($\bar\vartheta, E_1$). If we apply a constant bias E_C to the sample in the direction $\bar\vartheta = \pi$, then we can choose its value such that the absorption at the odd harmonics will vanish for definite energy deficits $\Delta\hbar\omega$ (Fig. 2a). Since the application of a constant bias increases the symmetry of the field inside the crystal, this leads to a decrease in the first harmonic. The second harmonic increases in this case (Fig. 2b), since the volume of the symmetric part of the constant field in the crystal increases.

On reversal of the direction of E_C the volume of the asymmetric part of the field increases, leading to an increase in the first harmonic (Fig. 2a) and a decrease in the second harmonic (Fig. 2b).

Investigation of the absorption of polarized light by samples of cylindrical shape showed that while $\Delta\alpha_2$ is practically independent of the orientation of the electric field relative to the crystallographic axes, the value of $\Delta\alpha_1$ clearly follows the symmetry of the crystallographic plane chosen (for example, (110), Fig. 3). To explain this, let us assume that the microfield intensities responsible for the generation of the second harmonic (at a definite $\Delta\hbar\omega$), are oriented in the direction of some definite polarity of the $\langle111\rangle$ axes. Two $\langle111\rangle$ axes lie

in the (110) plane ($k = 1, 2$), and two others ($k = 3, 4$) lie in a plane perpendicular to the given plane and project onto [100] in the direction opposite to the projections of the first two axes. Here it is also assumed that each specific microfield E_0 is oriented along one of the four $\langle 111 \rangle$ axes and that the contributions $\Delta V_{E_{0k}}$ of each of these axes is the same.

We first take into account only that fraction of the light absorption for which the polarization vector $e \parallel E_{0k}$. The absorption of light polarized along the k axis that takes place in the microfield E_{0k} can be represented in the form of a superposition of the components polarized along the axes of a Cartesian coordinate system (x, y, z). The amplitudes of these components ought to be proportional to the squares of the corresponding projections of the polarization vector $e_k(e_{kx}, e_{ky}, e_{kz})$. The absorption of the light polarized along the kth axis in the microfield connected with the kth axis will be proportional to the vector $n_k(e_{kx}^2, e_{ky}^2, e_{kz}^2)$.

We choose a set of coordinates such that $x \parallel E_1$, $y \parallel E_1$ and are in the (110) plane, and $z \parallel [x, y]$ and is directed along the direction of light propagation. In such a set of coordinates

$$n_1(\cos^2(\varphi+\varphi_0), \quad \sin^2(\varphi+\varphi_0), \quad 0),$$

$$n_2(\cos^2(\varphi-\varphi_0), \quad \sin^2(\varphi-\varphi_0), \quad 0), \qquad (12)$$

$$n_{3,4}(^1/_3\cos^2(\varphi+\pi/2), \quad ^1/_3\sin^2(\varphi+\pi/2), \quad ^2/_3),$$

where φ_0 is the angle between the [111] and [110] axes. Thus, if we take into account the absorption of light polarized only along the directions of asymmetry of the separate fields (i.e., along $\langle 111 \rangle$), then, following (7), we can write down the first absorption harmonic in the form

$$\Delta\alpha_1^{\parallel}(\varphi) = \left(-n_1\cos(\varphi+\varphi_0)+n_2\cos(\varphi-\varphi_0)-\frac{2}{\sqrt{3}}n_{3,4}\sin\varphi\right)\frac{\partial\alpha}{\partial E^2}E_0E_1. \quad (13)$$

Here $\Delta\alpha_{1x}$ determines the absorption of light with $e \parallel E_1$ and $\Delta\alpha_{1y}$ with $e \parallel E_1$.

If we take into account only the absorption of light with $e \perp E_{0k}$, then, similar to (12)–(13), we can obtain an expression for $\Delta\alpha_1^{\perp}(\varphi)$. If it is now assumed that the absorption in the electric field does not depend on the orientation of the plane of light polarization relative to the intensity of the electric field, then it is easy to show that

$$\Delta\alpha_{1x}^{\parallel}(\varphi) = -\Delta\alpha_{1x}^{\perp}(\varphi), \quad \Delta\alpha_{1y}^{\parallel}(\varphi) = -\Delta\alpha_{1y}^{\perp}(\varphi), \quad (14)$$

i.e., there should be no absorption at the first harmonic.

Thus, we can state that if the microfields are oriented along the $\langle 111 \rangle$ directions and that the weight of each orientation is the same, then the single condition (5) is insufficient for the generation of the odd harmonics. Satisfaction of the following condition is also necessary:

$$\Delta\alpha_1^{\parallel}(\varphi) \neq \Delta\alpha_1^{\perp}(\varphi). \quad (15)$$

As is well known,[17-19] down to very small values of $\Delta\hbar\omega$, when the contributions of light and heavy holes are equal, the electroabsorption in GaAs and CdTe is polarized, i.e., $\Delta\alpha^{\parallel} > \Delta\alpha^{\perp}$.

Taking (15) into account, it is easy to get the angular dependence of the absorption of polarized light at the fundamental from (13):

$$\Delta\alpha_{1x}(\varphi) \sim \cos^2\varphi \sin\varphi, \quad (16)$$

$$\Delta\alpha_{1y}(\varphi) \sim (1-3\cos^2\varphi)\sin\varphi. \quad (17)$$
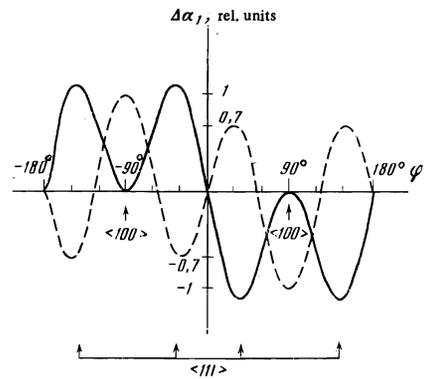
FIG. 5. Angular dependences of $\Delta\alpha_1$, constructed from Eqs. (16) and (17): solid line — for plane-polarized light with $e\parallel E_1$ (the function $\Delta\alpha_{1x}(\varphi)$); broken line — $e \perp E_1$ (the function $\Delta\alpha_{1y}(\varphi)$).

Figure 5 shows the functions $\Delta\alpha_{1x}(\varphi)$ and $\Delta\alpha_{1y}(\varphi)$. As is seen from the graphs, they are in qualitative agreement with the analogous experimental dependences (Figs. 3a and 3b).

CONCLUSION

As a result of the investigations described above, we can conclude the following:

1. The presence of odd harmonics of the absorption indicates the existence of asymmetric regions of constant field inside high-ohmic single crystals of GaAs and CdTe.

2. The intensity E_0 of these fields is greater than the intensities of the external fields $E_1 \approx 2 \times 10^4$ V/cm.

3. The observation of odd harmonics, i.e., the detection of asymmetric regions of constant fields in samples, is possible only at intensities E_1 that are larger than certain critical values. These critical values of E_1 depend on the energy deficit $\Delta\hbar\omega$.

4. The dependences of the absorption of plane-polarized light at the first harmonic on the orientation of the external field relative to the crystallographic axes indicates the orientation of the asymmetry of the microfields along the $\langle 111 \rangle$ axes.

5. If the microfields are directed along a definite polarity of the $\langle 111 \rangle$ axes or in any other symmetric fashion, then the first harmonic can arise only as a result of the fact that the absorption of light polarized along the microfield differs from that polarized in a direction perpendicular to it.

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Translated by R. T. Beyer
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