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Structure of acceptor-hydrogen and donor-hydrogen complexes in silicon from uniaxial stress studies

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We report the results of a uniaxial stress study of the vibrational absorption due to hydrogen-passivated boron acceptors and arsenic donors in silicon. Under stress, the H-related vibrational bands shift and split in frequency. From the observed splittings and intensity ratios we determine that the symmetry of both centers is trigonal. For B-H we observe a large sensitivity to stress which we speculate is due to the tendency of H to move off the ⟨111⟩ axis of the complex under compression. The two vibrational bands of As-H are confirmed to be due to stretching and doubly degenerate wagging modes. Our results provide support for the structural model proposed by Johnson *et al.* [Phys. Rev. Lett. **56**, 769 (1986)] for donor-H complexes in which the H is at the antibonding site of one of the donor's Si neighbors.

I. INTRODUCTION

Hydrogen in semiconductors has attracted much recent attention,[1,2] partly because hydrogen may be inadvertently introduced during several processes commonly used in the electronics industry. In many cases hydrogen neutralizes or passivates the electrical and optical activity of defects in the materials. Almost all of the basic properties associated with hydrogen, such as the mechanism for H diffusion or the structures of H-containing complexes, are controversial. In silicon, the passivation of the group-III acceptors, B, Al, and Ga, is well known[3,4] and has been studied extensively. The recently discovered passivation of group-V donors by hydrogen[5] is a weaker effect than the passivation of acceptors and is not as well studied or understood. The donor-H and acceptor-H complexes provide excellent model systems for the study of H-related defects. Here we examine these complexes with infrared absorption and uniaxial stress to gain insight into their structure and properties.

Of the models that have been proposed for B-H complexes, the model where the H atom is bonded to one of the boron's Si nearest neighbors [Fig. 1(a)] is gaining acceptance. This model was proposed by Pankove *et al.*[6] and was later confirmed to be the lowest-energy configuration in cluster calculations by DeLeo and Fowler.[7] Recent calculations by Chang and Chadi[8] and by Van de Walle *et al.*[9] also find that this configuration has lowest energy. Ion channeling and nuclear reaction analysis experiments[10,11] on B-donor complexes have determined that the donor occupies the bond-centered site between boron and one of its Si neighbors. Although we take the bond-centered configuration for the hydrogen to be correct and interpret our data accordingly here, we note that some groups still favor an antibonding configuration for the H in the acceptor-H complexes.[12-14]

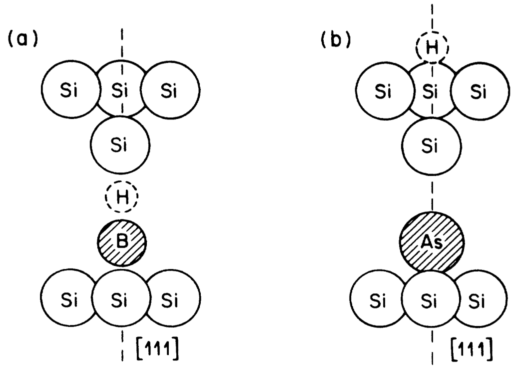

FIG. 1. Schematic models of (a) the B-H complex and (b) the As-H complex.

The vibrational frequencies of acceptor-H complexes[15] (1903 cm^{-1} for B-H, 2201 cm^{-1} for Al-H, and 2171 cm^{-1} for Ga-H complexes at liquid-He temperature) depend strongly on the acceptor species. Further, there are anomalously large shifts and broadenings of these bands with increasing temperature.[15-17] For example, the B-H band shifts to 1875 cm^{-1} at room temperature. In addition to large frequency shifts, the Al-H and Ga-H vibrational spectra show thermally populated sidebands to the low-energy side of the main vibrational bands.[17] Such sidebands are reminiscent of the rich vibrational structure associated with oxygen in Si.[18,19] Studies of these sidebands and knowledge of the previous work on the Si:O problem have led to the suggestion that for Al-H and Ga-H complexes, the hydrogen is pushed off the [111] axis by these larger acceptors.[17] Low-frequency motions of the H about the [111] axis would give rise to a ladder of closely spaced states for each H-stretching level (much as rotations add fine structure to vibrational levels in molecular spectroscopy) and hence the observed sidebands. For B-H complexes there are no sidebands resolved although the large-frequency shift with increasing temperature the similarity to Al-H and Ga-H.

Recent Hall-effect measurements by Johnson *et al.* have demonstrated the weak passivation of Si:P by H_2

plasma exposure.[5] The formation of donor-H complexes was inferred to be responsible for the passivation from the increase in the Hall mobility. Donor-H complex formation was confirmed by the observation of H vibrations at passivated P, As, and Sb donors by Bergman *et al.*[20] All three donor-H complexes give rise to absorption bands close to 1560 cm^{-1} and at 809 cm^{-1}, which indicates that they are very similar in structure. The 1560-cm^{-1} band was assigned to a stretching mode and the 809-cm^{-1} band to a doubly degenerate wagging mode. The absence of splitting for the wagging vibration suggests that the symmetry of the complex is trigonal or higher. In contrast to the bands due to acceptor-H complexes, the donor-H bands show no unusual temperature dependence or additional structure. The bands have widths of ~ 1 cm^{-1} at liquid-He temperature and shift ~ 5 cm^{-1} when the temperature is raised to room temperature.

On the basis of semiempirical calculations,[5] Johnson *et al.* put forward a structural model for the donor-H complex in which the H atom is attached to one of the donor's Si nearest neighbors in an antibonding position along the $\langle 111 \rangle$ axis [Fig. 1(b)]. Our previous ir-absorption results[20] support this model. The vibrational frequencies for donor-H complexes have little or no donor dependence suggesting that the H is not bonded to the donor directly. A doubly degenerate wagging mode is consistent with the trigonal symmetry of the proposed complex. Chang and Chadi[8] have recently performed *ab initio* pseudopotential calculations and have found that the structure shown in Fig. 1(b) is the most stable for P-H complexes. We note that neither the semiempirical[5] nor *ab initio*[8] calculations correctly predict the observed vibrational frequencies[20] of the P-H complex.

In this work, we have examined the absorption bands of B-H and As-H under uniaxial stress. The uniaxial stress results allow us to determine the symmetry of the complexes and to confirm our previous mode assignments[20] for donor-H complexes. There has been no direct determination of the symmetry of the donor-H complexes by other means. Further, the magnitude and sign of the stress-induced splittings for B-H and As-H bands provide additional insight into the structures of the complexes.

II. TRIGONAL CENTERS UNDER UNIAXIAL STRESS

Defects in Si with lower symmetry than tetrahedral can have several equivalent orientations. For example, a trigonal center can have its threefold axis along any of the $\langle 111 \rangle$ axes. In the absence of an applied stress, centers oriented along different axes are orientationally degenerate. The application of stress to the crystal in a general direction lifts this degeneracy, which results in a splitting of the spectral bands.[21] Symmetry puts consistency restrictions on the observed splittings for different directions of stress. The splittings can be expressed in terms of a few parameters, the higher the symmetry, the fewer the parameters needed. Furthermore, through the use of polarized light in uniaxial stress experiments, centers with different orientations can be selectively excited. The combination of the observed splitting pattern and intensity ratios of the stress split components makes possible a consistent assignment of spectral features and symmetry of the center. The literature provides comprehensive tables to which experimenters can compare their data. Our analysis is based upon the results of Kaplyanskii[22] and those of Hughes and Runciman.[23]

Our uniaxial stress results for the two complexes in this study are consistent with trigonal symmetry. For a trigonal center, the splitting of spectral bands due to A_1 stretching modes are described by two parameters, $\mathcal{A}_1$ and $\mathcal{A}_2$. The parameter $\mathcal{A}_1$ is proportional to the hydrostatic component of the stress, and gives rise to a stress direction-independent shift in frequency, whereas $\mathcal{A}_2$ induces a shift which depends on the orientation of the center. Expressions for the splittings and the intensity ratios for the absorption of polarized light for an A_1 mode of a trigonal center are given in Table I.

The case of a doubly degenerate E mode of a trigonal center is somewhat more complicated, since we also lift the vibrational degeneracy of the modes as well. Hence, two additional parameters, $\mathcal{B}$ and $\mathcal{C}$, are introduced in the analysis. $\mathcal{A}_1$ and $\mathcal{A}_2$ retain their meaning, and $\mathcal{B}$ and $\mathcal{C}$ lift the degeneracy of the two modes at each center. Expressions for the splittings and the intensity ratios for polarized absorption for an E mode of a trigonal center are given in Table II.

III. EXPERIMENTAL PROCEDURES

The samples for this study were cut from floating zone Si crystals oriented by Laue x-ray backscattering. The surfaces to which stress was to be applied were lapped flat and parallel. The crystals were cut into slabs, polished, and ion implanted with B or As. The energies were 30, 100, and 180 keV, each to a dose of 7×10^{14} cm^{-2}, for B, and 30 and 100 keV, each to a dose of 1×10^{15} cm^{-2}, for As. The implants were activated by a rapid thermal anneal at 1200 °C for 60 s. The slabs were cut into bars, typically $8 \times 2 \times 2$ mm^3, with the long axis parallel to the stress direction. The bar shaped samples were hydrogenated in a Technics Planar Etch II plasma reactor at 120 °C for 3 h on each side.

TABLE I. Theoretical expressions for the stress-induced frequency splitting Δ of a symmetrical A_1 vibrational mode of a trigonal center in Si. Intensity ratios of the stress-split components for different polarizations of absorbed light are also given. **F** is the applied force, σ the magnitude of the stress, and **E** the electric vector of the light.

		Intensity and polarization		
Direction of stress	Δ	$\mathbf{E}\parallel\mathbf{F}$		$\mathbf{E}\perp\mathbf{F}$
$\mathbf{F}\parallel[001]$	$\mathcal{A}_1\sigma$	4		4
$\mathbf{F}\parallel[111]$	$(\mathcal{A}_1+2\mathcal{A}_2)\sigma$	3		0
	$(\mathcal{A}_1-\frac{2}{3}\mathcal{A}_2)\sigma$	1		4
		$\mathbf{E}\parallel\mathbf{F}$	$\mathbf{E}\parallel[001]$	$\mathbf{E}\parallel[1\bar{1}0]$
$\mathbf{F}\parallel[110]$	$(\mathcal{A}_1+\mathcal{A}_2)\sigma$	4	2	0
	$(\mathcal{A}_1-\mathcal{A}_2)\sigma$	0	2	4

TABLE II. Theoretical expressions for the stress-induced frequency splitting Δ of a doubly degenerate *E* vibrational mode of a trigonal center in Si. Intensity ratios of the stress-split components for different polarization of absorbed light are also given. **F** is the applied force, σ the magnitude of the stress, and **E** the electric vector of the light.

Direction of stress	Δ	Intensity and polarization		
		$\mathbf{E}\parallel\mathbf{F}$		$\mathbf{E}\perp\mathbf{F}$
$\mathbf{F}\parallel[001]$	$(\mathcal{A}_1-2\mathcal{B})\sigma$	16		4
	$(\mathcal{A}_1+2\mathcal{B})\sigma$	0		12
$\mathbf{F}\parallel[111]$	$(\mathcal{A}_1+2\mathcal{A}_2)\sigma$	0		6
	$(\mathcal{A}_1-\frac{2}{3}\mathcal{A}_2-\frac{4}{3}\mathcal{C})\sigma$	16		1
	$(\mathcal{A}_1-\frac{2}{3}\mathcal{A}_2+\frac{4}{3}\mathcal{C})\sigma$	0		9
		$\mathbf{E}\parallel\mathbf{F}$	$\mathbf{E}\parallel[001]$	$\mathbf{E}\parallel[1\bar{1}0]$
$\mathbf{F}\parallel[110]$	$(\mathcal{A}_1+\mathcal{A}_2-\mathcal{B}+\mathcal{C})\sigma$	0	0	12
	$(\mathcal{A}_1+\mathcal{A}_2+\mathcal{B}-\mathcal{C})\sigma$	4	8	0
	$(\mathcal{A}_1-\mathcal{A}_2+\mathcal{B}+\mathcal{C})\sigma$	0	8	4
	$(\mathcal{A}_1-\mathcal{A}_2-\mathcal{B}-\mathcal{C})\sigma$	12	0	0

The spectra were measured with a Nicolet 60SX or a Bomem DA3.02 Fourier-transform spectrometer. Each instrument was equipped with a continuous-flow cryostat with KRS-5 windows, a globar ir source, and a Ge on KBr beamsplitter. The Nicolet instrument was equipped with a pyroelectric TGS detector, and the Bomem instrument with a Ge:Cu photoconductive detector. All measurements were made near liquid-helium temperature.

Stress was applied using a push rod system. The force was generated by a pneumatic cylinder. The stress was calculated from the gas pressure in the pneumatic cylinder and the areas of the cylinder and sample. The incident light was polarized with a wire-grid polarizer on a KRS-5 substrate.

IV. UNIAXIAL STRESS DEPENDENCE OF THE VIBRATIONAL ABSORPTION

Spectra of the 1903-cm^{-1} stretching mode of the B-H complex are shown in Fig. 2 for different stress orientations and polarizations of absorbed light. Both the stress induced splittings and the intensity ratios of the bands are consistent with an A_1 mode of a trigonal center. Figure 3 shows the stretching frequency as a function of stress for the high-symmetry stress directions.[24] The solid lines have been drawn using the expressions in Table I with values of the parameters $\mathcal{A}_1$ and $\mathcal{A}_2$ obtained from a numerical fit to the data. The parameters are given in Table III.

The stress dependence of the transition frequency of the 1561-cm^{-1} mode of the As-H complex is shown in Fig. 4. The solid lines represent fits to the data with the parameters given in Table III. The splittings and polarization data are well explained by an A_1 stretching mode of a trigonal complex. We note that the weaker component expected for [111] stress and $\mathbf{E}\parallel\mathbf{F}$ was not resolved because the stress splitting is smaller than the linewidth.

Spectra of the 810-cm^{-1} mode of the As-H complex are shown in Fig. 5 for different stress orientations. The features split into more components than the A_1 stretching modes. This is expected for an *E* wagging mode because the stress lifts the vibrational degeneracy as well as the orientational degeneracy as was discussed in Sec. II. In Fig. 6 the transition frequencies are plotted as a function of stress.[24] The solid lines represent fits of the expressions given in Table II to the data with the parameters given in Table III. The splittings and intensity ratios of the bands are entirely consistent with our previous assignment of the 810-cm^{-1} mode to the H wagging vibration of the As-H complex.

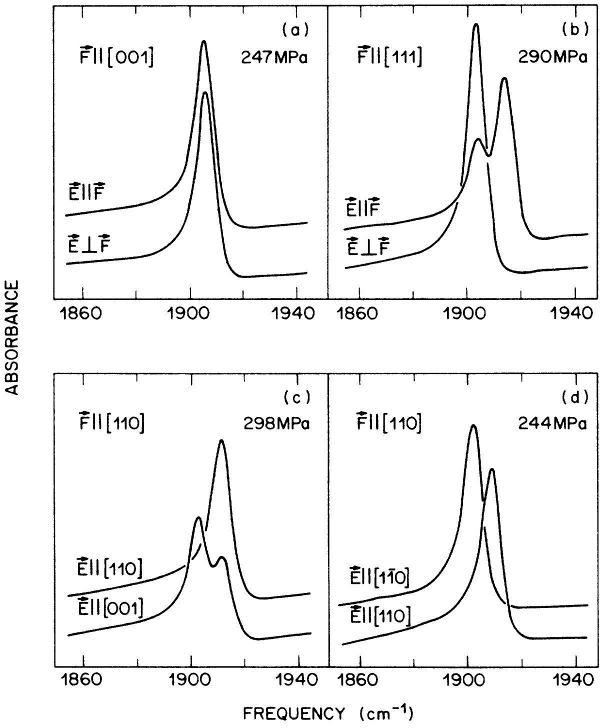

FIG. 2. Spectra taken at liquid-He temperature with polarized light of the Si:B-H stretching mode under stress. **F** is the applied force and **E** the electric vector of the light.

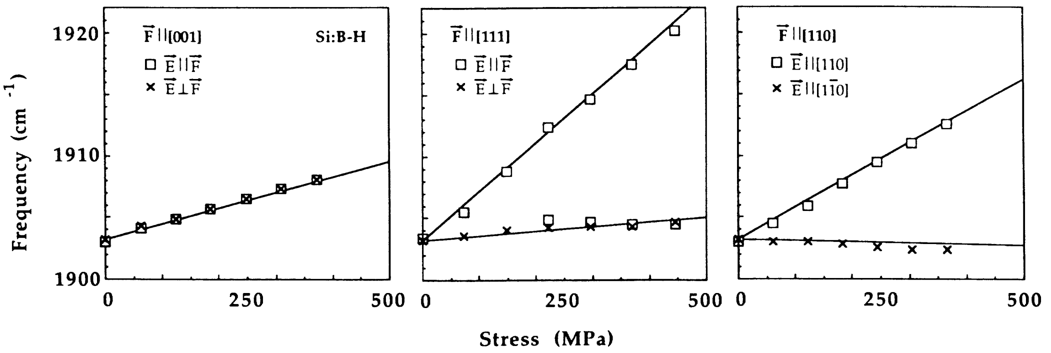

FIG. 3. Stress-induced shifts together with polarization characteristics of the Si:B—H stretching mode at 1903 cm^{-1} for different directions of stress. **F** is the applied force and **E** the electric vector of the light. The solid lines are drawn according to the theoretical expressions in Table I, using the parameters in Table III.

V. DISCUSSION

Our uniaxial stress results for the B-H and As-H vibrational modes are explained well by complexes with trigonal symmetry. For B-H, this symmetry assignment agrees with channeling results reported previously.[10,11] For As-H, this is the first experimental determination of the symmetry of the donor-H complexes of which we are aware. Our uniaxial stress results for the donor-H complexes confirm our previous assignment[20] of the 1561-cm^{-1} band to a longitudinal H-stretching vibration and the 810-cm^{-1} band to a transverse wagging mode.

The stress parameters $\mathcal{A}_1$ and $\mathcal{A}_2$ of B-H given in Table III are very large for local vibrational modes; for example, they are an order of magnitude larger than those of the 1561-cm^{-1} mode for As-H. We suggest that this large coupling to the stress can be qualitatively understood by considering the behavior of the larger acceptors, Al and Ga, when complexed with hydrogen. Al-H and Ga-H complexes have vibrational frequencies near 2200 cm^{-1}, ~300 cm^{-1} larger than the frequency of the B-H complex.[15] For these larger acceptors it has been suggested that the H has been squeezed off the [111] axis.[17] We recall that the H vibration of the B-H complex is essentially a Si—H stretching motion[6,7] and that this frequency is depressed from the ~2100–2300-cm^{-1} value expected for Si—H bonds[25] because of the crowded, bond-centered configuration. We propose that for the larger acceptors the H is pushed off axis to relieve the crowding, the Si—H bond is less compressed, and consequently the Si—H frequency increases to near its more common value. The H in the B-H complex might have a tendency to be pushed off axis by an applied compressive stress and the off-axis motion could then cause the vibrational frequency to increase toward the frequencies of the Al-H and Ga-H complexes. We note that the Al-H and Ga-H complexes have qualitatively different behavior than the B-H complex under stress supporting the notion that they might have a different (i.e., off-axis) zero-stress configuration.[26] The theoretical calculations of Chang and Chadi[8] and of Van de Walle *et al.*[9] show that H motions perpendicular to the B-H-Si axis in Fig. 1(a) do not significantly increase the total energy of the complex, further underscoring the importance of off-axis motions and distortions for the acceptor-H complexes.

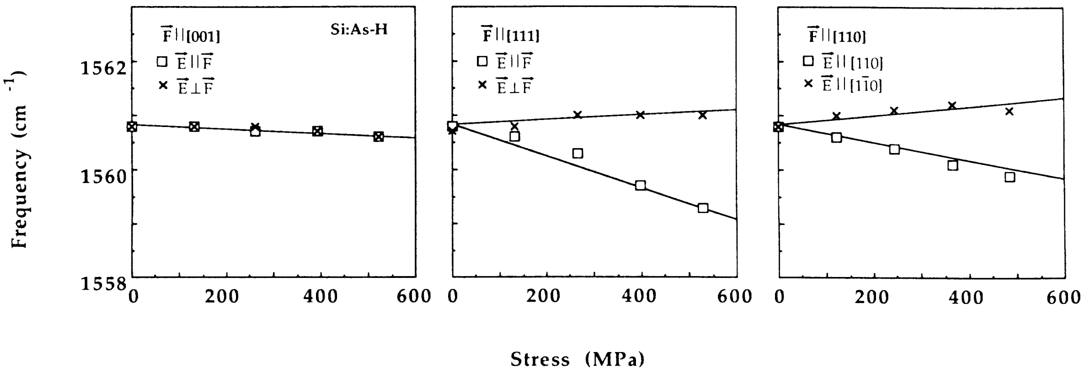

FIG. 4. Stress-induced shifts together with the polarization characteristics of the Si:As—H stretching mode at 1561 cm^{-1} for different directions of stress. **F** is the applied force and **E** the electric vector of the light. The solid lines are drawn according to the theoretical expressions in Table I, using the parameters in Table III.

TABLE III. Experimentally obtained values for the stress parameters of the vibrations studied in this work given in $\text{cm}^{-1}/\text{MPa}$. The values were obtained from numerical fits of the data to the expressions in Tables I and II.

Si:B—H
$\mathcal{A}_1 = 1.27 \times 10^{-2}$
$\mathcal{A}_2 = 1.35 \times 10^{-2}$

Si:As—H stretch
$A_1 = -4.0 \times 10^{-4}$
$\mathcal{A}_2 = -12.5 \times 10^{-4}$

Si:As—H wag
$\mathcal{A}_1 = 9.1 \times 10^{-4}$
$\mathcal{A}_2 = 6.4 \times 10^{-4}$
$\mathcal{B} = 7.6 \times 10^{-4}$
$\mathcal{C} = 31 \times 10^{-4}$

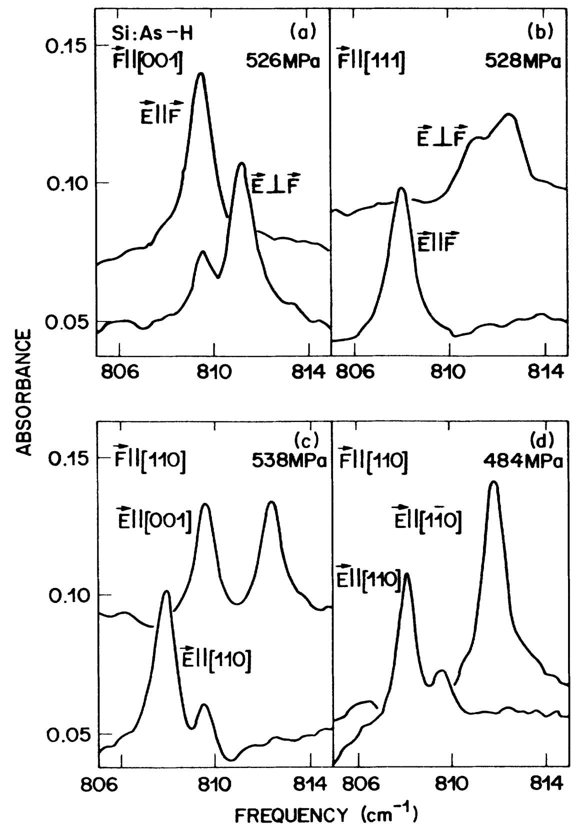

FIG. 5. Spectra taken at liquid-He temperature with polarized light of the doubly degenerate Si:As—H wagging mode under stress. **F** is the applied force and **E** the electric vector of the light.

The signs of the stress-coupling parameters provide additional insight into the structure of the As-H complex beyond the determination of symmetry. The parameter $\mathcal{A}_1$, which characterizes the hydrostatic component of the stress-induced shift to the center of gravity of a band, is negative for the 1561-cm^{-1} stretching mode and positive for the 810-cm^{-1} wagging mode. Hence, compressive stress reduces the frequency difference between the centers of gravity of the stretching and wagging bands. We consider these stress shifts in the context of the donor-H model with H in the antibonding configuration[5] shown in Fig. 1(b). If the H were vibrating in a tetrahedral environment then the distinction between stretching and wagging would disappear and these modes would be degenerate. Thus, the frequency difference between these bands is a measure of the deviation from tetrahedral toward trigonal symmetry. The applied compressive stress reduces this frequency difference in our experiments and the "axiality" of the complex. This is consistent with the stress pushing the antibonding H shown in Fig. 1(b) toward the tetrahedral interstitial site.

In Chang and Chadi's calculations[8] on the P-H complex, the H is near the tetrahedral interstitial site. We propose that its stable position is closer to the donor atom's nearest-neighbor silicon on the basis of the frequency difference between longitudinal and transverse H stretching frequencies (which are incorrectly predicted in the calculation) and the uniaxial stress results described above.

The Raman feature observed at 1906 cm^{-1} for the B-H complexes has been examined under uniaxial stress recently by Stutzmann and Herrero.[16] No splitting was observed for a ⟨111⟩ stress and a large splitting was ob-

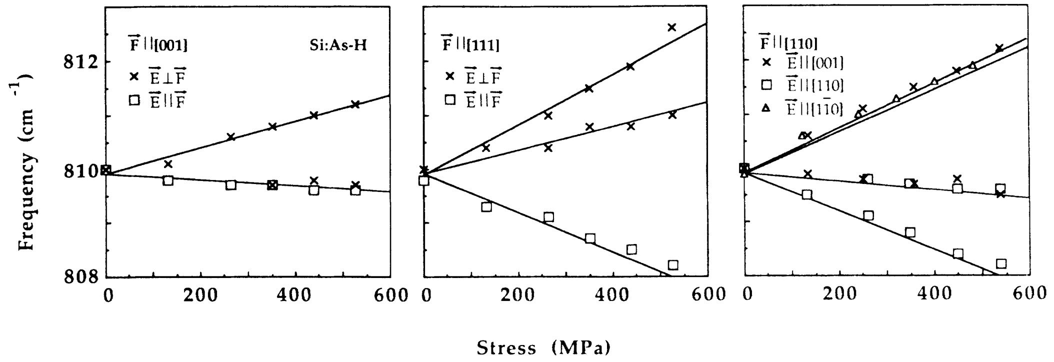

FIG. 6. Stress-induced shifts together with polarization characteristics of the doubly degenerate Si:As—H wagging mode at 810 cm^{-1} for different directions of stress. **F** is the applied force and **E** the electric vector of the light. The solid lines are drawn according to the theoretical expressions in Table II, using the parameters in Table III.

served for ⟨100⟩ stress. These results are in marked disagreement with the results we have reported here. At present, we have no explanation for this discrepancy.

VI. CONCLUSION AND SUMMARY

We have used infrared spectroscopy and uniaxial stress to study hydrogen-passivated B acceptors and As donors in Si. From the observed splittings and intensity ratios observed in the stress experiments we find that both the B-H and As-H complexes have trigonal symmetry.

The trigonal symmetry of the B-H complex is in agreement with several theoretical calculations[7–9,12] and recent channeling results for B-D complexes which favor the bond-centered configuration[10,11] shown in Fig. 1(a). We have found previously that the vibrational spectra of acceptor-H complexes are sensitive to the acceptor species and show large frequency shifts for the H stretching bands with temperature.[17] Here, the H stretching frequency has been reported to be unusually sensitive to stress for the B-H complex. We have proposed, in the context of the bond-centered model, that off-axis motions or distortions of the H might be responsible for these unusual characteristics of the acceptor-H complexes.

For As-H complexes, our previous assignments[20] of the 1561-cm^{-1} mode to longitudinal H stretching and the 810-cm^{-1} mode to transverse H wagging have been confirmed. The trigonal symmetry found here and the previous result that the vibrational frequencies are insensitive to the donor species provide support for the donor-H model shown in Fig. 1(b) that was proposed by Johnson *et al.*[5] Because the vibrational characteristics of the donor-H complexes for P, As, and Sb are similar we expect that they will all behave similarly under uniaxial stress.

Although both the B-H and As-H complexes are trigonal, their vibrational characteristics and behavior under stress are dramatically different. The crowded, bond-centered configuration of acceptor-H complexes gives rise to large frequency shifts with temperature and large stress coupling. The donor-H complexes have the H weakly bonded to Si in an antibonding configuration and have more ordinary, local-vibrational-mode properties.

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