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Atomic model for the *EL*2 defect in GaAs

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From an analysis of the electronic properties of the *EL*2 defect in its stable and metastable configurations, and of the optically or electrically stimulated transformation between these, we deduce atomic models for the two configurations. Our model of the stable configuration consists of a divacancy on one side of an As—on—Ga-site antisite defect. For the metastable configuration we propose that the antisite separates the two vacancies. We show that this model fits the electrical and optical observations in detail. We note that the model is also in accord with thermodynamic determinations and existent positron-annihilation data. We propose a test for the model by an additional positron experiment.

I. INTRODUCTION

The atomic identity of the deep-level defect know as *EL* 2 has been the subject of many investigations and much controversy for more than ten years.[1–36] *EL* 2 is technologically important because the state-of-the-art method to grow semi-insulating GaAs for use as a substrate for advanced GaAs electronic devices uses the liquid-encapsulated Czochralski (LEC) process with the As vapor pressure controlled so as to introduce a concentration of the *EL* 2 deep levels sufficient to compensate the residual dopants. [It is necessary to have semi-insulating substrates to allow for isolation of adjacent devices and to reduce parasitic capacitance in metal-semiconductor field-effect transistors (MESFET's).] There is also considerable scientific interest in the *EL* 2 defect; we will be able to cite only a small fraction of the vast number of interesting papers on the subject.

Although the atomic identity of *EL* 2 is controversial, there seems to be a consensus emerging among both experimentalists and theorists that *EL* 2 is a complex involving an As—on—Ga-site antisite defect, $\mathrm{As}_{\mathrm{Ga}}$, as one of its constituents. It has also been established[16] that in the stable configuration this antisite defect has four As nearest neighbors. Some workers have previously suggested that *EL* 2 is nothing more than the isolated $\mathrm{As}_{\mathrm{Ga}}$, but there is now convincing evidence that $\mathrm{As}_{\mathrm{Ga}}$ is only part of a more complicated complex.[16–21] There is also a growing consensus that *EL* 2 is not a unique complex,[22–27] but a family of related complexes. In this paper we will describe our model for the core of *EL* 2, i.e., the constituents we believe are present in all members of the family, as if that member were unique. However, we will also indicate the structure of other members that we believe contain one or more neutral antistructure pair in various configurations about the core.

One of the most interesting characteristics of *EL* 2 is the appearance of metastability in its properties at low temperatures, $T < 100$ K. This metastability of properties is evidently a consequence of metastability of the atomic configuration of each member of the *EL* 2 family. Effects evidencing metastability include photoconductivity,[6,8] photoluminescence,[28,29] photostimulated electron spin resonance (photo-ESR),[30,31] optical absorption,[32] and photocapacitance.[33–36] These effects are usually interpreted within the context of configuration coordinate (CC) diagrams with large lattice relaxations,[33,35,36] as with a Franck-Condon shift. The effect known as Auger deexcitation,[34] in which a free electron takes away the energy released by an intradefect configurational transformation without ever becoming localized, is also often invoked.

We suggest that analysis of these metastable properties is a powerful avenue of attack for the problem of atomic identification. In that vein we here offer an atomic model for *EL* 2, Fig. 1, which can account for the metastable properties of this defect complex. It is similar to that

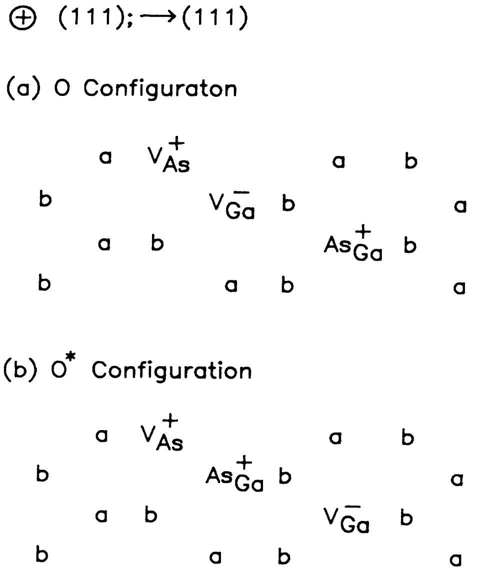

FIG. 1. Atomic model proposed for *EL* 2: (a) in the equilibrium configuration, *O*; (b) in the metastable configuration, O^*.

which we recently proposed[37] for the so-called *M* center in InP, which also displays metastable effects. For both we invoke vacancy nearest-neighbor hopping as the mechanism of the large lattice relaxation concomitant with the transformation between configurations. In the case of *EL* 2, the complex is now generally believed to have very much the same ionization-level scheme in both configurations.[35,38] This means that for the same position of the Fermi level, E_F, the complex will attain the same net charge in either configuration.

We denote the thermodynamically stable atomic configuration as O and the metastable configuration, O^*. The reader should not misinterpret this notation either as indicating that the *EL* 2 complex involves oxygen (as thought when this notation was coined) or that the net charge of the complex in these two configurations is zero. For our model of the complex and for the ionization levels of the three point defects from which it is composed, V_{Ga}, V_{As}, and $\mathrm{As}_{\mathrm{Ga}}$ (Fig. 2), which we have taken from what we regard as the best available empirical determinations,[30,31,39–41] the net charge may be -1, 0, $+1$, $+2$, or $+3$ depending on E_F. However, the absolute charge states of *EL* 2 are not firmly established by available experiments; charge differences between states in the gap are established. Furthermore, it seems that *EL* 2 must have both deep donor and deep acceptor states in the gap because it serves to compensate both acceptor and donor dopants. Hereafter we will discuss ionization levels in terms of the ionization levels shown in Fig. 2, the justification of which will be described below. Many of the conclusions we draw would not be changed if somewhat different ionization levels of the point defects were to be assumed and the net charge of the complex for various values of E_f were altered.

We note that the effect of vacancy nearest-neighbor hopping for both *EL* 2 and the *M* Center, or for any vacancy-related defect in a compound semiconductor, is that a vacancy on one sublattice is exchanged for a vacancy on the opposite sublattice together with the creation or annihilation of an antisite defect.[22,25,42–44]

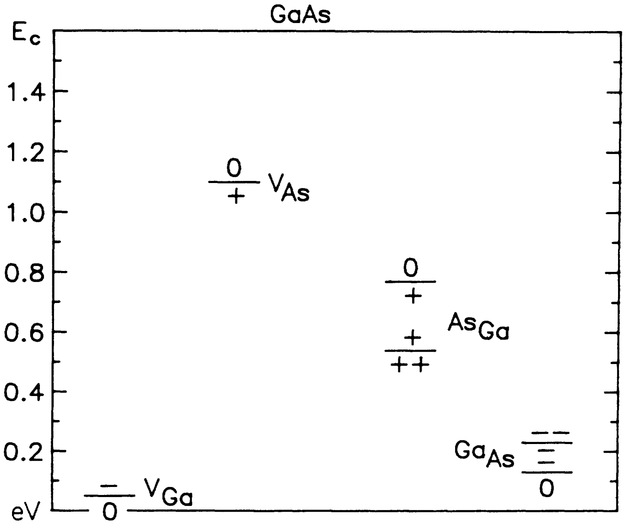

FIG. 2. Literature values for the ionization levels of vacancies and antisite defects in GaAs at $T=0$ K, where the band gap is 1.52 eV.

We must note that our conclusions are consistent with the thermochemical data for *EL* 2 as analyzed[9] by Zou and co-workers, who previously concluded that the complex involves a divacancy plus an As—on—Ga-site antisite defect. It is also consistent with the recent positron-annihilation data[18] of Dannefaer and Kerr, which confirms the presence of two-vacancy complexes in semi-insulating GaAs that contains *EL* 2 and gives strong evidence that their concentration is of the same order as that of *EL* 2. We are also in agreement with the conclusion[10] of Baraff and Schlüter, based on their local density approximation, Green's-function method calculations, that the actuator of the *EL* 2 transformations is the nearest-neighbor hopping of a Ga vacancy, V_{Ga}.

II. METASTABLE PROPERTIES OF *EL* 2

Levinson has succinctly summarized the metastable properties[38] of *EL* 2; we will draw from his presentation. The interesting metastable properties of *EL* 2 include the following.

(1) An effect known as persistent photocapacitance quenching[33–36] (PPCQ) is observed by cooling to $T < 100$ K an *n*-type GaAs-metal diode with a sufficient concentration of *EL* 2. The deep levels of the *EL* 2 complex are saturated with electrons so that the complex attains a net negative charge, -1 in our model (i.e., they compensate some of the donors). This is usually done by allowing the diode to stand without electrical bias. (This process is called a "zero-bias pulse" in the dark.) The diode is then subjected to a reverse bias, which brings E_F in the junction to a position a little above midgap. The junction capacitance is monitored under reverse bias while the sample is illuminated with light having $0.9 < h\nu < 1.35$ eV. The capacitance first increases, as would be expected if electrons are simply being detrapped from a donor level which is inferred to be part of the *EL* 2 complex. (If one of the acceptor levels that account for the net negative charge of *EL* 2 were being deionized, we would expect the capacitance to decrease.) Thus, the net charge of the complex changes to 0 (or to a less negative state). However, as the illumination continues, the capacitance begins to decrease and eventually regains its initial value. This is taken as evidence that the original ionization state, -1, is regained but now it is in the metastable O^* configuration. Further illumination does not produce any further change in junction capacitance. Thus, the photoinduced increase in capacitance expected of a material containing simple deep donors, the photocapacitance, is found to be eliminated (quenched) by *EL* 2 complexes and this condition persists for extended times at low temperatures.

The photoconductivity for photon energies above the band gap of the sample in the O^* configuration is 2 orders of magnitude less than for the O configuration.[6,8] Thus, the photoconductivity is also quenched by the same persistent reconfiguration of the *EL* 2 defect that eliminates the photocapacitance.

If the photon energy is set above 1.4 eV, the same experiment causes the junction capacitance to increase monotonically and to saturate. That is what should occur if isolated deep donors were simply ionized by the light.

It should be noted that the 0.9-eV threshold photon energy for the PPCQ process is definitely greater than a 0.77-eV threshold[30,31] to photoexcite an electron from the $\mathrm{As_{Ga}}$ ($\mathrm{As_{Ga}}$ $0 \to +$). Moreover, the upper bound photon energy, 1.35 eV, is distinctly less than the band gap[45] of GaAs, which is 1.52 eV for $T=0$ K.

It is also important to note that the O^* to O transformation is not photoexcited for photon energies less than the band gap.[35] (Values down as far as $h\nu=0.4$ eV have been tried.)

(2) Thermal regeneration: The metastable configuration O^* which quenches the photoconductivity and which may be produced as just described, will persist for hours if $T<100$ K is maintained. However, the stable state O, which permits strong photoconductivity, is regenerated by a thermally activated process at a rate[35]

$$R_{\mathrm{th}}=10^{11}\exp(-0.3\ \mathrm{eV}/k_BT)\ \mathrm{sec}^{-1}\ . \tag{1}$$

No change in junction capacitance is observed during the thermal regeneration of O from O^*. This is taken as strong evidence that O and O^* have the same net charge, as stated above when we adopted the notation.

(3) Auger deexcitation: The rate of regeneration of O from O^* may be accelerated above R_{th} by injecting free electrons into the junction.[34,35] There is evidently an interaction between the free electrons and those localized in the O^* state of the complex despite the fact that no net change in charge state accompanies the regeneration of O. This enhancement of the regeneration rate by electron injection has been ascribed to Auger deexcitation.

The Auger regeneration proceeds at a rate given by an interaction cross section σ for electrons that is thermally activated as[33]

$$\sigma=10^{-13}\exp(0.108\ \mathrm{eV}/k_BT)\ \mathrm{cm}^2\ . \tag{2}$$

It may well be that the regeneration process is not truly an Auger deexcitation process but does involve the transient localization of an electron at the complex. If so, then because there is evidently no change in the net charge of the complex for the total regeneration process, the initial capture of the electron into $EL\,2$ (with this thermally activated cross section) must be followed by the spontaneous reemission of an electron. It seems most likely that this occurs after the large lattice relaxation and may be from a different member of the complex.

The metastable properties of $EL\,2$ are often described in terms of a CC diagram such as that shown in Fig. 3, which is essentially the same as that given[35] by Vincent, Bois, and Chantre (their Fig. 7). The stable configuration O must have a net energy less than that of the metastable configuration O^* and the two must be separated with an activation barrier of 0.3 eV for indirect, phonon-assisted transitions.

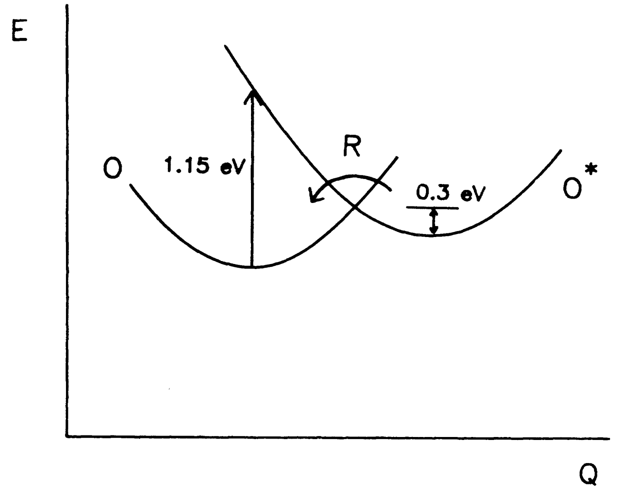

FIG. 3. Configuration coordinate diagram for $EL\,2$ in GaAs. The metastable configuration O^* is positioned using the thermal activation energy (0.3 eV) and the photon energy for maximum quenching efficiency (1.15 eV).

III. IONIZATION ENERGIES OF POINT DEFECTS IN GaAs

Any atomic identification of $EL\,2$ requires some estimate of the ionization energies of the point defects which may comprise the complex. We now justify as well as possible our best estimates of the ionization energies of the two antisite defects and the two vacancies in GaAs, which we have shown in Fig. 2.

Here we ignore the possibility that self-interstitials might play a role in $EL\,2$ for three reasons. First, no self-interstitial has been firmly identified in any III-V compound even after electron irradiation, which must have produced self-interstitials together with the vacancies that have been firmly identified.[46] Therefore, the attribution of any ionization level to either self-interstitial in GaAs would have to be pure speculation. Second, the same observation of the relatively greater stability of irradiation-induced vacancies, just noted, implies that the enthalpy of formation of self-interstitials in III-V compounds is too large for them to be important constituents of the deep-level complexes that determine the static properties of the material.[42] Third, recent positron annihilation studies show that intrinsic GaAs contains of order $1\times10^{17}\ \mathrm{cm}^{-3}$ two-vacancy complexes.[18] As a divacancy would afford the proper lattice site to annihilate a self-interstitial of either type, it is very difficult to believe that any substantial concentration of self-interstitials could persist for laboratory times.

The point defect ionization levels that have been best established are probably the two donor levels of $\mathrm{As_{Ga}}$. From photo-EPR measurements, Weber *et al.* report[13,30,31]

$$E_{0/+}(\mathrm{As_{Ga}})=E_c-0.77\ \mathrm{eV}\ , \tag{3}$$

where E_c denotes the conduction-band edge, and

$$E_{+/++}(\mathrm{As_{Ga}})=E_c-1.00\ eV\ . \tag{4}$$

These measurements were made at $T=6$ K, for which the

GaAs band gap[45] $E_{cv}=1.52$ eV.

For the As vacancy, V_{As}, we take the only ionization level to be that of a single donor with

$$E_{0/+}(V_{\mathrm{As}})=E_c-0.45\text{ eV}\ . \tag{5}$$

This is based on the work[39] of Thomas *et al.*, who found that LEC GaAs grown in a Ga-rich melt was *n* type provided the *B* concentration was kept low. Variable-temperature Hall effect measurements revealed a deep donor in the sample with an activation energy of 0.45 eV. (Thomas *et al.* suggest that this level might belong either to V_{As} or to Ga_i; we select the former option over the latter for the reasons stated at the beginning of this section.) This identification is in good agreement with the theory[47] of Bachelet *et al.* Other calculations[40,48–50] indicate the level should be somewhat nearer to E_c.

For the Ga vacancy V_{Ga}, we accept the semiempirical estimate[40] of Potz and Ferry that there is a single acceptor level at

$$E_{-/0}(V_{\mathrm{Ga}})=E_v+0.01\text{ eV}\ . \tag{6}$$

This value is in reasonable agreement with that[50] of Ho and Dow, who place it at $E_v-0.03$ eV. Lin-Chung and Reinecke estimate[49] this level 0.44 eV above E_v. Thus, although there is some controversy as to the exact energy of this acceptor level,[51] all estimates place it in the lower third of the band gap so that is should be filled in thermal equilibrium for all bias levels used in the experiments described above.

There is some controversy as to the ionization levels for $\mathrm{Ga}_{\mathrm{As}}$. There are two points of view. According to one,[52,53] there are two acceptor levels and both are near midgap;

$$E_{-/0}(\mathrm{Ga}_{\mathrm{As}})=E_v+0.40\text{ eV} \tag{7}$$

and

$$E_{--/-}(\mathrm{Ga}_{\mathrm{As}})=E_v+0.70\text{ eV}\ . \tag{8}$$

The other point of view[41,54–56] agrees that there are two acceptor levels but places them much closer to E_v:

$$E_{-/0}(\mathrm{Ga}_{\mathrm{As}})=E_v+0.078\text{ eV} \tag{9}$$

and

$$E_{--/-}(\mathrm{Ga}_{\mathrm{As}})=E_v+0.20\text{ eV}\ . \tag{10}$$

We believe these two points of view can be reconciled by taking account of the differences in the methods of preparation of the samples used by the different groups. Those who hold to the former, near midgap determinations prepared their samples in either of two ways: (1) Liquid-phase epitaxy (LPE) grown material was subjected to irradiation with 1-MeV electrons;[57] or (2) LPE GaAs was cooled rapidly (from 800 °C to 100 °C in less than one minute).[52] It was noted than when the same samples were cooled rather less rapidly, the midgap levels ascribed to $\mathrm{Ga}_{\mathrm{As}}$ could not be found. Those who hold to the latter, near E_v determination, used semi-insulating GaAs that had been grown by LEC under Ga-rich conditions (low As over pressure).

We propose that the acceptor levels of $\mathrm{Ga}_{\mathrm{As}}$ are indeed near E_v, as the latter group holds, if the $\mathrm{Ga}_{\mathrm{As}}$ is relatively distant from any vacancy or other antisite defects. We further propose that the near-midgap ionization levels ascribed to $\mathrm{Ga}_{\mathrm{As}}$ by the former group are characteristic of $\mathrm{Ga}_{\mathrm{As}}$ with a V_{Ga} in its ionized acceptor state, V_{Ga}^- on a nearest-neighbor site. Note that according to Eq. (6), the V_{Ga} acceptor ionization level is lower than those proposed for $\mathrm{Ga}_{\mathrm{As}}$. Therefore, when E_F is near either of the $\mathrm{Ga}_{\mathrm{As}}$ levels, it is above the acceptor level of V_{Ga}, so that the vacancy should be ionized. The Coulomb field of the electron already bound to the V_{Ga} will repel other electrons from any nearby $\mathrm{Ga}_{\mathrm{As}}$ and raise the position of E_F required to populate the acceptor states of the $\mathrm{Ga}_{\mathrm{As}}$. Thus, when $\mathrm{Ga}_{\mathrm{As}}$'s are formed by nearest-neighbor hopping of V_{As}'s, which is a common mode of self-diffusion in GaAs, there is simultaneously created a V_{Ga} on the nearest-neighbor site and the interaction between the two should raise the acceptor levels of the $\mathrm{Ga}_{\mathrm{As}}$ from those characteristic of the isolated point defect. The magnitudes of these two increases, about 0.3 eV for the more nearly-free-electron-like first level $E_{-/0}(\mathrm{Ga}_{\mathrm{As}})$ and about 0.5 eV for the deeper level $E_{--/-}(\mathrm{Ga}_{\mathrm{As}})$, are roughly in accord with the theory of such interactions[51,58,59] by Sankey and Dow. One can also rationalize this shift using a very simple point charge with dielectric screening approximation.[60,61] It is reasonable to suppose that there are many more isolated $\mathrm{Ga}_{\mathrm{As}}$'s in Ga-rich LEC material that have not been irradiated or quenched to produce an excess concentration of vacancies.

We further note that the assignment of $\mathrm{Ga}_{\mathrm{As}}$ levels in Ga-rich LEC material was done making use of photoluminescence and temperature-dependent Hall-effect measurements. The analysis of this data is somewhat complicated by the role of *B* that gets into LEC GaAs from the borosilicate glass used to encapsulate the material. We agree with the conclusions[56] of Dansas, who asserts the $E_v+0.078$ eV level is the first acceptor state of $\mathrm{Ga}_{\mathrm{As}}$, while another level found at $E_v+0.068$ eV is the first acceptor state of B_{As}, the antisite defect of the *B* component of the dilute alloy of GaAs with B_{As}. As *B* is a more electronegative element[62] than Ga, it is natural that, due to the central-cell correction to the effective-mass-approximation value for the hydrogenic first acceptor level, B_{As} should have a first acceptor ionization level slightly lower than $\mathrm{Ga}_{\mathrm{As}}$.

IV. AN ATOMIC MODEL FOR *EL* 2

We conclude that any model for *EL* 2 must be consistent with the following facts.

(1) $\mathrm{As}_{\mathrm{Ga}}$ is one component of *EL* 2 and in the *O* configuration it has four As nearest neighbors.

(2) For the same position of E_F, the stable *O* configuration and the metastable O^* configuration generally have the same net charge.

(3) The $O\rightarrow O^*$ transformation can be driven with photons of energy $0.9<h\nu<1.35$ eV for $T<100$ K.

(4) The $O^*\rightarrow O$ transformation cannot be induced by photons of energy $0.4<h\nu<1.5$ eV.

(5) Once formed, O^* has an activation barrier for

thermal transition back to O of 0.3 eV, so it is persistent at low temperatures.

(6) The $O^* \rightarrow O$ transition can also be driven nonthermally by injection of free electrons. The effective cross section σ for electrons in this process is thermally activated with a thermal activation energy of 0.108 eV.

(7) There is no net change in the charge state of $EL\,2$ accompanying the injection induced $O^* \rightarrow O$ transition so that effect must either involve capture of the free electron followed by spontaneous reemission after the large lattice relaxation has occurred or an Auger deexcitation process.

We also agree with almost all the literature that the two configurations must be separated by some atomic reconfiguration, commonly called a large lattice relaxation.

Returning to our model for $EL\,2$ in Fig. 1, we note that it contains an $\mathrm{As_{Ga}}$ [fact (1)] as required by the extensive ESR studies,[13,16,30,31] and that in the O configuration this antisite defect has four As nearest neighbors. Also note that our model has all the same point defects, one V_{Ga} and one V_{As} in addition to the $\mathrm{As_{Ga}}$, in both configurations; the configurations differ only as regards the position of the vacancies with respect to the $\mathrm{As_{Ga}}$. As the effect of the position of adjacent deep levels on the deep ionization level is only moderate,[51,58,59] it is clear that our model is in accord with fact (2), the ionization state of the two configurations is generally the same for any given E_F. It is also clear that the O configuration is the one of lower total energy due to the divacancy binding energy. The energy to separate a divacancy in GaAs, as occurs in our model for the O configuration, to a $V_{\mathrm{Ga}}+V_{\mathrm{As}}$ pair of neutral single vacancies has been estimated[61] to be 0.86 eV.

Consider now the photoexcited $O \rightarrow O^*$ transformation. We note that the cutoff value for the photon energy, 1.35 eV [fact (3)], for this transition is well below the band gap of GaAs. We hold that the only reasonable explanation for a cutoff photon energy well below the band gap is the following. The operative mechanism for the photoexcited transformation must be a local excitation of some part of the defect complex which, provided this electronic energy remains localized for sufficient time, may be transformed into sufficient kinetic energy of one (or more) of the adjacent atoms that it hops (they hop) as required for the transformation. Thus, the photoexcited transformation will be quenched if the final-state energy of the electron that is excited in the localized defect mode becomes so high that it overlaps a band of delocalized states. If that happens, the electron would rapidly be delocalized into the band and swept from the junction taking with it the excess energy that it obtained from the photon and which would be required to drive the transformation. In the same vein, the threshold photon energy is determined by the requirement that the local excitation must be able to transfer sufficient energy to the atom (or atoms) so that it can make the transformation.

Now we note that the empirical value of the cutoff photon energy, 1.35 eV, coincides closely with the energy to deionize the V_{Ga}^- by boosting its electron into the conduction band. To us, this strongly indicates that the operative mechanism is an internal excitation of the V_{Ga}^-. The threshold value of the photon energy is 0.9 eV, as noted in fact (3). According to the ballistic model (BM) for atomic migration,[63,64] the energy required for a V_{Ga} to hop to a nearest-neighbor site, i.e., for an As atom to hop into the V_{Ga}, is 0.96 eV. (The BM value for hopping of a Ga atom, i.e., V_{As} nearest-neighbor hopping is 0.89 eV.) Since we just noted the evidence that it is photoexcitation of a V_{Ga} that is the operative mechanism, it is most reasonable to assume the threshold energy is determined by the requirement that there be enough energy in the local excitation to transfer the required hopping energy to one of the surrounding As atoms. We regard the agreement between the BM estimate for this parameter of 0.96 eV and the empirical value of 0.9 eV as entirely satisfactory.

The nearest-neighbor hopping event leaves the complex in a rather unstable configuration, where a V_{As} has two nearest neighbors that are $\mathrm{As_{Ga}}$'s. It seems clear that the V_{As} will again hop to one of these nearest-neighbor sites in order to annihilate one of these antisite defects. Coulomb repulsion from the original V_{As}^+ will cause the hop illustrated in Fig. 4(d) to be favored so that the O^* configuration will result. (The other option would simply recreate the O configuration.)

We illustrate our model of the $O \rightarrow O^*$ transformation in Fig. 4. The photoexcitation of the V_{Ga} to an excited electronic state

$$V_{\mathrm{Ga}}^- + h\nu \rightarrow (V_{\mathrm{Ga}}^+)^* \tag{11}$$

is shown in Figs. 4(a) and 4(b). Provided that this excitation can transfer sufficient energy into the motion of one of the four nearest-neighboring As atoms, this may induce nearest-neighbor hopping, as illustrated in Fig. 4(c). However, the resulting configuration is unstable and the vacancy again hops to a nearest neighbor. In doing so it may create the O^* configuration (as we propose it) Fig. 4(d), or it might return to the O configuration.

It is worth noting that the cross section for what we identify as the intracenter transition (11) is an order of magnitude greater[65] than the cross section associated with the transformation to the metastable state (10^{-16} and 10^{-17} cm^2, respectively). This is consistent with the reasonable expectation that the probability that sufficient energy will be transferred from the electronic excitation into kinetic energy of one As atom is rather less than unity.

The photoexcitation mechanism described above requires a photon energy greater than that for the first ionization of As/Ga [i.e., $\mathrm{As_{Ga}}(0\rightarrow+)=0.77$ eV]. Also, the second ionization energy of $\mathrm{As_{Ga}}$ [i.e., $\mathrm{As_{Ga}}(+\rightarrow++)+1.00$ eV] is just above the threshold for this photoexcitation mechanism (0.9 eV). Thus, the photoionization process should compete with the internal excitation process that we invoke for the transformation. We expect that the photoionization process would occur with greater frequency. If so, then we must conclude that the rate of decrease of junction capacitance in the PPCQ experiment should be slow, which is indeed what is found experimentally.

Our interpretation of the intracenter transition differs from that of Kaminska *et al.*, who suggest[65,66] that the transition arises from the first ionization of As_{Ga} ($0 \rightarrow +$) and that this final state is resonant in the conduction band. In forming our interpretation, that it is the V_{Ga} that is excited and that the final state is not resonant until the photon energy exceeds 1.35 eV, whereupon the photoexcited transitions ceases, we reasoned as follows. If the final state were resonant in the conduction band, the most probable deexcitation path would be ionization of the excited electron, thermalization of the resultant free electron to the conduction-band minimum, and capture of an equivalent electron from that band-edge distribution by the ionized As_{Ga}. That process is illustrated in Fig. 5. If this were indeed the case, the maximum amount of energy that could be transferred from the intracenter deexcitation process to the atom or atoms that must move in the large lattice relaxation process would be just the recombination energy, 0.77 eV. But, if that were the case, then the photon energy threshold to induce the transformation would also be 0.77 eV, not the 0.9 eV that is observed.

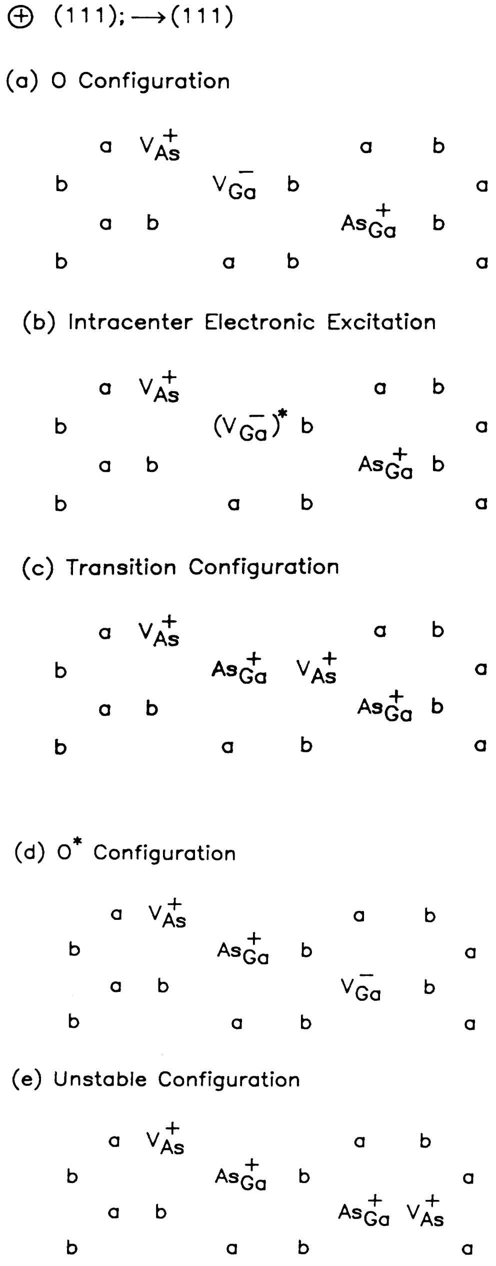

FIG. 4. Steps involved in the $O \rightarrow O^{*}$ transformation responsible for PPCQ: (a) equilibrium configuration, O; (b) intracenter electronic excitations; (c) transition configuration; (d) metastable configuration, O^{*}; and (e) unstable configuration.

Consider now fact (4), the $O^{*} \rightarrow O$ transformation cannot be photoexcited with photons of any energy. In our model the reason that the reverse of the photoexcited $O \rightarrow O^{*}$ transformation will not go is rather obvious. If from the O^{*} we excite the V_{Ga}^{-} to the same local excited state that is operative for the $O \rightarrow O^{*}$ transformation, we might also induce one of the neighboring As atoms to hop into that vacancy and produce a $V_{As}As_{Ga}$ pair. If it is the As atom that is also bonded to the As_{Ga} in the O^{*} configuration that hops, then the unstable configuration of Fig. 4(d) is regained. However, this will reconvert to the O^{*} configuration due to the repulsion of the V_{As}^{+} that was just noted in our description of the $O \rightarrow O^{*}$ transformation. If one of the other As atoms hops into the excited V_{Ga} of the O^{*} configuration, then the configuration shown in Fig. 4(e) will be created. It is easy to see that this configuration is marginally stable at best and tends to return to the O^{*} configuration, Fig. 4(d). An additional

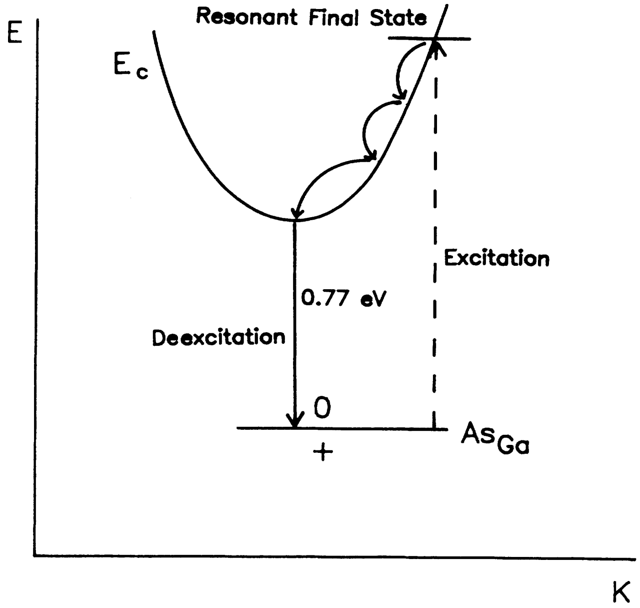

FIG. 5. Most probable deexcitation path if the intracenter electronic excitation be ascribed to the first ionization of As_{Ga} to a final state that be resonant in the conduction band.

reason that the $O^* \rightarrow O$ transition is not observed to be photoexcited may be the following. The $V_{\rm Ga}$ is part of a divacancy in the O configuration but has four As nearest neighbors in the O^* configuration. We suppose that the interaction of a $V_{\rm Ga}$ with the $V_{\rm As}$ serves to broaden and multiply the final states of the electronic transitions for the O configuration to the point that they are effectively continuous. (The local-density theory calculations of Baraff and Schluter seem to support[10] the contention that the $V_{\rm Ga}$ supports several excited states.) A $V_{\rm Ga}$ with no defect on nearest-neighbor sites may have well-separated final-state eigenvalues so that the photoexcitation can occur for only certain, discrete values of the photon energy.

Consider now fact (5), the 0.3-eV value of the thermal activation barrier against the reestablishment of the stable O configuration from the metastable O^* configuration. We have just noted in our explanation of fact (4), that the persistence of O^* at low T in the face of photon irradiation that is fully capable of driving the $O \rightarrow O^*$ transformation is the repulsion between the $V_{\rm As}^+$ at the far end of the complex and the $V_{\rm As}^+$ between the two $\mathrm{As}_{\rm Ga}^+$'s in the unstable, transition, configuration, that is illustrated in Fig. 4(d). A simple point-charge approximation of this Coulomb repulsion at the second nearest-neighbor separation with full, static dielectric screening ($\epsilon = 12$), gives this repulsion to be 0.30 eV. The agreement is better than experimental uncertainty and certainly well within what should be required of such a simple approximation. That this repulsion should define the empirical activation energy against reestablishment of the stable state follows from the fact that thermal excitation is constantly inducing vacancies to hop to nearest-neighbor sites so that both the $O \rightarrow O^*$ and the $O^* \rightarrow O$ transitions are being attempted. While the stability of the O configuration is mandated by its lower total energy, the rate of the net $O^* \rightarrow O$ reaction is determined by this barrier that biases the transition state, Fig. 4(d), toward the O^* configuration.

Consider now fact (6), the $O^* \rightarrow O$ transition can be induced by injection of electrons. If electrons are injected, they will have a probability to neutralize one of the two $V_{\rm As}^+$'s of the transition configuration shown in Fig. 4(c). In either case, this would turn off the Coulomb repulsion that biases the system to return to the metastable O^* configuration rather than the stable O configuration. Therefore, to the extent that electrons may be trapped at the complex in the transition configuration, the otherwise inhibited $O^* \rightarrow O$ transition should become allowed upon electron injection. The preexponential factor in the effective electron capture cross section for this process, 10^{-13} cm^2, is characteristic of the so-called giant cross sections for capture of a carrier in a trap with a Coulomb attractive potential, as between an electron and a $V_{\rm As}^+$, which has been described in the classic papers of Lax.[67,68] The activation energy for this cross section, 0.108 eV, is approximately twice the optic phonon energy in GaAs. It is necessary that the Coulomb potential energy of the interaction be dissipated by phonon emission before the thermal velocity, plus the added velocity due to the Coulomb attraction, carries the electron through the complex and away from the $V_{\rm As}$. Henry and Lang observed[7] this same activation energy and argued that it is a consequence of multiphoton emission concomitant to the trapping process at an attractive center. We therefore conclude that this, and not an Auger deexcitation process, is what occurs at $EL2$'s when carriers are injected into the junction.

Finally, we have fact (7): the complex returns to its normal charge state for the prevailing E_F after the electron-injection-induced transformation just described. This is a natural consequence of the transformation to the O configuration, which has the same component point defects as does the O^* configuration. The electron that was trapped to the $V_{\rm As}$ during the transformation will spontaneously be reemitted thermally because of the divacancy Coulomb interaction, which will push the $V_{\rm As}$ level closer to E_c than its position for an isolated $V_{\rm As}$.

V. OTHER EXPERIMENTS AND OTHER MODELS

We have shown that the divacancy plus As—on—Ga-site antisite model for $EL2$ is consistent with the critical seven facts regarding its electrical and optical properties that have been gleaned from a vast literature. We now consider other experiments that bear on the identity of $EL2$ and other models of the complex that have been proposed.

A very powerful tool for the identification of point defect complexes is thermochemical analysis of the relation between processing conditions and defect concentrations.[60,69] Over a period more than ten years, Zou and co-workers have developed[9] the case that $EL2$ is a complex consisting of one each of $V_{\rm Ga}$, $V_{\rm As}$, and $\mathrm{As}_{\rm Ga}$, which are the same constituents as in our model of the stable configuration. (Thermochemical analysis does not provide information regarding the configuration of the constituents.) In making their case, Zou *et al.* analyzed a wide range of data from many sources regarding the effects of strain, dislocations, temperature, and stoichiometry on $EL2$ concentrations. The reader may refer to Ref. 9 for a review of that analysis.

However, one of the present authors (J.A.V.V.) has long advocated a somewhat different model[5,22,25,42,69] of $EL2$, which is shown in Fig. 6 and contains two $V_{\rm Ga}$ plus $\mathrm{As}_{\rm Ga}$ in its stable configuration. Thus, it contains one more $V_{\rm Ga}$ and one less $V_{\rm As}$ than does the present model, that is supported also by the thermochemical analysis of Zou *et al.* In this model also the actuator of the transformation is nearest-neighbor hopping of a $V_{\rm Ga}$, as is now supported by the elaborate calculations[10] of Baraff and Schluter. The model of Fig. 6 was a prediction,[5] made when there was almost no data, of what should be the most numerous complexes formed as the crystal cools to room temperature from the isolated point defects that are grown in when the crystal is formed. It is not a simple matter to distinguish between the two models,[69] but we now prefer the present model largely because it gives a logical explanation of fact (4), the inability to phototransform the O^* configuration, and we see no good explanation for this fact within the model of Fig. 6.

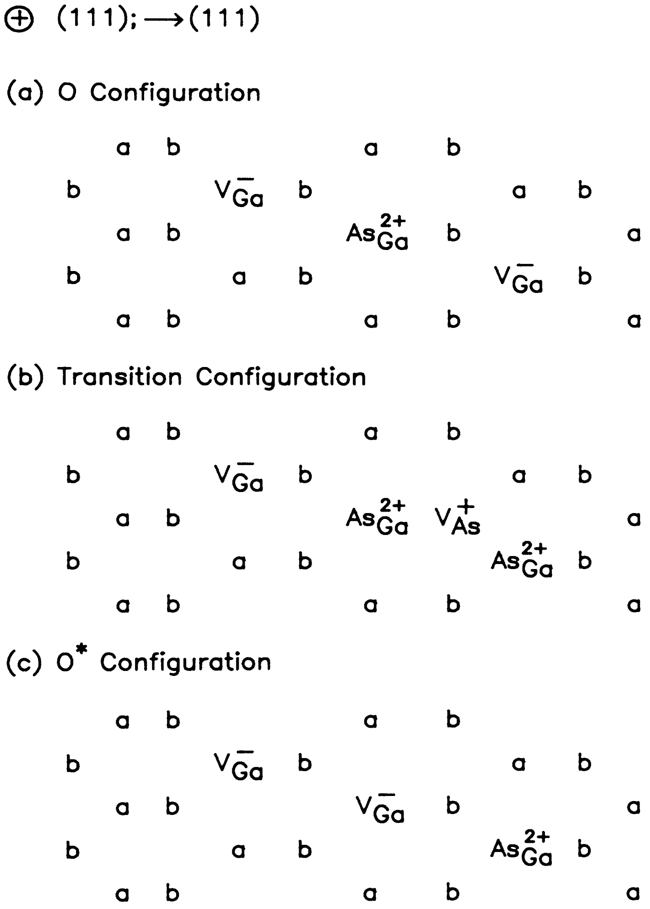

FIG. 6. Alternative model for $EL2$ previously proposed by Van Vechten in Ref. 5: (a) O configuration; (b) transition configuration; and (c) O^* configuration.

The model of Fig. 6 was supported by high-resolution, transmission electron microscopy (TEM) studies[2,3] which found 10^{17} cm^{-3} (typical $EL2$ density) defect complexes in the purest available GaAs, which were "rodlike," one atom in diameter but between 1.0 and 2.0 nm in length and oriented in $\langle 110 \rangle$ directions. The model of Fig. 6 provides a straight rod, 1.2 nm long, oriented in $\langle 110 \rangle$ directions, because the point defects occupy three sites on one sublattice and the two V_{Ga}^{-}'s on either side of the $\mathrm{As}_{\mathrm{Ga}}^{2}$ repel each other. The present model of $EL2$ is not quite as long as that of Fig. 6 and is not quite straight. In the absence of a substantial lattice distortion about the divacancy, the complex would bend from a $\langle 110 \rangle$ direction to a $\langle 111 \rangle$ direction at the V_{Ga}. To support the present model over that of Fig. 6 in the face of the direct lattice imaging TEM study[2,3] of Van der Sande and Peters, we must either conclude that they mistook a bent rod for a straight one or that there is indeed a substantial lattice distortion about the divacancy. The conclusion that there may in fact be a sufficient lattice distortion is supported by the theoretical work[10] of Baraff and Schluter, who predict a displacement of an As atom about a single V_{Ga} of as much as 30% of the bond length in some charge states. We will return to the question of lattice distortions below in a discussion of recent electron-nuclear double optical resonance (ENDOR) studies.[70]

The positron-annihilation measurements[18] of Dannefaer and Kerr provide very strong evidence that the $EL2$ complex contains two vacancies but, as yet, they do not clearly distinguish between the models of Figs. 1 and 6. A great utility of the positron-annihilation experiment in this context is that positron lifetimes are sensitive to free volume, as in vacancies and vacancy complexes, but not to interstitials, impurities, or antisite defects. The method cannot confuse vacancies with any of these other point defects, which is not true of most other methods. The positron decay spectrum[18] of GaAs is found to contain exponential components with characteristic lifetimes of 220 ps for decay in the bulk states, 265 ps for decay in single vacancies, and 295 ps for decay in a two-vacancy complex. These characteristic lifetimes may be compared with values for Si of 221 ps for the bulk, 271 ps for single vacancies, and 320 ps for decay in a divacancy.[71] The concentration of the two-vacancy complexes in intrinsic GaAs was determined[18] to be 1×10^{17} cm^{-3}, which is within experimental error the same as the electrically determined $EL2$ concentration and the concentration of rod defects observed in TEM direct lattice imaging. It is difficult to believe the positron signal comes from anything other than $EL2$. However, while the bulk and single-vacancy characteristic lifetimes are very similar in Si and in GaAs, the two-vacancy complex in intrinsic GaAs is rather less than for the divacancy in Si. This could be ascribed to the large lattice distortion at the divacancy in GaAs invoked to square the model of Fig. 1 with the TEM. It could be taken as support for the model of Fig. 6, where the positron wave function would be spread between two V_{Ga}'s and would annihilate at a rate not much faster than in the single vacancy as it would if there were a divacancy.

This leads us to propose what we regard as one further crucial experiment regarding the exact identification of $EL2$. It is possible to distinguish between the models of Figs. 1 and 6 by repeating the positron experiment when the $EL2$'s are in the O^* configuration and comparing the magnitudes of the two-vacancy characteristic lifetime to the values already obtained with the O configuration. This can be done by holding the sample at low T and illuminating it during the positron experiment. If the model of Fig. 1 is correct, the lifetime must go distinctly down because the divacancy of the O configuration is split in the O^* configuration. If the model of Fig. 6 is correct, the lifetime should go up because the two vacancies move closer together.

One other model of $EL2$ must now be discussed, that of von Bardeleben, Stievenard, Bourgoin, and Huber (BSBH), which proposes[72] that the complex consists of a $\mathrm{As}_{\mathrm{Ga}}$ plus an As interstitial, As_i. We feel that the positron annihilation results[18] clearly exclude this hypothesis. However, some support has been given to this model recently by ENDOR studies.[70] As mentioned above, previous EPR, ENDOR, and other measurements clearly indicate that $EL2$ contains an $\mathrm{As}_{\mathrm{Ga}}$ and that is has four As nearest neighbors in the *O* configuration. (Note that both the models of Figs. 1 and 6 satisfy that requirement.) The recent ENDOR data are interpreted to indicate an interaction of the $\mathrm{As_{Ga}}$ nucleus with a fifth As in an antibonding, $\langle 111 \rangle$ direction. The As_i of the BSBH model is in the tetrahedral interstitial position in the antibonding $\langle 111 \rangle$ direction, as required for this interpretation of the ENDOR data.

We feel that the model of Fig. 1 can account for the recent ENDOR data. If there were no lattice distortion about the divacancy, then in the *O* configuration, the $\mathrm{As_{Ga}}$ would have a normal $\mathrm{As_{As}}$ in the unique antibonding $\langle 111 \rangle$ direction toward the divacancy at its fifth-nearest-neighbor site. The interaction between the $\mathrm{As_{Ga}}$ and that particular $\mathrm{As_{As}}$ nucleus would be far stronger than normal both because of the void created by the divacancy and because of the negative charge spilling into that void to form the wave function of the electron bound in the acceptor state of the V_{Ga}. Thus, the fifth As nucleus that interacts with the $\mathrm{As_{Ga}}$ could simply be the normal $\mathrm{As_{As}}$ in that direction. Alternatively, if there is indeed a major lattice distortion about the V_{Ga} of the *O* configuration, then one of the As atoms participating in that distortion may move close enough to the antibonding axis to account for the ENDOR result. In the latter case, the distinction to the BSBH model would be partly semantic; what we, and Baraff and Schluter, regard as an As atom participating in a major lattice distortion about a V_{Ga} could also be called an As interstitial participating in the complex. This latter hypothesis might also serve to square the model of Fig. 6. with the ENDOR data.

Finally, we address the issue that there is a growing consensus that *EL* 2 is not a single, unique defect complex but is actually a family of similar defects. One explanation for this is the hypothesis that both the complex of Fig. 1 and that of Fig. 6 are present in substantial numbers and that their behavior is very similar. It is difficult to argue that both should not occur, as was indicated in Ref. 5. As was also argued in Ref. 5, as well as Refs. 22, 25, 42, and 69, an inevitable consequence of the nearest-neighbor hopping mode of single-vacancy migration is the presence of variable numbers of antistructure pairs, $\mathrm{As_{Ga}Ga_{As}}$, in the vicinity of complexes formed from vacancies. What we regard to be the central core of *EL* 2 is shown in Fig. 7(a). Another configuration involving an antistructure pair about the "father of the *EL* 2 family," according to the model of Fig. 1, is indicated in Fig. 7(b). These antistructure pairs tend to be neutral and have a moderate effect on ionization levels. They should have much more effect on cross sections. This is in general accord with observations of the distinctions among the members of the *EL* 2 family.[23,24] Another example of the *EL* 2 family is shown in Fig. 7(c) and involves a Si donor. Because the divacancy is neutral, we expect very little electrostatic pairing between *EL* 2 and the Si donor.

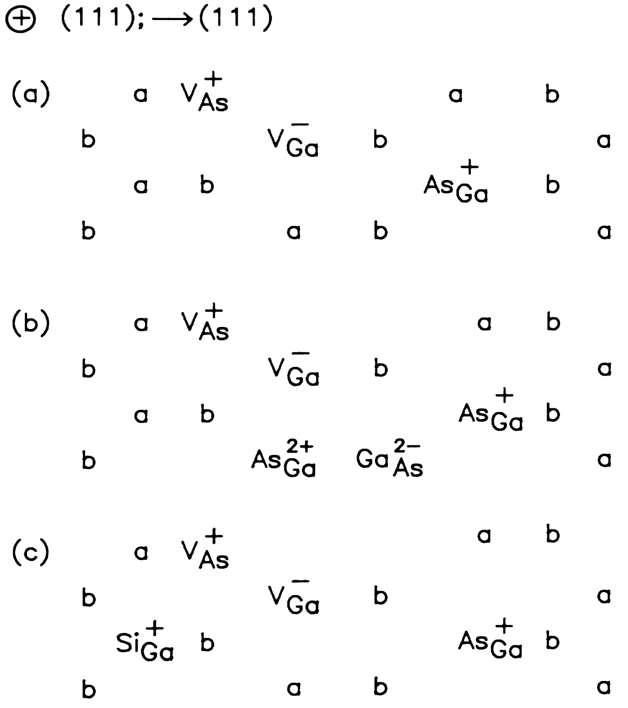

FIG. 7. Examples of other members of the *EL* 2 family: (a) the core; (b) the core and one antistructure pair in one of many configurations; (c) the core and an ionized Si donor.

VI. SUMMARY

We have shown that the model of Fig. 1 is capable of accounting in detail for all electrical and optical characteristics of *EL* 2 that we have been able to glean from the vast literature. The same model is in accord with a vast literature of thermochemical data.[9] It is also in accord with positron-annihilation results that seem to exclude all models that do not contain two vacancies.[18] We have argued that it is not inconsistent with either TEM or ENDOR data. It is consistent with the best available theoretical conclusions. Finally, we have proposed a critical experiment using positron annihilation to distinguish between the model of Fig. 1 and that of Fig. 6, which otherwise would behave in a very similar fashion in most experiments. While real samples likely contain substantial numbers of the complex shown in Fig. 6, we feel there is now a very strong case for identifying that of Fig. 1 as the "father of the *EL* 2 family."

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