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Electric-field-enhanced electron emission from isolated sulfur and selenium donors in silicon

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The electric field effect on the thermal electron emission of isolated sulfur and selenium centers was studied in silicon. Various degrees of field dependences were observed for all centers except for the neutral sulfur center, which exhibited almost no field effect. Taking the measured field dependences into account it is shown that the zero-field enthalpies are in good agreement with previously measured optical binding energies. However, experimental evidence suggests that field-effect studies are not always reliable measurement techniques to identify whether the defect studied is an acceptor or a donor level.

I. INTRODUCTION

In all cases where direct evidence of the exact lattice position and the chemical identity of a defect in a semiconductor cannot be provided experimentally, any theoretical model of such a defect will depend on the type of the defect, i.e., whether the defect is a donor or an acceptor. A typical example of such a dilemma is the *DX* center in $Al_xGa_{1-x}As$. Although several theoretical models have been proposed, the final decision as to which of these models is correct depends to a large extent on whether the center is a donor or an acceptor.[1] However, no such evidence has yet been provided for the *DX* center.

Evidence for the defect type is readily obtained for simple effective-mass-theory (EMT) -like centers exhibiting well-resolved line spectra in absorption or photoconductivity, such as the group-III acceptors and group-V donors in silicon. Similar straightforward results have been obtained for some of the chalcogen double donors in silicon.[2] In all these cases the line spectra provide the most convincing arguments for the type of defect, in agreement with other investigations which establish the defect type beyond any doubt.

The methods of establishing the defect type are less straightforward for centers such as the transition-metal-related defects in silicon which exhibit more complicated line spectra. However, it has been shown[3,4] that the defect type can still be determined fairly easily for these centers if the defect exhibits phonon-assisted Fano resonances, since the phonons involved in the resonances are characteristic of the defect type.

It is quite evident that such methods can only be used for establishing the defect type when the centers to be studied exhibit line spectra. In all other cases different analytical methods have to be applied. One such method which has been frequently explored is the application of the Poole-Frenkel effect.[5,6] When studying electric-field-assisted thermal ionization of bound charge carriers in electronic semiconductors and insulators it is normally assumed that defects show a field effect, and that the effect increases with the amount of charge bound at the center. Since acceptor and donor levels are expected to exhibit different field effects, it has been suggested[7] that the study of field-enhanced thermal ionization processes can be used to establish the defect type. Such studies have been performed in a variety of semiconductors for several different impurities and defects.[7–13]

The purpose of this paper is to present a comprehensive study of the electric-field-assisted thermal ionization of the single substitutional sulfur and selenium donors in silicon. The electronic structure of these centers is known in detail from high-resolution spectroscopic studies[2] and the defect type has been established and confirmed by several different methods, both experimental[14] and theoretical.[15] Most of the previous studies on double donors in silicon showed good agreement between optical and thermal data for the energy position of the centers.[16,17] Other investigations claimed considerable discrepancies between thermal and optical activation energies and these discrepancies were explained by taking into account the Poole-Frenkel effect.[8] It was therefore of interest to find out whether or not there are inconsistencies between optical and thermal data and, if this is the case, whether these inconsistencies are due to the Poole-Frenkel effect. If the centers show the Poole-Frenkel effect or any field-enhanced emission-related process, then it is important to determine how large the field effect for these impurities is and how characteristic the effect is for the determination of the defect type.

II. EXPERIMENTAL DETAILS

The samples used in this investigation were sulfur, selenium, and tellurium diffused silicon p^+-n diodes, similar to those employed in previous studies.[14,16,17] It is known that doping silicon with chalcogens could create a variety of chalcogen-related centers in silicon. Therefore, special care was taken to make sure that only the isolated impurity defects were studied in all three cases. It has already been pointed out in previous studies that the thermal capacitance transients in tellurium doped sam-

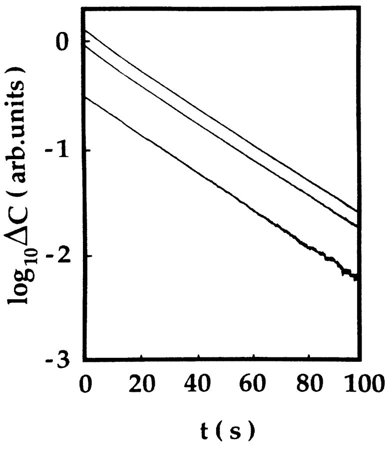

FIG. 1. Capacitance transients of the charged selenium level with a 2 V reverse bias. The upper transient corresponds to a 0.2-V pulse, the middle transient to a 0.7-V pulse, and the lower transient is the difference of the above two transients.

ples are nonexponential.[17] Since the capacitance transients in our tellurium doped samples were also nonexponential, no further studies related to these phenomena were performed on these diodes for reasons which will become clear later.

All studies were conducted using single shot measurements.[18] This aided us in the measurement of strictly exponential transients. A narrow range within the space-charge region was selected by taking the difference between two transients recorded at two slightly different pulse voltages V_1 and V_2 and keeping the reverse bias V_R constant. In this manner only thermal emission processes at a fairly constant electric field strength F were studied. F was varied by changing the reverse bias V_R. Typical transients obtained for V_1 and V_2 as well as the difference of these transients are presented in Fig. 1. It is evident from the figure that all three transients are indeed purely exponential.

Since the thermal emission rate for electrons, e_n^t, of deep donors depends strongly on the measurement temperature T, correct absolute values of e_n^t are obtained only if the sample temperature is known to within a high degree of precision. It is readily shown from an Arrhenius plot of $\ln e_n^t$ versus $1/T$ that an error of only 1 K in the sample temperature will change the thermal activation energy E_{th} of the charged selenium donor by about 5 meV. The temperature of our sample holder was therefore calibrated by replacing the samples with a silicon diode having an ideality factor of 2 and subsequently measuring the temperature from the change in the forward bias V at constant current I. The temperature dependence of the I-V characteristic of the silicon diode was carefully checked prior to these calibrations. It is worth mentioning that the difference between the temperature of the sample holder, when measured with the thermocouple, and the corrected temperature of the sample was found to be about 2.4 K.

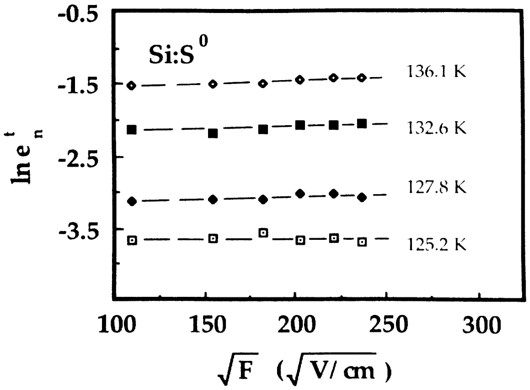

FIG. 2. Thermal emission rate of electrons as a function of the electric field for the neutral sulfur donor at different temperatures.

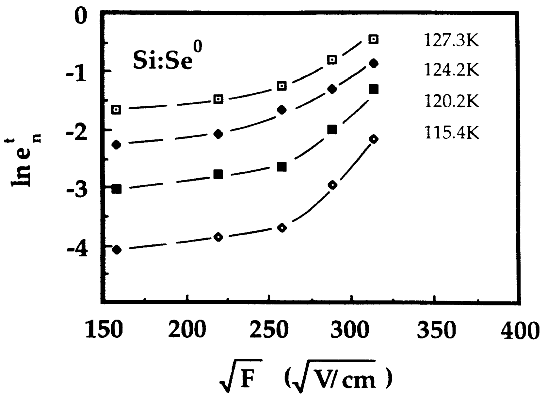

FIG. 3. Thermal emission rate of electrons as a function of the electric field for the neutral selenium donor at different temperatures.

III. EXPERIMENTAL RESULTS

The thermal emission rates for electrons as a function of the electric field are shown in Figs. 2 and 3 for the neutral sulfur and selenium donor, respectively, at different temperatures. Up to about 65 kV/cm there is little if any

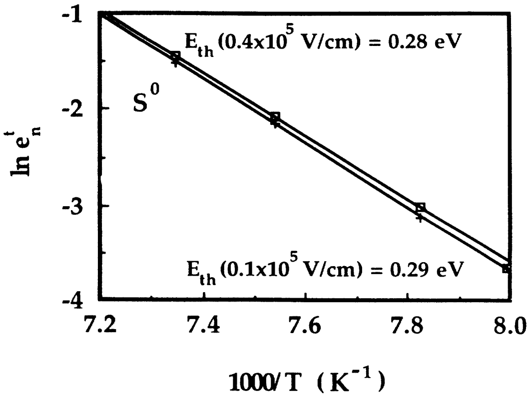

FIG. 4. Arrhenius plots of the thermal emission rate for the neutral sulfur donor at two different electric fields.

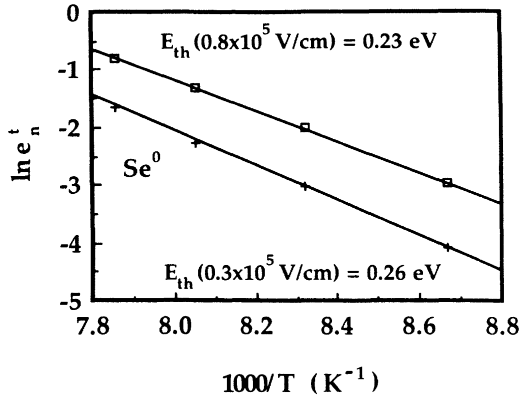

FIG. 5. Arrhenius plots of the thermal emission rate for the neutral selenium donor at two different electric fields.

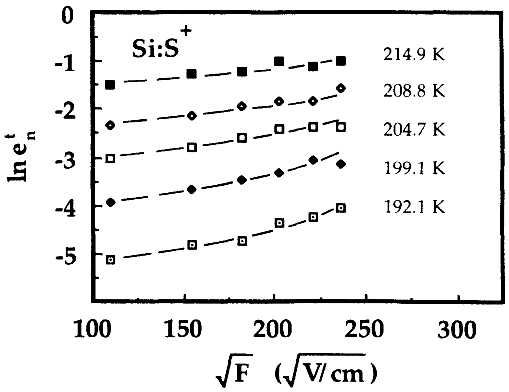

FIG. 7. Thermal emission rate of electrons as a function of the electric field for the charged sulfur donor at different temperatures.

field dependence observed in both cases. However, for the neutral selenium donor which could be studied at much higher field strength than the sulfur doped diode, a substantial field dependence is observed at field strengths greater than 65 kV/cm. The field sensitivity appears to decrease with increasing temperature, implying a decrease in the thermal activation energy as a function of increasing field strength.

This is more clearly seen when the logarithm of the emission rate is plotted versus $1/T$ for different electric fields (Figs. 4 and 5). Since for both impurities the electric field dependence of the emission rate does not vary much with temperature, up to field strengths of 65 kV/cm, only small changes, if any, in the activation energy are observed. Only the neutral selenium donor exhibits a considerable decrease in activation energy at field strengths above 65 kV/cm (Fig. 5). This demonstrates that the neutral Se exhibits a field dependence of the emission rate which is larger than that resulting from experimental errors. However, this is not the case for the sulfur donor. A comparison with previous studies[14,16] shows that within the experimental error these results are in good agreement with the results presented in this paper for lower field strengths.

The variation of the activation energy, E_{th}, with electric field is summarized in Fig. 6, which is a plot of the measured activation energies versus $F^{1/2}$ for the neutral sulfur and selenium donor. Considering that the accuracy of junction space-charge techniques (JSCT) is about ±10 meV in absolute values (but somewhat better in relative values) it is fair to say that almost no decrease in barrier height is observed for the neutral sulfur donor. The slight increase observed at the highest field strengths is probably due to a decrease in measurement sensitivity since the difference of the two transients needed for a measurement decreases with increasing reverse bias. In contrast to the neutral sulfur donor the selenium center shows a considerable field dependence of the activation energy resulting in a decrease of E_{th} with increasing field strength.

Quite different results were obtained when studying the charged sulfur and selenium donors in silicon. Plotting e_n^t versus $F^{1/2}$ (Figs. 7 and 8), a field dependence is observed for both centers. Although the field dependence is smaller for the charged selenium donor, a considerable change in the field dependence with decreasing tempera-

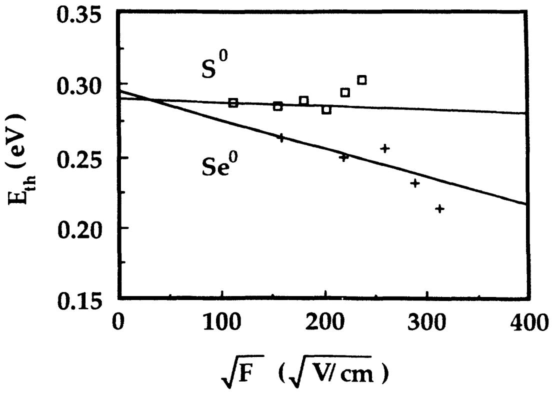

FIG. 6. Electric field dependence of the activation energy for the neutral sulfur and selenium donors.

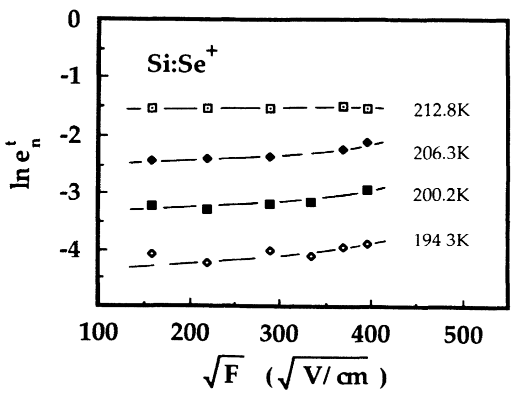

FIG. 8. Thermal emission rate of electrons as a function of the electric field for the charged selenium donor at different temperatures.

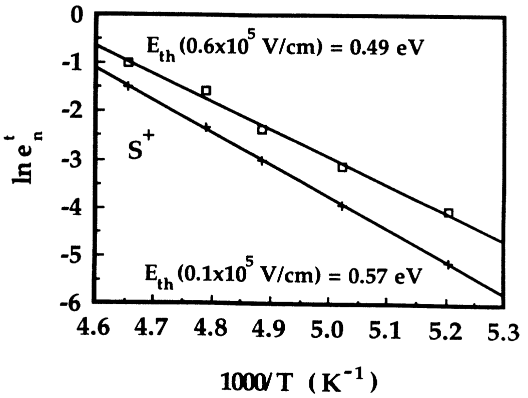

FIG. 9. Arrhenius plots of the thermal emission rate for the charged sulfur donor at two different electric fields.

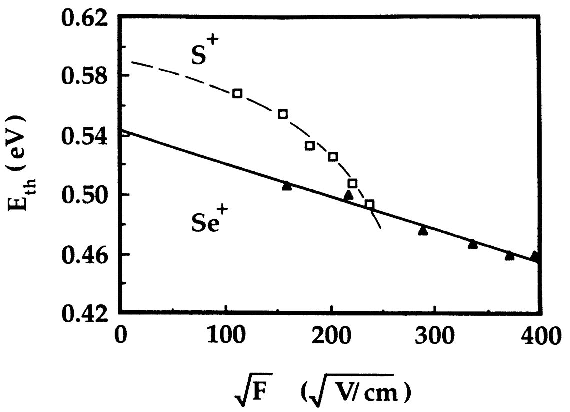

FIG. 11. Electric field dependence of the activation energy for the charged sulfur and selenium donors.

ture was observed in both cases. This implies that the activation energies obtained from Arrhenius plots of the thermal emission rate vary considerably with field strength for both centers (Figs. 9 and 10). A comparison shows that the activation energies obtained in Figs. 9 and 10 for low field strengths are in fair agreement with previously published results.[14,16] The various activation energies obtained at different field strengths are summarized in Fig. 11 for the charged sulfur and selenium donor, showing that for the charged selenium center the decrease in the activation energy seems to be proportional to $F^{1/2}$. A rather different field dependence is observed for the charged sulfur center. When the measured activation energies for the charged sulfur center are plotted versus F instead of $F^{1/2}$ (Fig. 12), the data are fairly well represented by a straight line suggesting that the lowering of E_{th} is directly proportional to F rather than $F^{1/2}$.

The electric field effect on the thermal electron emission may in principle arise due to different reasons. However, since previous measurements of chalcogens in silicon have been analyzed in terms of the Poole-Frenkel effect, we have used a similar presentation of our results in order to facilitate a comparison of our data with previously published results.

If the decrease in activation energy is due to the Poole-Frenkel effect, the energy lowering ΔE_{th} in the one-dimensional model by Frenkel[5] can be expressed as[19]

$$\Delta E_{th} = cF^{1/2} , \tag{1}$$

where c is a temperature-independent constant of about 2×10^{-4} eV(V/cm)$^{-1/2}$ for a singly charged center such as the neutral chalcogen donors before ionization. If e_{n0}^t is the emission rate without field, the emission enhancement is given by[19]

$$e_n^t = e_{n0}^t \exp(\Delta E_{th}/kT) . \tag{2}$$

Combining Eqs. (1) and (2), it is readily shown that the change in activation energy $\Delta E'$ between two Arrhenius plots at field strengths F_1 and F_2 can be written as

$$\Delta E' = c(F_2^{1/2} - F_1^{1/2}) = kT\ln(e_{n2}^t/e_{n1}^t) . \tag{3}$$

Hence, c is given by

$$c = kT\ln(e_{n2}^t/e_{n1}^t)/(F_2^{1/2} - F_1^{1/2}) . \tag{4}$$

Using Eq. (1) and the data presented in Fig. 6, a value of about 2×10^{-4} eV(V/cm)$^{-1/2}$ is obtained for c in the

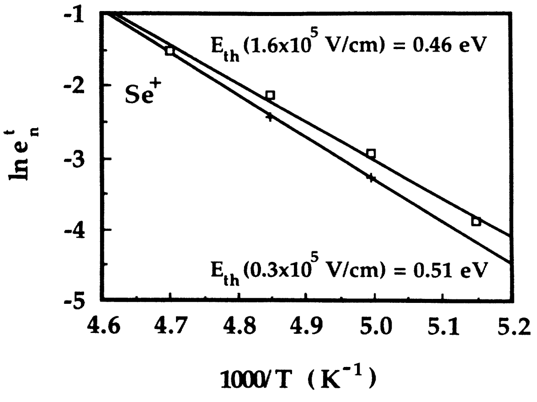

FIG. 10. Arrhenius plots of the thermal emission rate for the charged selenium donor at two different electric fields.

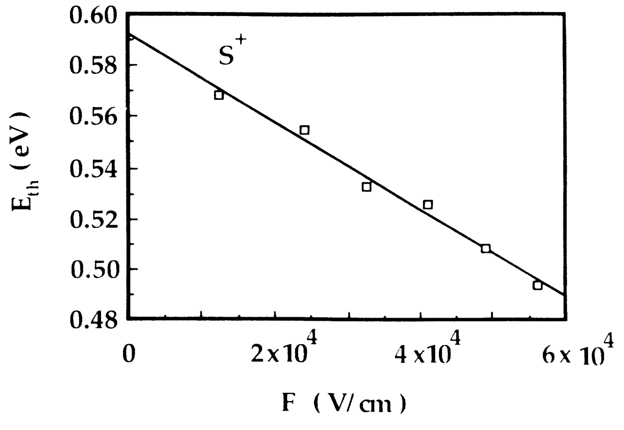

FIG. 12. Variation of the activation energy with electric field for the charged sulfur donor.

case of the neutral selenium center, whereas the corresponding value for the neutral sulfur center is smaller than 10^{-5} eV$\,(\mathrm{V/cm})^{-1/2}$. A similar analysis of the results presented in Fig. 11 gives a value of about 2.5×10^{-4} eV$\,(\mathrm{V/cm})^{-1/2}$ for the charged selenium center, which is about half of the value expected. In the case of the selenium centers this would suggest that the electric-field-enhanced electron emission is governed by the Poole-Frenkel effect. However, if Eq. (4) is used instead of Eq. (1), the data shown in Fig. 5 for the neutral selenium center give only a value of 0.8×10^{-4} eV$\,(\mathrm{V/cm})^{-1/2}$ for c, which is about three times smaller than the value obtained in Fig. 6. The value of c obtained from such an analysis depends on the pair of field strengths used. The apparent variation is explained by the fact that simple Poole-Frenkel barrier lowering does not accurately explain the neutral selenium data, as is evidenced by the significant deviation of three of the measuring points from the best fit line in Fig. 6. Still larger discrepancies are found for similar reasons for the charged selenium center when applying the same analysis to the results of Fig. 10. In this case a value of 0.3×10^{-4} eV$\,(\mathrm{V/cm})^{-1/2}$ is calculated for c. By applying Eq. (4) to the thermal emission data of the charged sulfur center (Fig. 9), a value of 1.2×10^{-4} eV$\,(\mathrm{V/cm})^{-1/2}$ is obtained for c which is reasonably close to the value expected for the Poole-Frenkel effect but nevertheless is irrelevant since Fig. 11 shows that ΔE_{th} is not proportional to $F^{1/2}$.

IV. DISCUSSION

The binding energies of the first and second electron of the isolated sulfur and selenium double donors in silicon are accurately known from high-resolution spectroscopic measurements.[20,2] The spectra clearly show that all centers have T_d point-group symmetry implying that they are not due to chalcogen-related complexes. The values of the different binding energies measured at low temperature are summarized in Table I. Using local Auger-effect studies,[21] the two levels observed for each center have been proven to be coupled, and due to different charge states. These results are in good agreement with doping experiments.[22]

In this paper the thermal emission rates are presented as Arrhenius plots of e_n^t and not e_n^t/T^2 in order to facilitate comparison with previously published data.[8,11,14,16] It should, however, be noted that the activation energy E_{th} obtained from such Arrhenius plots has a special physical significance. E_{th} is the change in enthalpy of the level studied provided the capture cross section of electrons, σ_n^t, is proportional to T^{-2}. In all other cases, E_{th} is an unspecified energy without a real physical meaning and certainly cannot be used for purposes of comparison with optical binding energies, which has been done[8,11] occasionally. A relevant energy, namely the change in enthalpy, ΔH_n, due to the ionization of the center is obtained when the activation energy E_c deduced from an Arrhenius plot of $\sigma_n^t T^2$ is added to E_{th} provided the capture cross section can be expressed as

$$\sigma_n^t=\sigma_0 T^{-2}\exp(E_c/kT)\ . \tag{5}$$

This procedure follows directly from the detailed balance relation, which can be written as[23]

$$e_n^t=\sigma_n^t v_{\mathrm{th}} N_c \exp(-\Delta G_n/kT)\ , \tag{6}$$

where ΔG_n is the change of Gibb's free energy given by

$$\Delta G_n=\Delta H_n - T\,\Delta S\ . \tag{7}$$

Here, v_{th} is the thermal velocity of electrons in the conduction band, N_c is the effective density of states in the conduction band, and ΔS is the change in entropy. Combining Eqs. (5)–(7), Eq. (6) can be rewritten as

$$e_n^t=A\ \exp[-(\Delta H_n-E_c)/kT]\ , \tag{8}$$

where $A=\sigma_0 T^{-2} v_{\mathrm{th}} N_c \exp(\Delta S/k)$ is a temperature-independent constant. The change in enthalpy is therefore given by

$$\Delta H_n=E_{\mathrm{th}}+E_c\ . \tag{9}$$

At low temperatures the optical binding energies are well approximated by ΔG_n, which in turn is comparable with ΔH_n. Hence, it is the change in enthalpy at zero field strength and not E_{th} which should be compared with optical binding energy at low temperatures.

In previous studies,[14,16] we have shown that it is difficult to determine the exact temperature dependence of the electron capture for the chalcogen double donors in silicon, since the experimental data could be fitted to different temperature dependences. It is, however, im-

TABLE I. A summary of the values of the different binding energies of the first and second electron of the isolated sulfur and selenium double donors in silicon measured at low temperatures.

Chalcogen donor	Optical binding energy (eV)	E_c (eV)[a,b,c]	Zero-field E_{th} (eV)	ΔH_n (eV)	E_{th} (eV)[a,b]	Zero-field E_{th} (eV)[d]
S^0	0.32	0.02	0.29	0.31	0.30	0.33
Se^0	0.31	0.01	0.29	0.30	0.29	0.32
S^+	0.61		0.59		0.59	0.55
Se^+	0.59	0.05	0.54	0.59	0.52	0.59

[a] Reference 14.
[b] Reference 16.
[c] Reference 24.
[d] Reference 11.

portant to note that the change in enthalpy obtained from a combined analysis of σ_n^t and e_n^t is independent of the assumed temperature dependence of σ_n^t as long as a proper analysis of e_n^t is performed, implying that Eq. (6) is not violated. Assumptions of the temperature dependence of σ_n^t are useful only for a physical interpretation of the different activation energies obtained and not for the evaluation of enthalpies.[14,16] Since different excitation processes give rise to different temperature dependences of the capture cross section, a careful analysis of the temperature dependence of the capture cross section may nevertheless give information on the excitation process, provided the capture cross section is studied over a wide range of temperature.

E_c values have been measured for the neutral sulfur[14] and selenium[16] donor as well as for the charged selenium[24] donor and are summarized in Table I together with the E_{th} values obtained for zero electrical field. The sum of these two values gives the change of enthalpy ΔH_n at zero-field strength. In all three cases these values are in good agreement with the optical binding energy, suggesting that the observed field dependences are reasonable. A similar conclusion may also be valid for the S^+ level since it is expected that the electron capture is some kind of cascade process and, hence, the temperature dependence of the capture cross section will give a positive E_c value. This implies that E_{th} of the charged sulfur level at zero field should be smaller than the optical binding energy.

A comparison with previously published data[14,16] shows that they are almost identical with the zero field values of E_{th} presented in this paper except for the charged selenium center for which our previous result is smaller. This agreement is explained partly by the small field effect of the neutral centers and partly by the fact that capacitance measurements favor excitation processes in the low field part of the junction, which in our case had a rather low free-electron concentration in the neutral *n* region. Experimental evidence indicates that a low free-carrier concentration is essential, since an increase in the free-carrier concentration by only 1 order of magnitude may result in a decrease of E_{th} by several tens of meV. E_{th} values are, therefore, not very useful in general for the characterization of defects since they may be considerably different from the change in enthalpy due to thermal capture barriers and/or due to electric-field-assisted thermal-ionization processes. The data published by Roos *et al.*[11] differ from our results obtained at zero-field strength by more than 30 meV (Table I). The authors compare their E_{th} values at zero-field strength with their optical binding energies and attribute the good agreement in the values of these energies as proof that previously observed differences between E_{th} and optical binding energies ought to be explained by the Poole-Frenkel effect and a one-step thermal emission process. We have already pointed out that changes in enthalpy and not E_{th} values should be compared with optical binding energies and that the earlier evaluation of changes in enthalpy[14,16] are not affected by the assumed emission processes.

The results presented in this paper suggest that the energy resolution of JSCT is not sufficient to determine the exact type of electric-field-enhanced emission process involved. It is therefore rather difficult to evaluate whether the defect studied is a donor or an acceptor since the field dependence of ΔE_{th} for an acceptor is not very different from the one for a donor. In the case of the one-dimensional Poole-Frenkel effect, ΔE_{th} is proportional to $F^{1/2}$ for a center with Coulomb potential whereas $\Delta E_{\text{th}} \sim F^{4/5}$ for a center with polarization potential.[19] However, at low field strengths JSCT should be sensitive enough to distinguish between the different field enhancements of thermal emission rates due to different potentials. Since the experimental results cannot be explained by one particular type of field effect, it is concluded that more than one kind of field effect is involved. The *c* factors calculated from Figs. 2 and 3 lie in the range $\leq 10^{-5}$ eV$\,(\text{V/cm})^{1/2}$ for S^0 and $\leq 0.1-3\times 10^{-4}$ eV$\,(\text{V/cm})^{1/2}$ for Se^0. Corresponding values calculated from Figs. 7 and 8 are $0.5-1\times 10^{-4}$ eV$\,(\text{V/cm})^{1/2}$ for S^+ and $\leq 0.3\times 10^{-4}$ eV$\,(\text{V/cm})^{1/2}$ for Se^+. Hence, depending on the temperature and range of field strength, the *c* value obtained can provide support in favoring or disregarding the Poole-Frenkel effect.[8] It is also interesting to note that in spite of the small field dependence of the thermal emission rates, quite appreciable field effects are observed for ΔE_{th} as, for example, in the case of the charged selenium center. The field dependence of ΔE_{th} would support the Poole-Frenkel effect for this center, whereas the field dependence of e_n^t would not. Furthermore, ΔE_{th} of the neutral sulfur center seems to be little affected by the field, whereas ΔE_{th} of the charged sulfur center is proportional to *F*. The E_{th} values of the neutral selenium center are plotted versus $F^{1/2}$ in Fig. 6, however it can be shown that a still better agreement is obtained when plotting the values versus *F*, although the ΔE_{th} values of Se^+ are obviously proportional to $F^{1/2}$. Therefore, the field dependences of E_{th} and e_n^t most probably suggest rather complicated electric-field-enhanced emission processes.

Obviously such properties are characteristic not only of isolated chalcogen donors but also of chalcogen related complexes. In a detailed study of a tellurium-related complex in silicon it was shown by Hofmann *et al.*[8] that the neutral version exhibited a negligible field dependence whereas the charged center was well described by the Poole-Frenkel effect. This suggests that the electric-field-enhanced emission process and in particular the Poole-Frenkel effect is not always a reliable technique for determining the defect type.[12] All results presented in the paper have been obtained from single shot measurements. This implies that we were able to check that all transients employed in the study were exponential. It is not certain that similar conditions are always observed when using deep-level transient spectroscopy (DLTS) or double deep-level transient spectroscopy (DDLTS) (Ref. 25) for this kind of study.

V. CONCLUSION

We have performed a comprehensive study of the electric field effect on the thermal emission of isolated sulfur and selenium centers in silicon. The study shows that previously published data[14,16] on ΔH_n are in good agreement with the results presented in this paper and that the analysis previously performed is still valid. The centers investigated exhibit various electric field effects. In particular, one of the centers showed a negligible field dependence. Furthermore, the field dependence of thermal emission rates and activation energies can support different models of field-enhanced excitation processes. It therefore seems that investigations of electric-field-enhanced emission processes do not always provide reliable information on the defect type.

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