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New EL2 structural model based on the observation of two sequential photoquenching processes

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Abstract. The $EL2$ photoquenching kinetics has been studied by measuring the transmittance of a 1.05 μm laser beam as a function of time. The photoquenching curve presents two time constants that we associate to a two-stage transition through an intermediate state to the final metastable state. This was verified by measuring the temperature dependence of the transmittance with the sample prepared in a stage of partial photoquenching. Besides being also metastable, we show that the intermediate state has an optical absorption-cross section that is temperature dependent. In order to explain the existence of such a state, we propose a new structural model for the $EL2$ in which the motion of an arsenic atom finds two metastable positions after photoexcitation. Our model is based on a mixture of features derived from the models proposed by Chadi and Chang [1] and more recently by Fukuyama et al. [2].

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1 Introduction

The $EL2$ is a gallium arsenide donor native defect [3] which has the property of becoming optically inactive to photons of ∼1.18 eV after undergoing a photoinduced internal transition to its metastable state, $EL2^*$, an effect called photoquenching [4,5]. Besides the quenching of the optical absorption, the transition to this metastable state also leads to the quenching of a number of other physical properties. A persistent metastable state can only be achieved when the sample is cooled below 90 K and excited with photons of energy between 1.0 and 1.3 eV [6]. The reverse path, i.e., the total recovery of the $EL2$ from the metastable state back to the normal state is achieved when the sample is heated above 120 K [7]. This reverse path is also known as thermal recovery. The recovery can also be induced by light with 0.75–1.0 eV photons and by the application of strong electric fields. [8–10] Various experimental methods, such as infrared absorption [4], photocurrent [11], photocapacitance [6] and electron paramagnetic resonance [12], among others [13], have been applied to the study of the $EL2$ photoquenching. Furthermore, experiments under hydrostatic and uniaxial pressure have indicated that the photoquenching $EL2^0 \rightarrow EL2^*$ transition is a plastic deformation of its structure in which the symmetry changes from tetrahedral to trigonal [14].

Despite the intensive and hard work done since the discovery of the $EL2$ center, up to this date its internal microscopic structure is still a matter of debate. Due to the location of the energy level, the first model considered it to be an oxygen impurity localized at a gallium site [13]. This model was discarded after the results of SIMS (Secondary Ion Mass Spectroscopy) [15] appointed a disagreement between the $EL2$ and oxygen concentrations. In addition, it should be expected that an increase of Ga_2O_3 at the growth solution would result in an increase of the $EL2$ concentration, but that have not been observed. The dependence of the $EL2$ concentration on stoichiometric parameters has shown that the $EL2$ is indeed a lattice defect, a fact accepted today. Holmes et al. [16] showed that the $EL2$ concentration increases when the growth solution is arsenic-rich and, for this reason, the $EL2$ was believed to be to a gallium vacancy (V_{Ga}). The gallium vacancy allows the possibility of motion of an arsenic atom from its lattice site towards the vacancy site, creating an arsenic antisite (As_{Ga}) and leaving behind an arsenic vacancy (V_{As}). Baraff e Schlüter [17] have studied theoretically the $V_{Ga} \rightarrow As_{Ga} + V_{As}$ model, and have calculated its configurational coordinate diagram. The calculation, however, did not predict all the observed properties and was considered incomplete. The main component of $EL2$ structure, a consensus nowadays, has been identified from EPR (Electron Paramagnetic Resonance) experiments. The first observation of the $EL2$ by EPR was done

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by Wagner et al. [12], and the signal was assigned to the charged arsenic antisite (As^{+}_{Ga}). The signal changes produced by photoexcitation had the same photoquenching properties of the $EL2$, but the temperature dependence of the thermal recovery was different.

During the 1980's and 1990's, many authors supported the isolated As_{Ga} model, while others, such as Meyer et al. [18], defended the association of an arsenic antisite and an arsenic interstitial (As_{Ga} – As_i).

Schick et al. [19] have done first principles calculation of a number of defect structures and verified that the As_i has an exceedingly large formation energy. Therefore, its presence close to an As_{Ga} after annealing is highly improbable.

Chadi and Chang [1], calculated that the isolated As_{Ga} could undergo a lattice relaxation with neighboring atoms. This could result in a new state with T_2 symmetry below the conduction band. He proposed that this new state was the metastable $EL2$.

For a number of years the community has been trying to associate the $EL2$ to the As_{Ga}, but today, evidences are growing that other associated centers participate in the metastable transition. Jimenez et al. [20] presented experimental evidence of an actuator level that works as a trigger to initiate the metastable transition.

Recently, Fukuyama et al. [2] have suggested that the $EL2$ was a three center complex defect: an As_{Ga}, a Ga_{As} and a V_{As}. In their model, the $EL2$ structure is stabilized by a coulomb attraction between the positively charged As_{Ga} site and the negatively charged Ga_{As} site that prevents the As atom to move towards the V_{As}, along the $\langle 111 \rangle$ direction. The transition to the metastable state is initiated by a photoionization which leads to a charge transfer between these sites that destroys the coulomb attraction, allowing the As atom to move to an interstitial position along the $\langle 111 \rangle$, as calculated by Baraff et al. [17]. The model explains naturally the presence of the actuator level which is assumed to be correlated with the Ga_{As}. The main criticism, perhaps, is that the overall structure lacks an As atom, which is difficult to justify since the concentration of $EL2$ is favored in As-rich samples. Despite not giving this criticism a deeper consideration the authors argued that Arsenic vacancies have been observed in As-rich samples.

This brief review shows that, in one hand, all models proposed so far are capable of explaining a number of properties, but, on the other hand, all have problems in some other aspect. Until now, a consensus on the $EL2$ microscopic structure has not been firmly established. The only consensus, perhaps, is that the As_{Ga} is an active component of the $EL2$ center.

In this work we present experimental evidence that the photoquenching process is composed of two subsequent transitions involving another intermediate metastable state. In order to explain our observations we propose a model which involves a mixture of features belonging to the As_{Ga} model proposed by Chadi and Chang [1], and to the three-center-complex proposed by Fukuyama et al. [2].

2 Experiment

The sample investigated in this work consisted of a nominally undoped $1 \times 1 \times 2$ cm^3 GaAs block containing 1.45×10^{16} cm^{-3} $EL2$ centers, as determined by optical absorption spectroscopy [21], and a carbon concentration of the order of $1{\times}10^{16}$ cm^{-3}, estimated from FFT-IR absorption [22]. The spectral analysis revealed that essentially all $EL2$ were in the normal state at room temperature. The lack of an appreciable concentration of $EL2^{+}$, together with a resistivity of $6.2{\times}10^{6}$ Ω cm may be an indication of a residual n-type doping. The experiment consisted of cooling down the sample to 30 K in the dark and, after a 10 minute stabilization period, monitoring the temporal change in transmittance when it was subjected to a $\lambda = 1.05$ μm cw radiation from an InGaAs laser. The beam was focused at the sample surface and the intensity was adjusted by the insertion of calibrated filters. The temperature was controlled by placing the sample in a cold finger liquid-helium cryostat, and was monitored by two sensors mounted on the top and bottom sides of the sample. Since the beam was positioned at the center of sample, the temperature at the irradiated spot was estimated to be the average of these two sensor readings. The error in this estimation is less than 4 K, which is half of the maximum temperature difference registered across the sample during a heating/cooling cycle.

3 Results

Figure 1 shows the well known photoquenching effect: the transmittance increases as the $EL2$ goes to the metastable state by the exposure to the 1.05 μm excitation. In this figure, the open circle indicates that, after this point, the laser power has been reduced by a factor of ∼400 to probe the transmittance without affecting the state of the system during the time-frame of the experiment. The solid circle indicates the point at which the filters have been changed in order to increase the power to allow the metastable state to be significantly populated, therefore inducing the photoquenching effect. The results show that while at the fundamental $EL2^{0}$ state, the normalized transmittance (normalized to the maximum value when absorption is neglected and multiple reflections taken into account, i.e, to the value of Eq. (4) with $\alpha = 0$) was 0.23 and after 1100 s of strong illumination it achieves nearly 0.95. In addition, we have checked that this final state is persistent by turning off the laser during 1 h while keeping the sample at low temperature. After this period of time no change in transmittance was detected.

In all photoquenching experiments we have carried out using this setup we noticed that the photoquenching curve presented two-time constants: an initial fast rise up to about 70% transmittance, followed by a much slower rise from then on up to about 97%. Our initial guess was that this behavior was an optical artifact, since the focused beam is inhomogeneous and the beam center should achieve maximum transparency faster than the edges. However, by varying the temperature, as discussed

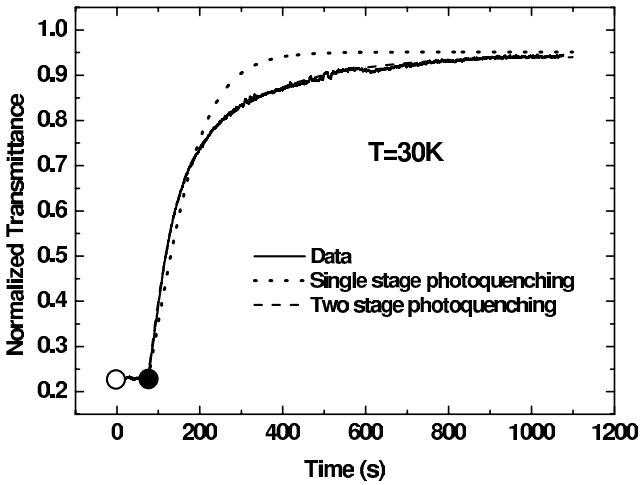

Fig. 1. Photoquenching curve of the $EL2$ at 30 K. The open circle indicates the use of a set of filters such as to keep a very low light intensity to probe the transmittance without affecting the state of the system. The filled circle indicates the use of higher power to induce the photoquenching effect. The increase of the transmittance reveals the $EL2$ transition from the normal to the optically inactive metastable state. The dotted curve is the prediction using a single stage photoquenching model, whereas the dashed curve, which is barely seen since it runs on top of the data, refers to the two-stage photoquenching model (see text).

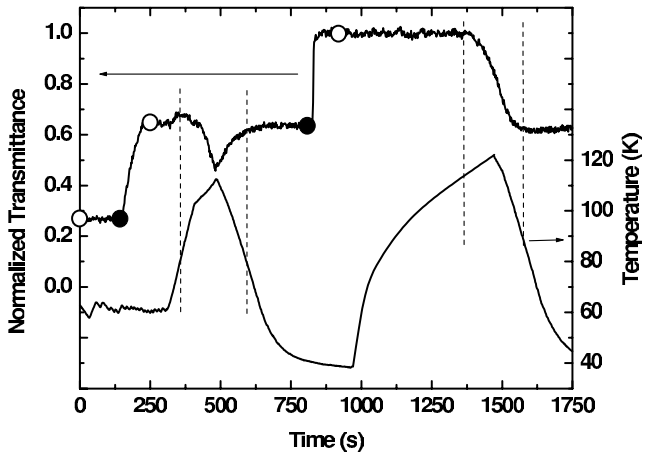

Fig. 2. Optical transmittance changes induced by temperature variation at different photoquenching stages. The open circle indicates the use of a set of filters such as to keep a very low light intensity to probe the transmittance without affecting the state of the system. The filled circle indicates the use of higher power to induce the photoquenching effect. The $EL2$ were initially in the normal state, then they were partially photoquenched until the transmittance reached 65%. The sample was then heated up to 112 K and quickly cooled down to about 40 K. Next, the photoquenching was completed and the temperature was increased again, up to 120 K, and quickly cooled down below 50 K. See discussion in the text.

below, we were convinced that these two time constants have a different physical origin.

Figure 2 summarizes the experiment in which we varied the temperature after having prepared the system at two different photoquenching stages: the first one with an incomplete photoquenching (transmittance nearly 65%), and the second one when the photoquenching was complete (transmittance nearly 100%). Again, the open circles in the figure indicate the use of probing light, whereas the filled circles indicate the use of stronger light to induce the photoquenching. The second open circle, at $t \sim 250$ s, marks the beginning of a heating/cooling cycle with an incomplete photoquenching. Notice that as the temperature rises beyond 100 K the transmittance begins to drop. Such a drop could indicate that a recovery process had started, with the metastable centers returning to the normal state. However, as we cooled down the sample, while still keeping a low probe light intensity, the transmittance nearly recovered its original value. It is unlikely that the recovered centers returned to the metastable state after cooling. It is certainly more likely that the metastable population was practically unchanged during the heating/cooling cycle, and the changes in transmittance were due to a temperature dependent absorption cross-section.

In a separate experiment we monitored the transmittance with low light intensity while cooling the sample from 300 K down to 30 K and only a monotonic change of 4% in transmittance was seen. Therefore, the absorption of the $EL2$ in the normal state ($EL2^0$) is essentially temperature independent. If the partial photoquenching state simply corresponds to a mixture of $EL2^0$, whose absorption is temperature independent, and $EL2^*$, whose absorption is bleached, then the absorption of the mixture should also be temperature independent, but that was not seen in the experiment with partial photoquenching. These observations lead to the conclusion that the temperature dependence of the absorption cross-section observed with partial photoquenching is an intrinsic property of this state.

Next, we used intense light to achieve the state of maximum transparency (97%) corresponding to total photoquenching (second filled circle in Fig. 2), and after reducing the power back to probe light intensity (second open circle in Fig. 2) we started another heating/cooling cycle. Notice that, as before, the transmittance dropped when the sample was heated up to 120 K, but this time it did not recover after the sample was cooled down to 40 K. This behavior corresponds to the usual thermal recovery process. Therefore we are convinced that the state of maximum transparency corresponds to the true metastable state. The stage of partial photoquenching, therefore, is not simply a state of superposition of normal and metastable centers, but in fact, it contains a different intermediate state.

4 The two-stage photoquenching model

Based on the experimental results discussed above we are convinced of the existence of an intermediate state, which we have called $EL2^*_a$, through which the system passes to achieve the final metastable state that we have called

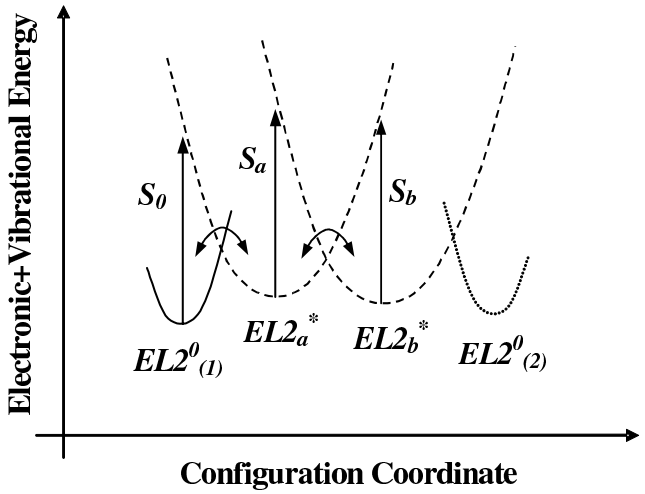

Fig. 3. Configurational coordinate diagram proposed for the $EL2$ system, incorporating an intermediate state. The system can pass from the intermediate state $EL2_a^*$ to the final $EL2_b^*$ by optical absorption or by thermal excitation over an energy barrier. There are two different configurations for the normal state, labeled $EL2_{(1)}^0$ and $EL2_{(2)}^0$ (see Sect. 5). The first one is considered the true normal state. The curves corresponding to the conduction and valence bands were omitted for clarity.

$EL2_b^*$. In order to discuss the optical and thermal properties of this state we propose the configurational coordinate diagram depicted in Figure 3.

The system, initially in the normal state $EL2_{(1)}^0$, is excited by photon absorption with a cross-section S_0 to the intermediate metastable state $EL2_a^*$ and, from this state it can be photoexcited to the final metastable state $EL2_b^*$, with an optical cross-section S_a. The structure discussed in Section 5 allows a different configuration for the normal state labeled $EL2_{(2)}^0$ that will be discussed later. Since there is a residual absorption after the final photoquenching state is reached, we have associated it to an optically induced recovery from the metastable state $EL2_b^* \rightarrow EL2_a^*$ with an optical cross-section S_b. Considering that the concentration of centers in the normal, intermediate metastable and final metastable $EL2$ states are given by N_0, N_a and N_b, respectively, the kinetics of photoquenching can be described by the following set of equations:

$$\frac{dN_a}{dt} = \frac{I}{h\nu}\big[(S_{n,0}+S_0)N_0 - (S_{n,a}+S_a)N_a + S_bN_b\big], \quad (1)$$

$$\frac{dN_b}{dt} = \frac{I}{h\nu}\big[S_aN_a - S_bN_b\big], \quad (2)$$

$$N^0 = N_T - N_a - N_b. \quad (3)$$

$$\alpha = (S_{n,0}+S_0)N_0 + (S_{n,a}+S_a)N_a + S_bN_b, \quad (4)$$

$$T = \frac{(1-R)^2\ e^{-\alpha L}}{1-R^2\ e^{-2\alpha L}}. \quad (5)$$

where N_T is the total $EL2$ concentration (1.45×10^{16} cm^{-3}), α is the absorption coefficient, $R = 0.28$ is the sample surface reflectivity, and T is the transmittance. We have included in equations (1) and (4) the absorption cross-sections of photoionization of the fundamental ($S_{n,0}$) and intermediate metastable ($S_{n,a}$) states. The dashed curve in Figure 1, which runs essentially on top of the experimental data, was obtained with the following parameters: $S_0 = 8.0 \times 10^{-17}$ cm^2, $S_a = 1.5 \times 10^{-17}$ cm^2, $S_b = 1.5 \times 10^{-18}$ cm^2, $S_{n,0} = 2 \times 10^{-17}$ cm^2 and $S_{n,a} = 4 \times 10^{-18}$ cm^2, at 30K. In order to explain the transmittance drop around $t \sim 500$ s in Figure 2, we assume that S_a increases when the temperature exceeds 80 K. Again, we emphasize that it is unlikely that there is a significant population change in this heating/cooling process. The temperature dependence of the absorption cross-section above 80 K suggests a strong coupling with optical phonons.

The cross-sections obtained with the fitting, indicate that in the final steady-state configuration, about 10% of the centers still remain in the intermediate state while the remaining 90% are found in the final metastable state (at 30 K). This is essentially dictated by the ratio of the cross-sections S_a and S_b. The configurational coordinate diagram proposed opens up the possibility of a thermal excitation from the intermediate to the final state. However, if the energy barrier between the two states is larger than 0.34 eV, which is the known barrier between the final metastable state and the normal state, we would not be able to observe a recovery to the original configuration $EL2_{(1)}^0$, since it would be more probable a transition to the alternative configuration $EL2_{(2)}^0$. On the other hand, if the barrier is lower than 0.34 eV, a thermal excitation from the intermediate to the final metastable state should be possible to observe. As a matter of fact, we have noticed during the heating process in some of our thermal recovery experiments, that the transmittance sometimes rises slightly above the bleached level (95–97%) just before the thermal recovery process begins. Such an increase could be the signature of this thermal excitation.

5 The new structure for the EL2 complex

As mentioned in the introduction, the existing $EL2$ models fail in some aspect when compared with accepted experimental data. Of the models proposed so far, the isolated As_{Ga} model is the simplest that possesses most of the desired properties. It lacks, however, the actuator mechanism that triggers the transition to the metastable state. As said in the introduction, the three-center-complex proposed by Fukuyama et al., resolved this problem with a gallium antisite (Ga_{As}) which stabilizes the structure with a coulomb interaction with the neighboring As_{Ga}. Neither of these models, however, display two metastable positions as required by our experimental data, and the latter model still has the problem of having a Ga-rich stoichiometry. In our model, we have tried to keep the main features of these two models and yet generating a structure with two different configurations for the metastable state, as discussed below.

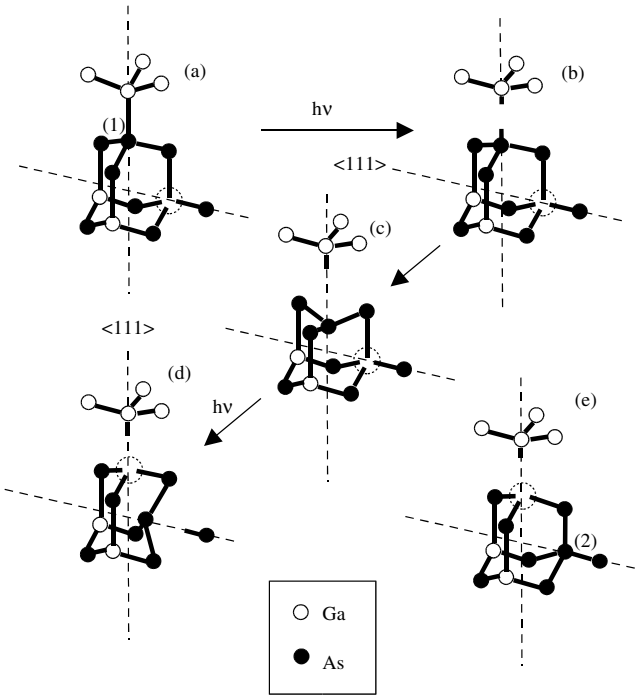

Fig. 4. Different stages of the $EL2$ center. (a) The normal $EL2^0$; (b) after photoexcitation there is a charge transfer between the Ga^{-}_{As} and the As^{+}_{Ga} that leads to the Ga_{As}-As_{Ga}; (c) the structure relaxes with the motion of the As atom and the flipping down of the three remaining bonds; (d) after a new photoexcitation, the atom migrates to an equivalent site along a different $\langle 111 \rangle$ direction towards the V_{Ga}, and rearranges the bonds with the nearest neighbor atoms of this site. This corresponds to the motion of the vacancy to the original As_{Ga}; (e) this configuration may be one of the possibilities of a thermal recovery. The other configuration is the reverse path and the choice between the two will depend on the energy barriers.

The basic $EL2^0$ state in our proposal is shown in Figure 4a. The As_{Ga}, initially in the position labeled as (1), is responsible for the metastability, whereas the Ga_{As} controls the transition to the metastable state via the coulomb interaction with the neighboring As_{Ga}. Instead of a V_{As}, as proposed by Fukuyama et al., our structure incorporates a V_{Ga}, which is more easily justified considering that it leads to an As-rich structure. Also V_{Ga} are more likely to be found in n-type samples compared with isolated As_{Ga} [23]. In the following we have labeled this state as $EL2^0_{(1)}$ to differentiate it from the similar state $EL2^0_{(2)}$ depicted in Figure 4e, where there the As_{Ga} is the position labeled as (2). As already proposed [2], the transition to the metastable state begins with a photoexcitation of the As_{Ga} with a charge transfer to the Ga_{As}, resulting in the breaking of the As_{Ga}-Ga_{As} bond (Fig. 4b). This effect loosens the As_{Ga} atom and, similarly to what has been calculated for the isolated As_{Ga}, the excited states relaxes via an Jahn-Teller effect to the configuration shown in Figure. 4c [1,24]. Next, we propose that, under a new photoexcitation, a further migration of the As atom is possible. It moves to a position along another $\langle 111 \rangle$ direction, towards the V_{Ga}. The bonds are then reestablished with the nearest neighbor atoms of this site and the released ones constitute a new V_{Ga} site, located where the As_{Ga} was originally. A question now arises about the thermal recovery. As the temperature rises, the As atom in this latter configuration has two choices: either it makes the reverse path back to (a) ($EL2^0_{(1)}$) or it relaxes to the position described by (e) ($EL2^0_{(2)}$) in Figure 4. The choice will depend on the energy barriers of both ways. The known 0.34 eV energy barrier for the $EL2$ recovery, in the isolated As_{Ga} model, is explained as the energy cost to "squeeze" the As atom through the space formed by three other As atoms in the structure [24]. In this case we should expect the energy barrier between the configurations (d) and (e) ($EL2^*_b \rightarrow EL2^0_{(2)}$) in Figure 4 to be approximately the same to return from (c) to (b) ($EL2^*_a \rightarrow EL2^0_{(1)}$). These two values should be slightly different since the electronic environments of these configurations are not equal. Also, we expect the barrier between the two metastable configurations (d) and (c) ($EL2^*_b \rightarrow EL2^*_a$) to be less than 0.34 eV because, otherwise, the system would not be able to undergo a thermal recovery back to the original state, ending up in the configuration (e) of Figure 4 ($EL2^0_{(2)}$).

The two possibilities for the As_{Ga} in this structure can lead to two final states during the recovery in a non-adiabatic heating process. This may explain why the properties of system are so dependent on its thermal history. The initial conditions can be reestablished by heating the system up to 300 K for a prolonged time. Therefore we expect the final configuration (e) ($EL2^0_{(2)}$) to have a higher energy than the configuration (a) ($EL2^0_{(1)}$) to where the system relaxes at high temperatures. These conclusions are summarized in the configurational coordinate diagram in Figure 3.

6 Conclusion

We have observed two time constants in our photoquenching experiments that have been fit by assuming two sequentially accessed metastable states. The experiments motivated the proposal of a new microscopic structure through which an arsenic atom moves in going from the normal state to an intermediate metastable state, and from this to a final metastable state when optically excited. The structure allows for two stable and slightly different metastable configurations. The model incorporates the following aspects of previous models which are worth mentioning: 1) a Ga_{As}, which responds for the actuator level reported in the literature; 2) the coulomb coupling between an As_{Ga} and Ga_{As} that is part of the actuator mechanism, as proposed by Fukuyama et al. [2]; 3) the mechanism of metastability described by Chadi and Chang [1], including the barrier 0.34 eV barrier for the thermal recovery; 4) a V_{Ga} that makes the structure As-rich; and 5) the possibility of a different and more energetic $EL2^0$ structure that helps in explaining the multiplicity experimental results of experiments carried out in far from thermal equilibrium conditions.

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