

Absence of dichroism for the DX optical-bleaching transients in $Al_{0.35}Ga_{0.65}As:Te$

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We have attempted to observe preferential ground- to metastable-state conversion by polarized light of a subgroup of DX centers in $Al_{0.35}Ga_{0.65}As:Te$. The expected effect was calculated for transition moments either parallel or perpendicular to the defect symmetry axes for C_{3v} , D_{2d} , C_{2v} , and C_{1h} defect symmetries. The experiment was performed by monitoring the transient transmission through an optically thick specimen for various polarization sequences. We found no statistically significant effect, and the theoretical expectations fall well outside the limits determined from the experimental uncertainty. We conclude that the transition moment for the optical conversion of the DX center is very nearly isotropic, which leads to one of two possibilities. Either both parallel and perpendicular dipoles accidentally contribute simultaneously to the DX -center ionization band, or there is no large symmetry-lowering distortion at the DX center.

I. INTRODUCTION

The electrical properties of $Al_xGa_{1-x}As$ doped with the conventional substitutional group-IV (Si,Ge,Sn) or group-VI (S,Se,Te) donors are observed to change abruptly in the compensation region $x > 0.22$.^{1,2} For $x < 0.22$, the donors are characterized by simple shallow effective-mass-like states. For $x > 0.22$, a deeper state emerges into the gap, trapping and removing free carriers, and dominating the electrical properties of the material. This deep level—the so-called DX center—has been a subject of intense interest and controversy during the past decade.

It is now well established² that DX centers in $Al_xGa_{1-x}As$ arise from the isolated substitutional n -type dopants and do not involve a second impurity or defect nearby as originally proposed.³ The deep DX level's optical ionization energy is much larger than its thermal one. The shallow level can be metastably occupied below ~ 100 K by optical excitation, resulting in persistent photoconductivity (PPC). Recapture by the deep level is a thermally activated, multiphonon-emission process; optical capture does not occur. These properties appear best explained in terms of a large lattice relaxation model in which a substantial change in lattice configuration occurs in the transformation between the shallow and deep states.¹⁻⁴ The extent, symmetry, and microscopic nature of the relaxation is still in question, however.

Recent calculations^{5,6} by Chadi and Chang have produced detailed predictions for a large lattice relaxation at the DX . For group-IV donors, which occupy a group-III lattice site, the donor is predicted to break its bond with a nearby As neighbor and relax in the opposite $\langle 111 \rangle$ direction. For group-VI donors, which reside on group-V lattice sites, it is the nearby group-III host atom that breaks away in a $\langle 111 \rangle$ direction from the donor. This prediction has subsequently been duplicated for the Si donors by Dabrowski, Scheffler, and Strehlow.⁷ Also, in an earlier work, Morgan⁸ found that DX centers and their related phenomena could be explained as the displacement of substitutional donors from their normal,

centered lattice positions, and he argued that a trigonal distortion would be the most probable. In contrast, the calculations of Yamaguchi, Shiraishi, and Ohno⁹ found the trigonal distortion to be unstable; rather, the deep state was found to be associated with the simple substitutional donor having T_d symmetry.

An early attempt to probe experimentally the local distortion associated with the DX center was made by Narayanamurti, Logan, and Chin,¹⁰ who studied ballistic phonon absorption in liquid phase epitaxy (LPE) $Al_xGa_{1-x}As$ ($0.3 < x < 0.5$) doped with either Sn or Te. A comparison with phonon absorption in undoped material provided evidence for a $\langle 111 \rangle$ axial distortion in the case of Sn and a $\langle 110 \rangle$ distortion in the case of Te when the donors were in their shallow metastable states. No difference in phonon absorption between doped material cooled to 1.5 K in the dark and undoped material was found, so their experiment provided no information about the symmetry of a large lattice relaxation in the deep DX state.

Kitano and Mizuta^{11,12} investigated the local structure of Se impurities in metal-organic chemical-vapor deposition-grown $Al_{0.38}Ga_{0.62}As$ by extended x-ray-absorption fine structure (EXAFS). No difference between shallow metastable and deep DX states was found in the nearest-neighbor bond length around the Se, suggesting that if a large lattice relaxation exists at the DX , it is not a breathing-mode distortion.

In a similar experiment, Hayes *et al.*¹³ investigated the local structure of Sn impurities in GaAs and in metal-organic vapor-phase epitaxy (MOVPE)-grown $Al_{0.3}Ga_{0.7}As$ by EXAFS. With the deep DX levels populated in the alloy, they found no large difference for the two materials in the Sn-to-nearest-neighbor separation. Here, too, the conclusion was that no large breathing-mode distortion exists at the DX center.

In contrast, Rowe *et al.*¹⁴ performed EXAFS on $Al_xGa_{1-x}As$ ($x = 0.45$ or 0.35) doped with either S or Si, and they found a large breathing-mode contraction associated with S and indications that Si is nonsubstitutional. However, neither the data nor the analysis for Si were included in their paper, which also failed to discuss, for ei-

ther center, which state was being probed—shallow metastable or deep DX —so any conclusions in this work pertaining to the deep DX state must be regarded as inadequately supported.

Gibart *et al.*¹⁵ investigated MOVPE-grown $\text{Al}_x\text{Ga}_{1-x}\text{As}:\text{Sn}$ by Mössbauer spectroscopy and found that for $0.3 < x < 0.4$ a pair of resonances, isomer shifted with respect to the substitutional shallow donor Sn resonance, could be associated with the DX center. Three possible interpretations were proposed, but the simplest requires a large lattice relaxation with a substantial deviation from cubic symmetry at the Sn DX center.

In contrast, Yu *et al.*¹⁶ combined the techniques of particle-induced x-ray emission and ion-beam channeling in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ ($x = 0.41, 0.42$) to deduce that Te and Sn DX centers have no off-center displacement from the substitutional site larger than 0.14 \AA .

Mooney, Theis, and Calleja¹⁷ attributed multiple DX -center emission rates found in Si-doped $\text{Al}_x\text{Ga}_{1-x}\text{As}$ to the influence of different local atomic configurations around the donors in the deep DX state. (Only a single DX emission rate is found in pure GaAs.) Baba *et al.*¹⁸ proposed a similar interpretation for the multiple-emission rates observed in a GaAs- $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layered structure. The signature of these multiple-emission rates are three¹⁷ to four^{18,19} discrete peaks which appear in the deep-level transient spectroscopy (DLTS) spectrum. The number of peaks observed can be neatly explained by the Chadi and Chang distortion scenario. Again, in this picture, the Si donor goes off its group-III site in a $\langle 111 \rangle$ direction into an interstitial site such that its nearest neighbors are a vacancy and three group-III atoms. Then the four DLTS peaks would correspond to replacement of the three Ga atoms by zero, one, two, or three Al atoms. The Chadi-Chang theory is further supported²⁰ by the observation that the lattice-relaxation energies, which vary considerably among group-IV donors, are nearly identical for all group-VI donors, for which the prediction is that not the impurity but one of its group-III neighbors moves into the interstitial site.

Leszczynski, Suski, and Kowalski²¹ studied the linewidth of one of the x-ray diffraction Bragg peaks as a function of DX -level occupancy in LPE $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}:\text{Te}$. The peak broadened significantly when the sample was cooled from room temperature to 77 K in the dark, but it narrowed again when the DX level was depopulated via optical illumination. This was taken as evidence for a distortion in the deep state of the unit cell by $0.1\text{--}0.4 \text{ \AA}$. However, no information on the symmetry of the distortion was presented.

Noge and Ikoma²² attempted to observe in $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}:\text{Si}$ a dependence on polarization angle of capacitance transients induced by polarized light. They calculated the time evolution of the deep DX -level occupancy as a function of polarization with the ionization probability proportional to $|\mathbf{d} \cdot \mathbf{E}|^2$, where \mathbf{d} is the transition dipole moment and \mathbf{E} is the electric field. The dipole \mathbf{d} was taken to be parallel to the defect symmetry axis. Equivalent orientations were summed over for the simple cases of $\langle 100 \rangle$, $\langle 110 \rangle$, or $\langle 111 \rangle$ dipoles, and a large anisotropy was predicted for their experimental geometry,

except in the $\langle 110 \rangle$ case. However, the time-dependent DX -level occupancy determined from their measured capacitance transients did not reveal an anisotropy larger than the experimental uncertainty (about 10%). The results suggested that the transition dipoles are parallel to neither $\langle 111 \rangle$ nor $\langle 100 \rangle$, but the signal-to-noise ratio was insufficient to rule out the $\langle 110 \rangle$ possibility.

These incomplete and often conflicting results from a variety of different probes of the local DX environment, together with the conflicting theoretical predictions for the DX -center large lattice relaxation, indicate that more experiments which address this issue are needed. Our recent report^{23,24} of optical-bleaching transmission transients associated with DX ground- to metastable-state conversion suggests a simple test, which can be viewed as an extension and refinement of the experiment of Noge and Ikoma.²² If the dipole moment for the ionization transition has some special direction, as might be expected in the case of a large off-center distortion, then preferentially bleaching with polarized light a subgroup of configurationally equivalent dipoles might reveal a substantial dichroism in the DX -bleaching transients. If, on the other hand, the transition moment is isotropic, no dichroism should be observed. It is the purpose of this paper to present the results of our attempt to observe such dichroism in the DX -bleaching transients.

The results of this work may be briefly summarized as follows. The expected effect is calculated in terms of a dichroic ratio which can be directly compared with experiment. These calculations were done for transition moments either parallel or perpendicular to the symmetry axes for C_{3v} , D_{2d} , C_{2v} , and C_{1h} defect symmetries. Large deviations are predicted for the dichroic ratio from the value 1, the value expected for isotropic moments. Experimentally, however, the dichroic ratio is found to be 1 within an experimental uncertainty which allows us to place tight limits on the size of the effect. The theoretical predictions fall well outside these limits. In other words, we find no statistically significant evidence for the preferential bleaching by polarized light of a subgroup of configurationally equivalent dipoles, and this result leads us to conclude that the transition moment for the DX optical ionization transition is very nearly isotropic.

This paper is divided into five sections. Section II gives experimental details. Section III presents the experimental results. Section IV describes theory. Discussions and conclusions are presented last in Sec. V.

II. EXPERIMENTAL DETAILS

Our sample was a single crystal of $\text{Al}_{0.35}\text{Ga}_{0.65}\text{As}$ doped with Te, and grown by the LPE method on a GaAs substrate by Hitachi Cable Co., Ltd. It was the same sample studied by us previously in magneto-optical experiments.^{23,24} The Te doping level was specified as $1.2 \times 10^{18} \text{ cm}^{-3}$. The substrate was lapped off prior to our measurements. The sample was in the form of a rectangular parallelepiped with the dimensions $3 \times 1 \times 0.37 \text{ mm}$.³ The long axis of the sample was oriented along the $[1\bar{1}0]$ crystallographic direction, the intermediate axis along $[110]$, and the face was the (001) plane. The sample

was mounted on a quartz rod at the end of a long stainless-steel tube, with the long axis of the sample parallel to the axis of the tube. The tube was then inserted vertically into the bore of an optical-access cryostat with quartz windows. Following each measurement of an optical *DX*-bleaching transient, the tube could be lifted into a warmer part of the cryostat for repopulation of the deep *DX*-state levels in the dark and then returned for further optical studies.

The transmission transients were measured at $1\ \mu\text{m}$ (where the *DX* absorption displays a broad maximum^{23,24}) with the sample immersed in pumped liquid helium at 1.7 K. The light source was a tungsten-halogen lamp, whose luminosity could be changed by adjusting the current through the filament. A Jarrel-Ash Mark X, $\frac{1}{4}$ -meter monochromator (600 lines/mm, $1\text{-}\mu\text{m}$ blaze grating) selected wavelength with a colored-glass filter to eliminate unwanted orders. The beam was passed through a linear polarizer, attached at the output slit of the monochromator to a cylindrical mount. The mount, ruled in degrees, could be rotated about an axis collinear with the beam, so that any desired angle of the polarization direction with respect to the vertical could be accurately and reproducibly obtained. An image of the output slit was focused onto the sample, and the image was larger than the sample to ensure uniform sample excitation. A liquid-nitrogen-cooled Ge detector was used to monitor the transmitted light. A mask smaller than the sample's shadow was placed in front of the detector to ensure that only the light transmitted through the sample was being detected. The light incident on the sample was modulated using a mechanical chopper at 200 Hz and the transmitted signal was synchronously lock-in amplified.

III. EXPERIMENTAL RESULTS

Figure 1 presents a typical example of a bleaching transient in our experiment. The sample is first cooled to 1.7 K in the dark. At $t=0$, polarized light with $1\text{-}\mu\text{m}$ wavelength is admitted to the sample. In the case of Fig. 1, the light is initially polarized with $\mathbf{E} \parallel [110]$. At $t=t_1$, chosen to be the time at which the transmission has increased by 50% of its ultimate total change, the shutter is closed. The polarization is then rotated to $\mathbf{E}' \parallel [1\bar{1}0]$, the shutter is opened at $t=t_2$, and the transient is allowed to proceed until all *DX* centers have been converted to the metastable state. This is verified by closing the shutter, rotating to the original polarization, and observing that the transmission has indeed reached a saturated, steady-state level, as shown in Fig. 1.

The bleached-state transmission levels for the two polarizations are not equal because the light exiting the monochromator and incident on the polarizer is already partly polarized. This effect is corrected for by multiplying the $\mathbf{E}' \parallel [1\bar{1}0]$ part of the transient by a scaling factor determined from the ratio of the steady-state transmission levels for the two polarizations. These corrected data are the trace labeled "scaled" in Fig. 1. The error introduced by this procedure is about the same magnitude as the noise observed on the steady-state part of the transmission.

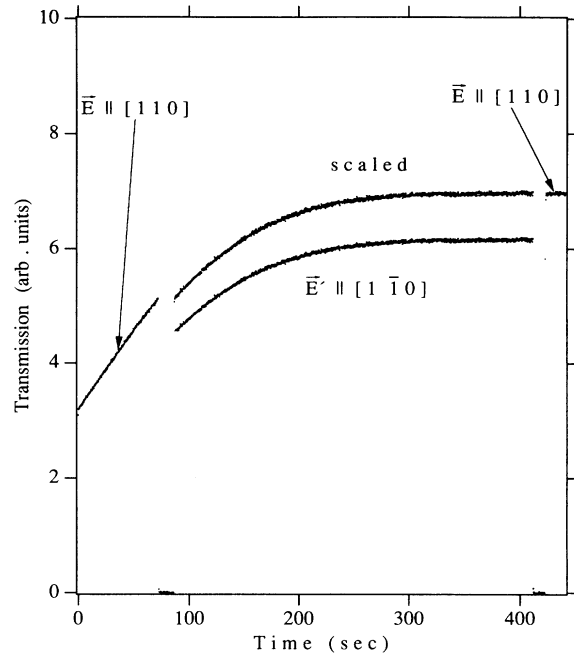


FIG. 1. Typical polarized bleaching experiment. Transmitted intensity vs time is recorded. The sequence of polarizations is $\mathbf{E} \parallel [110] \rightarrow \mathbf{E}' \parallel [1\bar{1}0] \rightarrow \mathbf{E} \parallel [110]$. The lower $\mathbf{E}' \parallel [1\bar{1}0]$ curve is the raw data, and the upper curve has been scaled to correct for polarization by the grating monochromator.

It is not necessary to assume in the foregoing procedure that the free-carrier absorption generated by the ionization of *DX* centers into the PPC state is isotropic. Free-carrier absorption depends on the effective mass and the relaxation time and therefore may depend in principle on the direction in which the free carriers are accelerated by the photon field. However, the polarization is always rotated between directions of equivalent symmetry—either $\mathbf{E} \parallel [110] \rightarrow \mathbf{E}' \parallel [1\bar{1}0]$ or $\mathbf{E} \parallel [100] \rightarrow \mathbf{E}' \parallel [010]$ —so we may neglect this consideration.

As shown in Refs. 23 and 24, the time-dependent part of the absorption coefficient at any time t may be determined according to

$$\alpha(t) - \alpha_m = -\frac{1}{d} \ln \left[\frac{I(t)}{I(\infty)} \right], \quad (1)$$

where $I(t)$ is the transmitted intensity at time t , $I(\infty)$ is the steady-state level, and d is the sample thickness. At $t=0$, Eq. (1) takes the value $\alpha_g - \alpha_m$, where α_g is the absorption coefficient due to *DX* centers all in the ground state and α_m is the absorption after they have all been converted to the metastable (free-carrier) state.

We define a dichroic ratio D as the ratio of Eq. (1) at the times t_1 and t_2 according to

$$D \equiv \frac{\alpha(t_1) - \alpha_m}{\alpha(t_2) - \alpha_m}. \quad (2)$$

The data in Fig. 1 give $D = 0.997 \pm 0.020$, where the statistical uncertainty is determined from the noise. Repeated measurements and a weighted average give

$D = 1.0042 \pm 0.0064$. A similar procedure for the polarization sequence $\mathbf{E} \parallel [100] \rightarrow \mathbf{E}' \parallel [010]$ gives $D = 1.0016 \pm 0.0079$.

IV. THEORY

We assume that all DX centers are initially in their deep ground-state levels at low temperature in the dark. Each center i will have an optical transition dipole moment \mathbf{d}_i , leading to an optical absorption cross section $\sigma_{gi} = \sigma_{g0} |\mathbf{d}_i \cdot \mathbf{E}|^2$. We take \mathbf{d} and \mathbf{E} normalized throughout, so that σ_{g0} is the maximum cross section. Then we divide the N centers evenly into the w groups of defect orientations equivalent by symmetry (three for D_{2d} , four for C_{3v} , etc.), in each of which the σ_{gi} are the same, giving

$$\alpha(t) - \alpha_m = \sum_{j=1}^w N_j(t) (\sigma_{gj} - \sigma_m). \quad (3)$$

In Eq. (3), $N_j(t)$ is the number of DX centers in group j remaining in the ground state at time t , and σ_m is the metastable state absorption cross section. Since the metastable state corresponds to free carriers in the conduction band, σ_m is independent of j . The $N_j(t)$ will decay exponentially according to

$$N_j(t) = \frac{N}{w} \exp(-\phi \eta \sigma_{gj} t). \quad (4)$$

In Eq. (4), ϕ is the photon flux and η a ground- to metastable-state conversion efficiency. Making the substitutions $f_j = |\mathbf{d}_j \cdot \mathbf{E}|^2$, $\sigma_{gj} = f_j \sigma_{g0}$, $\tau = \phi \eta \sigma_{g0} t$, and $q = \sigma_{g0} / \sigma_m$, we have

$$D = \frac{\sum_{j=1}^w (qf_j - 1) \exp(-f_j \tau)}{\sum_{j=1}^w (qf'_j - 1) \exp(-f'_j \tau)}. \quad (5)$$

Here $f'_j = |\mathbf{d}_j \cdot \mathbf{E}'|^2$ where \mathbf{E}' is the polarization beginning

at $t = t_2$. We have assumed that there is no conversion or reorientation while the shutter is closed and the polarization rotated, so that $N_j(t_2) = N_j(t_1)$.

The quantity τ in Eq. (5) must be determined from the conditions for each experiment. It is possible to obtain it from the $0 \leq t \leq t_1$ part of the transient without having to know ϕ , η , or σ_{g0} according to

$$G \equiv \frac{\alpha(t_1) - \alpha_m}{\alpha(0) - \alpha_m} = \frac{\sum_{j=1}^w (qf_j - 1) \exp(-f_j \tau)}{\sum_{j=1}^w (qf_j - 1)}. \quad (6)$$

In our experiments, we have selected t_1 such that the transmission has increased by 50% of its ultimate total change. For the transmission at $1 \mu\text{m}$ of our 0.37-mm sample ($\alpha_g d \sim 0.9$), this corresponds to $G = 0.40 \pm 0.01$, and we use this value in solving for τ from Eq. (6).

We must also know q , the ratio of the maximum ground-state- and metastable-state-absorption cross sections. This may be determined by

$$q \equiv \frac{\sigma_{g0}}{\sigma_m} = \left[\frac{1}{w} \sum_{j=1}^w f_j \right]^{-1} \frac{\alpha_g}{\alpha_m}. \quad (7)$$

The ratio of absorption coefficients may be obtained from Refs. 23 and 24. At $1 \mu\text{m}$ it has the value of approximately 10. The factor involving the sum takes the values 3 or $\frac{3}{2}$ for the symmetries considered in this paper, so we will use values for q of either 30 or 15, respectively.

Table I gives expressions for G and D in terms of q and τ for the various symmetries and polarization sequences considered. Transition moments parallel or perpendicular to the defect symmetry axis are considered separately, since if they contribute simultaneously to the absorption there will be no effect ($D = 1$). The polarization sequence $\mathbf{E} \rightarrow \mathbf{E}'$ and the squared projections f_j and f'_j are also given.

Table II gives calculated values of q , τ and D for each

TABLE I. Expressions for G and D in terms of q and τ for the various defect symmetry groups and polarization sequences.

Group	\mathbf{d}	\mathbf{E}	f_j	\mathbf{E}'	f'_j	G	D
C_{3v}	\parallel	[110]	$0, \frac{2}{3}$	$[1\bar{1}0]$	$\frac{2}{3}, 0$	$\frac{(2q-3)e^{-2\tau/3} - 3}{2q-6}$	$\frac{(2q-3)e^{-2\tau/3} - 3}{(2q-3) - 3e^{-2\tau/3}}$
C_{3v}	\perp	[110]	$\frac{1}{3}, 1$	$[1\bar{1}0]$	$1, \frac{1}{3}$	$\frac{(q-3)e^{-\tau/3} + 3(q-1)e^{-\tau}}{4q-6}$	$\frac{(q-3)e^{-\tau/3} + 3(q-1)e^{-\tau}}{3(q-1)e^{-\tau/3} + (q-3)e^{-\tau}}$
D_{2d}, C_{2v}	\parallel	[100]	1, 0, 0	[010]	0, 1, 0	$\frac{(q-1)e^{-\tau} - 2}{q-3}$	$\frac{(q-1)e^{-\tau} - 2}{(q-2) - e^{-\tau}}$
D_{2d}	\perp	[100]	1, 0, 1	[010]	1, 1, 0	$\frac{2(q-1)e^{-\tau} - 1}{2q-3}$	$\frac{2(q-1)e^{-\tau} - 1}{(q-2)e^{-\tau} + q - 1}$
C_{2v} C_{1h}	$\left. \begin{array}{l} \perp \\ \parallel \end{array} \right\}$	[100]	$\frac{1}{2}, 0, \frac{1}{2}$	[010]	$\frac{1}{2}, \frac{1}{2}, 0$	$\frac{(q-2)e^{-\tau/2} - 1}{q-3}$	$\frac{2(q-2)e^{-\tau/2} - 2}{(q-4)e^{-\tau/2} + q - 2}$
C_{2v} C_{1h}	$\left. \begin{array}{l} \perp \\ \parallel \end{array} \right\}$	[110]	$0, 1, \frac{1}{4}, \frac{1}{4}, \frac{1}{4}, \frac{1}{4}$	$[1\bar{1}0]$	$1, 0, \frac{1}{4}, \frac{1}{4}, \frac{1}{4}, \frac{1}{4}$	$\frac{(q-1)e^{-\tau} + (q-4)e^{-\tau/4} - 1}{2(q-3)}$	$\frac{(q-1)e^{-\tau} + (q-4)e^{-\tau/4} - 1}{q-1 - e^{-\tau} + (q-4)e^{-\tau/4}}$

TABLE II. Predicted values of q , τ and D for each defect symmetry and polarization sequence.

Group	\mathbf{d}	E sequence	$q \sim$	τ	D
C_{3v}	\parallel	$[110] \rightarrow [1\bar{1}0]$	30	1.26	0.39
C_{3v}	\perp	$[110] \rightarrow [1\bar{1}0]$	15	1.14	0.66
D_{2d}, C_{2v}	\parallel	$[100] \rightarrow [010]$	30	0.82	0.39
D_{2d}	\perp	$[100] \rightarrow [010]$	15	0.86	0.56
C_{2v} C_{1h}	\perp \parallel }	$[100] \rightarrow [010]$	30	1.73	0.55
C_{2v} C_{1h}	\perp \parallel }	$[110] \rightarrow [1\bar{1}0]$	30	1.67	0.47

symmetry and polarization sequence. Note that in each case, D is significantly smaller than 1 and well outside the experimental limits determined from the statistical uncertainty.

The case of dipoles perpendicular to a C_{1h} symmetry axis requires special attention. A representative normalized transition moment in a (110) symmetry plane may be expressed as $a(0,0,1) + b(-1,1,0)/\sqrt{2}$, where a and b are constants subject to the normalization condition $a^2 + b^2 = 1$. When light is incident in the $[001]$ direction

of the crystal and is polarized parallel to $[100]$, we have three values for the f_j : $b^2/2$, $b^2/2$ and $1 - b^2$, corresponding to the three pairs of $\{110\}$ defect symmetry planes for which $|\mathbf{d}_j \cdot \mathbf{E}|^2$ differ. Rotating the polarization to $[010]$ transforms these to $b^2/2$, $1 - b^2$, and $b^2/2$, respectively. When $b^2 = \frac{2}{3}$, the dipoles lie along $\langle 111 \rangle$ directions, all the f_j are equal, and consequently no dichroism is expected ($D = 1$). We find for the dichroic ratio in the general case

$$D = \frac{(qb^2 - 2)\exp(-b^2\tau/2) + (q - 1 - qb^2)\exp[-(1 - b^2)\tau]}{(q - 2 - \frac{1}{2}qb^2)\exp(-b^2\tau/2) + (\frac{1}{2}qb^2 - 1)\exp[-(1 - b^2)\tau]} \quad (8)$$

The quantity τ is determined from

$$G = \frac{(qb^2 - 2)\exp(-b^2\tau/2) + (q - 1 - qb^2)\exp[-(1 - b^2)\tau]}{q - 3} \quad (9)$$

Equations (8) and (9) show that both τ and D depend on the orientation of the dipole in the $\{110\}$ planes. Only the square of b appears, so we need only consider values in the range $0 \leq b \leq 1$. Solutions for τ vs b are obtained graphically from the intersection of Eq. (9) and $G = 0.40$. The appropriate value for q is $3(\alpha_g/\alpha_m) \sim 30$.

Figure 2 gives a plot of Eq. (8) for D vs b . For $b = 0$ ($\langle 100 \rangle$ dipoles), we expect to find the maximum effect ($D = 0.39$). Except when b^2 is near $\frac{2}{3}$ ($\langle 111 \rangle$ dipoles), the theoretical expectations fall well outside the experimental limits, shown as the dashed lines in Fig. 2.

We next consider the $[110] \rightarrow [1\bar{1}0]$ polarization sequence. Here we have $f_j = 0, b^2, f_+, f_+, f_-,$ and f_- , and $f'_j = b^2, 0, f_-, f_-, f_+,$ and f_+ , where $f_{\pm} = (a/\sqrt{2} \pm b/2)^2$. The dichroic ratio is

$$D = \frac{-1 + (qb^2 - 1)\exp(-b^2\tau) + 2(qf_+ - 1)\exp(-f_+\tau) + 2(qf_- - 1)\exp(-f_-\tau)}{qb^2 - 1 - \exp(-b^2\tau) + 2(qf_- - 1)\exp(-f_+\tau) + 2(qf_+ - 1)\exp(-f_-\tau)} \quad (10)$$

We determine τ from the intersection of $G = 0.40$ and

$$G = \frac{-1 + (qb^2 - 1)\exp(-b^2\tau) + 2(qf_+ - 1)\exp(-f_+\tau) + 2(qf_- - 1)\exp(-f_-\tau)}{2q - 6} \quad (11)$$

Equations (10) and (11) are symmetrical under the interchange of the subscripts $+$ and $-$. Thus D does not depend on the relative sign of a and b , and we can make the substitution $a = +\sqrt{1 - b^2}$ and limit b to the range

$0 \leq b \leq 1$. In other words, all possible values of D are realized in the $a \geq 0, b \geq 0$ quadrant. The appropriate value of q is again 30.

Figure 3 gives D vs b for the $[110] \rightarrow [1\bar{1}0]$ polariza-

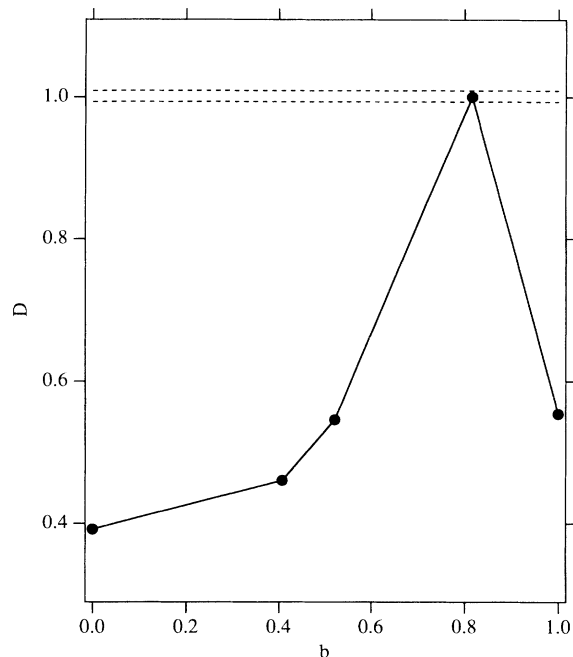


FIG. 2. D vs b for dipoles in the reflection plane of a defect with C_{1h} symmetry, with $G=0.40$. The polarization sequence is $\mathbf{E} \parallel [100] \rightarrow \mathbf{E}' \parallel [010]$. The solid circles are theoretical values. The dashed lines give the range of experimental values.

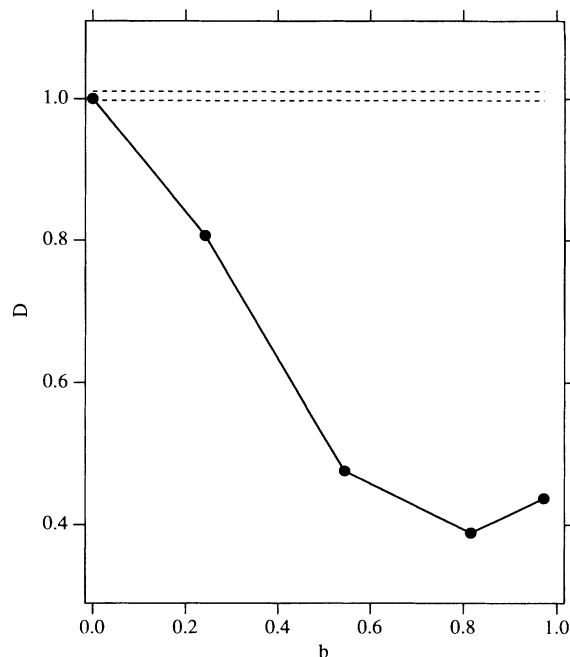


FIG. 3. D vs b for dipoles in the reflection plane of a defect with C_{1h} symmetry, with $G=0.40$. The polarization sequence is $\mathbf{E} \parallel [110] \rightarrow \mathbf{E}' \parallel [1\bar{1}0]$. The solid circles are theoretical values. The dashed lines give the range of experimental values.

tion sequence. The solid circles are values calculated from Eq. (10). The theoretical expectations fall well outside the experimental limits (dashed lines) except when b has a value near 0. However, Figs. 2 and 3 together show that no value of b gives a dichroic ratio in agreement with all the experiments.

V. SUMMARY AND CONCLUSIONS

We have calculated the expected effect on the DX optical transmission transients of preferentially bleaching subgroups of DX centers with polarized light. The effect was quantified in terms of a dichroic ratio D , which could be directly compared to the results of experiment. The calculations were done for transition moments parallel or perpendicular to the defect symmetry axis in the case of C_{3v} , D_{2d} , C_{2v} , and C_{1h} symmetries. Large deviations of D from 1 were expected on these theoretical grounds.

The results of experiment, however, were that $D = 1$ to

high precision. Experimental standard deviations allowed us to place tight limits on the allowed values of D . The theoretical expectations fell well outside these limits in all cases. Hence we conclude that the dipole moment for the DX optical ionization transition is very nearly isotropic.

One of two conclusions may be drawn from these results. If a large symmetry-lowering distortion exists at the DX center, both parallel and perpendicular transition moments must accidentally be contributing simultaneously to the DX -center optical ionization band at $1 \mu\text{m}$. Alternatively, the DX center undergoes no large symmetry-lowering distortion.

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