

and  $V_{KA}$ <sup>5</sup>) of color centers with impurities have been with smaller impurities (e.g.,  $\text{Li}^+$  in sodium halides and  $\text{Li}^+$  and  $\text{Na}^+$  in potassium halides). Past attempts<sup>5</sup> to produce an association between the self-trapped hole and  $\text{Na}^+$  impurities in LiF have been unsuccessful.

Since an associated  $H$  center was not expected, the LiF crystals studies were not deliberately doped with  $\text{Na}^+$  impurities. However, since attempts to produce  $H$  centers in high-purity Harshaw LiF were unsuccessful, the samples used were grown from reagent-grade material. Attempts<sup>8</sup> have been made without success to produce  $H$  centers in high-purity LiF by x rays at 4.2°K and by bombardment with 4.5-MeV electrons at liquid-nitrogen temperatures (sample temperature less than 90°K during bombardment).

Since we have not produced the isolated  $H$  center without the presence of the  $\text{Na}^+$  impurity in LiF, we plan to examine KCl where the production of the  $H$  center apparently occurs

in high-purity material.

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### ELECTRONIC VOLUME EFFECT IN SILICON\*

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Volume contraction of a silicon crystal under intense illumination by laser light has been observed and an explanation of the phenomenon in terms of an electronic effect is proposed.

The generation of elastic waves in solids and liquids due to the absorption of intense, short-duration light pulses available from Q-switched lasers has been discussed previously.<sup>1</sup> The energy of the light absorbed causes heating of a surface layer; because of thermal expansion a compression pulse is propagated into the sample. For a free surface, the boundary conditions require that the compression be followed by a tension pulse of equal magnitude.

In the present experiments, the laser consists of a neodymium-doped glass rod Q switched by a rotating prism to produce pulses of about 25-nsec full width at half-amplitude. For low-intensity light of this wavelength silicon has an absorption constant of  $\alpha = 40 \text{ cm}^{-1}$ . The laser output is monitored by a calibrated ITT FW-114 planar photodiode. The samples are cylindrical single crystals of phosphorus-doped n-type silicon of 30- to 50- $\Omega$ -cm resistivity,

with (111) faces lapped optically flat to 3 fringes and parallel to 30 secs. To measure the amplitude of the stress pulse that has traveled through the specimen, an X-cut quartz stress gauge, 1.3 cm in diameter and 0.6 cm thick, is bonded to the back face of the sample with a thin layer of Nonaq stopcock grease.<sup>2</sup> The piezoelectric current gives a record of stress with time, which is displayed on the oscilloscope. The operation and calibration of such stress gauges have been described by Graham, Neilson, and Benedick.<sup>3</sup>

In Fig. 1(a) is shown the shape of a typical laser pulse incident on the sample, as monitored by the photodiode, on a time scale of 40 nsec/cm. The resulting stress pulse registered by the quartz gauge is depicted in Fig. 1(b); the time scale is 200 nsec/cm and the gauge is oriented such that an upward deflection on the oscilloscope screen indicates tension, while down-

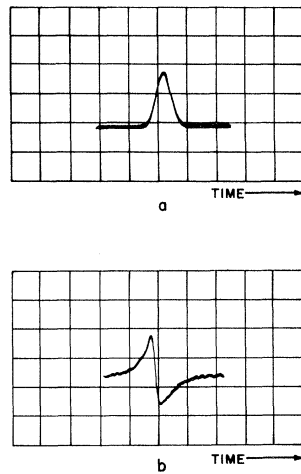


FIG. 1. (a) Shape of laser pulse incident on sample. Time scale 40 nsec/cm. (b) Stress pulse registered by quartz gauge. Tension indicated by upward and compression by downward deflection. Time scale 299 nsec/cm.

ward indicates compression. The polarity of the observed response is opposite to that expected for a lattice dilation due to thermal expansion, and thus implies that a lattice contraction occurs initially. The plot of Fig. 2 indicates that the peak tension increases linearly with incident light intensity. This is true only in the relatively low-energy-density regime ( $<0.1$  J/cm<sup>2</sup>), below the level where visible surface damage takes place. It should be noted that light pulses from a laser incident on filter glass with an optical absorption constant of 40 cm<sup>-1</sup> do produce stress pulses of the polarity expected for thermal expansion.<sup>4</sup>

A lattice contraction can be interpreted in

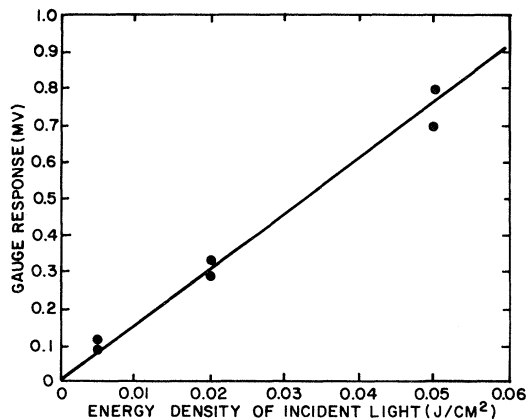


FIG. 2. Peak of tension pulse as a function of incident light intensity.

terms of a semiconductor crystal's volume dependence on the occupation of the electronic energy levels. Keyes<sup>5</sup> showed that a density  $n$  of excess electrons in the conduction bands leads to a dilation

$$\Delta c/c = -n(3\Xi_d + \Xi_u)/9B, \quad (1)$$

and that the corresponding change due to a concentration  $p$  of holes in the valance band is

$$\Delta c/c = -p(3\Xi_d')/9B, \quad (2)$$

where  $c$  is the lattice constant,  $B$  is the bulk modulus, and  $\Xi_d$ ,  $\Xi_u$ , and  $\Xi_d'$  are the appropriate deformation potential constants. The pressure derivative of the energy gap can be written

$$dE_G/dP = -(3\Xi_d + \Xi_u + 3\Xi_d')/3B, \quad (3)$$

and since for electron-hole pairs  $n=p$ , the total change due to the addition of carrier pairs is simply

$$\frac{\Delta c}{c} = \frac{dE_G}{dP} \frac{n}{3}. \quad (4)$$

Thus the sign of the effect is the same as that of the pressure coefficient of the energy gap, which is positive for germanium but negative for silicon. Photoexcitation of electron-hole pairs therefore should cause the germanium lattice to expand, while silicon should contract under the same conditions. For germanium the effect has been demonstrated experimentally by Figielski,<sup>6</sup> who measured the length increase of  $n$ -type germanium samples into which excess pairs of current carriers were introduced by monochromatic illumination. North and Buschert<sup>7</sup> interpreted their length-change measurements of electron-irradiated germanium by taking into account the same electronic effect.

If a semiconductor is illuminated with light of a wavelength corresponding to photon energies greater than the band gap, electron-hole pairs will be produced throughout a layer whose thickness depends on the optical absorption properties of the material. According to the above calculation a silicon lattice will contract in this region, producing a tensile stress pulse which propagates into the sample, followed, for the case of a free surface, by a compression of equal magnitude. The tensile stress can be observed if the competing effect of surface heating is low enough. This is the case in the pres-

ent experiment, since the wavelength of the incident light,  $1.06\mu$ , corresponds to a photon energy of 1.17 eV, which is just above the 1.11-eV indirect band gap of silicon at room temperature. At this wavelength a large portion of the absorbed energy may be expected to produce electron-hole pairs. In interpreting the results quantitatively, however, one must consider the change of the optical properties of the sample when large numbers of free carriers are generated. It has been observed, for example, that the reflectivity of silicon increases linearly with light intensity under laser illumination and may more than double before visible surface damage occurs.<sup>8</sup> Expressing the reflectivity in terms of the refractive index  $N$  and extinction coefficient  $K$ ,

$$R = [(N-1)^2 + K^2] / [(N+1)^2 + K^2]^{-1}, \quad (5)$$

one finds that a doubling of the low incident intensity value of  $R = 0.32$  is associated with an increase of the optical absorption constant from  $40 \text{ cm}^{-1}$  to  $\sim 5 \times 10^5 \text{ cm}^{-1}$ , if  $N$  is assumed to remain constant. This implies a large increase in free-carrier absorption, and hence thermal energy deposit, after the first portion of the light pulse has excited free carriers. The relative amounts of the total energy contributing to carrier excitation and to thermal heating depend on the change with time of the optional properties while the sample is being illuminated.

From Fig. 2 it is seen that an incident light energy density of  $0.03 \text{ J/cm}^2$  corresponds to a gauge response of 0.5 mV, indicating a stress of about 0.5 bar.<sup>3</sup> Possible losses in the bond between sample and gauge and distortions in the gauge itself are neglected here, but the difference in acoustic impedance of sample and gauge is considered. For one-dimensional strain in the  $\langle 111 \rangle$  directions, the change in lattice constant is

$$\frac{\Delta c}{c} = \frac{3\sigma}{c_{11} + 2c_{12} + 4c_{44}} = -2.9 \times 10^{-7}, \quad (6)$$

where  $c$  is the lattice constant,  $\sigma$  is the stress, and  $c_{ij}$  are the elastic constants. The observed strain is the resultant of the electronic volume effect and of thermal expansion,

$$\frac{\Delta c}{c} = \left( \frac{\Delta c}{c} \right)_{\text{el}} + \left( \frac{\Delta c}{c} \right)_{\text{th}}. \quad (7)$$

To calculate the magnitudes of the two effects, consider the dilation of a uniformly illuminat-

ed volume of a sample. From the analysis given by Keyes<sup>5</sup> and Figielski,<sup>6</sup> the change of lattice constant for the electronic volume effect in silicon can be written

$$\left( \frac{\Delta c}{c} \right)_{\text{el}} = -1.0 \times 10^{-24} n, \quad (8)$$

where  $n$  is the concentration of electron-hole pairs. The dilation due to thermal heating can be approximated by

$$\left( \frac{\Delta c}{c} \right)_{\text{th}} = \frac{\rho(h\nu)n'}{C} = 0.3 \times 10^{-24} n', \quad (9)$$

where  $\rho$  is the coefficient of linear thermal expansion,  $h\nu$  the photon energy,  $C$  the specific heat, and  $n'$  the effective density of photons whose energy contributes entirely to thermal expansion. From the measured value of the stress [Eq. (6)] and from expressions (8) and (9) an estimate can be made of the relative amounts of the total deposited energy that contribute to electron-hole pair excitation and to thermal expansion. If, as a first approximation, the simplifying assumption of uniform optical absorption throughout a surface layer of the sample is made, the calculated partition of energy is nearly independent of the assumed penetration depth. For values of the absorption constant from 40 to  $40 \times 10^3 \text{ cm}^{-1}$ , from 22 to 30 % of the photon energy must be used for carrier excitation, and the remainder deposited thermally, to account for the measured stress. The large thermal effect is plausible in view of the above discussion of free-carrier absorption.

For a better quantitative correlation between experiment and theory, i.e., a direct check of Eq. (8), an independent determination of the ratio of ionizing absorption to free-carrier absorption in silicon at high carrier concentrations is in progress.

In the present experiment the electronic volume effect dominates and has, in silicon, the negative sign predicted by theory.

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### ISOMER SHIFTS IN THE $2^+ \rightarrow 0^+$ ROTATIONAL TRANSITION OF $\text{Yb}^{170}$

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We have carried out Mössbauer measurements of isomer shifts on the 84-keV,  $2^+ \rightarrow 0^+$  rotational transition of  $\text{Yb}^{170}$  with the objective of measuring  $\delta\langle r^{-2} \rangle$ , the difference in the mean-square charge radius between the  $2^+$  and  $0^+$  ground-state rotational levels. Such measurements are very sensitive to nonadiabatic effects in rotational nuclei and have a bearing on such interesting questions as whether or not particle-particle forces in rotating nuclei are dependent upon the angular velocity.

In a previous work,<sup>1</sup> in which an isomer shift between rotational levels was measured, the estimate of the chemical factor which appears in the theoretical expression for the isomer shift was not of sufficient accuracy to draw definite conclusions about the nuclear states. In the measurements reported here we have taken advantage of the relatively regular features of rare-earth salts and intermetallic compounds to obtain a result for  $\delta\langle r^{-2} \rangle$  of sufficient accuracy to be of theoretical interest. For the first time the sign of  $\delta\langle r^{-2} \rangle$  has definitely been established.

The measurements were made at 4.2°K with a source of irradiated  $\text{TmAl}_2$ . The absorbers consisted of Yb metal, various intermetallic compounds, and salts with Yb ions of valencies 2+ and 3+. All the absorbers used showed an unsplit absorption line narrower than four times the minimum possible width ( $2\Gamma_n$ ). Previous measurements with other  $\text{Yb}^{3+}$  salts all showed complex spectra. All the absorption lines were symmetrical, so that isomer shifts could be accurately determined. Each isomer shift determination was made from a number of spectra, some of which are shown in Fig. 1. Throughout the measurements frequent checks were made on the energy calibration. For most absorbers, spectra were also taken with the energy scale expanded by a factor of 2. All the results, which are listed in Table I, are referred to the  $\text{YbAl}_2$  absorber. The errors quoted are larger than the statistical errors and take into account possible instrumental instabilities.

The main results were these:

(1) The maximum shift between 3+ and 2+ ions occurred between  $\text{YbGa}$  garnet and  $\text{YbSO}_4$  and was  $+0.63 \pm 0.10$  mm/sec.

(2) The maximum shift between Yb metal and salts containing  $\text{Yb}^{2+}$  ( $\text{YbSO}_4$ ) was  $+0.36$

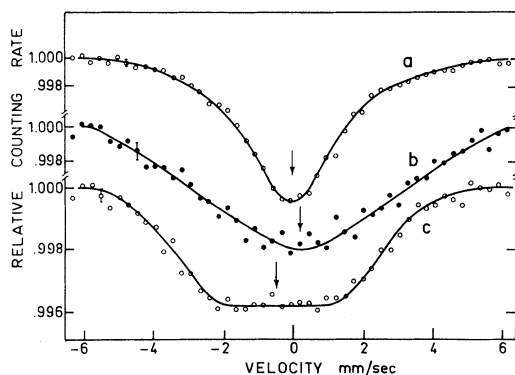


FIG. 1. The absorption of the 84-keV gamma rays of  $\text{Yb}^{170}$  at 4.2°K as a function of the relative velocity between source and absorber. (a)  $\text{YbAl}_2$  absorber. (b)  $\text{YbGa}$  garnet absorber. (c)  $\text{YbSO}_4$  absorber.

Table I. Summary of isomer-shift results.

Compound	$E$ (mm/sec)
$\text{YbCl}_2$	$-0.25 \pm 0.07$
$\text{YbSO}_4$	$-0.39 \pm 0.07$
Yb metal	$-0.03 \pm 0.07$
$\text{YbAl}_2$	0.00
$\text{YbSi}_2$	$+0.02 \pm 0.07$
$\text{YbCl}_3$ anhydrous	$+0.11 \pm 0.07$
$\text{YbGa}$ garnet	$+0.24 \pm 0.07$