

DYNAMIC EFFECTS OF ELECTRO- AND PIEZOREFLECTANCE OF LIGHT BY CRYSTALS

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1. INTRODUCTION

WE can distinguish two facts that explain the especial interest that has been shown in the recently-discovered effects in solid-state physics, electro- and piezoreflectance of light. The first and major of these involves the essence of the topic: in recent years people have begun to solve the problem of complete reconstruction of the energy spectrum of the electrons in the higher energy bands in a crystal (see, e.g. [1,2]). Naturally, the experimental studies along this line are not based on using transfer phenomena, as in studying the Fermi surfaces of metals. Rather, they are based on applying various optical methods (ordinary optical spectroscopy, magnetooptics, and photoelectric emission) involved with electronic states rather far from the Fermi level. Consequently, the discovery and use of new physical effects or new experimental methods of this type has become all the more interesting. The second fact involves the methodological side of the problem: discovery of the effects of electro- and piezoreflectance of light proved possible because of a marked breakthrough in the experimental technique of determining small changes in the intensity of reflected light. Use of dynamic methods of measurement has made it possible to detect signals corresponding to $\Delta R/R \sim 10^{-6}$. Here ΔR is the change in the reflected-light intensity R that appears when a periodically-varying magnetic field, [3] electric field, [4] or elastic stress [5] is applied to the specimen. Naturally, the increase in sensitivity by 2-3 orders of magnitude attained by using dynamic methods opens up fundamentally new possibilities of studying the band structure of solids by the methods of electro- and piezoreflectance of light.

By principle, using any optical effect to study the band structure of a solid is analogous to the methods of atomic spectroscopy. The experimental curves of the effect as a function of the frequency of the transmitted or reflected light reveal certain features, which can then be identified with certain interband transitions, and one can get quantitative and qualitative information on the electronic energy spectrum of the upper filled and nearest unfilled bands. The essence of the dynamic optical methods amounts to "shifting" or varying the amplitude of the given feature by some periodic external agent, and reducing the problem of detecting it to the detection by a light receiver of an alternating signal having the assigned frequency and phase. This greatly extends the possibilities of the optical methods with respect to the number of observable features, their fine structure, the effect of external fields, the temperature, etc. As applied to the electro- and piezoreflectance effects, I can illustrate this with the example of features involving the existence of Van Hove singularities.

The notion of critical points on the curve of the density of quasiparticle states in a crystal was introduced by Van Hove, [6] and Phillips [7] has applied these ideas to the electronic energy spectrum of a crystal, and has made a complete classification of critical points. We shall see what effect is exerted by different types of Van Hove singularities on the frequency-dependence of the optical characteristics of a crystal in the region of the direct interband transitions.

In its general form, the expression for the imaginary component of the dielectric constant $\epsilon = \epsilon_1 + i\epsilon_2$ arising from interband transitions can be written as follows: [8]

$$\epsilon_2(\omega) = \frac{e^2 \hbar^2}{8\pi^2 m} \sum_{jj'} \int_S \frac{f_{jj'}(\mathbf{k})}{E_{jj'} |\nabla_{\mathbf{k}} E_{jj'}|} dS.$$

The surface S is one of the isoenergetic interband surfaces fixed by the condition:

$$\hbar\omega = E_{j'}(\mathbf{k}) - E_j(\mathbf{k}) = E_{jj'}.$$

The summation is performed over all pairs of bands between which transitions are allowed. If we assume that the oscillator strength for a given pair of bands is independent of \mathbf{k} , we get

$$\epsilon_2(\omega) = \frac{2\pi \hbar e^2}{m\omega} \sum_{jj'} f_{jj'} \left(\frac{dN}{dE_{jj'}} \right)_{\hbar\omega = E_{jj'}}.$$

where

$$\frac{dN}{dE_{jj'}} = \frac{1}{(2\pi)^3} \int \frac{dS}{|\nabla_{\mathbf{k}}(E_{j'} - E_j)|}.$$

Evidently, the critical points of the interband density of states $dN/dE_{jj'}$, and correspondingly of $\epsilon_3(\omega)$, are determined by the relation

$$\nabla_{\mathbf{k}}(E_{j'} - E_j) = 0.$$

The analytic behavior of $dN/d\omega$ near the Van Hove singularities can be found by expanding in series $E_{j'}(\mathbf{k}) - E_j(\mathbf{k})$ or $\omega_{jj'}(\mathbf{k})$ in the neighborhood of \mathbf{k}_0 under the condition $|\nabla_{\mathbf{k}} \omega_{jj'}(\mathbf{k})|_{\mathbf{k}=\mathbf{k}_0} = 0$:

$$\omega_{jj'}(\mathbf{k}) = \omega_c + \sum_{\alpha=1}^3 a_{\alpha} \epsilon_{\alpha} \Delta k_{\alpha}^2, \\ \epsilon_{\alpha} = \pm 1, \Delta k = k - k_0.$$

A critical point of type M_0 (minimum density of states) arises from the condition $\epsilon_1 = \epsilon_2 = \epsilon_3 = 1$, and the density of states near M_0 is determined by the relations

$$\frac{dN}{dE_{jj'}} = \begin{cases} c + O(\omega - \omega_c) & \text{when } \omega < \omega_c, \\ c + A(\omega - \omega_c)^{1/2} + O(\omega - \omega_c) & \text{when } \omega > \omega_c. \end{cases}$$

Correspondingly, an M_3 point (maximum density of states) is defined by:

$$\epsilon_1 = \epsilon_2 = \epsilon_3 = -1,$$

$$\frac{dN}{dE_{jj'}} = \begin{cases} c + A(\omega_c - \omega)^{1/2} + O(\omega - \omega_c) & \text{when } \omega < \omega_c, \\ c + O(\omega - \omega_c) & \text{when } \omega > \omega_c. \end{cases}$$

The behavior in the neighborhood of saddle or hyperbolic points of the type M_1 ($\epsilon_1 = \epsilon_2 = -\epsilon_3 = 1$) and

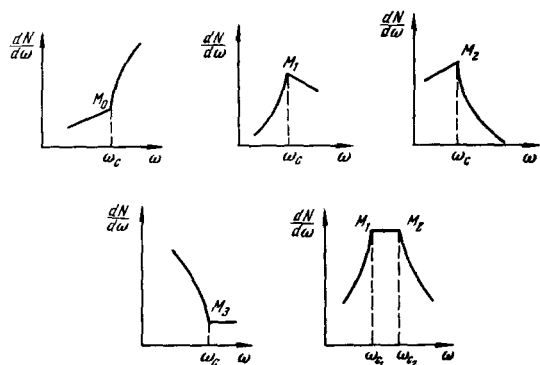


FIG. 1. Density of states near critical points of different types.

M_2 ($\epsilon_1 = \epsilon_2 = -\epsilon_3 = -1$) is given respectively for M_1 by:

$$\frac{dN}{dE_{jj'}} = \begin{cases} c - A(\omega_c - \omega)^{1/2} + O(\omega_c - \omega) & \text{when } \omega < \omega_c, \\ c + O(\omega - \omega_c) & \text{when } \omega > \omega_c, \end{cases}$$

and for M_2 by:

$$\frac{dN}{dE_{jj'}} = \begin{cases} c + O(\omega - \omega_c) & \text{when } \omega < \omega_c, \\ c - A(\omega - \omega_c)^{1/2} + O(\omega - \omega_c) & \text{when } \omega > \omega_c. \end{cases}$$

Figure 1 shows graphically the type of behavior of the density of states near the critical points. If we assume, for example, that an external electric field E or an elastic stress U affects the position of the critical point, i.e., $d\omega_c/dE$ or $d\omega_c/dU \neq 0$, this effect will give rise to corresponding features on the curves of $\epsilon_2(\omega)$ plotted against E or U . For example, in the vicinity of an M_1 critical point with $\omega < \omega_c$, we can write the $\epsilon_2(\omega)$ relation in which we are interested in the form:

$$\epsilon_2 = a(\omega_c - \omega)^{1/2},$$

Hence, the quantity

$$\frac{d\epsilon_2}{dU} = \frac{a}{2} \frac{d\omega_c}{dU} (\omega_c - \omega)^{-1/2},$$

which is one of the components of the strain potential, must have a singularity near M_1 . In turn, this will give rise to a singularity in the reflectance $\Delta R = \alpha\Delta\epsilon_1 + \beta\Delta\epsilon_2$. The variation $\Delta R(\omega)/R(\omega)$ when an alternating electric field is applied is the dynamic electroreflectance effect, while the corresponding elastic effect is the piezoreflectance effect.

2. ELECTROREFLECTANCE SPECTRA

The dynamic electroreflectance effect was first observed by Seraphin and Hess.^[4] The purpose of this study was to detect by this method a Franz-Keldysh effect at the Van Hove singularities situated above the fundamental absorption edge. The experiment was performed with light normally incident on the surface of a germanium crystal. The voltage was applied to a transparent electrode insulated from the germanium by a thin layer of dielectric. The alternating modulating electric field at 250 Hz frequency was of the order of 10^5 V/cm. The receiving instrument, a photomultiplier or a PbS photoelement with a narrow-band phase-sensitive amplifier, made it possible to measure a value of $\Delta R/R$ of the order of 5×10^{-6} (this limit was

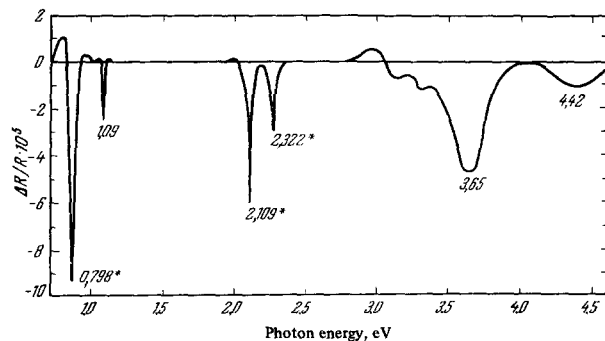


FIG. 2. Electroreflectance spectrum of germanium. The peaks marked with asterisks correspond to effects an order of magnitude larger than is indicated in the scale at the left.

later reduced to about $(1-3) \times 10^{-6}$). The optical part of the apparatus provided a spectral resolution of at least 0.003 eV. Fig. 2 gives the electroreflectance spectrum of germanium in the range 0.7–4.5 eV. The curve shows peaks in the regions of all the interband transitions that had been observed previously in light reflection and absorption experiments and had been identified from the existing calculations of the band structure of germanium. However, even in its first application, the electroreflectance method showed substantial advantages. The 1.09-eV line near the fundamental absorption edge was observed for the first time in reflected light. The quadruplet in the region 2.8–4.0 eV was resolved for the first time. The spin-orbital splitting of the valence band in the 2.2-eV region shows up very well.^[9] Depending on the conditions of preparation of the specimen surface, the absolute sizes of the observed peaks can vary by more than an order of magnitude, but their relative values vary considerably less, while their positions on the frequency scale remain fixed to an accuracy of ~ 0.005 eV. An electroreflectance effect was found in^[10] in p-type silicon, and then also in other semiconductors. Along with the new experimental data, theoretical studies also have begun to appear. They provide a basis for more exact identification of the observed interband transitions. In discussing the problem of the effect of an electric field on hyperbolic Van Hove singularities, Phillips proved a so-called duality theorem.^[2] According to it, near an M_1 saddle point ϵ_2 must be decreased by an electric field lying along the axis of the isoenergetic cone of the interband density of states. This will produce a blue shift in the corresponding absorption edge, instead of the red shift occurring in the Franz-Keldysh effect. Thus one can determine the type of the corresponding Van Hove singularity from the sign of the electroreflectance effect (other conditions being the same, since we have to take into account the effect of $\Delta\epsilon_1$ on the electroreflectance). In fact, hyperbolic-type singularities were first found in this way. For example, in the triplet^[10,11] observed in silicon in the 3.3–3.5 eV region (Fig. 3), the long-wavelength line I was identified with the fundamental absorption edge (Franz-Keldysh effect), while the short-wavelength line III, which is of opposite sign, was identified with a hyperbolic singularity corresponding to the L'_2-L_1 transition (anti-Franz-Keldysh effect). The occurrence of the intermediate line II was ascribed^[11] to the pos-

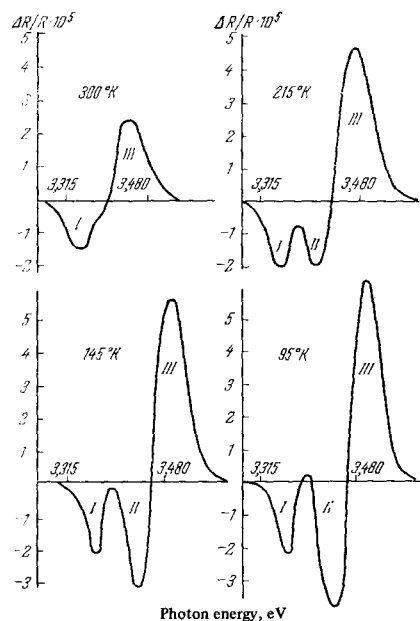


FIG. 3. Electroreflectance spectra of silicon at different temperatures.

sibility of exciting so-called hyperbolic excitons.^[2] However, the question of the existence of hyperbolic excitons is still under debate.^[12,13]

Further study of the electroreflectance effect in silicon has shown that the observed regularities are more complex than they seem at first glance. In n-type silicon specimens of about the same resistance as the previously studied p-type specimens, lines II and III changed sign (phase), while line I did not. They also observed considerable frequency shifts in lines, an effect of the surface potential on the peak positions, etc. Elucidation of the nature of these regularities requires a deeper understanding of the physics of surface effects in semiconductors.

An electroreflectance line has also been found in silicon at 4.2 eV.^[14] It was identified with a hyperbolic-type X_4-X_1 transition. Two groups of electroreflectance lines have been observed in GaAs^[15] in

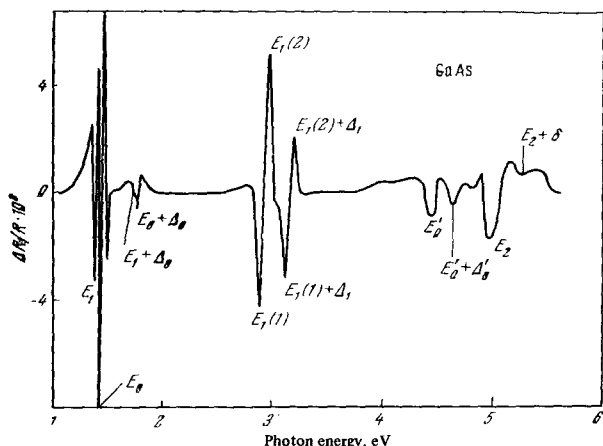


FIG. 4. Electroreflectance spectrum of n-type GaAs obtained by the electrolyte method.

the 1.5 and 3.0 eV regions, and were identified with $\Gamma_{15}-\Gamma_1$ and $\Lambda_3-\Lambda_1$ interband transitions, and respectively, with their spin-orbit components. The spin-orbit splittings were measured to high accuracy for the first ($\Delta = 0.348 \pm 0.002$ eV) and the second ($\Delta = 0.232 \pm 0.002$ eV) groups. The signs of the effects and also the signs of the line shifts produced by an electric field permitted them to suggest on the basis of the duality theorem that the lines of the first group correspond to parabolic-type transitions, and those of the second group to hyperbolic. Immediately following the first study on the electroreflectance effect,^[4] a communication appeared^[16] on the observation of an electroreflectance effect by the method of polarizing an electrolyte in contact with the surface of the semiconductor being studied. The electrolyte method is based on the fact that the dipole moments of the polarized molecules create a strong electric field at the surface of the semiconductor. Although the voltage on the condenser plates is relatively small (~ 2 eV), the field acting on the surface layer of the semiconductor is larger than in the ordinary Seraphin-Hess method,^[4] as judged by the size of the observed electroreflectance effect. In addition, the electrolyte method can be applied in the ultraviolet; it makes fewer demands on the quality of the semiconductor surface, and the specimen can be cleaned in the same electrolyte without exposing it to the air. Precise measurements of the spin-orbit splitting in various semiconductors of the IV, III-V, and II-VI groups have been made by the electrolyte method, and also certain hyperbolic-type transitions have been identified.^[16,17]

The potentialities of this method can be illustrated by the electroreflectance spectrum of n-type GaAs from^[18], as reproduced in Fig. 4. The identification of the lines in the electroreflectance spectrum proposed by the authors^[18] can be seen in the diagram of interband transitions in GaAs (Fig. 5). This same work studied the electroreflectance spectrum of GaP and nine alloys in the GaAs-GaP system. They present and discuss the curves giving the positions of the lines E_0 , E_1 , E'_0 , and E_2 and the spin-orbit parameters Δ_0 and Δ_1 as functions of the concentrations of the alloys.

Aspnes^[19] has developed further the theory of the electroreflectance effect as a consequence of the Franz-Keldysh effect. He studied not only the relation of the

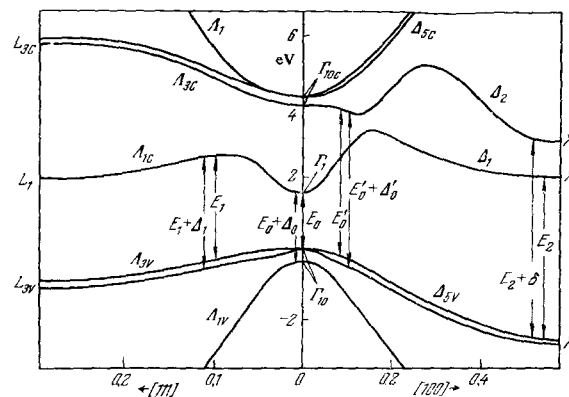


FIG. 5. Band structure of GaAs, with the identification of the electroreflectance spectrum in Fig. 4 proposed by the authors.^[18]

effect to the type of singularity, but also the anisotropy of the electroreflectance effect, which should considerably simplify the identification of transitions. However, I should note here that thorough study of the shapes and positions of electroreflectance lines and comparison of these data with electroabsorption measurements has cast doubt on the assumption that the Franz-Keldysh effect as canonically understood plays the major role in generating electroreflectance lines. Handler and his associates^[20] (see also^[21]) have advanced the hypothesis that effects of suppression of exciton absorption of light by the electric field play the major role in generating electroreflectance lines, at least in the region of the direct-transition absorption edge, and possibly for the other lines.

Feinleib^[22] has recently observed an electroreflectance effect in metals for the first time. He performed the measurements on silver and gold by the electrolyte method. Even though the depth of penetration of the static electric field into the metal is at least two orders of magnitude smaller than that of the light wave, the observed effect proved to be rather large, of the order of 0.5% at the maxima. The position of the absorption edge was found to be correlated with the features of the interband transitions by comparison with other experimental methods. Nevertheless, the piezoreflectance method apparently shows more promise for studying the band structure of metals, and we shall proceed now to discuss it.

3. PIEZOREFLECTANCE SPECTRA

The piezoreflectance effect was discovered by Engeler and his associates^[5] immediately after the appearance of the first study on electroreflectance. The method of measurement is analogous in principle to that described in the last section, except that the external electric field is replaced by an alternating elastic stress produced by a piezoelectric transducer. The approach to the problem of studying the band structure of solids by the piezoreflectance method also does not differ: it involves detecting and identifying Van Hove singularities. In particular, the piezoreflectance method has also permitted confirmation of the existence of hyperbolic-type singularities in silicon.^[23]

The piezoreflectance method has proved especially effective in studying the band structure of metals. Engeler and his associates^[24] have studied the piezoreflectance spectra of the noble metals. They made the measurements on film specimens directly sputtered onto a piezoelectric transducer. They determined experimentally the frequency-dependences of the relative variation of the reflected-light intensity $\Delta R/R$ caused by the elastic stress. Then they determined the corresponding phase changes $\Delta\theta$ in the reflected light by using the Kramers-Kronig relations. By using the formulas of metalloptics for the reflectance of a metal, one can easily derive relations between the experimental values $\Delta R/R$ and $\Delta\theta$ and the variations in the real and imaginary components $\Delta\epsilon_1$ and $\Delta\epsilon_2$ of the dielectric constant:

$$\Delta\epsilon_1 = \frac{1}{2} [n(\epsilon_1 - 1) - k\epsilon_2] \frac{\Delta R}{R} + [k(\epsilon_1 - 1) + n\epsilon_2] \Delta\theta,$$

$$\Delta\epsilon_2 = \frac{1}{2} [k(\epsilon_1 - 1) + n\epsilon_2] \frac{\Delta R}{R} - [n(\epsilon_1 - 1) - k\epsilon_2] \Delta\theta.$$

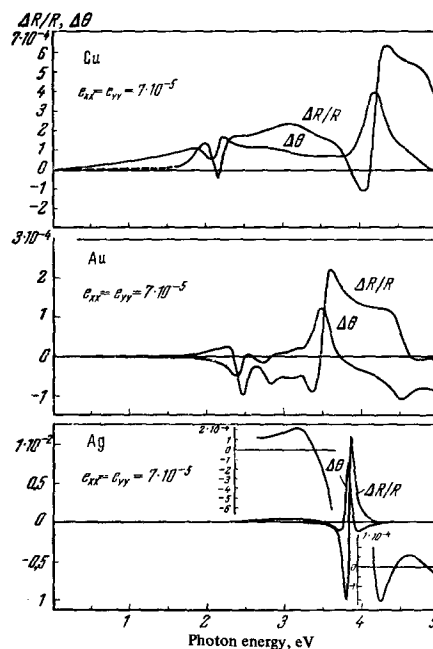


FIG. 6. Piezoreflectance spectra of copper, gold, and silver, and the calculated curves of the phase-shift $\Delta\theta$ of the reflected light.

Figure 6 shows the curves for $\Delta R/R$ and $\Delta\theta$ obtained in this way, and Fig. 7 shows the $\Delta\epsilon_1$ and $\Delta\epsilon_2$ curves for copper, gold, and silver. The authors identified the observed features on the $\Delta\epsilon_2$ curves as follows. The peak at 4.2 eV for copper and at 3.5 eV for gold involve a Van Hove singularity corresponding to transitions near X_5-X_4' , while the $\Delta\epsilon_2$ minimum at 4.5 eV for gold involves a singularity of type $L_{2,1}-L_{12}$. The features of the $\Delta\epsilon_1$ and $\Delta\epsilon_2$ spectra above 2.1 eV for copper and 2.35 eV for gold correspond to the fun-

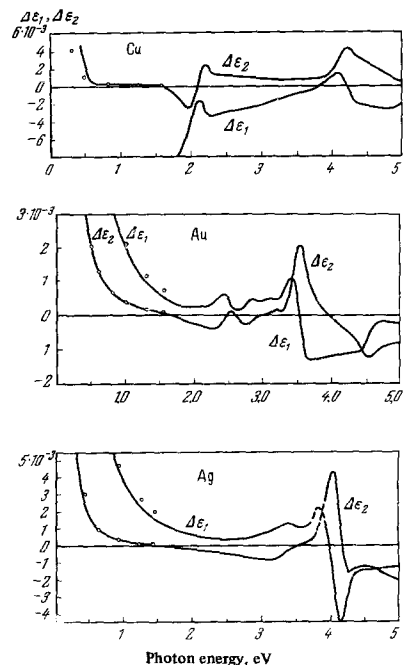


FIG. 7. $\Delta\epsilon_1$ and $\Delta\epsilon_2$ of copper, gold, and silver, as obtained by the piezoreflectance method.

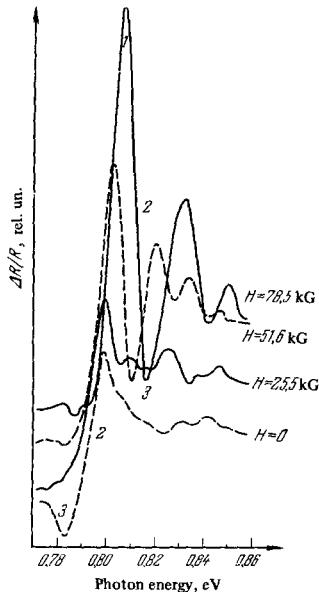


FIG. 8. Piezoreflectance spectra of germanium near the absorption edge for different values of the external magnetic field.

damental absorption edge for direct interband transitions, according to the existing calculations of the band structures of these metals. The behavior of $\Delta\epsilon_1$ and $\Delta\epsilon_2$ for silver in the 4-eV region apparently involves plasma resonance. The authors made the very interesting statement that the negative contribution to $\Delta\epsilon_2$ in the range 1.7–2.1 eV for copper, 1.5–3.5 eV for silver, and 1.6–2.35 eV for gold indicates the existence of interband transitions in the noble metals at energies lying below the fundamental absorption edge. It has not yet been possible to detect such features in the spectra of the noble metals by other experimental methods. Perhaps these low-energy anomalies have something in common with analogous anomalies recently discovered by Mayer in the alkali metals.^[25] Hence, study of them can be used to elucidate the nature of interband optical transitions in metals, to relate the roles of direct and indirect transitions, and to test the hypothesis of the existence of the so-called “metallic” excitons, etc.

4. COMBINED AND EXOTIC EFFECTS

New potentialities are opened up in the study of the electronic energy spectrum of crystals by studying electro- and piezoreflectance spectra while an external static magnetic or electron field or a static elastic stress is applied. Lax and his associates^[26] have obtained the piezoreflectance spectrum of germanium in the presence of a static magnetic field (magnetopiezoreflectance). The magnetic fields up to 90 kilogauss, oriented normal to the reflecting surface, were produced by a Bitter magnet. The light was almost normally incident on the specimen, and the relative deformation of the specimen amounted to 5×10^{-5} . Figure 8 shows the magnetopiezoreflectance spectra of germanium. The observed oscillations correspond to transitions between Landau levels in the valence band and the conduction band. The quantitative splitting

parameters are found to agree well with previously performed static measurements of magnetoabsorption of light in germanium. It was convincingly shown in^[26] that application of the dynamic magnetopiezoreflectance method is very promising, since it permits one to observe even at room temperature the details of structure of the interband transitions that remained outside the experimenter's field of view in the static methods. A communication has recently appeared on measurement of magnetopiezoreflectance in germanium at low temperatures.^[27]

The electroreflectance spectra of Ge, GaSb, and InSb in the presence of a magnetic field (magnetoelectroreflectance) have been measured in^[28], where they also got good agreement with static magnetoreflectance measurements. Magnetoelectroreflectance study of semiconductors is of interest particularly because the theory of light absorption in a semiconductor in crossed electric and magnetic fields, with account taken of the modification of the selection rules in this case, has been well developed.^[29]

Frova and Boddy^[30] have studied the electroreflectance spectra of the ferroelectric materials BaTiO₃, KTaO₃, and KTaO₃–KNbO₃. They made the measurements by the electrolyte method over the range 2–6.5 eV. The electroreflectance effect for the ferroelectric materials proved to be very large: one or two orders of magnitude larger than in typical semiconductors. The electroreflectance spectra showed many features, which the authors tried to identify with Van Hove singularities on the basis of the existing band calculations. The physical nature of the electroreflectance effect in ferroelectric materials is not yet clear. The authors showed that the electroreflectance is not due to the Franz-Keldysh effect, and proposed that it involves lattice polarizability (displacement of the transition ions), which in turn causes a change in the band structure and a shift in the critical frequencies.

Berglund^[31] has tested a new dynamic modulation method of studying the absorption edge of semiconductors. It consisted in studying the effect of a periodic temperature variation of the specimen on optical absorption near the absorption edge. He performed the measurements on silicon, gallium arsenide, and other materials, and drew conclusions on the contributions of direct and indirect transitions to the absorption.

Apparently one can also observe an analogous effect in reflected light upon direct variation of the specimen temperature (thermoreflectance). Batz^[32] has carried out the first experiments of this type. While he measured the periodic variation of the intensity of reflected light when dc current pulses were passed through the specimen, the author considered one of the fundamental causes of the observed effect to be the variation in the temperature of the specimen due to Joule losses. He made the measurements on an n-type germanium plate of dimensions 1.5 cm \times 0.4 cm \times 0.002 cm at a frequency of 13 Hz under conditions that provided for dissipation of the heat in the intervals between the current pulses. The reflection spectra showed features at 0.8 and 2.2 eV, which correspond to previously-identified interband transitions in germanium.

We can also consider the recently-published study of Bloembergen and his associates,^[33] in which they

found an effect of an electric field on the intensity of second-harmonic generation in reflection of light from silicon and silver. They made the measurements by the electrolyte method with a ruby laser ($\lambda = 6940 \text{ \AA}$) with the light incident at an angle of 45° . They found a non-linear dependence of the second-harmonic amplitude on the electric field intensity. They note a considerable role played by surface effects, and also effects of the semiconductor-electrolyte and metal-electrolyte contacts, although the mechanism of action of the electric field in this case is not completely clear.

In conclusion I should note that the purpose of this article has not been to review systematically and to discuss the results of studying the electronic energy spectrum of crystals by means of electro- and piezoreflectance and related methods. This cannot be done, first of all because measurements of this type have just begun: the first studies on electro- and piezoreflectance appeared in 1965, and there are many unclearities and even contradictions in the interpretation of the obtained results. However, even the first experiments have shown that the new methods possess extremely high sensitivity and resolving power, and are universal for studying different types of substances: semiconductors, metals, and ferromagnetic and ferroelectric materials. This explains the appearance of the large number of studies on this problem and the intensive search for new methods, and it provides sufficient grounds for drawing the attention of physicists studying the electronic structure of solids to the promise of this new line of approach.

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Note added in proof. Please note the following recent articles:

³⁴M. Cardona, K. L. Shaklee, and F. H. Pollak, Phys. Rev. 154, 696 (1967) (summarized data on electroreflectance of semiconductors).

³⁵W. J. Scouler, Phys. Rev. Letts. 18, 445 (1967) (thermoreflectance of gold).

³⁶J. Hanus, J. Feinleib, and W. J. Scouler, Phys. Rev. Letts. 19, 16 (1967) (thermoreflectance of ferromagnetic nickel).

³⁷U. Gerhardt, D. Beaglehole, and R. Sandrock, *ibid.* 19, 309 (1967) (piezoreflectance of single crystals of copper).

³⁸V. Rehn and D. S. Kyser, Phys. Rev. Letts. 18, 848 (1967); A. Prostak and W. N. Hansen, Phys. Rev. 160, 600 (1967) (electroreflectance of metals).

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