Stress splitting of the $EL2$ zero-phonon line: Need for reinterpretation of the main optical transition

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(Received 18 December 1989)

The T_2 final state of the main optical transition of $EL2$ can couple to lattice distortions of a, ϵ, and τ symmetry. The stress splitting of the zero-phonon line (ZPL) observed by Kaminska *et al.* gives information about the strength of the coupling between the lattice and the T_2 final state. We have obtained that information and have shown that the lattice relaxation associated with the τ mode used by Kaminska *et al.* to account for their results cannot be larger than about 70 meV, and is probably much closer to 45 meV. We find that the lattice relaxation associated with the ϵ modes is negligible, and we estimate, from analysis of hydrostatic-pressure experiments, that the lattice relaxation associated with the a modes is about 90 meV. Thus, the separation of 140 meV between the peak in the optical absorption at 1.18 eV and the ZPL at 1.04 eV (a separation commonly taken to be due to lattice relaxation) can be accounted for. If this agreement is interpreted as strengthening the case that the ZPL and the peak in the optical absorption are related to each other by lattice relaxation, it exacerbates well-known problems in understanding the optical absorption. Among these problems are the intensity of the ZPL, the fact that it moves in a direction opposite to that of the main peak under hydrostatic pressure, and the fact that it rides up over the beginning of the main peak when hydrostatic pressure is applied. Our calculation does make it clear that there must be at least *two* local modes, one of a symmetry and the other of τ symmetry, in addition to any of the perfect-crystal phonon modes usually considered. Although this potentially adds a new feature to the analysis, it does not by itself resolve the problems with the absorption. We discuss the role that ionization and recapture may play in resolving these problems and suggest an experiment that might clarify this issue.

I. BACKGROUND

$EL2$ is a native midgap donor defect in GaAs which is of interest in large part because of its optically induced metastability. It is known that As_{Ga}, the arsenic antisite defect, is a main constituent of $EL2$.[1] There is active controversy over whether $EL2$ is the isolated As_{Ga} or whether, in addition, $EL2$ contains another defect complexed with the As_{Ga}.[2] The present work, however, will deal only with aspects of the optical absorption resulting from the $A_1 \rightarrow T_2$ transition at the As_{Ga} and not at all with the question of an additional defect being present in $EL2$. In the following, we shall describe some calculations which were prompted by the need to explore certain inconsistencies in the interpretation of the optical transition, particularly, aspects of it which arise because of coupling between the electronic state of the defect and the relaxation of the lattice in which it is embedded.

Let us review the most prominent features of the optical absorption. The main optical absorption of $EL2$ has been identified as being an A_1 to T_2 transition at the neutral As_{Ga}. Experimentally, it is characterized by three features:[3] (i) A smoothly rising photoionization cross section starting at about 0.77 eV. This is ascribed to the transfer of an electron from a midgap A_1 level of the As_{Ga} to the conduction bands. (ii) Superimposed on this is a broad peak of half-width about 0.15 eV centered on $h\nu = 1.18$ eV. This feature is strongly associated with the transition to the metastable state and has, in most analyses, been regarded as an intracenter A_1 to T_2 optical transition. Its broadening is ascribed to lattice relaxation, with 1.18 eV being the energy of the "vertical transition" in which the configuration of the lattice is the same in the initial and final electronic states. (iii) Below this, at 1.04 eV, is a zero-phonon line (ZPL) with multiphonon transitions spaced above it at intervals of 11 meV.

If the structure at 1.18 eV is the vertical transition, the Franck-Condon shift d_{FC} is 1.18–1.04 eV = 140 meV. In addition, *if* the 11-meV phonon is the lattice relaxation phonon, the Huang-Rhys factor $S^{HR} = d_{FC}/\hbar\omega$ is of order 13. In such a case, a ZPL of relative intensity $e^{-13} \approx 10^{-6}$ would be unobservable. This has prompted the suggestion that the lattice relaxation phonon is really 24 or 33 meV, approximately two or exactly three of the observed 11-meV phonons.[4] Both of these phonons are allowed in GaAs since its phonon spectrum extends up to about 35 meV.[5] Either would be close to the 29-meV value[6] needed to account for the 0.15-eV half-width of the optical absorption, at least in the simplest model.[7] However, recent measurements of the optical absorption under hydrostatic pressure have shown that the 1.18-eV feature shifts to lower energy while the 1.04-eV feature shifts to higher energy when pressure is applied.[8] Moreover, and perhaps more difficult to explain away, under hydrostatic pressure, the ZPL is observed to ride up over the onset of the broad peak.[8] This raises the possibility

that the two features are not simply the vertical transition and the associated zero-phonon line, even though there is ample evidence that they are both part of the same $A_1 \rightarrow T_2$ transition at the As_{Ga} defect. The proper interpretation of both features is left open by this possibility.

Baj and Dreszer[8] have, in fact, suggested that the two features might be optical transitions to two different final states of the *EL2* defect. In the present calculation, the fact that we find the lattice relaxation to be very close to the observed 140 meV weakens the case for Baj and Dreszer's proposal. This is unfortunate, because from the standpoint of intellectual simplicity, their suggestion is the cleanest way out of the difficulties. There is still the possibility that, due to the inherent uncertainties in our method of analysis, we have overestimated the lattice relaxation and that the Baj and Dreszer proposal will indeed, as we hope, turn out to be correct. For this reason, we discuss the implications of their proposal on our understanding of the optical spectrum and we propose an experiment whereby it might be tested.

Part of the background for the work we shall describe is to be found in an earlier paper[9] which was concerned with the stress splitting of the $A_1 \rightarrow T_2$ transition at the As_{Ga} in the presence of another defect. In carrying out those calculations, we rederived the formulas used successfully by Kaminska *et al.*[2(a)] to describe the results of their stress-splitting experiments. The underlying assumption was that the final T_2 state is coupled to a τ-mode lattice distortion which could represent, e.g., the displacement of the central As atom from its lattice site. We noted that one of the parameters used by Kaminska *et al.* in their fitting, namely, the tunnel splitting parameter Δ, was really a function only of S^{HR} and $\hbar\omega$, and we noted that the value of Δ demanded by the fitting required that S^{HR} be no greater than 2. Since Δ is an exponentially decreasing[10] function of S^{HR}, use of the value $S^{HR}=13$ obtained from $d_{FC}/\hbar\omega$ would result in a value of Δ far too small to account for the results of Kaminska *et al.* However, the value $S^{HR}=2$ leads to the following serious problem: Since the maximum phonon energy in the system cannot exceed 35 meV, d_{FC} cannot exceed 70 meV, whereas in the conventional interpretation, d_{FC} is 140 meV. Rather than pursue this contradiction, we had suggested that it might be overcome if one took into account the displacement of the surrounding atoms as the As_{Ga} moves. The lattice distortion associated with the four nearest neighbors gives rise to an a mode, an ϵ mode, and a τ mode, all of which can couple to the T_2 electronic state. The large fitted value of Δ might not demand such a small and restrictive Huang-Rhys factor, because both the a and ϵ modes would reduce the relative size of the tunneling barrier [see Eq. (9.3), Ref. 9] whose existence gives rise to the exponentially small Δ. Another way of saying this is that Δ could be as small as the Jahn-Teller calculation limits it to be while the additional lattice relaxation needed to make up the full 140 meV might be contributed by the other modes.

In the present paper we explore the suggestion that coupling to the next shell of neighbors might supply the extra relaxation needed and show that indeed it does. We have evaluated the couplings to the two other lattice distortions (that of a symmetry and that of ϵ symmetry), and we find that they are close to 90 meV in total, so that if the τ-mode relaxation is at its maximum value, the total relaxation is about 160 meV. However, a more detailed look at the τ-mode contribution suggests that 70 meV is an upper bound to a relaxation which is probably closer to 45 meV. This brings the total estimate of lattice relaxation close to the observed 140 meV. As we have already noted, this is somewhat unfortunate, in that by strengthening the case that the feature at 1.04 eV and the peak in the absorption are indeed the ZPL and its associated vertical transition, it exacerbates the other well-known problems associated with the optical absorption, problems which could be resolved if, as Baj and Dreszer had suggested, these two features really arose from different final states.

For estimating the contribution of the ϵ modes, we proceed as follows: It is known that for a given value of S^{HR}, the largest value of Δ is to be found when the coupling to the τ and ϵ modes are of equal strength.[11] We calculate the stress splitting of the final state in this situation and find that there is no choice of parameters that can make the calculated result simulate the observed stress splitting. We conclude that for the calculated stress splitting to resemble the observed stress splitting, the coupling to the ϵ mode must be much less than that to the τ mode.

We then make a model which allows us to estimate the a-mode lattice relaxation. A model is made necessary because the a-mode coupling does not affect the stress splittings, and so there is neither any restriction on its strength nor any way to learn about it from the stress-splitting experiments. Input to this model is obtained from an analysis of hydrostatic pressure measurements. The model gives an a-mode relaxation of about 90 meV.

It is clear, from the fact that the total lattice relaxation can be accounted for only by coupling to both a and τ modes, that there must be at least two local phonons involved in the relaxation process, one of a symmetry and one of τ symmetry. (By local phonon is meant either a discrete lattice mode or a resonance whose vibrational amplitude is greatest in the immediate neighborhood of the defect. There is no need to distinguish here between the two possibilities, although in the strictest sense, it is only a resonance that can transport vibrational energy away from the neighborhood of the defect and lead thereby to lattice relaxation.) The possibility of more than one independent local phonon being involved in the lattice relaxation has not been seriously considered before, although the phonons above the ZPL have been described in terms of a single localized phonon plus such other phonons as the perfect-crystal supports. The implications of the presence of two or more local modes of distortion is a problem for future study. In the meantime, we should entertain the proposal that, in spite of the calculated 135 meV of lattice relaxation, the Baj and Dreszer suggestion is correct, and that the peak in the optical absorption arises from some final state other than the state that gives rise to the ZPL. One possibility is that it is a feature of

the ionization from the A_1 level to the bands. This ionization would be followed by subsequent recapture into the T_2 state of the ionized As_{Ga} if there is no electric field present. In this way, the peak in the ionization cross section would eventually reflect itself as a peak in the rate of capture into the T_2 level, and from there, as a peak in $\sigma^*(h\nu)$, the optical cross section for transfer of $EL2$ to the metastable state. This last possibility would go far towards resolving some of the puzzles connected with the optical absorption of $EL2$.

II. CALCULATION ASSUMING EQUAL ϵ- AND τ-MODE COUPLING

It has been argued that a calculation is not necessary to verify that the coupling to the ϵ lattice mode must be considerably weaker than that to the τ mode. In its simplest form, the argument is as follows: The coupling parameters, which are fitted in order to describe the stress-splitting experiments, are products of electronic stress-coupling parameters and Ham reduction factors.[12,13] The sizes of the Ham factors are in turn determined by the strengths of the couplings between the T_2 state and the various modes of the lattice. The small size of the fitted ϵ-mode stress-coupling parameter[2(a),2(b),9] comes about because the ϵ-mode Ham reduction factor is much smaller than the τ-mode Ham factor. This must mean that the Jahn-Teller coupling to the ϵ-mode lattice distortion is much weaker than that to the τ-mode distortion. However, making such an argument ignores the possibility that it is the ϵ-mode electronic stress-coupling parameter that is small while the ϵ- and τ-mode Ham factors are similar in size. In the Appendix, we show that a simple estimate of the electronic coupling to the lattice does give a small value of ϵ electronic stress-coupling parameter and so one has no *a priori* information about the relative sizes of the ϵ- and τ-mode Ham factors. It is for this reason that the calculation described below is necessary to the conclusion we have reached.

The background for this section of the paper is to be found in a series of papers by O'Brien where the problem of a T_1 or a T_2 electronic state equally coupled to a τ and an ϵ lattice mode is discussed.[11,14,15] We draw only on results of the first paper in the series.[11] In that paper, it is shown that the eigenstates of the system are of the form

$$\Psi_l^m(Q,q)=\psi_l^m(\theta,\phi)(\sin\theta\cos\phi|x\rangle+\sin\theta\sin\phi|y\rangle+\cos\theta|z\rangle) . \quad (1)$$

Here $|x\rangle$, $|y\rangle$, and $|z\rangle$ are the electronic wave functions of the T_2 final state, θ and ϕ are angles which O'Brien introduced to parametrize the lattice distortion Q, and ψ_l^m is a spherical harmonic. Only odd values of l are allowed. Thus the ground state ($l=1$) is triply degenerate and, for the case of interest here, has T_2 symmetry. The first excited state ($l=3$) is sevenfold degenerate and, for the case considered here, contains an A_1 state, a T_1 state, and a T_2 state. It is within this ten-state manifold (composed of $l=1$ and 3 spherical harmonics) that we must calculate the stress splitting of the lowest three states.

The calculation proceeds in exact analogy to the one we presented in Ref. 9 and so we need only to mention the differences between the present calculation and the earlier one. Here, the basis states are taken in the following order: $A_1, T_{1x}, T_{1y}, T_{1z}$ and $T_{2x}, T_{2y} T_{2z}$, these referring to the excited state, and then T_{2x}, T_{2y}, T_{2z}, these last referring to the ground state. It is convenient to label the second repetition of the T_2 representation by the symbol T_3. The unperturbed Hamiltonian is $H_0=H_i\delta_{ij}$ where $H_i=\Delta$ for $i=1,2,\ldots,7$ and $H_i=0$ for $i=8,9,10$.

The next part of the calculation is setting up a 10×10 stress matrix for each of the three stress directions. The *form* of each matrix is known in terms of the group coupling coefficients,[9,12,16] but the evaluation of each matrix requires knowledge of the Ham reduction factors. Evaluating them is the only difficult part of the calculation. We denote the Ham factors by the symbol $K_{\alpha\beta}^{\gamma}$ where γ refers to the symmetry of the operator and where $\alpha\beta$ refers to the symmetry of the representations involved. Let us define these factors precisely.

Using the notation $\Omega\equiv\theta,\phi$, we rewrite Eq. (1) as

$$\Psi_l^m(Q,q)=\psi_l^m(\Omega)\sum_\nu f_\nu(\Omega)|\nu\rangle , \quad (2)$$

where $\nu\rangle=|x\rangle,|y\rangle$, or $|z\rangle$, and where the $f_\nu(\Omega)$ are defined by comparing Eqs. (1) and (2). [The normalization of Eqs. (1) and (2) is such that integrating over Q is carried out by integrating Ω over the full unit sphere even though in doing so, the lattice distortion Q is carried twice over its allowed range of values.[11] This has no effect on what follows.] The wave functions (1) and (2) transform like the spherical harmonics because the other factor in Eq. (2) transforms according to the A_1 representation. What is needed, however, are states which transform like irreducible basis functions for the group T_d, the group under which the Hamiltonian of the isolated As_{Ga} is invariant. A unitary transformation $T_\mu^\Gamma(l,m)$ can be found to produce them:

$$\Psi_\mu^\Gamma(Q,q)=\sum_{l,m} T_\mu^\Gamma(l,m)\Psi_l^m(Q,q) . \quad (3)$$

Γ takes on the labels A_1, T_1, T_2, and T_3. The convention we are using, that T_3 arises only from the $l=1$ manifold while A_1, T_1, and T_2 arise out of the $l=3$ manifold, is easily incorporated into the choice of unitary matrix T.

Now consider a purely electronic operator $U_\lambda^\gamma(q)$ which, under the group T_d, transforms as the λ partner function for the irreducible representation γ. It has nine matrix elements within the three electronic states $|\nu\rangle$. As is well known, there are linear relations among these elements that arise because the states $|\nu\rangle$ are themselves partner functions for the T_2 irreducible representation of T_d. As a consequence these nine elements can be written under the form

$$\langle\nu|U_\lambda^\gamma|\nu'\rangle=V^\gamma\langle T_2,\nu|\theta_\lambda^\gamma|T_2,\nu'\rangle , \quad (4)$$

where $\langle T_2,\nu|\theta_\lambda^\gamma|T_2,\nu'\rangle$ are the group coupling coefficients tabulated by Koster *et al.*[16] They are completely independent of the operator $U_\lambda^\gamma(q)$. The only number that depends on the operator U_λ^γ specifically is

V^γ, which is independent of λ, ν, and ν'. Thus, given the θ^γ_λ matrices, all the many elements described by Eq. (4) are trivially obtained once one of them has been calculated.

In this particular example, the states $|\nu\rangle$ and $|\nu'\rangle$ were drawn from the same T_2 manifold. In general, the states $|\nu\rangle$ might have been partner function for any irreducible representation, say Γ, while the states $|\nu'\rangle$ might have been partners for any other irreducible representation, say Γ'. The same sort of linear relations among the matrix elements will arise. For the case of interest here (γ taking the labels A_1, E, and T_2 while Γ and Γ' take the labels A_1, T_1, and T_2), the relationship analogous to (4) is

$$\langle \Gamma\mu|U^\gamma_\lambda|\Gamma'\mu'\rangle = V^\gamma(\Gamma,\Gamma')\langle \Gamma\mu|\theta^\gamma_\lambda|\Gamma'\mu'\rangle \ . \tag{5}$$

Again the group coupling coefficients appear on the right. The only numbers which depend on the specific operator U^γ_λ are $V^\gamma(\Gamma,\Gamma')$. As before, these are independent of λ, μ, and μ'. Given the θ^γ_λ matrices as tabulated by Koster *et al.*, all the matrix elements for a given γ, Γ, and Γ' are again known once one of them is calculated.

Consider now the special case where the states appearing in (5) are the vibronic states defined in (3). The matrix element in (5) can be evaluated explicitly using (2) and (4) as

$$\langle \Gamma_\mu|U^\gamma_\lambda|\Gamma'\mu'\rangle = V^\gamma \sum_{l,m,\nu}\sum_{l',m',\nu'} [T^\Gamma_\mu(l,m)]^* T^{\Gamma'}_{\mu'}(l',m')\langle T_2,\nu|\theta^\gamma_\lambda|T_2,\nu'\rangle \int d\Omega\,[\psi^m_\lambda(\Omega)]^* f_{\nu'}(\Omega)\psi^{m'}_{l'}(\Omega) \ . \tag{6}$$

Comparing (5) with (6), it is clear that the quantity

$$K^\gamma_{\alpha,\beta} \equiv V^\gamma(\alpha,\beta)/V^\gamma \tag{7}$$

can be evaluated by equating (5) and (6), taking care only to choose a set of values of λ, μ, and μ' for which $\langle \alpha\mu|\theta^\gamma_\lambda|\beta\mu'\rangle$ does not vanish. The evaluation will involve only quantities which are known and independent of the specific operator $U^\gamma_\lambda(q)$. That such a development would be possible in general was first noted by Ham.[13]

The task of evaluating the unitary matrix and carrying out the needed integrations for all the cases we shall need ($\gamma = A_1, E$, and T_2; Γ and $\Gamma' = A_1$, T_1, T_2, and T_3) is straightforward but tedious. It probably could have been simplified by using some of the deeper symmetry properties inherent in the relationship between Q and Ω, properties that O'Brien specifically built into the parametrization of the lattice distortion.[11] However, we evaluated the Ham factors by carrying out the integrations for all needed combinations of γ, Γ, and Γ', checking at the same time to verify that the results were independent of λ, μ, and μ'. The results we obtained are as follows: $K^{A_1}_{\alpha\beta} = \delta_{\alpha\beta}$. For $\gamma = E$,

$$K^E_{\alpha\beta} = \begin{array}{c} \\ T_1 \\ T_2 \\ T_3 \end{array} \begin{array}{c} \begin{array}{ccc} T_1 & T_2 & T_3 \end{array} \\ \begin{pmatrix} 0 & -2\sqrt{5}/15 & \sqrt{\frac{3}{35}} \\ -2\sqrt{5}/15 & 4/15 & \frac{3}{5}\sqrt{\frac{3}{7}} \\ \sqrt{\frac{3}{35}} & \frac{3}{5}\sqrt{\frac{3}{7}} & 2/5 \end{pmatrix} \end{array} \tag{8a}$$

and for $\gamma = T_2$,

$$K^\gamma_{\alpha\beta} = \begin{array}{c} \\ A_1 \\ T_1 \\ T_2 \\ T_3 \end{array} \begin{array}{c} \begin{array}{cccc} A_1 & T_1 & T_2 & T_3 \end{array} \\ \begin{pmatrix} 0 & 0 & -\frac{2}{3}\sqrt{\frac{2}{5}} & \sqrt{\frac{6}{35}} \\ 0 & -\frac{1}{3} & -\sqrt{\frac{1}{135}} & -2/\sqrt{35} \\ -\frac{2}{3}\sqrt{\frac{2}{5}} & -\sqrt{\frac{1}{135}} & -\frac{1}{15} & -\frac{2}{5}\sqrt{\frac{3}{7}} \\ \sqrt{\frac{6}{35}} & -2/\sqrt{35} & -\frac{2}{5}\sqrt{\frac{3}{7}} & \frac{2}{5} \end{pmatrix} \end{array} \ . \tag{8b}$$

III. RESULTS FOR EQUAL ϵ- AND τ-MODE COUPLING

With these values of $K^\gamma_{\alpha\beta}$, and using Eqs. (5) and (7), one can evaluate the stress matrices in terms of S^{A_1}, S^E, and S^{T_2}. These are three parameters which, when multiplied by σ, the magnitude of the stress, describe the stress coupling in the electronic T_2 manifold in the same way as does V^γ in Eq. (4). These parameters and Δ are the same four parameters as were needed in the earlier analysis (coupling to the τ mode alone) but their values in fitting this new situation may be different from those obtained for the earlier one.

The 10×10 Hamiltonian matrix is diagonalized numerically and the energy of the lowest three states is studied as a function of the magnitude of the uniaxial stress for each of the three stress directions indicated. Figures 1 and 2 give the results of the calculations for two choices of the four important parameters. These two figures are sufficient to illustrate the method of fitting the parameters. Figure 3 is the result of fitting, assuming coupling to the τ mode alone, and it can be taken as an excellent description of the experimental data.[2(a),2(b)]

First, we describe the fitting procedure. As was the case earlier, the $\sigma\|[100]$ spectrum depends on S^{A_1} and S^E and on Δ, but because of the very small stress splitting, S^E must be very small. This has the consequence

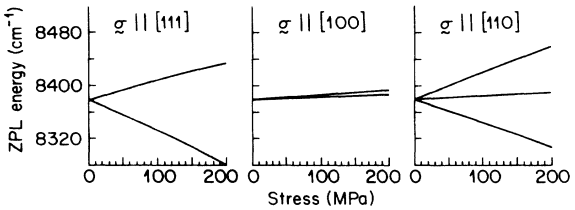

FIG. 1. Stress splitting of the zero-phonon line of an $A_1 \rightarrow T_2$ transition with the T_2 final state equally coupled to τ- and ϵ-mode lattice distortions. The tunneling parameter Δ has been taken to be 1000 cm^{-1} and the other parameters have been adjusted to fit the observed stress splitting as much as possible.

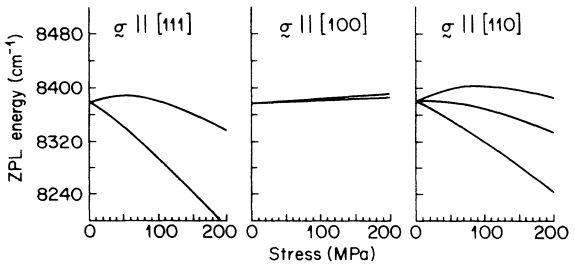

FIG. 2. Same as Fig. 1 except that $\Delta=100\ \mathrm{cm}^{-1}$.

that the coupling to the higher states is virtually zero and the resulting spectrum is virtually independent of Δ. Fitting the energy of the two lines at $\sigma=0$ and at $\sigma=200$ MPa completely determines S^{A_1}, S^E, and E_0, where E_0 is the energy of every transition at zero stress. The values of these three parameters (or more properly, of $K^{A_1}S^{A_1}$, K^ES^E, and E_0) are identical to the corresponding parameters as fitted in the earlier analysis where coupling was to the τ mode only.

This leaves S^{T_2} and Δ to be fixed. For $\sigma\|[111]$, the experimental value of the stress splitting at $\sigma=200$ MPa is $150\ \mathrm{cm}^{-1}$. Satisfying this constraint establishes a relation between Δ and S^{T_2}. The arbitrary choice $\Delta=1000\ \mathrm{cm}^{-1}$ with the corresponding value of S^{T_2} gives the spectra plotted in Fig. 1. This value of Δ was chosen only because it gives the correct end points of the $\sigma\|[111]$ spectrum. There is now very little interaction between the $l=1$ and 3 manifolds. All lines that do interact show approximately the same mild curvature. The result here is unsatisfactory in that it fails to reproduce the pronounced curvature of the line labeled $E5$ in Fig. 3. In addition, the value of Δ is unphysically large by at least an order of magnitude, at least according to the formula $\Delta=10(\hbar\omega)^2/12E_{\mathrm{JT}}$ given in Ref. 11. (E_{JT}, the Jahn-Teller relaxation energy, is analogous—in this problem of relaxation of a degenerate state—to d_{FC}, the Franck-Condon shift, which is the relaxation energy of a nondegenerate state.) This value of Δ implies a Jahn-Teller coupling so small as to invalidate the derivation of Eq. (1), but its use does serve to establish the nature of the difficulty the whole fitting procedure is going to encounter.

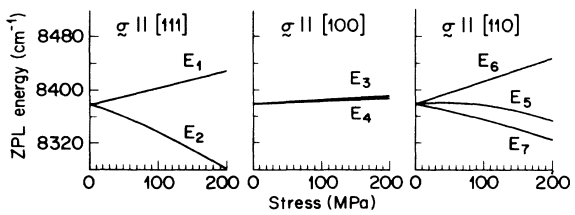

FIG. 3. Stress splitting of the zero-phonon line of an $A_1\rightarrow T_2$ transition with the T_2 final state coupled only to the τ-mode lattice distortion. The parameters have been chosen so as to provide a near perfect fit to the observed stress splitting.

In Fig. 2, we have taken a more realistic value of Δ, namely, $\Delta=100\ \mathrm{cm}^{-1}$. This increases the interaction between the $l=1$ and 3 manifolds and captures the approximate shape of $E5$. At this point, we are within the range of values for which the equations we are using will be valid, and we are in a parameter range which could also be physical interesting. Now however, although we have obtained approximately the right slope and curvature for $E5$, the change does violence to the shape of four other lines in the spectra. In particular, note how low are their values at $\sigma=200$ MPa compared to those in Fig. 3. There is no way out of this situation: in order to bring the other lines back to a better fit, we must give up the pronounced curvature of $E5$. This situation arises becauses there are now so many representations in the $l=3$ manifold that most lines in the $l=1$ manifold are coupled to higher states. This was not the case for the earlier analysis (τ coupling only) where there was only one state, A_1, in the excited state manifold and where only a few of the lower lines are coupled to the excited state. We conclude from the failure to fit the experimental data that the ϵ-mode coupling must be far less than that of the τ mode which, by itself, fits the data perfectly.

IV. ESTIMATE OF THE a-MODE RELAXATION

We now turn to an estimate of the a-mode lattice relaxation associated with the $A_1\rightarrow T_2$ electronic transition. We can no longer make use of stress-splitting information so instead, we make a simple linear model for the lattice relaxation: Let Q be the coordinate for the a-mode lattice distortion. $Q=0$ will refer to the equilibrium configuration with no electrons in the A_1 state or in the T_2 resonance. Let K be the spring constant and let F_{A_1} or F_{T_2} be the force constants for adding an electron to the A_1 or T_2 state, respectively. Then the lattice energies for various electronic states are

$$E^{2+}(Q)=\tfrac{1}{2}KQ^2\ , \tag{9a}$$

$$E^{+}(Q)=\tfrac{1}{2}KQ^2-QF_{A_1}\ , \tag{9b}$$

$$E^{0}(Q)=\tfrac{1}{2}KQ^2-2QF_{A_1}\ , \tag{9c}$$

$$E^{*}(Q)=\tfrac{1}{2}KQ^2-Q(F_{A_1}+F_{T_2})\ . \tag{9d}$$

Charge states 2+, 1+, and 0 have zero, one, and two electrons, respectively, in the A_1 state. The excited state $E^*(Q)$ has one of its localized electrons in the A_1 state and the other in the T_2.

Each of these energies is of the form

$$E^{\alpha}(Q)=\tfrac{1}{2}K(Q-Q^{\alpha})^2-E_0^{\alpha}\ . \tag{10}$$

When the defect undergoes an electronic transition from state i, where the lattice coordinate was Q_i, to state f, where the equilibrium lattice coordinate will be Q_j, the amount by which the energy exceeds its minimum value in state f (the lattice relaxation energy) is

$$\Delta E_{if}=\tfrac{1}{2}K(Q^i-Q^f)^2\ . \tag{11}$$

For example, using

$$Q^{2+}=0 , \tag{12a}$$

$$Q^{+}=F_{A_1}/K , \tag{12b}$$

$$Q^{0}=2F_{A_1}/K , \tag{12c}$$

$$Q^{*}=(F_{A_1}+F_{T_2})/K , \tag{12d}$$

the relaxation energy associated with transfer of an electron between the A_1 state and conduction band is

$$\Delta E_c=F_{A_1}^2/2K \tag{13a}$$

while that associated with the $A_1 \rightarrow T_2$ transition is

$$\Delta E^{*}=\tfrac{1}{2}K(Q^{*}-Q^{0})^2=(F_{T_2}-F_{A_1})^2/2K . \tag{13b}$$

This gives us

$$\Delta E^{*}=(x-1)^2\Delta E_c , \tag{14a}$$

where

$$x \equiv F_{T_2}/F_{A_1} . \tag{14b}$$

Equation (14) captures the intuitive idea that if the charge distribution in the T_2 state is not too different from that in the A_1 state (with regard to force on the a mode), then the lattice relaxation associated with transferring an electron from the A_1 to the T_2 state will be less than that produced by removing the electron from the A_1 state altogether.

There are several experimental estimates of ΔE_c. By fitting the deep-level optical spectroscopy (DLOS) data on the "0" level (now known to be the A_1 state of the As_{Ga}) Chantre, Vincent, and Bois[17] arrived at $\Delta E_c=0.12$ eV. Makram-Ebeid[18] obtained the value 0.115 ± 0.05 eV from the temperature dependence of the electron-capture cross section using a theory by Ridley.[19] Makram-Ebeid and Lannoo[20] obtained the slightly higher value 0.14 ± 0.01 eV by interpreting emission kinetics data according to a theory of phonon-assisted tunneling.

Let us now consider the evaluation of x: The coordinate Q for the a-mode lattice distortion is the change of bond length between the central As_{Ga} and any of its four nearest neighbors. Therefore, the value of Q can be altered by applying hydrostatic pressure to the system. Using the definition that the levels $\epsilon_{A_1}(Q)$ and $\epsilon_{T_2}(Q)$ are the energies to add an electron to the $1+$ state of the system when the lattice coordinate has the value Q, we have

$$F_{A_1} \equiv d\epsilon_{A_1}(Q)/dQ=(d\epsilon_{A_1}/dP)/(dQ/dP) , \tag{15a}$$

$$F_{T_2} \equiv d\epsilon_{T_2}(Q)/dQ=(d\epsilon_{T_2}/dP)/(dQ/dP) , \tag{15b}$$

$$x \equiv F_{T_2}/F_{A_1}=(d\epsilon_{T_2}/dP)/(d\epsilon_{A_1}/dP) . \tag{16}$$

The pressure dependences of the two energies can be extracted from experimental measurements: Dreszer and Baj[21] found that the motion of the *EL2* main donor level (the A_1 state) was 8.7 meV kbar relative to the Γ minimum. This minimum itself has a pressure derivative of 11.8 meV kbar relative to the valence band,[22] which means that the A_1 state moves at $11.8-8.7=3.1$ meV kbar relative to the valence band. On the other hand, Baj and Dreszer[8] found that the pressure derivative of the ZPL is 2.4 meV kbar, which means that the final T_2 state of this transition is moving at a rate of $3.1+2.4=5.5$ meV kbar relative to the valence band. On this basis, we obtain $x=5.5/3.1=1.8$.

Referring back to the experimental estimate, we take $\Delta E_c=0.14$ eV as a reasonable value. On this basis, Eq. (14a) gives $\Delta E^{*}=90$ meV as an estimate of the a-mode contribution to the lattice relaxation of the T_2 electronic state after an $A_1 \rightarrow T_2$ electronic transition.

V. DISCUSSION

In Sec. I of this paper, we showed why the lattice relaxation associated with the τ mode can be 70 meV at most. We argue now that the actual lattice relaxation is substantially below this value. The reasoning here is that the relationship between Δ and $\hbar\omega$, as obtained from the numerical calculations of Caner and Englman,[10] shows that the value of E_{JT} drops rapidly with $\hbar\omega$ for a fixed value of Δ. The value of $E_{JT}=70$ meV was obtained for $\hbar\omega=35$ meV and $\Delta=60$ cm^{-1}. Keeping this same value of Δ (as is required by the fit to the experiments) and letting the phonon energy drop to 30, 25, 20, and 10 meV, we find values of E_{JT} of 54, 40, 24, and 3 meV, respectively.

The localized τ mode of interest here must have an energy below the maximum of the GaAs phonon band structure. To estimate that energy, we have calculated vibrational frequencies using a Keating model in a cluster of 123 atoms. We adjusted the Keating parameters to reproduce the phonon band structure of GaAs, and then altered them to represent the physics at the antisite.[23] The alteration consisted of replacing the mass of the gallium atom at the center of the cluster with the mass of the arsenic atom, and weakening the bond stretching constant between the central arsenic atom and its four nearest neighbors by 25%. This is to simulate the effect of having two electrons in an antibonding state at the defect where there would otherwise be eight electrons in four bonds. The τ mode which most strongly involves the central atom drops in energy from 35 to 28 meV under this change. Such a phonon energy corresponds to an E_{JT} of about 45 meV. This is a much more reasonable estimate of E_{JT} than is the maximum value we have discussed.

The same Keating-model calculation gives two a-mode phonons which couple strongly with the T_2 state. These two modes have the strongest motion of the nearest-neighbor atoms. Their energies are 29 and 18 meV. The numbers obtained here, 28 meV for the τ mode and 29 and 18 for the a mode, are subject to such uncertainties as both the Keating model itself introduces, and those that our representation of the force change simply as a 25% reduction in bond strength have caused. Nonetheless, the fact that there is one τ mode whose energy is significantly below the top of the spectrum, and two strongly coupled a modes of relatively high energy may prove useful in eventually understanding the fine structure of the absorption spectrum.

It may be a coincidence, but these energies of 29 and 28 meV are close to what was mentioned in Sec. I as being needed to account for the 0.15 eV half-width of the optical absorption. One should perhaps not take this agreement too seriously because the theory under which that 0.15-eV width was analyzed is one in which the electronic state involved in the relaxation is nondegenerate. The details of the relaxation process change when, as here, a degenerate state is involved in the relaxation. Nonetheless, a rough estimate of the strength of the zero-phonon line relative to the main peak is $\exp(-E_a/\hbar\omega_a - E_\tau/\hbar\omega_\tau)$ where E_i and ω_i are the lattice relaxation and the phonon associated with mode i. In this case, this estimate is that the ZPL has an intensity $\exp(-0.15/0.029)$ or 0.006 of the total.

In Sec. III, we showed that the coupling to the ϵ mode must be small, implying that the associated lattice relaxation must be much smaller than that of the τ modes.

Finally, in Sec. IV, we obtained an estimate of 90 meV for the lattice relaxation associated with the a modes. These relaxations add up to a difference of about 135 meV between the vertical transition and the associated ZPL.

Is it significant that we have found a lattice relaxation very close to the observed one when there are features of this situation which differ from those assumed in the standard treatments of the Jahn-Teller effect? These treatments all assume electronic states which are discrete and fully localized. The situation here is that the final T_2 state is a resonance, close to the bottom of a band before relaxation occurs, and below it after. For this reason, the question is hard to answer without a complete theory of lattice relaxation when the electronic state involved is a degenerate resonance lying in a region of rapidly varying background density of states, which is the situation here.

It should be noted, however, that breaking the conventional relationship between the 1.18- and 1.04-eV features can resolve some of the puzzles concerning the optical absorption of $EL2$, as Baj and Dreszer[8] have already discussed.

If we assume that the 1.04-eV feature is the zero-phonon line, it leaves open the question of what the peak at 1.18 eV represents if it is not the vertical transition associated with the ZPL. One clue suggests that the peak could be ionization of an electron from the A_1 midgap level to the conduction band. That clue is the fact that the 1.18-eV feature also shows up in the DLOS spectrum of Chantre *et al.*[17] This spectroscopy is sensitive to electrons transferred from the deep level to the extended states, and as such, it should not give signals arising from internal transitions such as the A_1 to T_2 we are discussing here. In the original interpretation of the DLOS results, the features were interpreted as transitions from the localized level to the Γ, L, and X point conduction-band minima. It is true that this interpretation fell into disuse when the optical-absorption measurements of Kaminska *et al.*[24] showed that the photocurrent was less than the optical absorption in a spectral region which corresponded to the spectrum for activating $EL2$ to its metastable state. This was taken to mean that the transition of $EL2$ from its normal to its metastable state was driven by an internal optical transition, not an ionizing one. There is no reason to doubt that. What we are suggesting however, is the possibility that the vertical internal optical transition may be masked by more intense photoionization which coincidentally takes place at the same energy.

This speculation needs to be tested experimentally of course. One way of doing so might be a careful comparison of the optical cross section for transferring $EL2$ from its normal to its metastable state in two types of experiments. One type of experiment would be photocapacitance experiment similar to those performed by Vincent and Bois[25] who discovered the metastability. These experiments took place in a junction where any electrons ionized to the conduction band are swept away immediately. As a consequence, the only electrons available to drive the transition to the metastable state are those which undergo the A_1 to T_2 internal transition. The other experiment would be photobleaching experiments where there is no internal electric field.[26,27] In such case, electrons ionized to the conduction bands could be recaptured by the ionized As_{Ga}'s and, judging from the energetics, that recapture would most likely be via states near the L point into the T_2 resonance. Once the electron is in the T_2 state, the transition to the metastable is triggered.

The idea of two cross sections for these two paths of entry into the T_2 state is illustrated in Fig. 4. Among the

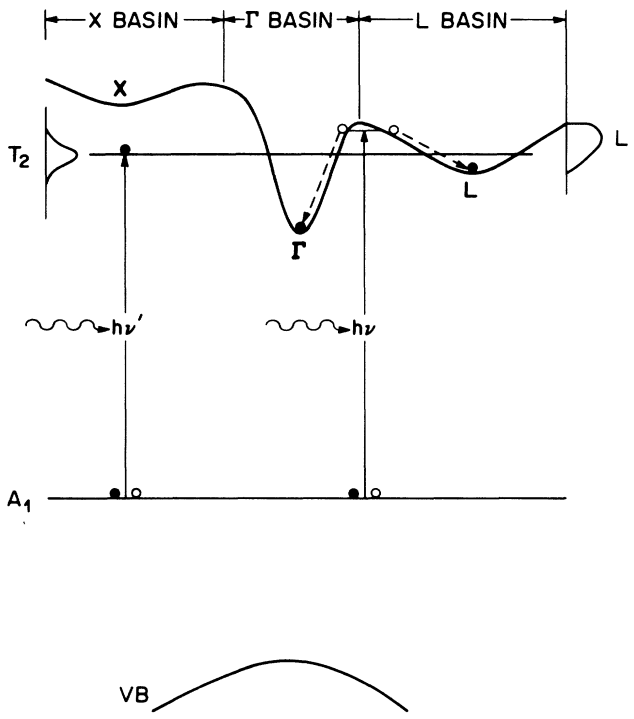

FIG. 4. Sketch of the channels by which an electron, ionized from the A_1 level, can be captured into the T_2 level. One channel is via an internal optical transition. The other is via ionization to an L-valley catch basin in the conduction band, thermalization to the bottom of the valley, and multiphonon capture to the T_2 state. The cross section for this latter process is dependent on the conduction-band structure in the catch basin, and not on the exact energy of the T_2 state.

ideas being presented here are that one can think of the lowest conduction band as being separated into eight different "catch basins" which together cover the entire Brillouin zone. Each catch basin consists of a contiguous region of k space such that if an electron were released into that region, it would (in the absence of strong momentum changing collisions) thermalize down into one of the eight conduction-band minima, arriving thereby at Γ, or at one of the three X points or at one of the four L points. The unrelaxed T_2 electronic state lies very close to the L band minimum, so multiphonon capture will be very efficient for electrons in this minimum and not for electrons in the other minima.

Calculation of the rate at which electrons are captured into the T_2 state by multiphonon emission clearly involves knowledge of the number of electrons which have been ionized out of the A_1 level into the catch basins for the L points. This number varies with the energy of the incident light, and we have indicated that variation by the cross section versus energy sketch placed to the right of the L valley in Fig. 4. The shape of that curve depends on the k distribution in the A_1 state and on the detailed shape and location of the L-point catch basin, but does not depend on the energy of the T_2 state.

There is also the possibility of direct optical transfer from the A_1 state to the T_2 state. The cross section for this process is sketched to the left of the T_2 state in Fig. 4. This cross section *does* depend on the energy of the T_2 state of course. In an experiment in which a strong electric field is present, as in a junction where the electrons are swept away from the defect, the only way for an electron to get into the T_2 state is via this direct internal transition. On the other hand, where there is no electric field, the electrons will thermalize into the various minima and will be attracted to the ionized defects. There are now two paths into the T_2 state, the direct one and also multiphonon capture from the L-point catch basins. The cross section for the total rate of entry to the T_2 state will be the sum of the two separate cross sections.

Suppose that, for some reason, we have overestimated the amount of lattice relaxation and that the vertical transition really lies beneath the peak in the optical absorption. Then the cross section for transferring $EL2$ to the metastable would have its maximum in the photocapacitance measurements somewhat below where it would be in the optical bleaching measurements, where the peak is at 1.18 eV.[26,27] Although $\sigma^*(h\nu)$ has been measured using photocapacitance by Vincent *et al.*[28] who found that the peak was at 1.13 eV, we cannot consider this to be a confirmation of the suggestion made here until such measurements are repeated with special care to determine how much of the shift of the peak to an energy below 1.18 eV is caused by the different temperature at which the different types of experiments are performed. Alternatively, the effects of pressure on these two types of cross section might reveal pressure coefficients of different magnitude or sign.

Another possibility, suggested by Skowronski,[29] is that the 1.04-eV feature is not the ZPL at all but is instead an effective-mass state associated with the L-point conduction-band minimum. The energy is correct for such an identification, and as von Bardeleben has very recently noted, the pressure coefficient of the final state of the 1.04-eV feature also matches that of the L point.[30] This possibility would allow the 1.18-eV feature to be the vertical transition of the internal transition and would imply that the ZPL has simply not been observed against the background of absorption caused by ionizing the A_1 level to the conduction bands.

The calculation presented in this paper has demonstrated that we can account for the full energy separation between the 1.18-eV optical absorption peak and the 1.04-eV ZPL using the existing theories of lattice relaxation and Jahn-Teller coupling. Our desire to explain the problems in the optical spectrum by retaining an explanation that the final state of the ZPL and the final state of the peak in absorption are different can be accommodated only under two different possibilities. One is that the conventional theories really do not apply to the situation encountered here where the relaxing state is a resonance not far from a band edge. The other possibility is that either the 1.18-eV feature has been wrongly understood to be the vertical transition associated with the ZPL or the 1.04-eV feature has been wrongly understood to be the ZPL. Sorting out which of these is correct should be both interesting and possible.

ACKNOWLEDGMENTS

I should like to thank Professor Frank S. Ham for calling to my attention the beautiful work of M. C. M. O'Brien on the $T_2\times(\epsilon+\tau)$ Jahn-Teller problem and for his help in clearing up some of the questions it raised while the present calculations were being carried out. I should also like to thank Professor H. J. von Bardeleben for sending me a copy of Ref. 30 prior to publication and Dr. W. Kuszko for interesting correspondence and for sending me a copy of Ref. 27 prior to publication.

APPENDIX

We shall now show that the electronic coupling constant S^E is inherently much smaller than the electronic coupling constant S^{T_2}. Our starting point is to assume, as was done in Ref. 9, that the T_2 symmetry electronic states can be regarded as linear combinations of the four antibonding orbitals which join the central As_{Ga} to each of its four nearest neighbors, as shown in Fig. 5. We write the A_1 and T_2 linear combinations of these four as

$$|s\rangle=\frac{1}{2\Lambda'}(\phi_1+\phi_2+\phi_3+\phi_2)\ , \tag{A1a}$$

$$|x\rangle=\frac{1}{2\Lambda}(\phi_1-\phi_2-\phi_3+\phi_4)\ , \tag{A1b}$$

$$|y\rangle=\frac{1}{2\Lambda}(\phi_1-\phi_2+\phi_3-\phi_4)\ , \tag{A1c}$$

$$|z\rangle=\frac{1}{2\Lambda}(\phi_1+\phi_2-\phi_3-\phi_4)\ . \tag{A1d}$$

Λ' and Λ are normalization factors which would equal 1 if the overlap of the ϕ_i orbitals were zero.

Now consider the matrix elements of a 100 stress in the

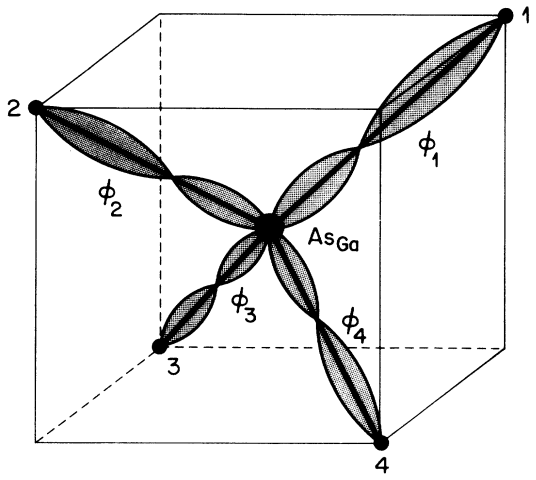

FIG. 5. Diagram showing the As_{Ga}, its four nearest-neighbor arsenic atoms designated $i=1,2,3,4$, and the four antibonding orbitals of which the T_2 states are linear combinations.

ϕ_i basis. The axis lies along the x direction. Since the 111 axes of all four orbitals make the same angle with x, the expectation value of the stress is the same for all four ϕ_i. We denote that expectation value as σA, where σ is the magnitude of the stress. Off-diagonal elements of the (100) stress, however, depend on how *pairs* of axes are related to the x direction. As one can see from Fig. 4, the two pairs of orbitals, ϕ_1-ϕ_4 and ϕ_2-ϕ_3, have one relation to the x direction while the other pairs of orbitals, ϕ_1-ϕ_2,ϕ_1-ϕ_3,ϕ_2-ϕ_4, have a different relationship to the x direction. Thus, the form of the matrix for a (100) stress is

$$M_{ij}(100)=\sigma\begin{bmatrix} A & \beta & \beta & \alpha \\ \beta & A & \alpha & \beta \\ \beta & \alpha & A & \beta \\ \alpha & \beta & \beta & A \end{bmatrix} . \tag{A2}$$

The stress potential is a local operator. Off-diagonal elements have a nonzero value only if there is a region of space near the As_{Ga} where two different orbitals ϕ_i and ϕ_j have appreciable amplitude. For this reason, we can anticipate that both α/A and β/A are small, of order of the overlap between different orbitals.

We transform Eq. (A2) to an $|s\rangle,|x\rangle,|y\rangle,|z\rangle$ basis, working to lowest order in the overlap—that is, using Eq. (A1) with $\Lambda=\Lambda'=1$. The result is

$$M_{\mu\nu}(100)=\sigma\begin{bmatrix} A+\alpha+2\beta & 0 & 0 & 0 \\ 0 & A+\alpha-2\beta & 0 & 0 \\ 0 & 0 & A-\alpha & 0 \\ 0 & 0 & 0 & A-\alpha \end{bmatrix} . \tag{A3}$$

The (100) stress is seen to be diagonal in the T_2 representation [the 3×3 matrix in the lower right-hand corner of Eq. (A3)]. Compare this with the matrix for the 100 stress as given by Eq. (4.6a) of Ref. 9:

$$\langle\cdot|V^{\text{stress}}|\cdot\rangle=\frac{\sigma}{3}\begin{bmatrix} S^{A_1}+2S^E & 0 & 0 \\ 0 & S^{A_1}-S^E & 0 \\ 0 & 0 & S^{a_1}-S^E \end{bmatrix} . \tag{A4}$$

Comparing (A3) and (A4), we get

$$-\alpha=2\beta , \tag{A5}$$

$$S^{A_1}=3A , \tag{A6a}$$

$$S^E=3\alpha . \tag{A6b}$$

If the orbitals ϕ_i and ϕ_j had not overlapped at the origin, then α would have been zero and S^E would have vanished.

This is not the case for S^{T_2}. It remains finite even if there is no orbital overlap. To see this, consider a stress along the 111 direction, and consider its matrix in the ϕ_i basis. The axis of the ϕ_1 orbital is parallel to the stress direction while the axes of like other three orbitals all make the same (nonzero) angle with the stress direction. In the absence of orbital overlap, the stress will have no off-diagonal elements in the ϕ_i basis, and we have a matrix of the form

$$M_{ij}(111)=\sigma\begin{bmatrix} B & 0 & 0 & 0 \\ 0 & C & 0 & 0 \\ 0 & 0 & C & 0 \\ 0 & 0 & 0 & C \end{bmatrix} . \tag{A7}$$

Upon transformation to the $|s\rangle,|x\rangle,|y\rangle,|z\rangle$ basis, this becomes

$$M_{\mu\nu}(111)=\sigma\begin{bmatrix} D & E & E & E \\ E & D & E & E \\ E & E & D & E \\ E & E & E & D \end{bmatrix} , \tag{A8a}$$

where

$$D\equiv(B+3C)/4 , \tag{A8b}$$

$$E\equiv(B-C)/4 . \tag{A8c}$$

Again, compare the lower right-hand 3×3 block of this matrix with the matrix for a (111) stress taken with respect to the states $|x\rangle,|y\rangle,|z\rangle$. As given by Eq. (4.6c) of Ref. 9, it is

$$\langle\cdot|V^{\text{stress}}|\cdot\rangle=\frac{\sigma}{3}\begin{bmatrix} S^{A_1} & S^{T_2} & S^{T_2} \\ S^{T_2} & S^{A_1} & S^{T_2} \\ S^{T_2} & S^{T_2} & S^{A_1} \end{bmatrix} . \tag{A9}$$

On comparing (A8) and (A9), we obtain

$$S^{A_1}=3(B+3C)/4 , \tag{A10a}$$

$$S^{T_2}=3(B-C)/4 . \tag{A10b}$$

In order to relate A, B, and C, we use the fact that the

expectation value of the stress, taken with respect to an orbital which is rotationally symmetric about some axis, is proportional to the square of the cosine of the direction between the axis and the stress. Using that relationship, we expect that

$$A:B:C=\tfrac{1}{3}:\tfrac{1}{1}:\tfrac{1}{9} \ . \tag{A11}$$

This makes Eq. (A6a) agree with Eq. (A10a), while in (A10b), it gives

$$S^{T_2}=2A \ . \tag{A12}$$

Finally, from (A6b) and (A12), we have

$$S^{E}/S^{T_2}=3\alpha/2A \tag{A13}$$

a quantity which is of the order of the overlap between orbitals ϕ_i and ϕ_j—i.e., small.

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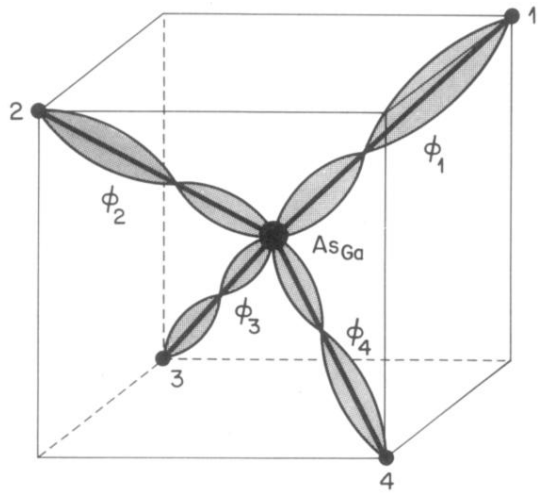

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