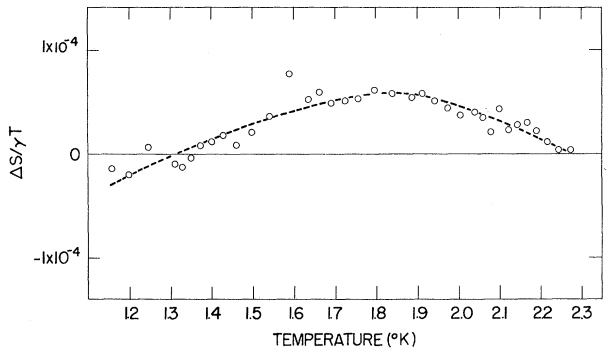

FIG. 2. Values of ΔS derived from the data shown in Fig. 1, expresses as a fraction of the normal electronic entropy.

our estimate of the uncertainty in stray heat over the course of a run. The significant features of Fig. 2 are the very small upper limit to ΔS in comparison with the normal electronic entropy ($\Delta S/\gamma T \lesssim 6\times10^{-5}$), and the corresponding absence of any substantial discontinuity or sudden change of slope near 1.7°K.

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PHOTOACTIVITY IN BISMUTH GERMANIUM OXIDE

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Bismuth germanium oxide,[1] $Bi_{12}GeO_{20}$, is an electro-optic[2] semiconductor which belongs to the cubic point group 23.[2] It is also piezoelectric,[1,3] photoconductive,[1,4] optically active,[2] and has unusually good ultrasonic properties.[5] The dark resistivity, ρ, is of the order of 10^{10} Ω cm and it was found[2] that the optical activity can be changed by an externally applied electric field. During an investigation of these effects, we have observed very large electric-field-biased, photoinduced, static changes in optical polarization using low-intensity monochromatic polarized light as an optical probe. We have also observed transiert and oscillatory effects of a similar nature. Since the major axis of the resultant elliptically polarized light is rotated by the photoinduced charge carriers, the effect can be called photoactivity.

These modifications in polarization may be induced under various conditions of excitation and are explained in terms of the charge carrier initiation of high electric field domains. In the absence of light, bismuth germanium oxide is an insulator. Since it is also photoconductive, the illumination of a small spot or region will result in the generation of free electrons, holes, or hole-electron pairs. Under the influence of the applied static field either or both kinds of charge carriers will be displaced, setting up a high electric field domain. Examination of static and slowly moving electric field domains also has been possible in other high-resistivity crystals. For example, the Franz-Keldysh effect has been used[6] in CdS to determine the regions of field inhomogeneity, and a scanning light spot has been used[7] in GaAs to map the regions of changes in photoconductivity response produced by field inhomogeneities.

The experimental arrangement employs a polarizing microscope in which the source light passes through a narrow-band optical filter before being transmitted through the crystal. Several types of compensators are employed as required. In bismuth germanium oxide, for a dc voltage applied parallel to either the

[110] or [001] direction and the probing monochromatic polarized light beam propagating parallel to the [$\bar{1}$10] direction, optical polarization may be changed in regions of the crystal by polychromatic side illumination. This side light may be any small lamp such as a microscope illuminator. Monochromatic side illumination also can be used if the wavelength is such that sufficient photoconductivity is induced. The area illuminated by the side light becomes conductive and the region which is not illuminated is nonconductive, thus producing a high electric field domain in the unilluminated section. The area affected is adjacent to one or both electrodes depending on the angle of the side light with respect to the electrode, as in Fig. 1(a). Results were the same using either silver or indium-mercury-amalgam electrodes. There was no evidence of non-Ohmic behavior. If either the side light or the electric field is removed, the affected region tends to remain intact for a relatively long period of time (seconds or minutes). Application of a magnetic field of about 1000 Oe increases the memory time of the effect by two to three times, depending on the sample used. The effect may be immediately erased by illumination with intense white light collinear with the probing beam. The action of the erasing light is to cause the entire sample to become conducting. The electric field domain moves rapidly to one electrode and the electric field gradient becomes uniform across the sample. Thus it is seen from the resultant polarization properties that the optical properties of the regions adjacent to the electrodes are changed.

Figure 2 shows data taken on a bismuth-germanium-oxide crystal whose length is ~1.55 mm in the direction of the applied electric field [110]. The width is ~1.78 mm and the thickness in the direction of propagation of the probing light [$\bar{1}$10] is ~3.78 mm. The natural optical activity (rotation of the plane of polarization) is given in the lower curve. The azimuth of the major axis of the elliptically polarized light resulting from application of electric field parallel to the [110] direction is shown by the central curve. Polarization is now elliptical since the crystal symmetry becomes noncubic and linear birefringence is also introduced.[2] The upper curve is the azimuth of the major axis of the ellipse in the photoactive region result-

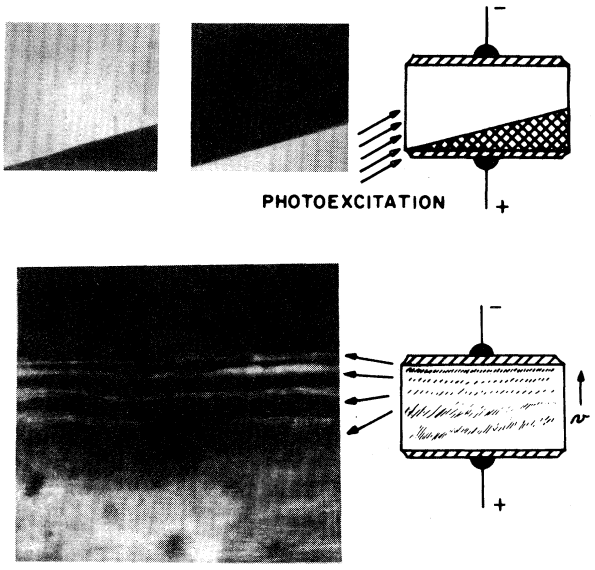

FIG. 1. (a) In the photomicrograph on the right, the analyzer was set for extinction of light over the entire sample in the absence of photoexcitation, while in the photomicrograph on the left under the same conditions the analyzer was set for maximum optical transmission. The photoactive region is the small triangle in lower right. (b) Photomicrograph of moving electric field domains. The domains form in the lower part of the crystal and move toward the top becoming sharper and more well defined.

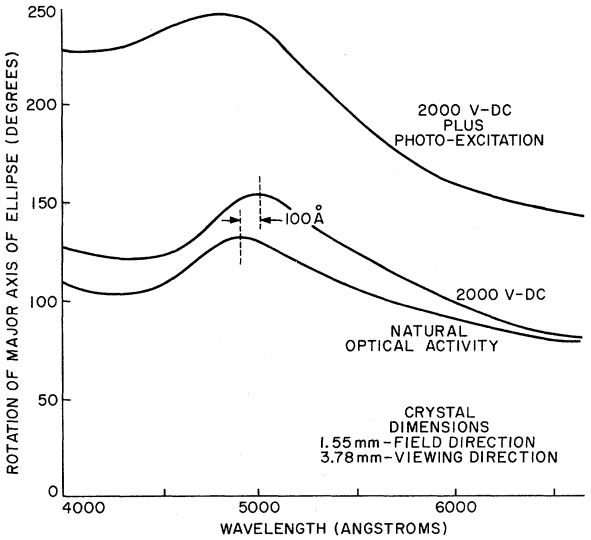

FIG. 2. Rotation of the major axis of the resultant elliptically polarized light as a function of wavelength. Lower curve: Natural optical activity (rotation of the plane of polarization). Middle curve: Rotations of major axis of ellipse with 2000 V dc applied. Upper curve: Rotation of major axis with bias field and photoexcitation.

ing from application of electric field and polychromatic side illumination. Since the wavelength dependence of this curve follows closely that of the photoconductivity curve,[4] the photoconductive origin is suggested. In these first available crystals it has not been determined whether the photoconductivity is intrinsic or is caused by extrinsic mechanisms, such as impurities or lattice defects. It is also seen from Fig. 2 that the rotation maximum has been shifted by 100 Å with the application of 2000 V dc (13 kV/cm). Since the anomalous rotatory dispersion from 4000 to 5000 Å is associated with an optical-absorption peak in the ultraviolet, measurements subsequently were made of the absorption-band edge with applied electric fields. Using a dual beam spectrometer and fields of 6 kV/cm we observed no shift of the band edge. Care was taken to make certain that the spectrometer source light was transmitted through the center of the crystal avoiding the regions near the electrodes. Further quantitative experiments are being carried out as a function of crystallographic orientation.

If it is assumed that the principle of superposition[8] holds, then a normally incident plane wave splits up into two elliptically polarized components of phase difference $\Delta = (2\pi/\lambda_0)(n_1 - n_2)$, where n_1 and n_2 are the indices of refraction for the two waves and λ_0 is the wavelength of light in a vacuum. The magnitude of Δ is given by the quadratic relation $\Delta^2 = \delta^2 + (2\rho)^2$, where δ is the phase difference with no optical activity and is related to the linear birefringence by $\delta = (2\pi/\lambda_0)(n' - n'')$, n' and n'' being the refractive indices of the crystal if optical activity were absent. In bismuth germanium oxide, with no electric field applied, $n' = n''$ and the linear birefringence is zero. The phase difference in the absence of linear birefringence is 2ρ, where the optical rotatory power is $\rho = \pi G/\lambda_0 \bar{n}$. G is the optical activity parameter or gyration, and $\bar{n} = (n'n'')^{1/2}$ is the mean refractive index. The circular birefringence, $n_l - n_r$, is related to ρ by $G/\bar{n} = n_l - n_r$, and n_l and n_r are the refractive indices of the left and right circular components, respectively. For an applied electric field, E, we may then write $\Delta^2(E) = \delta^2(E) + 4\rho^2(E)$. By suitable measurements of the transmitted light using a polarization analyzer to determine the azimuth of the resultant ellipse and various optical compensators to determine ellipticity, the electric-field-induced linear birefringence can be separated from the electric-field-induced change in optical activity.[2] When, in addition to the biasing electric field, a side light is applied, the polarization state is changed again and can be analyzed as before to separate the photoinduced linear birefringence from the photoinduced change in optical activity. For the combined effect, i.e., the electric-field-biased, photoinduced change in linear and circular birefringence, expressed in terms of Δ, we suggest the name photoactivity.

Transient photoinduced effects have been observed with dc voltage applied to either the [001] or [110] direction, a side light to generate charge carriers, and the probing beam parallel to the [$\bar{1}$10] direction. Directionality of the side light is not critical. The probing beam is plane polarized and a polarization analyzer is used to establish either a bright or a dark viewing field. The effect is seen as a series of bright or dark bands propagating slowly across the sample from the positive to the negative electrode, as shown in Fig. 1(b). In general the bands are initiated only when the voltage is first turned on with the side light already on or vice versa. It was found that the characteristics of the bands are most sensitive to pressure. By applying a small amount of pressure in the direction of the E field, a cyclic progression of bands is observed. The origin can be explained on the basis of the excitation of a band of charge carriers which, under the influence of the applied voltage, results in the formation of high electric field domains.[7,9] The band forms near the center or one end of the crystal and is of low contrast at that time. As it moves toward the negative electrode, the sides become sharper, the width becomes narrower, and the contrast increases strongly. Away from the negative electrode, as observed under a microscope, the band extends across the crystal with sharp flat sides parallel to the electrodes. As the terminal is approached, there is apparently a fringing of electric fields, and the band tends to intensify toward the center of the electrode. Observations are usually made with an electric field across the sample of about 5×10^3 V/cm. In these first experiments evidence of a negative resistance has been found in the high-resistance bismuth germanium oxide. At field strengths greater than ~10 kV/cm considerable noise and various oscillatory signals appeared on

the crystal current. That such an effect exists is also consistent with the stated observations on the well-formed electric field domains. That is, a limiting velocity of the charge carriers, and a decrease in that velocity with an increase in the electric field, would be a sufficient mechanism to stabilize the width of the domains.

With a polychromatic polarized probing light the bands appear as alternate colors. The photoinduced region of the static effect previously described also displays a different color from the unaffected area of the sample in a white probing light. The various colors illustrate the dispersion inherent in optically active materials. This very effectively demonstrates the differences in polarization of adjacent areas.

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INFLUENCE OF BAND STRUCTURE AND INTERATOMIC EXCHANGE ON SPIN FLUCTUATIONS IN METALS

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Theoretical work of Berk and Schrieffer[1] and of Doniach and Engelsberg[2] has suggested the importance of electron-paramagnon (spin-fluctuation) interactions in metals which are nearly ferromagnetic. Recent experiments on Ni-Rh alloys by Bucher et al.[3] and on Ni-Pd alloys by Schindler[4] give striking confirmation of these effects.

Initially, the theory was carried out for a spherical one-band model with a contact-exchange interaction. This model predicts exceptionally large many-body corrections, e.g., a mass enhancement of order 8 for Pd. From augmented-plane-wave[5] and relativistic augmented-plane-wave (RAPW)[6] calculations on Pd, and the observed specific heat, one finds a factor closer to 2. In addition, the alloy experiments exhibit a considerably smaller mass enhancement than this model predicts. In an attempt to account for this discrepancy, Doniach[7] added Hund's rule (intra-atomic) exchange to the model and found a reduction of the predicted enhancement, although it appears that the reduction is insufficient to bring theory and experiment into agreement.[3]

We report here on the importance of (a) band-structure effects and (b) interatomic exchange interactions in the problem, and show that they can lead to large additional changes in the many-body corrections.

Band-structure calculations[5,6] show that the heavy-hole energy contours in Pd can be roughly represented by three cylindrical sub-bands, each of length equal to the (100) reciprocal lattice vector and having their axes along x, y, and z, respectively. The Bloch functions $\psi_{k\alpha}$ for these sub-bands can be represented in terms of Wannier functions w_α by

$$\psi_{k\alpha} = (N)^{-1/2}\sum_j \exp(i\vec{k}\cdot\vec{R}_j) w_\alpha(\vec{r}-\vec{R}_j), \qquad (1)$$

$$\alpha = (x, y, z),$$

where N is the number of cells in the crystal. Recent theoretical work[8] shows that for d bands, the w_α behave like tight binding functions, and we expect matrix elements of short-ranged (screened) potentials taken between w's located on different sites will decrease rapidly with the site separation. Thus, we expand the Bloch matrix element $\langle \vec{p}+\vec{q}, \alpha; \vec{k}, \beta' | V | \vec{k}+\vec{q}, \beta; \vec{p}, \alpha\rangle$ in Wannier functions and retain only the one- and two-center terms. The importance of in-

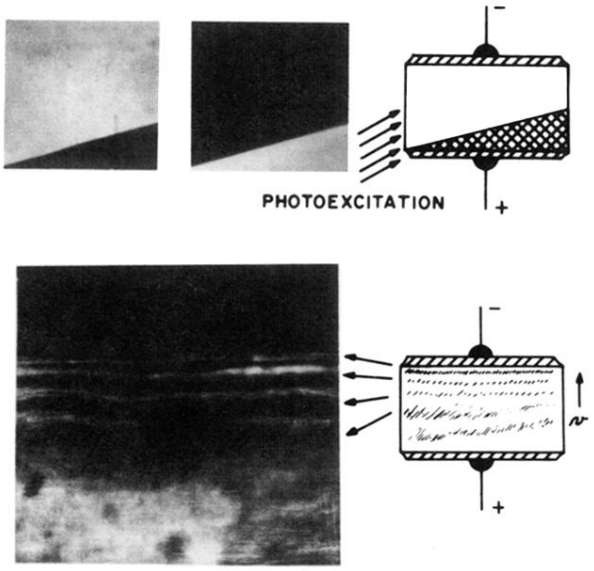

FIG. 1. (a) In the photomicrograph on the right, the analyzer was set for extinction of light over the entire sample in the absence of photoexcitation, while in the photomicrograph on the left under the same conditions the analyzer was set for maximum optical transmission. The photoactive region is the small triangle in lower right. (b) Photomicrograph of moving electric field domains. The domains form in the lower part of the crystal and move toward the top becoming sharper and more well defined.