Eur. Phys. J. B **60**, 9–14 (2007)
DOI: 10.1140/epjb/e2007-00318-9

THE EUROPEAN PHYSICAL JOURNAL B

Trap-recharging waves versus damped, forced charge-density oscillations in hexagonal silicon carbide

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Received 14 August 2007 / Received in final form 9 October 2007
Published online 23 November 2007 –

Abstract. Resonant excitation of space-charge waves (SCW) by means of an oscillating light pattern has been investigated in hexagonal silicon carbide with 4H and 6H stacking sequence, respectively. The experimental data set can be well explained by the existence of *trap recharging waves* for the 4H sample and allows to determine the product of mobility and lifetime $\mu\tau = (5.7 \pm 0.6) \times 10^{-7}$ cm^2/V, and the effective trap density $N_{\text{eff}} = (8.0 \pm 1.0) \times 10^{13}$ cm^{-3}, respectively. The data set of the 6H polytype indicates a comparably smaller effective trap density, but an unambiguous assignment to the existence of trap recharging waves fails. Taking into account the general classification of material parameters which provides the existence for SCW, the particular case of *damped, forced charge-density oscillations* can be concluded.

PACS. 42.65.-k Nonlinear optics – 42.70.Nq Other nonlinear optical materials; photorefractive and semiconductor materials – 71.45.Lr Charge-density-wave systems

1 Introduction

Space-charge waves (SCW) are eigenmodes of spatial-temporal oscillations of a space-charge density, which can appear in semi-insulating materials in the presence of an external electric field. The existence of eigenmodes of SCW, associated with the trap recharging process in semiconductors, was theoretically predicted in 1972 [1]. Hence, they are well-known as *trap-recharging waves* (TRW). Detailed experimental investigations of SCW started with the development of proper methods for the optical excitation and were mostly performed with photorefractive crystals of the sillenite family [2,3]. In recent years, SCW studies were extended to *classical* high resistive semiconductors like InP:Fe [4], CdTe:Ge [5,6] and hexagonal silicon carbide (SiC) [7].

The fundamental properties of SCW, such as their dispersion law, are determined by the charge-carrier mobility μ and life time τ, the dielectric constant ϵ as well as the effective trap density N_{eff} of the material. The latter is defined by $N_{\text{eff}} \approx N_A(N_D - N_A)/N_D$, where N_D is the number density of donors and N_A that of acceptors.

The main criterion for the existence of SCW is the magnitude of the quality factor $Q = d(1-\beta)/(1-\beta d^2)$, which must exceed unity if we deal with an eigenmode of SCW rather than with forced oscillations. Here, the factor $\beta = E_0/(E_q d) = \epsilon\epsilon_0(\mu\tau e N_{\text{eff}})^{-1}$ comprehends the key material parameters and the electron charge e. The experimental conditions are covered by $d = \mu\tau E_0 K$ with the applied electric field E_0 and the spatial frequency K. The so-called trap saturation field is represented by $E_q = eN_{\text{eff}}/(\epsilon\epsilon_0 K)$ [8]. In accordance with references [9,10], the general physical description of SCW in semiconductors can be roughly classified by three specific cases depending on the combination of the material parameters and the experimental conditions:

(1) the first case corresponds to a high quality factor $Q > 1$ and $\beta \ll 1$. A high Q can be provided by selecting proper experimental conditions, i.e., E_0 and K must satisfy the requirements that $d > 1$ and $E_0/E_q < 1$. In this case, the SCW dispersion reads $\Omega_K \sim 1/K$, where Ω_K is the eigenfrequency of the SCW, and the quality factor can be simplified to $Q = (d^{-1} + E_0/E_q)^{-1}$. For experimental convenience, it is assumed that the magnitudes of $\mu\tau$ and N_{eff} are large enough, simultaneously. These waves are also called ordinary TRW and are typical for sillenites [2,3] and InP:Fe [4];

(2) the second case corresponds to the condition $\beta \gg 1$. It is assumed that the magnitudes of $\mu\tau$ and N_{eff} are simultaneously small enough for experimental convenience. In this case, the magnitude $Q > 1$ is

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provided if $d < 1$ and $E_0/E_q > 1$. Here, the dispersion law of the SCW has a linear character ($\Omega_K \sim K$) and $Q \approx E_0/E_q$. This type of SCW was found in reference [5] in CdTe:Ge;

(3) the third case occurs when $\beta \approx 1$. In this situation, $Q < 1$ for any experimental conditions and there are no eigenwave modes at all. Here, only damped, forced charge-density oscillations can exist.

This classification does not take into account some specific factors such as the diffusion process, bipolar conductivity or a large ratio n/N_{eff} with the density of free charge carriers n. The latter can approach large values for the case of optical interband excitation of charge carriers. These factors can modify the numerical criterion, but do not change the basic concept. This classification implies that the SCW concept can be well applied for material and defect analysis [11].

In this paper the excitation of SCW is investigated in two polytypes of hexagonal silicon carbide having a stacking sequence of 4H and 6H, respectively. The experimental dependences point out the presence of *trap recharging waves* according to case (1) for the 4H sample and are found in full accordance with the results of reference [7] obtained for a sample from a different source. The experimental data set for the 6H sample indicates a comparably low effective trap density which results in a small quality factor $Q < (0.36 \pm 0.18)$ and $\beta = (0.5 \pm 0.2)$ by our analysis. Therefore, the conditions $Q > 1$ and $\beta \ll 1$ are not satisfactorily fulfilled, and the particular case of *damped, forced charge-density oscillations* according to case (3) must be concluded. According to these considerations the experimental accuracy, the validity of prerequisites, as well as the applicability of the simplified model of the theoretical approach are discussed.

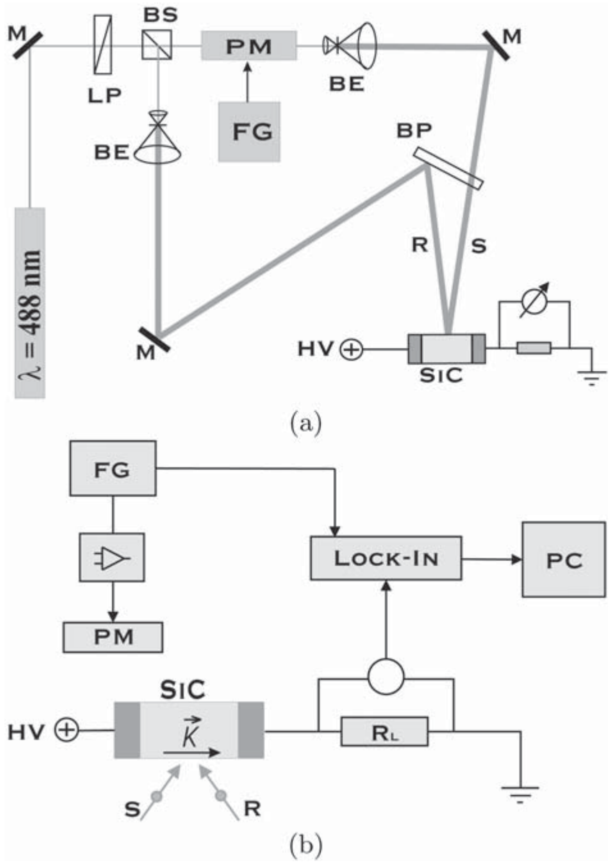

Fig. 1. (a) Scheme of the experimental setup, LP: combination of a half-wave plate and a polarizer, BS: beam splitter, R: reference beam, S: signal beam, PM: phase modulator, FG: function generator, BE: beam expander, BP: beam splitter plate, M: mirror, HV: high-voltage source. An Ar^+-laser at $\lambda = 488$ nm served as pump source. (b) Principle of electric detection of SCW in an external circuit; $\mathrm{R_L}$: loading resistor.

2 Samples and experimental setup

We performed measurements with two samples of hexagonal silicon carbide with 4H and 6H stacking sequence, respectively. These investigations allow us to compare the reproducibility of the SCW data for different samples having the rather similar crystallographic structure. Further, we can compare the data set with a sample from another source investigated in reference [7]. The studied samples differ with respect to the hexagonality which is 50% for the 4H-SiC and 33% for the 6H-SiC polytype. The hexagonality is defined as the ratio between the number of atoms in hexagonal positions and the total number of atoms in one unit cell. These two polytypes have different band gaps of 3.26 eV (4H-SiC) and 3.0 eV (6H-SiC), respectively. Both samples are quite transparent in the visible spectral range which indicates a high purity, i.e., a low trap concentration $(N_D - N_A) < 10^{16}$ cm^{-3} [12]. The dimensions of the samples are $4.4 \times 7.4 \times 0.52$ mm^3 (4H-SiC) and $3.7 \times 6.5 \times 0.62$ mm^3 (6H-SiC), respectively. For application of the electric field, electrodes were painted onto the samples surface with silver paste in a distance of $L = 4$ mm. Excitation of SCW was performed by illumination of the silicon carbide samples with a light interference pattern oscillating around an equilibrium position. Since the crystals exhibit photoconductivity and an external electric field is applied, the oscillating interference pattern generates a static and two counterpropagating space-charge gratings. Resonant excitation occurs if the oscillation frequency of the interference pattern coincides with the eigenfrequency of the SCW mode which possesses a spatial frequency equal to the light pattern.

The optical setup is shown in Figure 1(a). The light of an Ar^+-laser of $\lambda = 488$ nm is split into signal and reference beams. The phase of the signal beam is modulated with an electrooptic phase modulator, where the modulation frequency Ω and modulation amplitude Θ are adjusted by a function generator. The two beams are overlayed at the sample forming an oscillating interference pattern. Beam expanders in both arms of the two-beam interferometer provide the complete illumination of the investigated sample. The modulation depth m of the interference pattern can be varied by changing the intensity ratio of the two beams. Detailed information about the

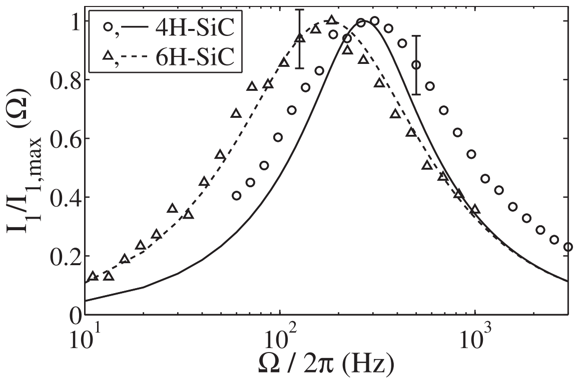

Fig. 2. Frequency dependence of the ac current for 4H-SiC and 6H-SiC with a total light intensity $W_0 = 125$ mW/cm^2, $E_0 = 3.5$ kV/cm, $K = 1.7 \times 10^3$ cm^{-1}, $\Theta = 0.3\pi$, and $m = 0.6$. The circles (○) and the triangles (△) are experimental data. The lines correspond to theoretical approximations in accordance with equation (1): solid line 4H-SiC; dashed line 6H-SiC. The fitting parameters used to calculate the theoretical curves are given in Table 1. Note that these parameters were used consistently in any of the following plots.

Table 1. The fitting parameters for 4H-SiC and 6H-SiC.

	4H-SiC	6H-SiC
$\mu\tau$	$(5.7 \pm 0.6) \times 10^{-7}$ cm^2/V	$(4.5 \pm 0.5) \times 10^{-7}$ cm^2/V
τ_M	$(1.8 \pm 0.2) \times 10^{-4}$ s	$(5.2 \pm 0.2) \times 10^{-4}$ s
N_{eff}	$(8.0 \pm 1.0) \times 10^{13}$ cm^{-3}	$(2.5 \pm 0.5) \times 10^{13}$ cm^{-3}

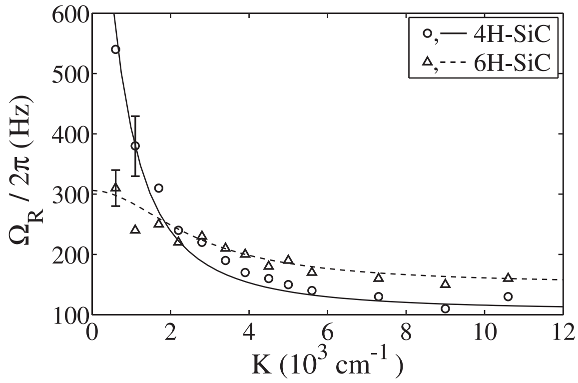

Fig. 3. Resonance frequency as a function of the spatial frequency K for 4H- and 6H-SiC with $E_0^{4H} = 3.5$ kV/cm, $E_0^{6H} = 1.0$ kV/cm, $W_0 = 125$ mW/cm^2, $\Theta = 0.3\pi$, and $m = 0.6$. The circles (○) and the triangles (△) are experimental values and the lines are theoretical approximations in accordance with (4).

principles of optical SCW excitation can be found in references [2,7]. The interaction of a running space-charge wave with the static periodical electric field formed by the static space-charge grating results in the appearance of spatial rectification. Hence, an alternating current arises which can be detected in an outside electric circuit. The advantage of this detection technique is, that the presence of the Pockels effect for an optical detection via light diffraction as used in photorefractive sillenites is not required [2]. Figure 1(b) shows the electric circuit for SCW detection via spatial rectification. The alternating current in the sample arising from spatial rectification is detected with the help of a loading resistor. The signal is fed into a Lock-In amplifier and is recorded as a function of the modulation frequency.

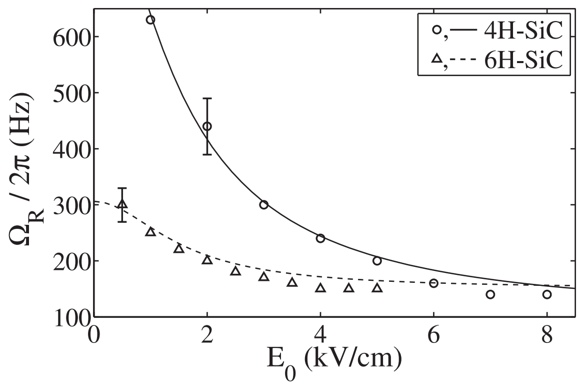

Fig. 4. Resonance frequency as a function of the applied electric field E_0 for 4H- and 6H-SiC with $W_0 = 125$ mW/cm^2, $K = 1.7 \times 10^3$ cm^{-1}, $\Theta = 0.3\pi$, and $m = 0.6$. The circles (○) and the triangles (△) are experimental values and the lines are theoretical approximations in accordance with (4).

3 Experimental results

Figure 2 shows typical resonance signals for the 4H-SiC and for the 6H-SiC sample, respectively. The amplitude of the ac current is normalized to its maximum value and is plotted as a function of the phase modulation frequency. The symbols denote experimental data whereas the lines are theoretical calculations in accordance with equation (1). The used fitting parameters are listed in Table 1. A dielectric constant of $\epsilon = 10$ was assumed for the calculations. The dependence of the resonance frequency on the spatial frequency K is shown for both polytypes in Figure 3. An external electric field of $E_0 = 3.5$ kV/cm (4H-SiC) and of $E_0 = 1.0$ kV/cm (6H-SiC) was applied. In Figure 4 the dependence of the resonance frequency on the applied electric field E_0 is shown for both samples. Both measurements were performed with a spatial frequency of $K = 1.7 \times 10^3$ cm^{-1}. Obviously, the resonance frequency decreases with increasing values of K as well as of E_0 for both samples investigated. This is in a good agreement with (4). The amplitude of the ac current at resonance is normalized to its maximum and plotted against the spatial frequency in Figure 5. The applied fields correspond to those in Figure 3. The appearing maximum at low values of K is caused by a low number of traps and allows to determine the effective trap density N_{eff}. Figure 6 shows the dependence of the normalized amplitude at the resonance frequency on the applied electric field. The same spatial frequency like in Figure 4 is chosen. The amplitude

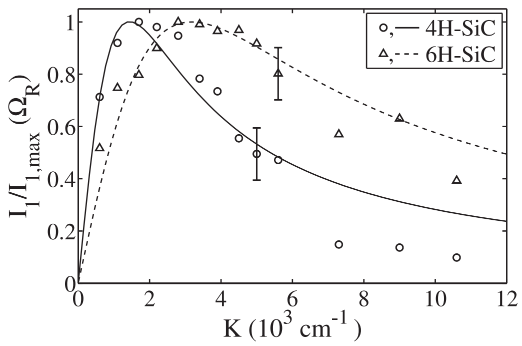

Fig. 5. Dependence of the ac current amplitude at the resonance frequency on the wave number K for 4H- and 6H-SiC with $E_0^{4H} = 3.5$ kV/cm, $E_0^{6H} = 1.0$ kV/cm, $W_0 = 125$ mW/cm^2, $\Theta = 0.3\pi$, and $m = 0.6$. The circles (○) and the triangles (△) are experimental values and the lines are theoretical approximations in accordance with (5).

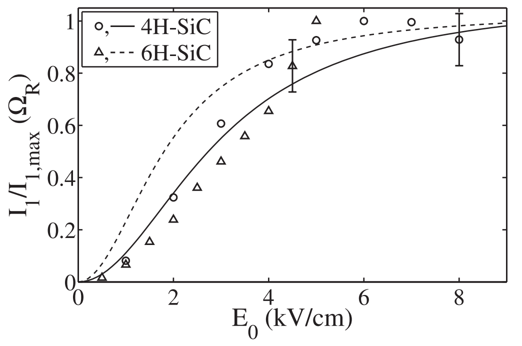

Fig. 6. Dependence of the ac current amplitude at the resonance frequency on the applied electric field E_0 for 4H- and 6H-SiC with $W_0 = 125$ mW/cm^2, $K = 1.7 \times 10^3$ cm^{-1}, $\Theta = 0.3\pi$, and $m = 0.6$. The circles (○) and the triangles (△) are experimental values and the lines are theoretical approximations in accordance with (5).

of the 4H-SiC sample increases with increasing field, but shows a saturation for $E_0 > 7$ kV/cm. This behavior correlates with the theoretical calculation, equation (5). The amplitude of 6H-SiC shows no saturation, but increases still at the highest applied field. The agreement with the theoretical curve is very poor.

4 Theoretical background

The experimental data show that the resonance frequency drops with increasing K for both samples. For a start we can assume that we are dealing with trap-recharging waves according to case (1). We therefore use the approximation $\beta \ll 1$ for our analysis, ignore the diffusion effect, consider the ratio $n/N_{\text{eff}} \ll 1$, ignore any screening effects, and consider $\tau/\tau_M \ll 1$, where τ_M is the Maxwell relaxation time. In this case, the detected current as a function of the modulation frequency is expressed by

$$I_1(\Omega) = \frac{\sigma E_0 m^2 \Theta \Omega}{2\omega\sqrt{[(1-\Omega/\omega)^2 + Q^{-2}][(1+\Omega/\omega)^2 + Q^{-2}]}}. \tag{1}$$

Here, σ is the conductivity.

$$\omega = \frac{1-\beta}{\tau_M d(1+d^{-2})}, \tag{2}$$

$$Q = \frac{d(1-\beta)}{1+\beta d^2}. \tag{3}$$

Additionally, in equation (1) the term correcting the frequency dependence at high frequencies is omitted for simplicity of our calculations (this correction is not important in the vicinity of the resonance). It follows from equations (1), (2) that the resonance frequency is described by

$$\Omega_R = \omega\sqrt{1+Q^{-2}}. \tag{4}$$

At the resonance frequency, the current reaches its maximum value with

$$I_1(\Omega_R) = \frac{1}{4}\sigma m^2 \Theta E_0 Q. \tag{5}$$

It follows from equations (3) and (5) that the intensity of the output signal has to exhibit a maximum as a function of K if

$$\beta d^2 = 1. \tag{6}$$

Equation (1) is very similar to equation (2) in reference [7]. However, unfortunately there are some misprints in [7]. Formulae (1)–(5) are correct for each of the three cases of the classification mentioned above, if the applied field is less than the saturation field. However, if the applied field is higher than the saturation field, the right part of the relationship (1) must be multiplied by a factor $1/(1+(E_0/E_q)^2)$ in accordance with the relationship (40) in [10]. This is of particular importance in the case $\beta \gg 1$. Here, the modulus of the frequencies and the quality factor must be used in (2)–(4).

5 Discussion

We will first discuss the results of the hexagonal silicon carbide sample with 4H stacking sequence. The experimentally determined dependences on the grating spacing and electric field are found in a good agreement with the presented theory and indicate the presence of TRW belonging to case (1). This assumption is confirmed by the estimated values for $\beta = (0.12 \pm 0.04) \ll 1$ and for the quality factor $Q = (1.26 \pm 0.19) > 1$, which fulfill the presumptions of the theoretical approach for ordinary TRW. The discrepancies for high K in Figure 5 can be explained by the mentioned presumptions ($d > 1$ and $E_0 < E_q$), which limit the interval for the existence of TRW to 0.5×10^3 cm^{-1} $< K < 4.4 \times 10^3$ cm^{-1} at a field of $E_0 = 3.5$ kV/cm. Furthermore, the material parameters $\mu\tau$, τ_M and N_{eff} deduced from our present analysis coincide within the experimental error very well with those obtained for a different 4H-SiC sample in reference [7]. In that publication, unambiguous dependences owing to TRW according to case (1) were found. So, we can state that TRW of case (1) are excited in the investigated silicon carbide sample with 4H stacking sequence. Moreover, it means that material parameters between samples from different sources show-up no significant difference. Therefore, the excitation of TRW with an inverse dispersion law could be expected for 4H silicon carbide of other sources as well.

A little bit more complicated is the case of the 6H-SiC sample. The dependences of the resonance frequency Ω_R on the spatial frequency K (see Fig. 3) and on the electric field E_0 (see Fig. 4) hint to an inverse dispersion behaviour. Further, the theory for ordinary TRW taking into account the effect of trap saturation describes very well the experimental data of this sample. In addition, significant similarities are obvious in the comparison between the data set for the 4H- and 6H-SiC samples: a clear resonance maximum (cf. Fig. 2) with a good signal-to-noise ratio showing an inverse dependence on K and E_0 can be detected in both samples. The conclusion of TRW of case (1) – which was unambiguously drawn for the 4H-SiC – therefore could be considered for the 6H-SiC sample, as well. Nevertheless, the obtained material parameters yield $\beta = (0.5 \pm 0.2) < 1$ and a maximal value $Q = (0.36 \pm 0.18) < 1$. The result $\beta < 1$ rather than $\beta \ll 1$ does not severely contradict the postulation for ordinary TRW of case (1), especially if the experimental error is considered. But the determined value of the quality factor does not reflect the necessary condition of $Q > 1$ for the existence of SCW, i.e., we have to exclude the existence of SCW. The experimentally determined signal therefore should be assigned to the presence of *damped, forced charge-density oscillations* according to case (3), and not to TRW of case (1).

This discrepancy between the interpretation at first sight and the quality factor less than unity enforces us to discuss the validity of the assumptions made to derive the formula set. First, only one type of charge carriers (electrons) was assumed to participate in the formation of TRW. This monopolar model looks quite reasonable for silicon carbide since it describes perfectly the experimental results. Furthermore, at the used wavelength ($E_{photon} < E_g$) excitation of two kinds of carriers might happen, but a monopolar conductivity is much more probable. In the case of band-to-band photoexcitation, the case of a bipolar conductivity definitely has to be taken into account for analysis. Then, the maximum of the resonance signal can exist not only due to the trap saturation effect, but as well due to the competition of two kinds of carriers (holes and electrons). However, this can be well excluded for SiC. A second assumption was to neglect diffusion processes. The contribution of this process can be estimated. At the highest spatial frequency used ($K = 10 \times 10^3$ cm^{-1}) the value of the diffusion field at room temperature is equal to $E_D \approx 250$ V/cm. For the same conditions, the trap saturation field is $E_q \approx 1.4$ kV/cm and the applied fields exceed 1 kV/cm. So, the diffusion field is less than E_q and E_0, and therefore it can be ignored in the first approximation. Of course at higher K, diffusion can play a more important role which reduces the quality factor. For 6H-SiC it is not such crucial, because even for $K = 3 \times 10^3$ cm^{-1} the quality factor is less than unity. A third assumption was to exclude the presence of the screening effect. Such phenomena are well-known for SCW due to non-ohmic contacts and were particularly found in reference [4] with InP:Fe. However, for SiC we can well exclude an influence of the screening effect, because the theoretical function making use of this assumption describes the experimental data with high accuracy, particularly for the 6H-SiC sample (cf. Fig. 2). The fourth assumption made is to assume $\tau/\tau_M \ll 1$. This assumption seems fulfilled with the determined values for the $\mu\tau$-product, τ_M, and common values of μ in 6H-SiC in the order of 450 cm^2(Vs)$^{-1}$ [12].

Taking these considerations into account and that the theoretical presumptions are fulfilled in a strict sense, we have to remark that the determined material parameters for the 6H-SiC sample represent valuable bench marks. The determination of these parameters with higher accuracy enforces to accommodate the formula set by considering the determined tendencies of the material parameters, i.e., a reduced effective trap density.

Taking into account the set of results on SiC from the present study together with our previous investigations with 4H-SiC in reference [7] we deduce a completed picture on the properties of SCW in SiC: obviously, the study of space-charge waves yields comparable results between SiC samples having the same stacking sequence, but taken from different sources. This is of interest from the point of view of material analysis since the parameters $\mu\tau$ and N_{eff} can be determined with an accuracy which is sufficient for many applications in semiconductors. However, a more careful analysis of the data set is required with SiC samples having a different stacking sequence, as it is demonstrated with the 6H-SiC sample. Although the existence of trap recharging waves from the data set might be concluded at a first sight, we recommend a careful analysis. We stress that the quality factor Q and the material parameter β represent necessary criteria. For an adequate statement, however, the assumptions made to derive the formula set for analysis should be checked in detail.

6 Conclusions

Excitation and analysis of space-charge waves were studied in silicon carbide with 4H and 6H stacking sequence. The effective trap concentrations and the $\mu\tau$ product were estimated from the experimental data considering the presence of trap recharging waves with an inverse dispersion law. The analysis remarkably yielded a quality factor less than unity for the 6H-SiC sample. The existence of space-charge waves therefore has to be excluded and only damped, forced charge-density oscillations can be exited in this material. A general classification of the material parameters providing the existence of trap-recharging SCW in semiconductors is presented.

Financial support from the Deutsche Forschungsgemeinschaft (DFG, project Nos. GRK 695 'nonlinearities of optical materials' and 436 RUS 17/15/07) is gratefully acknowledged.

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