

Effect of Polarized Light on the 1.8-, 3.3-, and 3.9- μ Radiation-Induced Absorption Bands in Silicon

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The effect of polarized light on the 1.8-, 3.3-, and 3.9- μ radiation-induced absorption bands in silicon, which have been correlated with the divacancy, has been studied. It is found that an illumination with polarized light of energy corresponding to the 1.8- or 3.3- μ band reduces the absorption coefficient of the band at a temperature below a critical temperature (32.5°K for the 1.8- μ band and 23°K for the 3.3- μ band). The phenomenon is attributed to the electronic reorientation of the divacancy achieved through an excited state by absorbing a photon. A net alignment of the defect can be observed only at a temperature at which the defect in the ground state is usually "frozen" in a particular orientation. From the temperature dependence of the recovery of the aligned defect in the dark, the activation energies of 0.076 and 0.056 eV are found to be required for the reorientation of the defects in the ground states causing the 1.8- and 3.3- μ bands, respectively. The results indicate that the 1.8- μ band arises from the neutral divacancy only. An analysis of the temperature dependence of the reorientation effect has been made. It is indicated that the activation energy required for the reorientation in the excited state is very small, probably in the order of the thermal energy at $\sim 10^\circ\text{K}$. No similar effect has been observed in the 3.9- μ band, which is consistent with the fact that this band contributes photoconductivity.

1. INTRODUCTION

THE divacancy in irradiated silicon has been extensively studied by electron paramagnetic resonance, infrared absorption, and photoconductivity.¹⁻⁸ In silicon, there are four equivalent atomic orientations for the divacancy and three electronic configurations for each atomic orientation. A divacancy exists in only one of the configurations. At a low temperature, a divacancy can be "frozen" in a particular configuration for a relatively long time, since insufficient thermal energy is available to overcome the potential barrier between the configurations. It is known that the 1.8-, 3.3-, and 3.9- μ radiation-induced absorption bands in silicon are correlated with the divacancy.⁴ Thus, a divacancy can become excited for a length of time after absorbing a photon with an energy corresponding to these bands. If the activation energy required for the electronic reorientation process in the excited state is lower than that in the ground state, and if the lifetime of the excited state is not too short, an electronic reorientation of the defect can occur under an illumination with light even at a temperature at which the unexcited divacancy is usually frozen. A net align-

ment of the defect can be obtained under an illumination with polarized light at such a low temperature, if the crystal is properly aligned with respect to the electric vector of the light. The advantage in measuring this phenomenon is not only to provide a useful tool for the study of the electronic reorientation of the unexcited defects without the use of a complicated stress cryostat, but also to yield some important information about the excited states of the defects which cannot be obtained by other means.

In this paper, we shall report the results from a study of the effect of illumination with polarized light on the 1.8-, 3.3-, and 3.9- μ bands, from which some important information about the divacancy in silicon has been deduced.

2. EXPERIMENTAL PROCEDURE

Rectangular samples (19×9.5×6.4 mm) were cut from commercially available silicon ingots which had been grown by the floating-zone technique. The orientations of the crystals were determined by means of the growth marks on the ingots.⁹ The resistivity of the sample before irradiation was about 0.05 Ω cm for P-doped crystal and about 0.1 Ω cm for B-doped crystal. The surface of the sample through which infrared light was transmitted had been polished with diamond compounds.

Figure 1 shows the arrangement of the optical apparatus used in our experiment. A LiF prism was mounted in the monochromator. The light was chopped at a frequency of 13 cps, and the signal, detected by a thermocouple detector, was amplified by a lock-in phase-sensitive amplifier. A liquid-He cryostat capable of controlling the sample at any temperature in the range 11–77°K with an accuracy of $\pm 0.1^\circ\text{K}$ was used.

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¹ G. D. Watkins and J. W. Corbett, *Phys. Rev.* **138**, A543 (1965).

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³ G. Bemski, B. Szymanski, and K. Wright, *J. Phys. Chem. Solids* **24**, 1 (1963).

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⁶ L. J. Cheng, in *Radiation Effects in Semiconductors*, edited by F. L. Vook (Plenum Press, Inc., New York, 1968), p. 143.

⁷ L. J. Cheng and J. Lori, *Phys. Rev.* **171**, 856 (1968).

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⁹ R. D. Hancock and S. Edelman, *Rev. Sci. Instr.* **27**, 1082 (1956).

A copper-constantan thermocouple pressed between the sample and the sample holder was used to measure temperature.

The irradiations were performed inside a water-cooled annulus of enriched uranium fuel at 50°C in the E-3 hole of the NRX reactor at the Chalk River Nuclear Laboratories. The fission neutron flux was $5.7 \times 10^{12} \text{ n cm}^{-2} \text{ sec}^{-1}$, measured by the $\text{Ni}^{58}(n,p)\text{Co}^{58}$ reaction by use of an effective cross section of 90 mb for fission neutrons.¹⁰

3. EXPERIMENTAL RESULTS

A sample of 0.05- Ω -cm P-doped silicon was irradiated with a fluence of 4.3×10^{16} fission neutrons cm^{-2} . The 1.8- and 3.3- μ bands were observed (see the solid curves in Fig. 2). It should be noted that the Fermi level of the sample must be below $E_c - 0.21$ eV, above $E_c - 0.21$ eV, and below $E_V + 0.25$ eV for the appearance of the 1.8-, 3.3-, and 3.9- μ band, respectively. The simultaneous observation of the 1.8- and 3.3- μ bands in the sample is due to the inhomogeneity of the neutron damage. The orientation of the sample is also shown in Fig. 2. It was found that the transmittance of the sample as measured by polarized light of energy corresponding to the 1.8- μ band increased with time to a certain saturation value, when the sample temperature was less than 32.5°K. A similar effect on the 1.8- μ band has been observed by using polarized white light.¹¹ It was found that the 3.3- μ band exhibited the same phenomenon, when the sample was at a temperature lower than 23°K.

An increase of the transmittance of a sample must correspond to a decrease of its absorption coefficient. The change of absorption coefficient is measured in the following way: The sample temperature is stabilized at a certain value lower than the critical temperature mentioned above. The wavelength of the light from the monochromator is adjusted to a certain desired value. The polarizer is rotated into the position where the electric vector of the light is parallel to the [011] direction of the sample, and the intensity of the transmitted light becomes saturated in time. Then the polarizer is rotated 90° (now $\mathbf{E} \parallel [0\bar{1}\bar{1}]$), and the light intensity (I_1) is measured immediately. Then the intensity becomes saturated and the value (I_2) is measured. Since $I/I_0 = e^{-\alpha x}$, where I_0 is the intensity of incident light, I the intensity of transmitted light, α the absorption coefficient, and x the sample thickness, we have the relation between the maximum change of absorption coefficient and the measured light intensities (I_1 and I_2) as

$$\alpha_1 - \alpha_2 = (1/x) \ln[(I_2 - I_1)/I_1 + 1]. \quad (1)$$

It should be noted that the value of $\alpha_1 - \alpha_2$ is twice the

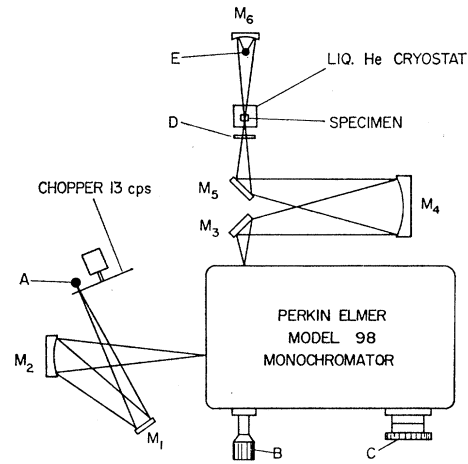


Fig. 1. Optical schematic diagram of the experimental setup. (A) SiC globular, (B) slit micrometer, (C) wavelength drum, (D) Perkin Elmer wire grid polarizer on rotatable frame, (E) thermocouple detector. M_1, M_3, M_5 —plane mirrors; M_2, M_4 —spherical mirrors; M_6 —ellipsoid mirror. The monochromator is used with a LiF prism.

value of the change of the absorption coefficient caused by the illumination, if the divacancies are equally populated among all the possible configurations at the beginning.

In Fig. 2, the dashed curve represents the maximum change of absorption coefficient in wavelength range from 1.2 to 4 μ at 15.1°K calculated from the data using Eq. (1). The figure shows clearly that the spectrum of the change of the absorption is similar to the absorption spectrum itself, indicating that only an illumination with polarized light of energy corresponding to the absorption energy of the band can cause the change. We found that 1.8- μ light cannot cause the change in the 3.3- μ band and vice versa. We also found that after the change at a certain wavelength within a band became saturated, no additional change at a different wavelength within the band was observed,

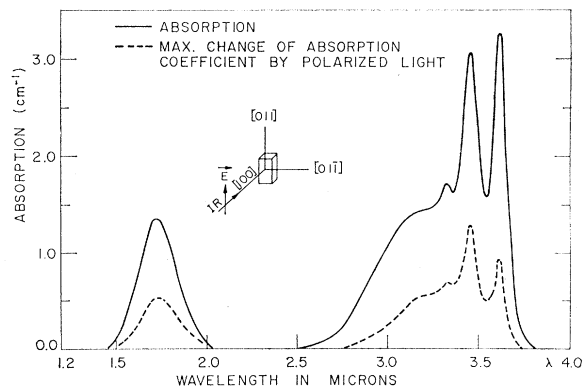


Fig. 2. 1.8- and 3.3- μ absorption bands and the spectral dependence of their maximum change ($\alpha_1 - \alpha_2$) induced by polarized light at 15.1°K.

¹⁰ A. W. Boyd, H. W. J. Connor, and J. J. Pieroni, Atomic Energy of Canada Limited Report No. 2203, 1965 (unpublished).

¹¹ G. D. Watkins (private communication).

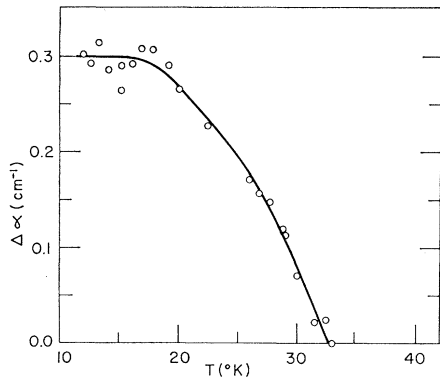


FIG. 3. Temperature dependence of the maximum change of absorption coefficient, $\Delta\alpha \equiv \frac{1}{2}(\alpha_1 - \alpha_2)$, at the peak of the 1.8- μ band.

if the time between the two measurements was very short.

The polarization efficiencies, defined as the maximum change of the absorption coefficient ($\alpha_1 - \alpha_2$) divided by the absorption coefficient, for the 3.61-, 3.45-, and 3.32- μ sub-bands are 0.28, 0.42, and 0.41, respectively. It is interesting that there is a substantial difference between the value for the 3.61- μ sub-band and the value for the other two. The polarization efficiencies at various wavelengths within the 1.8- μ band are roughly the same as indicated in Fig. 2.

The temperature dependence of the maximum change of absorption coefficient, $\Delta\alpha \equiv \frac{1}{2}(\alpha_1 - \alpha_2)$, at the peak of the 1.8- μ band, is shown in Fig. 3. The polarization effect appears at a temperature lower than 32.5°K. The value increases monotonically with decreasing temperature and reaches a saturation value at low temperatures. Figure 4 shows similar data for the 3.45- and 3.61- μ sub-band of the 3.3- μ band. Both sub-bands are similar, although the polarization efficiency of the 3.61- μ sub-band is only about $\frac{2}{3}$ that of the 3.45 sub-band. It is shown clearly in Fig. 5 that the curves become saturated at low temperatures.

The recovery of the change in absorption coefficients of the 1.8- and 3.3- μ bands in the dark was studied at various temperatures. The amount of the recovery of the change in the dark increases with time, roughly following the relation of $k(1 - e^{-t/\tau})$, where k and τ are constants, depending on the experimental conditions. The time required for 50% recovery of the changes for the two bands versus the reciprocal of measuring temperature are plotted in Fig. 5. The figure indicates clearly that the recovery has an exponential dependence at higher temperatures. From these results, we have found that the activation energies for recovery are 0.076 and 0.058 eV for the 1.8- and 3.3- μ bands, respectively. The corresponding frequency factors are 4.9×10^{11} and 7.4×10^{11} sec⁻¹. We have found no significant difference in the recovery between the 3.61- and 3.45- μ sub-bands. At lower temperatures, the

recovery times of both bands were found to become saturated (e.g. see Fig. 5 for the 3.3- μ band). The saturation effect may be attributed to the exposure of the sample to unpolarized light from the environment, since the sample in the cryostat is always exposed to some radiation through the windows, such as room-temperature radiation, even though the room is darkened during the experiments.

A sample of 0.1- Ω -cm B-doped silicon which was cut in the same orientation as the one shown in Fig. 3 was irradiated with a fission neutron fluence of 4.1×10^{16} n cm⁻². The 1.8- and 3.9- μ bands were observed in the sample. The same polarization effect of the 1.8- μ band

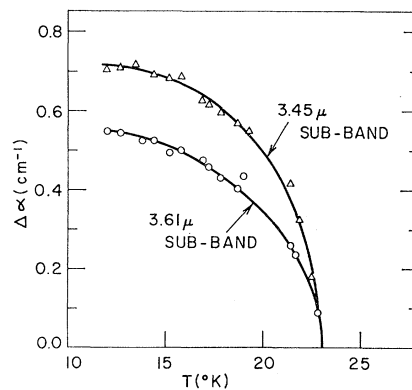


FIG. 4. Temperature dependence of the maximum change of absorption coefficients, $\Delta\alpha \equiv \frac{1}{2}(\alpha_1 - \alpha_2)$, at the peaks of the 3.45- and 3.61- μ sub-bands.

as mentioned above was observed. An attempt to observe the effect of illumination with polarized light of various wavelengths (1-5 μ) on the 3.9- μ band in a temperature range down to 11°K was made. No effect was found.

4. DISCUSSION

A. Defect Reorientation by Light

A model of the divacancy deduced from the EPR studies¹ is shown in Fig. 6. The adjacent vacancies are designated by the dashed circles c and c' . The nearest-neighbor atoms to vacancy c' are labeled a , b , and d ; those to vacancy c are labeled a' , b' , and d' . The symmetric divacancy belongs to the D_{3d} symmetry point group. The experimental results^{1,4-6,8} have shown the existence of a Jahn-Teller distortion which lowers the symmetry to C_{2h} . More pictorially, one may say that pair-wise bonding of vacancy neighbors occurs. There are three equivalent ways to arrange such bondings. A divacancy exists in only one of the three equivalent electronic configurations. It has been experimentally verified^{1,4-6,8} that an applied uniaxial stress can lift this degeneracy and, provided sufficient thermal energy is available to overcome the reorientation activation energy, the configurations can redistribute,

yielding a net alignment of the defect in the favored configuration. It has been found in these previous studies that the activation energy required for electronic reorientation in the divacancy is rather low. According to Watkins and Corbett,¹ the activation energy is 0.056 eV in the singly negatively charged divacancy and 0.073 eV in the singly positively charged divacancy. The corresponding frequency factors are 1.1×10^{13} and 5.4×10^{12} sec⁻¹. Therefore, a divacancy can be frozen in a particular configuration for a relatively long time at a low temperature.

In unstressed silicon, all the three configurations are equally populated. After a divacancy is excited by absorbing a photon, it can deexcite to its ground state with the same configuration it had before or to one with different configuration. The latter process becomes possible only when sufficient thermal energy is available to overcome the reorientation activation energy of the excited defect. If the reorientation activation energy of the excited defect is smaller than that of the unexcited one, an electronic reorientation of the defect at a temperature at which the defect is usually frozen can be achieved through the excited state by means of photon absorption. Under an illumination with polarized light of energy corresponding to the absorption

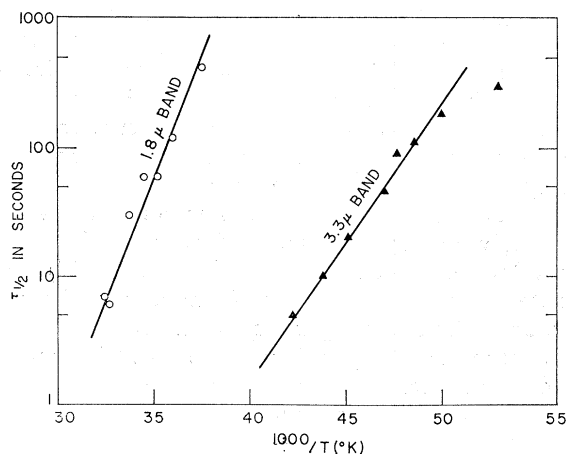


FIG. 5. Time required for 50% recovery of the light-induced changes for the 1.8- and 3.3- μ bands versus the reciprocal of measuring temperature.

energy at a temperature at which the above condition is fulfilled, the number of the defect with the transition dipole moment in the direction closer to the electric vector should decrease and the one with the moment further from the vector should increase, because the absorption rate in the former case is larger than that in the latter. Therefore, the illumination should cause a decrease of the absorption coefficient of the sample to the illuminating light. At the same time, the absorption of the light with the electric vector perpendicular to the electric vector of the illuminating light should increase. In other words, a net alignment of the defect should be

observed. This is what we have observed for the 1.8- and 3.3- μ bands. Therefore, our results show that the defects causing the two bands can be electronically reoriented by absorbing photons.

We have not observed a similar polarization effect on the 3.9- μ absorption band, which has been correlated with the singly positively charged divacancy. The following is an interpretation of the phenomenon. The 3.9- μ absorption band is associated with the 3.9- μ photoconductivity band.^{5,6,12} A new mechanism for the formation of photo-excited carriers was proposed in order to explain the shape of the photoconductivity band.^{5,6} In the new mechanism, a photo-excited carrier is not created by a direct excitation from the defect level to the valence band, but by a local excitation followed by an emission of a hole from the empty ground state of the excited defect. The hole-emission rate has to be much higher than the deexcitation rate of the excited defect in order to create an appreciable number of photo-excited carriers. This would make the lifetime of the excited defect (without an extra electron) very short, so that the defect could not have enough time for the reorientation, even if other conditions were favorable. During the emission of a hole, the excited defect gains one extra electron. The lifetime of the defect in this state might be quite long. A reorientation process may be possible. However, our experimental results indicate that the defect with the extra electron cannot reorient itself at a faster rate than that without the electron at the temperatures of our experiment. Therefore, a net alignment of the defect causing the 3.9- μ band cannot be observed. It should be noted that the above interpretation can also be applied for the process in which the photo-excited carrier is formed by a direct transition from the defect level to the carrier band.

Similar photo-induced dichroism in color centers of alkali-halide crystals, e.g., the *M* center in KCl, has been observed.¹³ The dichroism has been attributed to

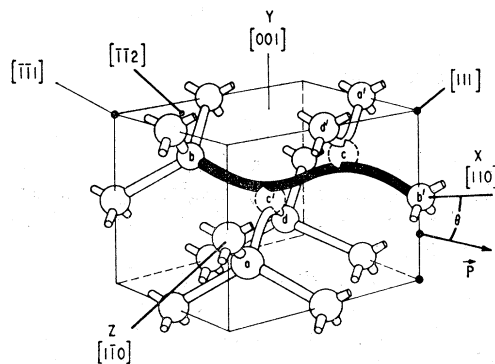


FIG. 6. Model of the divacancy deduced from EPR studies.

¹² H. Y. Fan and A. K. Ramdas, *J. Appl. Phys.* **30**, 1127 (1958).

¹³ T. J. Turner, R. DeBatist, and Y. Haven, *Phys. Status Solidi* **11**, 267 (1965); **11**, 535 (1965).

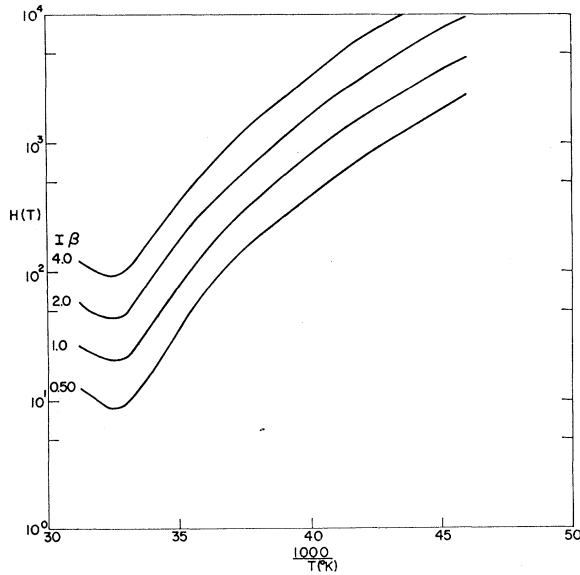


FIG. 7. Temperature dependence of calculated $H(T)$ for the 1.8- μ band.

the atomic (ionic) reorientation, but not to the electronic reorientation reported in this paper.

B. Defect Randomization

After a net alignment of the defect has been achieved by means of illuminating the sample with polarized light, the alignment disappears gradually in the dark. The disappearance is due to the randomization of the aligned defect. The process becomes possible when sufficient thermal energy is available to overcome the reorientation activation energy. The rate of reorientation may be written as $f_0 e^{-E/kT}$, where E is the activation energy, f_0 the frequency factor, k Boltzmann's constant, and T the temperature. It is seen from Fig. 5 that the experimental data on the reorientation rate in the dark monitored by the 1.8- and 3.3- μ bands do follow the above relation, even though we have experienced some background radiation problem at lower temperatures. For a divacancy in silicon, the measured lifetime τ can be written as

$$1/\tau = 3f_0 e^{-E/kT}. \quad (2)$$

The factor of 3 is due to the fact that there are three possible configurations. By neglecting the entropy contributions to the process, f_0 may be considered as the average vibration frequency of a certain normal mode in the defect which is responsible for the reorientation. The values of f_0 for the 1.8- and 3.3- μ bands are found to be $\sim 1.6 \times 10^{11}$ and 2.5×10^{11} sec $^{-1}$, respectively. The frequency factor for a solid at a temperature T can be approximately written as $f' = kT/h$, where h is Planck's constant. For $T = 30^\circ\text{K}$, $f' = 6.3 \times 10^{11}$ sec $^{-1}$ and, for 20°K , 4.2×10^{11} sec $^{-1}$. Our experimental values of f_0 are smaller than such values.

The relaxation of the atoms around the two adjacent vacant sites may cause the lowering of the vibration frequency. However, because of the large error in the measurements, we should only conclude that the experimental and calculated values are satisfactorily in agreement with each other.

The activation energy for the reorientation of the defect causing the 1.8- μ band is about 0.076 eV, which is a little higher than, but comparable with, the value of 0.073 eV for the singly positively charged divacancy reported by Watkins and Corbett.¹ However, the corresponding frequency factor for the 1.8- μ band is about one order of magnitude smaller than that for the singly positively charged divacancy. Certainly the values of E and f_0 for the 1.8- μ band are far from the values for the singly negatively charged divacancy.¹ There has been some argument that the 1.8- μ band might be due to the divacancies of three different charge states, i.e., singly positive, neutral, singly negative.⁴ Our results have certainly ruled out the possibility that the 1.8- μ band arises from the singly negatively charged divacancy. Because of the large difference in the frequency factor between the 1.8- μ band and the singly positively charged divacancy, it seems unlikely that the 1.8- μ band arises from the singly positively charged divacancy. Therefore, our results indicate that the 1.8- μ band only arises from the neutral divacancy. This conclusion does not contradict the experimental fact that the 1.8- and 3.9- μ bands were observed simultaneously in neutron-irradiated *p*-type Si,¹² because of the inhomogeneous distribution of the divacancy in neutron-irradiated *p*-type silicon reported recently by Cheng and Lori.⁷ The activation energy, not the frequency factor, for the 3.3- μ band agrees with that of the singly negatively charged divacancy. From its Fermi-level dependence,¹² the 3.3- μ band could be attributed to arise from the doubly negatively charged divacancy. A very intriguing question comes from the above argument: Why is there no observed absorption band arising from the singly negatively charged divacancy? We have no answer.

5. TEMPERATURE DEPENDENCE

The maximum fractional change of the absorption coefficient due to the divacancy caused by an illumination with polarized light at various experimental conditions can be derived by using the model of the divacancy shown in Fig. 6. In the Appendix, we have derived the maximum fractional change only for the condition that the incident light beam is along the [100] direction of the sample and the electric vector is parallel to the [110] direction (see Fig. 2). The result shows that the change is a function of the reorientation rate L in the ground state, the ratio of the de-excitation rate C to the reorientation rate M in the excited state, the direction of the transition dipole moment, the efficiency of the polarizer, and the light intensity. From

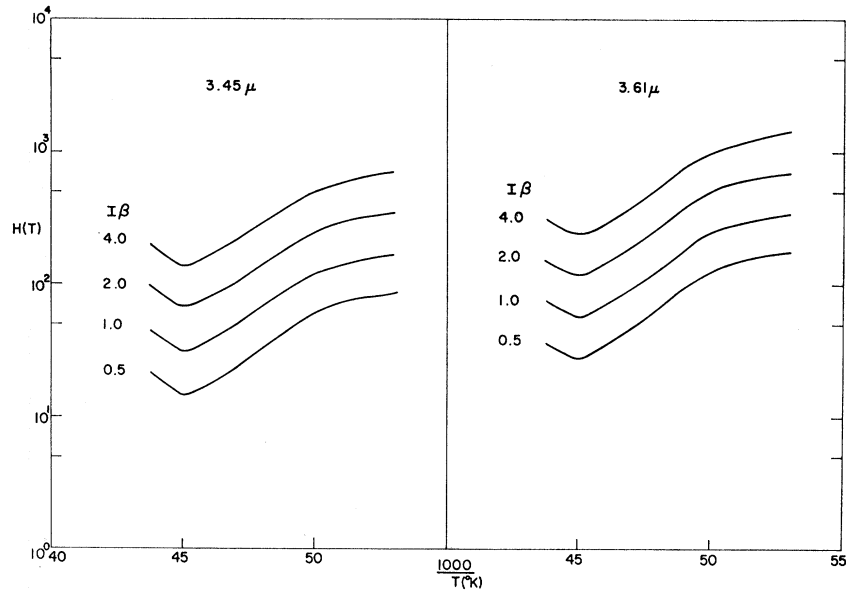


FIG. 8. Temperature dependence of calculated $H(T)$ for the 3.45- and 3.61- μ sub-bands.

previous experiments, we know that the value of θ (see Fig. 6 for definition) is about 10° for both the 1.8- and 3.3- μ bands. According to the manufacturer, the efficiency of the polarizer used is about 97.5% at 2 μ and 99% at 3.5 μ . The value of L at various temperatures can be obtained from the experiments measuring the characteristic time for the randomization of aligned defects in the dark (Fig. 5). Figures 3 and 4 show the experimental values of $\Delta\alpha$ for the 1.8- μ band and the 3.45- and 3.61- μ sub-bands at various temperatures. It is clear from Eq. (A14) in the Appendix that the value of $H(T) \equiv C/M$ at various temperatures can be obtained from our data. The results are shown in Figs. 7 and 8. Because we are unable to measure the absolute light intensity, the results have to be presented as families of curves, depending on the value of βI , where β is the atomic absorption coefficient at a particular wavelength and I the light intensity.

From Figs. 7 and 8, it is clear that the $H(T)$'s for both bands behave similarly. We have no quantitative explanation for the behavior of $H(T)$. Qualitatively, we may argue as follows. In a defect, there are two possible deexcitation processes, radiative and non-radiative. Thus, C may be written as

$$C = P + Qe^{-E'/kT}, \quad (3)$$

with E' as the activation energy in the nonradiative process.¹⁴ If we assume that the reorientation rate in the excited state of the defect can be written as $f''e^{-E''/kT}$, where E'' is the activation energy and f'' the frequency factor, then we have

$$H(T) = \frac{P + Qe^{-E'/kT}}{f''e^{-E''/kT}} = \frac{P}{f''}e^{+(E''-E')/kT} + \frac{Q}{f''}e^{(E''-E')/kT}. \quad (4)$$

¹⁴ D. C. Dexter, *Solid State Phys.* **6**, 406 (1958).

At lower temperatures, the radiative transition process usually dominates. Then $H(T) \cong (P/f)e^{E''/kT}$, i.e., $H(T)$ increases exponentially with $1/T$, roughly in agreement with our results. The slopes of the curves in Figs. 7 and 8 tend to become smaller as the temperature decreases. Because of the inconsistency of the data for L at lower temperatures, presumably caused by background radiation, we cannot obtain proper values for E'' . The fact that the value of $\Delta\alpha$ increases monotonically with the decrease of temperature down to 11°K (see Figs. 4 and 5) may suggest that E'' is very small, probably in the order of the thermal energy at $\sim 10^\circ\text{K}$. At higher temperatures, the nonradiative transition process should be important. Then $H(T) \cong (B/f'')e^{(E''-E')/kT}$, i.e., $H(T)$ increases exponentially with the increase of temperature if $E'' < E'$, which agrees roughly with our results. The condition of $E'' < E'$ is justified since, according to our experimental observation, the reorientation process is rapid at a temperature around 11°K.

We have observed that the polarization efficiency in the 3.61- μ sub-band is about 30% smaller than that of the 3.45- μ sub-band. Because of the same characteristic time for randomization observed in the two sub-bands, the data suggest that the defect has several excited states with different values of $H(T)$. It seems unlikely that the 3.45- and 3.61- μ sub-bands are due to the same transition and that the former is assisted by a phonon emission. However, it is possible that the 3.32- and 3.45- μ sub-bands have the same final state, since their polarization efficiencies are the same.

6. SUMMARY

The following is a summary of the results obtained from our experiments:

(1) An illumination with polarized light of energy corresponding to the 1.8- or 3.3- μ band reduces the absorption coefficient of the band at a temperature below a critical temperature (32.5°K for the 1.8- μ band and 33°K for the 3.3- μ band). The phenomenon is attributed to the electronic reorientation of the divacancy achieved through an excited state by absorbing a photon. A net alignment of the defect can be observed only at a temperature at which the defect in the ground state is usually frozen.

(2) From the temperature dependence of the recovery of the aligned defect in the dark, the activation energies of 0.076 and 0.056 eV are found to be required for the reorientation of the defects in the ground states, causing the 1.8- and 3.3- μ bands, respectively.

(3) A qualitative analysis of the temperature dependence of the reorientation effect indicates that the activation energy required for the reorientation in the excited state is very small, probably in the order of the thermal energy at $\sim 10^\circ\text{K}$.

(4) The results indicate that the 1.8- μ band arises from the neutral divacancy only.

(5) No similar effect has been observed on the 3.9- μ band, which is consistent with the fact that this band contributes photoconductivity.

ACKNOWLEDGMENTS

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APPENDIX

Here we present the derivation of the maximum change in absorption coefficient of a divacancy absorption band by using the model shown in Fig. 6.

In silicon, there are four equivalent atomic orientations for the divacancy. There are three electronic configurations for each atomic orientation, caused by the Jahn-Teller distortion. Each divacancy can be in only one of the three configurations. In unstressed silicon, all the 12 configurations are equally populated. In our experiments, polarized light is incident along the [100] direction of the sample and the electric vector of the light is parallel to the [011] or the [01 $\bar{1}$] direction. Then, the absorption coefficient can be written as

$$\alpha_{11} = \beta \sum_{ij} n_{ij} (P_{ij})_{11}^2,$$

where β is the atomic absorption coefficient of the defect, n_{ij} the number of defects in i th atomic orientation and j th electronic configuration, and $(P_{ij})_{11}$ the component of the transition dipole moment P of the defect in the ij configuration in the direction parallel to the

electric vector of the light. From the Appendix in Ref. 4, we have

$$\alpha_{11} = \beta [n_{11}A + (n_{12} + n_{13})B^+ + n_{21}A + (n_{22} + n_{23})B^- + (n_{32} + n_{33})B^+ + (n_{42} + n_{43})B^-],$$

where

$$A = \cos^2\theta$$

and

$$B^\pm = \frac{1}{4}(\cos\theta \pm \sqrt{2} \sin\theta)^2.$$

Since $n_{12} = n_{13}$, $n_{22} = n_{23}$, $n_{32} = n_{33}$, and $n_{42} = n_{43}$ for our experimental conditions, we have

$$\alpha_{11} = \beta [(n_{11} + n_{21})A + 2(n_{12} + n_{32})B^+ + 2(n_{22} + n_{42})B^-]. \quad (\text{A1})$$

Under an illumination with polarized light of energy corresponding to the absorption energy, the rates of the changes of the numbers of the defects in the three configurations of the $\langle 111 \rangle$ atomic orientation can be written as

$$dn_{11}/dt = \beta AKIn_{11} + Cn_{11} + 2(n_{12} - n_{11}')L, \quad (\text{A2})$$

$$dn_{11}'/dt = \beta AKIn_{11} - Cn_{11}' - 2(n_{11}' - n_{12}')M, \quad (\text{A3})$$

$$dn_{12}'/dt = (n_{11}' - n_{12}')M + \beta I [KB^+ + (1-K)B^-]n_{12} - Cn_{12}', \quad (\text{A4})$$

$$dn_{12}/dt = cn_{12}' - \beta I [KB^+ + (1-K)B^-]n_{12} - (n_{12} - n_{11})L, \quad (\text{A5})$$

where L is the reorientation rate of the defect in the ground state, M the reorientation rate of the defect in excited state, C the deexcitation rate of the defect, K the efficiency of the polarizer used, and the prime representing excited state. All the transitions involved are illustrated in Fig. 9. We have neglected the equations for n_{13} , because $n_{12} = n_{13}$. At an equilibrium condition (i.e., for maximum change), we have

$$dn_{11}/dt = dn_{11}'/dt = dn_{12}'/dt = dn_{12}/dt = 0.$$

Then, from Eqs. (A2)–(A5), we have

$$n_{12} = \frac{N}{4} \frac{A' + F(T)/\beta}{2A' + D^+ + 3I(T)/I\beta} \quad (\text{A6})$$

and

$$n_{11} = \frac{N}{4} \frac{D^+ + F(T)/I\beta}{2A' + D^+ + 3I(T)/I\beta}, \quad (\text{A7})$$

where

$$A' = KA, \quad D^+ = KB^+ + (1-K)B^-, \quad F(T) = L(3 + C/M),$$

T is temperature, and N is the total defect concentration in the sample. Similarly, we have

$$n_{22} = \frac{N}{4} \frac{A' + F(T)/I\beta}{2A' + D^- + 3F(T)/I\beta}, \quad (\text{A8})$$

$$n_{21} = \frac{N}{4} \frac{D^- + F(T)/I\beta}{2A' + D^- + 3F(T)/I\beta}, \quad (\text{A9})$$

for the $\langle 1\bar{1}\bar{1} \rangle$ atomic orientation;

$$n_{32} = \frac{N}{4} \frac{A'' + F(T)/I\beta}{2A'' + D^+ + F(T)/I\beta}, \quad (\text{A10})$$

$$n_{31} = \frac{N}{4} \frac{D^+ + F(T)/I\beta}{2A'' + D^+ + F(T)/I\beta}, \quad (\text{A11})$$

for the $\langle \bar{1}11 \rangle$ atomic orientation; and

$$n_{42} = \frac{N}{4} \frac{A'' + F(T)/I\beta}{2A'' + D^- + F(T)/I\beta}, \quad (\text{A12})$$

$$n_{41} = \frac{N}{4} \frac{D^- + F(T)/I\beta}{2A'' + D^- + F(T)/I\beta}, \quad (\text{A13})$$

for the $\langle \bar{1}\bar{1}\bar{1} \rangle$ atomic orientation; where

$$A'' = (1-K)A$$

and

$$D^- = KB^- + (1-K)B^+.$$

Now we have derived the values for all the n 's at the equilibrium condition. The absorption coefficient mea-

$$\frac{\Delta\alpha}{\alpha_0} = 1 - \frac{3}{2} \frac{1}{(A+2B^++2B^-)} \frac{AD^++2B^+A'+(A+2B^+)F(T)/I\beta}{2A'+D^++3F(T)/I\beta} + \frac{AD^-+2B^-A'+(A+2B^-)F(T)/I\beta}{2A'+D^-+3F(T)/I\beta} + \frac{2B^+A''+2B^+F(T)/I\beta}{2A''+D^++F(T)/I\beta} + \frac{2B^-A''+2B^-F(T)/I\beta}{2A''+D^-+F(T)/I\beta} = f\left(L, \frac{C}{M}, I\beta, \theta, k\right), \quad (\text{A14})$$

where L and C/M are functions of temperature. The experimental values of $\Delta\alpha/\alpha_0$ at various temperatures for the 1.8- and 3.3- μ bands can be obtained from Figs. 4 and 5. The values of L can be obtained from Fig. 6. The efficiency of the polarizer at various wavelengths are known (see Sec. 5). The values of θ for the 1.8- and 3.3- μ bands are about 10° .⁴ Therefore, we have been able to obtain families of curves for the temperature dependence of $H(T) \equiv C/M$ of the two bands, in which $I\beta$ acts as the parameter. The results are shown in Figs. 7 and 8.

It should be noted that, in the above calculation, we have neglected the effect of the polarizer efficiency on the measured absorption coefficient, which should be

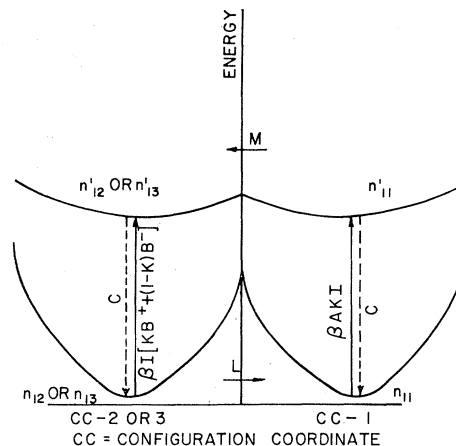


FIG. 9. Schematic configuration diagram to illustrate all the transitions in a $\langle 111 \rangle$ divacancy under an illumination with polarized light of its electric vector parallel to the $[011]$ direction.

sured without polarizer should be

$$\alpha_0 = \frac{1}{6} \beta N (A + 2B^+ + 2B^-).$$

It is easy to see that

$$\Delta\alpha/\alpha_0 = (\alpha_0 - \alpha_{11})/\alpha_0.$$

Then from Eqs. (A1) and (A6)-(A13), we have

written as

$$\alpha = \alpha_{11} \left(1 + \frac{\alpha_1 I_1}{\alpha_{11} I_{11}} \right) \frac{I_{11}}{I_0},$$

where I_1 and I_{11} are the intensities of light with its electric vector perpendicular to and parallel to the polarization direction of the polarizer. Since the efficiency of the polarizer used is rather high (e.g., 0.975 at 2 μ and 0.99 at 3.5 μ), we have used

$$\alpha = \alpha_{11}$$

in our calculation.

In our calculation, we have found that, for a fixed value of $H(T)$, the calculated value of $\Delta\alpha/\alpha$ [Eq. (A14)] is independent of θ in the range $\theta < 45^\circ$.