

# Distinction between the Poole-Frenkel and tunneling models of electric-field-stimulated carrier emission from deep levels in semiconductors

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The enhancement of the emission rate of charge carriers from deep-level defects in electric field is routinely used to determine the charge state of the defects. However, only a limited number of defects can be satisfactorily described by the Poole-Frenkel theory. An electric field dependence different from that expected from the Poole-Frenkel theory has been repeatedly reported in the literature, and no unambiguous identification of the charge state of the defect could be made. In this article, the electric field dependencies of emission of carriers from DX centers in  $\text{Al}_x\text{Ga}_{1-x}\text{As:Te}$ , Cu pairs in silicon, and Ge:Hg have been studied applying static and terahertz electric fields, and analyzed by using the models of Poole-Frenkel and phonon assisted tunneling. It is shown that phonon assisted tunneling and Poole-Frenkel emission are two competitive mechanisms of enhancement of emission of carriers, and their relative contribution is determined by the charge state of the defect and by the electric-field strength. At high-electric field strengths carrier emission is dominated by tunneling independently of the charge state of the impurity. For neutral impurities, where Poole-Frenkel lowering of the emission barrier does not occur, the phonon assisted tunneling model describes well the experimental data also in the low-field region. For charged impurities the transition from phonon assisted tunneling at high fields to Poole-Frenkel effect at low fields can be traced back. It is suggested that the Poole-Frenkel and tunneling models can be distinguished by plotting logarithm of the emission rate against the square root or against the square of the electric field, respectively. This analysis enables one to unambiguously determine the charge state of a deep-level defect.

## II. INTRODUCTION

The standard method to detect and analyze deep centers, in particular to determine binding energies, is deep-level transient spectroscopy (DLTS), which is applied in various modifications. However, DLTS does not allow to determine the charge state of a deep-level defect directly. The field-enhanced emission of carriers is frequently taken as an indication that the defect is ionized after the emission, and that the sign of its charge state is opposite to that of the emitted carrier. However, this criterion may be misleading. Generally speaking, the application of strong electric fields to semiconductors with deep centers yields stimulation of ionization or capture processes due to the Poole-Frenkel effect, the phonon-assisted tunneling, and the direct tunneling. The Poole-Frenkel effect occurs for charged impurities only, whereas phonon assisted tunneling (and direct tunneling in the strong electric fields) is possible for impurities in all charge states. Hence, carrier emission rate can increase in an electric field due to carrier tunneling even in the case of a neutral defect. In this paper, we present a method to unambiguously distinguish between the Poole-Frenkel and phonon assisted tunneling mechanisms of field-enhanced carrier emission, and report its application to several well-known defects in silicon, germanium, and  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ .

## III. IONIZATION OF DEEP CENTERS BY ELECTRIC FIELDS

The experimental investigations of carrier emission as a function of the electric field performed in the past may be classified in two groups. In one group the emission has been studied by applying field strengths below 1 kV/cm mainly on bulk samples. In this low field case the electric field stimulation of thermal emission has been attributed to the Poole-Frenkel effect which is caused by the lowering of the Coulomb barrier.<sup>1</sup> In the other group, mainly DLTS measurements, emission of charge carriers from defect levels has been investigated in fields up to  $10^5$  V/cm. The observed field stimulated emission and capture have been discussed in terms of Poole-Frenkel effect,<sup>2–4</sup> phonon assisted tunneling,<sup>5</sup> and a combination of both phenomena.<sup>6–10</sup> It is frequently emphasized that at high-field strengths the characteristic electric field dependence of the Poole-Frenkel effect does not fit well the experimental data.<sup>5–9,11</sup>

The well-known Poole-Frenkel effect describes the increase of the thermal emission rate of carriers in an external electric field due to the lowering of the barrier associated with their Coulomb potential [Fig. 1(a)]. The theory of this effect was developed by Frenkel,<sup>12</sup> who showed that the ionization probability increases in an electric field like

$$e(E)/e(0) \propto \exp(\varepsilon_{PF}/k_B T), \quad (1)$$

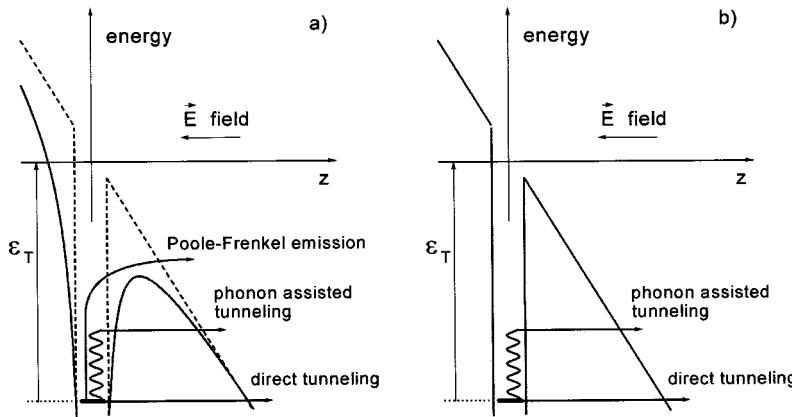


FIG. 1. Potential barrier for the emission of an electron from a deep-level defect in external electric field for: (a) charged impurities and (b) neutral impurities. The arrows show different ionization processes.

where the lowering of the barrier  $\varepsilon_{PF}$  is given by

$$\varepsilon_{PF} = 2 \sqrt{\frac{Zq^3 E}{\chi}}. \quad (2)$$

Here,  $Z$  is the charge of the center,  $q$  is the electron charge,  $E$  is the electric-field strength, and  $\chi$  is the dielectric constant. This expression yields an exponential increase of the ionization probability with the square root of the electric field. There are, however, several disagreements between the experiment and the Frenkel theory. In particular, experimental studies showed that the slope of  $\ln[e(E)/e(0)]$  versus  $\sqrt{E}$  is only about one half of that derived from Eqs. (1) and (2) and that at very low electric field strengths the emission rate becomes practically constant. These discrepancies are resolved by more realistic theoretical approaches which consider the emission of carriers in three dimensions,<sup>10,13</sup> take into account carrier distribution statistics,<sup>14–16</sup> or are based on the Onsager theory of dissociation.<sup>16,17</sup> For this study, it is sufficient to say that the proportionality, given by Eq. (1), is valid in a wide range of electric fields for both, the classical model of Frenkel, and more sophisticated models referenced above.

The theory of phonon-assisted tunneling in static electric fields was first considered quantum-mechanically. Makram-Ebeid and Lanoo carried out the most extensive numerical studies.<sup>18</sup> A semiclassical theory of phonon assisted tunneling was presented by Karpus and Perel<sup>19</sup> yielding analytical solutions of the problem, which we will use here. Both theoretical approaches are in good agreement. The emission rate increases exponentially with the square of the electric-field strength according to

$$e(E)/e(0) = \exp\left(\frac{E^2}{E_c^2}\right), \quad (3)$$

where  $E_c$  is a characteristic field strength, given by

$$E_c = \sqrt{\frac{3m^*\hbar}{e^2\tau_2^3}}, \quad (4)$$

where  $m^*$  is the effective mass of charge carriers,  $e$  is the elementary charge of the electron, and  $\tau_2$  is the tunneling time, given by (Ref. 20)

$$\tau_2 = \frac{\hbar}{2k_B T} \pm \tau_1, \quad (5)$$

where the plus and minus signs correspond to the adiabatic potential structures of substitutional impurities and auto-localized centers, respectively,<sup>21</sup> and  $T$  is the temperature. The time constant  $\tau_1$  is on the order of the inverse local impurity vibration frequency.<sup>21</sup>

A comparison of Eq. (1) with Eq. (3) suggests a simple criterion to distinguish the Poole-Frenkel and the phonon assisted tunneling mechanisms of ionization. Namely, the dependence of emission rate on electric field is given by  $\ln(e) \propto \sqrt{E}$  for the case of Poole-Frenkel model, while for the phonon assisted tunneling the electric field dependence is given by  $\ln(e) \propto E^2$ . Hence, a simple way to distinguish these two models is to plot the logarithm of emission rate against  $E^2$  and  $\sqrt{E}$  to compare which of the two plots can be fitted by a straight line. Surprisingly, to the best of our knowledge, this simple criterion has never been suggested or used before. In this article we will demonstrate the application of this analysis technique to three common defects in semiconductors.

In this paper, we used not only static electric fields, but also alternating fields with frequencies up to the terahertz range.<sup>21</sup> High-frequency fields may act like static fields as long as the oscillation period is longer than the characteristic time of the carrier emission process.<sup>22</sup> In the case of phonon assisted tunneling this characteristic time is the tunneling time  $\tau_2$ , and the quasi-static condition is given by  $\omega\tau_2 \ll 1$  (see Ref. 23 for the details). In this regime, the ionization probability is independent of the frequency, while in the high-frequency regime,  $\omega\tau_2 \gg 1$ , the ionization probability becomes frequency dependent.

The application of high frequency electric fields gives two important advantages compared to static fields. On the one hand, strong terahertz electric fields can be applied in a contactless manner to any kind of sample using the intense radiation of a high-power-pulsed far-infrared laser. Since photon energies in the far infrared are much smaller than the binding energies of deep impurities, one-photon ionization (i.e., photoionization) does not occur. On the other hand, the ionization probability may easily be determined by detecting photoconductivity, which is a very sensitive method with a high-dynamic range.

Applying the terahertz technique, the electric-field dependence of the ionization probability can be measured over

several orders of magnitude of the electric-field strength. Application of this technique to positively charged Ge:Hg enabled us to observe the Poole-Frenkel effect at low-electric field strengths and to trace the transition to phonon-assisted tunneling, which dominates carrier emission at high fields. In contrast, emission from DX centers in  $\text{Al}_x\text{Ga}_{1-x}\text{As}:\text{Te}$  was well described by the phonon-assisted tunneling model in the whole electric-field range. Cu-pairs in silicon could not be studied by the terahertz technique since only a very small fraction of diffused copper (usually on the order of 0.1% or less) forms Cu-pairs. Therefore, emission from Cu pairs in silicon was studied using DDLTS technique, which cannot be used to measure the emission rate at electric fields lower than approximately  $10^4$  V/cm due to the sensitivity limitations. This limitation hindered unambiguous determination of the charge state of this center. However, we found that in the range of electric fields accessible to DDLTS, emission from Cu pairs could be well described by phonon assisted tunneling.

#### IV. SAMPLES AND EXPERIMENTAL TECHNIQUE

The infrared measurements were carried out on *p*-type Ge(Hg) having an ionization energy of  $E_T = 90$  meV and DX centers in  $\text{Al}_x\text{Ga}_{1-x}\text{As}:\text{Te}$  (thermal ionization energy  $\varepsilon_T = 140$  meV, optical ionization energy  $\varepsilon_{\text{opt}} = 850$  meV). The doping concentration of the Ge:Hg samples was between  $10^{14}$  and  $10^{15}$  cm $^{-3}$ . Ionization of Hg impurities corresponds to a transition from a neutral ground state to a singly charged final state. Measurements of ionization of DX centers in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  were performed on thin films of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  ( $x = 0.35$ ) grown by MOVPE on semiinsulating (100)-GaAs substrates. The typical layer thickness was 30  $\mu\text{m}$ . All layers were doped uniformly with Te. The doping concentration was  $2 \times 10^{18}$  cm $^{-3}$ , as determined from secondary-ion-mass spectroscopy and Hall effect measurements. Measurements of electric-field dependence of ionization of Cu pairs ( $\varepsilon_T = 0.10$  eV) in Si were carried out on *p*-type boron-doped float-zone silicon samples with free carrier concentration of  $1.5 \times 10^{15}$  cm $^{-3}$ . Diffusion of copper was performed in a vertical furnace at temperature of 650 °C and was terminated by a quench in ethylene glycol. The effect of static fields on the emission rate from Cu pairs has been investigated by double deep level transient spectroscopy (DDLTS). Schottky diodes for DDLTS measurements were fabricated by aluminum evaporation. Temperature scans were performed using a Sula Technologies DLTS spectrometer, equipped with a helium cryostat.

Terahertz electric fields have been applied using high power far-infrared (FIR) laser pulses. The FIR laser was a molecular line-tunable NH<sub>3</sub>-laser optically pumped by a high power TEA-CO<sub>2</sub>-laser (URANIT 204).<sup>21</sup> The FIR laser produced linearly polarized radiation pulses of 40 ns duration. Electric-field strengths up to about 40 kV/cm ( $\approx 5$  MW/cm $^2$ ) could be achieved in the semiconductor samples in the frequency range from 5 to 50 THz. The corresponding photon energies ranging from 3.4 to 34 meV, respectively, are much smaller than the ionization energy of the impurities investigated. The samples were placed in a temperature-controlled optical cryostat. Light in the medium infrared and in the visible ranges was blocked by *z*-cut crystalline quartz plates

and 1 mm-thick black-polyethylene sheets, respectively. Samples of Ge:Hg, Si:Cu, and  $\text{Al}_x\text{Ga}_{1-x}\text{As}:\text{Te}$  were cooled so that at thermal equilibrium practically all carriers were bound to deep impurities ( $T = 30$  to 150 K, depending on the type of impurity). As the free carrier absorption is proportional to the free carrier concentration, which under these conditions is extremely small, even bulk samples can be illuminated homogeneously by far-infrared radiation. The electric field strength has been calculated from the laser radiation intensity inside the sample. The latter could be varied by using calibrated Teflon and polyethylene attenuators. The enhancement of the thermal ionization probability due to the electric field of the radiation, which is proportional to the change in the free carrier concentration, has been detected as photoconductive signal. The ratio of conductivity under illumination,  $\sigma_l$ , and dark conductivity,  $\sigma_d$ , has been determined from peak values of photoconductive signals. For laser pulses shorter than the carrier capture time, as it is the case here,  $\sigma_l/\sigma_d$  is equal to  $e(E)/e(0)$ , where  $e(E)$  is the emission rate as a function of the electric-field strength  $E$  (Ref. 21). Note that the FIR response in the case of DX-centers corresponds to the detachment of electrons from the defect yielding persistent photoconductivity.<sup>24</sup>

#### V. EXPERIMENTAL RESULTS AND DISCUSSION

In  $\text{Al}_x\text{Ga}_{1-x}\text{As}:\text{Te}$ , phonon assisted tunneling has been observed over the entire range of available electric field strength. This conclusion follows from the quadratic dependence of the logarithm of the ionization probability, as shown in Fig. 2. At relatively high temperatures, e.g., 100–150 K, and in the range of frequencies used in this study, the emission rate  $e(E)$  is independent of the radiation frequency proving that the electric field of far infrared radiation acts like a static field. The temperature dependence of  $\tau_2$  is in agreement with Eq. (5) with the minus sign, as it is expected from the adiabatic configuration potential of DX centers, and  $\tau_1 = 3.3 \times 10^{-15}$  s.

DDLTS data on the dependence of hole emission rate from Cu pairs in Sr are presented in Fig. 3. The good fit to a straight line in the  $\log(e)$  versus  $E^2$  plot demonstrates that in the electric field range of the measurement, between 10 and 40 kV/cm, the carrier emission is dominated by phonon assisted tunneling. Hence, no definite conclusion about the charge state of Cu pairs can be done in this case. Nevertheless, the conclusion about the dominance of phonon-assisted tunneling, done in this study, is an important progress in understanding of this defect in silicon.<sup>11</sup> The slope of  $\ln[e(E)]$  as a function of  $E^2$  gives the characteristic field strength  $E_c$  [Eq. (1)]. Evaluating this field the tunneling time  $\tau_2$  has been determined in the temperature range from 30 to 150 K. For both defects which could be described by phonon-assisted tunneling model,  $\text{Al}_x\text{Ga}_{1-x}\text{As}:\text{Te}$  and Si:Cu,  $\tau_2$  was found to be of the order of  $\hbar/(2k_B T)$ .

The electric-field dependence of emission from charged impurities cannot be fitted by the phonon assisted tunneling model alone. This is because in the case of charged impurities, the Poole-Frenkel and phonon assisted tunneling are two competing mechanisms of emission, as it is illustrated in Fig. 1(a). Since the width of the barrier for tunneling depends drastically on the field strength, one may expect that tunnel-

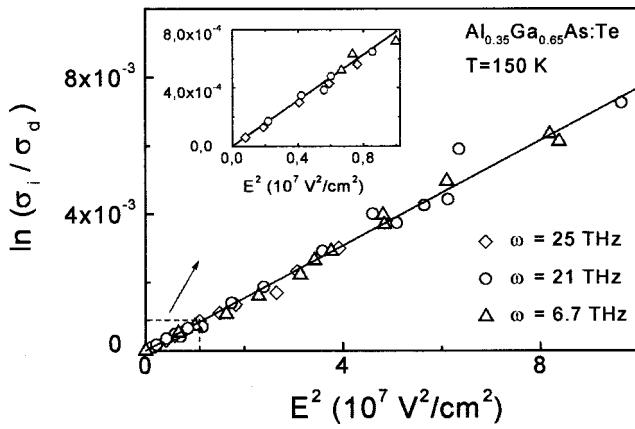


FIG. 2. Logarithm of the ratio of irradiated and dark conductivity,  $\sigma_i/\sigma_d = e(E)/e(0)$ , as a function of the square of the electric field  $E^2$  for DX centers in  $\text{Al}_x\text{Ga}_{1-x}\text{As:Te}$  measured at  $T=150 \text{ K}$  for radiation frequencies 6.7, 21, and 25 THz. The straight line corresponds to  $\exp(E^2/E_c^2)$ . The data for low-electric fields (box in the lower left corner) are shown on larger scale in the inset.

ing will be very inefficient, as compared to the Poole-Frenkel effect, in weak electric fields. In contrast, in strong electric fields the contribution of phonon-assisted tunneling may become substantial. This indeed was observed in our experiments on Ge:Hg, presented in Fig. 4. The emission rate is observed to grow exponentially with  $\sqrt{E}$  for electric field strengths below about 1 kV/cm (inset in Fig. 4). This relation between emission rate and electric field is characteristic for the Poole-Frenkel effect. Also in this case the emission rate is independent of radiation frequency. Increasing the electric field strength above 1 kV/cm leads to a change of the field dependence of the emission rate from  $\ln(e) \propto \sqrt{E}$  to  $\ln(e) \propto E^2$ . This change is clearly observable in Fig. 4 where  $\ln(\sigma_i/\sigma_d)$  is plotted as a function of  $E^2$ . In other words,  $\sigma_i/\sigma_d$  is proportional to  $\exp(E^2/E_c^2)$  for high-electric field, similarly to the case of neutral impurities. However, in contrast to neutral impurities, the straight line is shifted along

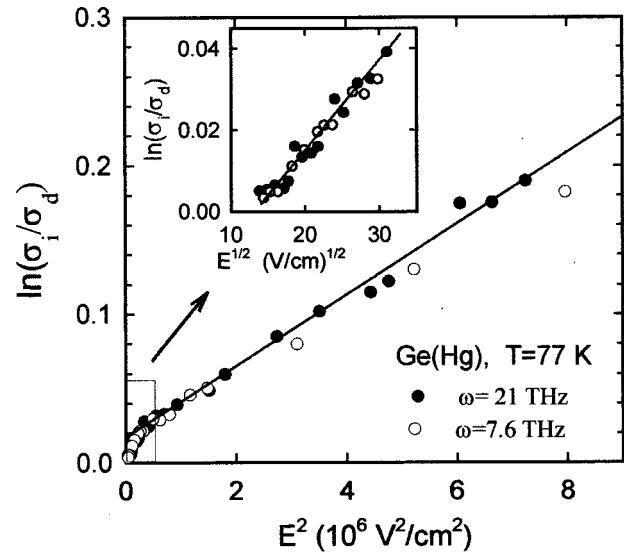


FIG. 4. Logarithm of  $\sigma_i/\sigma_d = e(E)/e(0)$ , as a function of  $E^2$  for Ge:Hg at  $T=77 \text{ K}$  measured with two frequencies of the field. The straight line corresponds to  $[\text{const} \times \exp(E^2/E_c^2)]$ . In the inset the data of the box in the lower left corner are replotted as a function of  $\sqrt{E}$ . The straight line shows that in this range of low-electric field strengths ( $< 1 \text{ kV/cm}$ ),  $\sigma_i/\sigma_d = e(E)/e(0)$ , increases exponentially with the square root of  $E$ .

the ordinate to higher values, which is in good agreement with the semiclassical theory.<sup>25</sup> This increase of phonon-assisted tunneling probability for charged impurities is caused by the lowering of the potential barrier height due to the Coulomb potential of the impurity. Hence, there are two criteria that can be used to identify the charge state of a defect from the dependence of carrier emission rate on applied electric field: (i) observation of the Poole-Frenkel effect at low fields and (ii) the shift of the straight line in the  $\ln[e(E)/e(0)]$  vs  $E^2$  diagram, corresponding to phonon assisted tunneling at higher electrical fields. For the second criterion it is essential to normalize the dependence  $e(E)$  by the emission probability at zero electric field,  $e(0)$ . Neither the Poole-Frenkel effect nor the shift have been observed for DX<sup>-</sup> centers in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ .

This procedure can be easily applied to reexamine the deviations of electric field emission from Poole-Frenkel dependence, reported in literature for various defects. For instance, Buchwald and Johnson<sup>9</sup> reported a deviation of electric field dependence of emission of electrons from EL2 center in GaAs in fields ranging from  $2.5 \times 10^5$  to  $3.8 \times 10^5 \text{ V/cm}$ . Although they discussed transition to tunneling in strong electric fields as a possible explanation for the observed effect, they did not provide a convincing proof for that. Replotting their data from  $\ln[e(E)]$  vs  $\sqrt{E}$  to  $\ln[e(E)]$  vs  $E^2$  gives a straight line (not shown), proving that phonon-assisted tunneling is indeed the dominant mechanism for carrier emission from EL2 in high-electric fields.

Finally, we note that the electric field range where phonon-assisted tunneling dominates deep impurity ionization is limited to field strengths  $E \leq E^* = \sqrt{2m^*E_T/(e\tau_2)}$  (Ref. 19). At higher fields, the carrier emission process is determined by direct tunneling (see Fig. 1), which is characterized by a slower increase of the emission rate with the

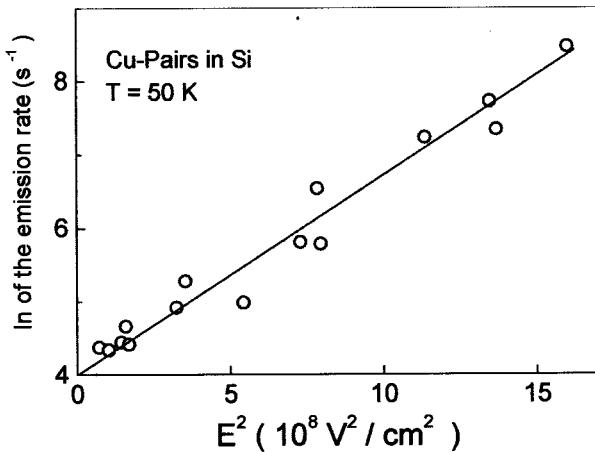


FIG. 3. Dependence of the logarithm of the emission rate  $e(E)$  on square of static electric field in the space charge region of a Si-Schottky diode with copper pairs at  $T=50 \text{ K}$ . The straight line corresponds to  $\exp(E^2/E_c^2)$ .

increasing electric field.<sup>19,21</sup> Direct tunneling requires much higher electric-field strengths than used in this work.

## VI. CONCLUSIONS

In summary, we have shown that the square root electric field dependence of the logarithm of emission rate usually used to identify the Poole-Frenkel effect can only be observed for charged impurities at sufficiently low-electric field strengths. At higher field strengths the Poole-Frenkel effect is outweighed by phonon-assisted tunneling whose emission rate exponentially grows with the square of the electric field strength. The lowering of the potential barrier height in an external electric field due to the Coulomb potential of charged impurities, which yields the enhanced emission rate at low fields (Poole-Frenkel-effect), gives only a correction to the phonon-assisted tunneling ionization rate at high fields. This correction does not alter the electric field dependence of phonon assisted tunneling, which therefore can easily be identified by plotting logarithm of the emission rate versus electric field squared for both charged and neutral impurities. Therefore, the determination of the slope of the power law of  $\ln(e)$  versus  $E$  for small fields is an easy and

unambiguous way to determine the nature of the field enhancement of the carrier emission. If phonon-assisted tunneling prevails even for low fields, the carrier is emitted from a neutral impurity. Since an enhancement of carrier emission rate in electric field is observed for both charged and neutral impurities, the observation of the enhancement of emission rate in an electric field alone is not sufficient to conclude on the charge state of an impurity. The latter can be inferred from a plot of the logarithm of the emission probability  $e(E)$  as a function of  $E^2$  at high fields and  $\sqrt{E}$  at low-field strength or from plotting the logarithm of the normalized emission probability  $e(E)/e(0)$  versus  $E^2$  in the electric-field range of phonon-assisted tunneling.

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