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Evidence for capture of holes into resonant states in boron-doped silicon

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The variation of hole population in the resonant states of B-doped Si excited by sequences of short electric-field pulses has been investigated by the technique of time-resolved step-scan far-infrared spectroscopy. From the variation of the $p_{3/2}$ absorptions, we find that the hole population in the ground state decreases continuously with the sequential electric pulses, as a result of the breakdown delay and hole accumulation in long-lived excited states. The measured time-varying spectra of the $p_{1/2}$ series have been analyzed and attributed to a significant variation of the hole population in the resonant states. We have also observed a new absorption line at 676 cm^{-1} which is probably caused by the electric-field induced mixing of the resonant states. © 2004 American Institute of Physics. [DOI: 10.1063/1.1795985]

I. INTRODUCTION

The electronic states of an impurity in semiconductors are regarded as resonant states when they are degenerate in energy with one or more continua of the band structure, a notion originating from Fano.¹ One of the most common types of resonant states is the classical one of acceptor states that are associated with the spin-orbit $p_{1/2}$ bands and degenerate with the continua of the $p_{3/2}$ bands in cubic semiconductors. The existence of such resonant acceptor states was first verified by Zwerdling *et al.* more than four decades ago by observation of the $p_{1/2}$ excitation spectrum from B- and Al-doped Si.² With the advance in techniques of sample preparation and infrared spectroscopy the excitation spectra have been resolved more accurately and reassigned for the $p_{1/2}$ resonant states as well as the $p_{3/2}$ localized states of various Group III acceptors (B, Al, Ga, and In) in Si.^{3–6} Theoretical formulations for the resonant states have been developed and so far the calculated results can supply a quantitative interpretation for the measured absorption lines.^{7,8}

Although the problem of resonant states in semiconductors seems not new, the possibility of terahertz (THz) emission in doped semiconductors has renewed interests of researchers to it in the last decade since the THz radiation can be connected with the electric-dipole transition from one of the resonant states to a localized state.^{9,10} The stimulated emission and lasing operation of THz waves have been real-

ized from p type uniaxially deformed Ge with the electric field and the external stress being in parallel^{10–12} and also in crossing¹³ configurations. The THz gain has been attributed to the population inversion of holes between the resonant states attached to the heavy-hole bands and the localized states attached to the light-hole bands, where the capture of holes from continuous band states into the resonant states plays a key part.^{9,14} Based on an analogous mechanism, possibilities of THz radiation from p -doped strained SiGe (Ref. 15) and quantum wells with resonant impurity states¹⁶ have been proposed without the external applied stress. So far, however, there has not yet been any direct experimental evidence for the hole capture by the resonant states.

In this paper, we report an experimental evidence for capture of holes into the $p_{1/2}$ resonant states in B-doped silicon under sufficiently high electric-field pulses. The main idea of the experiment is to observe the time-varying absorptions of both the $p_{1/2}$ and $p_{3/2}$ series for B-doped Si excited by sequences of electric pulses, using the technique of time-resolved (TR) step-scan spectroscopy. This observation can reveal information on the evolution of hole populations in the various types of acceptor states. The reasons for using B-doped Si are as follows. The binding energy of the ground state of B ($\epsilon_B=45\text{ meV}$) is the smallest among those of the Group III acceptors in Si, rendering the holes in B-doped Si more likely to be ionized in a moderate electric field at a low temperature. Secondly, the oscillator strength of transitions associated with the $p_{1/2}$ lines is the largest for B compared with other Group III species in Si.⁸ Furthermore, the optical phonon energy ($\epsilon_{op}=63\text{ meV}$) for Si is larger than the spin-

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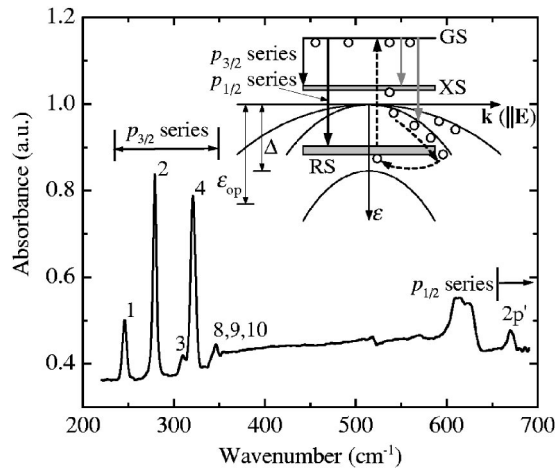


FIG. 1. The absorption spectrum for boron in silicon taken in the absence of external fields at 20 K. The inset illustrates the hole excitation and acceleration by the electric field, the hole capture into the resonant states, and the transitions for the $p_{1/2}$ and $p_{3/2}$ absorptions, where the abbreviations GS, XR, and RS stand for the ground states, the localized excited states, and the resonant states, respectively, of acceptors.

orbit split-off energy ($\Delta=44$ meV) between the $p_{1/2}$ and the $p_{3/2}$ bands. It follows that with a low impurity concentration and at a low temperature, there is a high probability for the ionized holes to gain sufficient kinetic energy ε in a high electric field to reach the energy of the resonant states before emitting an optical phonon, as illustrated in the inset of Fig. 1. In this situation the hole distribution in the momentum space will strongly displace from an isotropic one as a consequence of cyclic streaming motion in which optical phonon scattering is predominant.¹⁷ The average energy of the holes would be high enough and a considerable amount of holes can have kinetic energy in the range $\varepsilon_B < \varepsilon < \varepsilon_{op}$ so that they can play parts in the impact ionization of holes from the localized states. It is therefore possible for holes in B-doped Si under an appropriate electric field to achieve an ideal distribution for population inversion in such a way that the distribution function be narrow with its maximum at the energy of the resonant state while the localized states remain depopulated.^{9,14} Naturally, one would expect accumulation of holes in the resonant states through resonance scattering or other paths. Such a picture is difficult to achieve for Si doped with other species of acceptors that have a higher binding energy of holes. It is also hardly possible for p -type Ge that has a large spin-off energy Δ because the optical phonon scattering will prevent carriers from gaining enough kinetic energy to reach the energy of the resonant state. For polar semiconductors, strong acoustic phonon interaction would cause the distribution of carriers to be broadening in the momentum space, making them also unfit for the population inversion.

II. EXPERIMENT

The experiment was performed using samples with dimensions of 6×12 mm² cut along the [001] crystallographic axes from a 0.5 mm thick (100) B-doped Czochralski Si wafer of a room-temperature resistivity of 8Ω cm. The acceptor concentration in the samples is estimated to be about 1.5

$\times 10^{15}$ cm⁻³.¹⁸ Two contacts with area of 6×4 mm² were made using Al on one surface of the samples, leaving a central 6×4 mm² window for spectroscopic measurement. One of the samples was then treated for Ohmic contacts with annealing at 350°C for 30 min. The I-V curves measured from the sample in a pulse mode ensure that the contacts are Ohmic up to 1000 V. Each of the metallic contacts was then soldered together with a piece of $1 \times 18 \times 6$ mm³ copper using pure indium to improve the cooling efficiency of the sample in exchange gas of the continuous flow cryostat. Special design and tools were used to keep the indium thickness of being 0.45 mm. One of the copper holders was fixed to the finger of the cryostat while the other was left free. Such a scheme of making contacts and fixing the sample was used to alleviate the Joule heating and possible mechanical strain in Si. Rapid-scan spectra with resolution of 2.5 cm⁻¹ were taken from the sample in the aforementioned scheme and from another one mounted to the cryostat finger with a thin teflon film. No difference was found between the spectra in the absence of electric fields.

The cryostat with the sample was put into an IFS66v/S Fourier transform infrared (FTIR) spectrometer in such a way that the sample was capable of being irradiated by light from the FTIR source through the window between the contacts. In front of the sample was put symmetrically a screen with a 5×2.5 mm² hole, made of stainless foil covered by black paper to prevent the light from passing through the boundary sides into the sample. A He-cooled Si bolometer was used as a detector. Equipped with a polyethylene filter, a Mylar-3.5 μ m beam splitter, and KRS5 windows for the cryostat, the spectrometer allows one to measure spectra of interest in the range from 240 to 685 cm⁻¹.

To avoid overheating of the sample the electric field was applied in the burst mode. There were 50 pulses in a burst. The pulse duration was 0.2 μ s and the duty cycle was 0.01. The duration of each burst is thus 1 ms. The repetition rate of the bursts was 15.6 Hz. The Joule heating by the pulses led to a moderate increase in the environmental temperature from a temperature T_0 (measured in the absence of electric fields) to a higher temperature T^* . These temperatures were registered by a Ge thermoresistor with a time constant of 100 ms mounted on one of the copper pieces.

To increase the signal-to-noise ratio, 100 sets of data were recorded and coadded for each interferogram using the TR step-scan technique. There were 884 points in the interferogram demanded for an optical resolution of 2.5 cm⁻¹ and a phase resolution of 15 cm⁻¹. These parameters were a consequence of compromise between the resolution for distinguishing the absorption lines of interest and the time consumption for the measurement. Just before starting a real ac coupled TR measurement, we took a quick dc coupled TR interferogram in the absence of electric fields for further phase correction of the measured AC spectra. The obtained single-channel TR spectra $\Delta S(\nu, t)$ were further processed to get the corresponding delta absorbance spectra ΔA according to the formula

TABLE I. Some of parameters for three sets *A*, *B*, and *C* of spectra: the temperature (T_0) in the absence of electric fields, the average temperature (T^*), the applied voltage (V), the electric field (E), and the time resolution ($\Delta\tau$). The other parameters such as the pulse width ($0.2 \mu\text{s}$), the duty circle (0.01), and the number of pulses in a burst (50) are the same for the three sets of spectra.

Spectra	T_0 (K)	T^* (K)	V (V)	E (V/cm)	$\Delta\tau$ (μs)
A	20	27	740	1850	40
B	24	27	530	1300	40
C	45	53	500	1250	20

$$\Delta A(\nu, t) = -\ln \left[1 + \frac{\Delta S(\nu, t)}{S_0(\nu)} \right], \quad (1)$$

where $S_0(\nu)$, serving as a reference, is the ac coupled rapid-scan single-channel spectrum measured at T^* but in the absence of electric fields.

III. RESULTS AND DISCUSSION

The absorption line $2p'$ and lines 1, 2, 3, and 4 (in notation of Onton *et al.*³) which are of main interest in the present work can be well resolved. Figure 1 shows the spectra measured at 20 K in the absence of electric fields. The positions of the lines are in agreement with the data of Onton *et al.*³

We took three sets, *A*, *B*, and *C*, of electroabsorption spectra in the experiment. In Table I are listed some of the parameters adopted for measuring the three sets of spectra, such as the temperatures T_0 and T^* , the applied voltage V , the electric field E , and the time resolution $\Delta\tau$. The other parameters which are not listed, such as the pulse width ($0.2 \mu\text{s}$), the duty cycle (0.01), and the number of pulses (50), are the same for the three sets of spectra. The period of time slices at which the data were taken is set to equal the time resolution $\Delta\tau$. We took $\Delta\tau=40 \mu\text{s}$ for spectra *A* and *B*, and $20 \mu\text{s}$ for spectra *C*. It should be pointed out that in spite of the very short time duration $\Delta\tau$ compared with the time constant ($\sim 1 \text{ ms}$) of the bolometer, the high sensitivity of the bolometer and the wide dynamic range of a 16-bit analog-to-digital converter in FTIR allow us to observe the variation of optical signals with the time slices. The resulting electroabsorption spectra are shown in Fig. 2, where we show from the two sets *A* and *B* the spectra recorded at the first 11 time slices (slices 0–10) but from the set *C* the spectra at the first 11 each-third time slices (slices 0, 3, 6, ..., 30). Here, each of the first time slices, labeled with 0, was synchronized with the beginning of the bursts. We can see from the spectra that the values of delta absorbance ΔA of $p_{3/2}$ lines 1 (at 245 cm^{-1}), 2 (at 278 cm^{-1}), and 4 (at 319 cm^{-1}) are positive at the first several slices (slices 0–3 for spectra *A* and *B* and slices 0–9 for spectra *C*). This means that the absorptions measured at the first several time slices are larger than the reference absorption measured at T^* in the absence of electric fields. The larger absorption may be attributed to a higher concentration N_G of holes in the ground state or a lower concentration N_{LX} of holes in the localized excited states since the absorption is proportional to the difference $N_G - N_{LX}$ for the optical transition from the ground state to

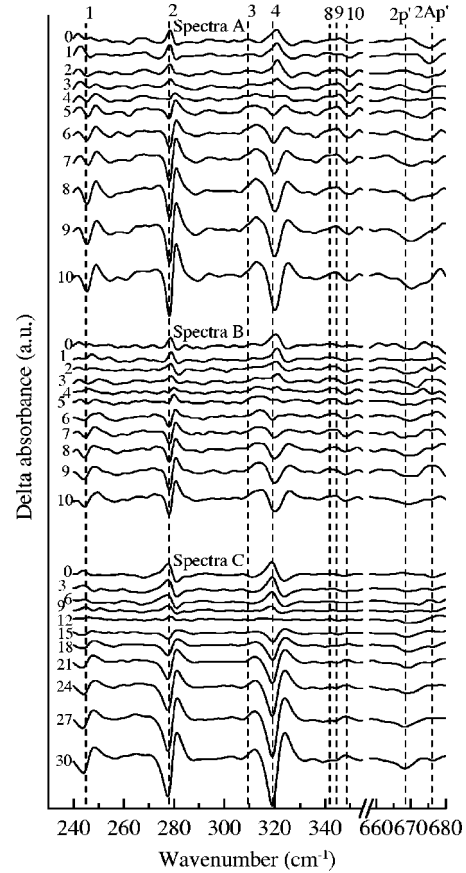


FIG. 2. The three sets, *A*, *B*, and *C*, of electroabsorption spectra taken at different time slices for boron in silicon excited by sequential electric-field pulses. The parameters for the three sets of spectra are listed in Table I.

the localized excited states. As will be demonstrated later, the lattice temperature T_L is dropping during the initial time slices as well as before the beginning of the bursts. This temperature dropping increases N_G while reduces N_{LX} before the first electric pulse is applied. The successive electric pulses can then remove holes from the localized acceptor states to shallower ones or to the continuous band states. Holes bound to shallower states are more likely ionized through either the Zener tunneling or the impact process when the electric field is applied. Only a smaller fraction of holes from the deeper ground state are excited by each of the short pulses of electric fields ($E=1850$, 1300 , and 1250 V/cm for spectra *A*, *B*, and *C*, respectively) due to the large separation in energy from the excited states.¹⁹ As a consequence, at the initial time slices when the free hole concentration p is small and thus the hole capture from the continuous band states is neglected, more holes are removed from a shallower acceptor state than from a deeper one by the electric pulses. The argument described above can also be supported by the difference in the magnitude of the delta absorbance of lines 1, 2, and 4 at the first time slices, where $\Delta A_4 > \Delta A_2 > \Delta A_1$ (ΔA_i being the delta absorbance of line i).

It is also a common feature of the spectra that the $p_{3/2}$ absorptions of lines 1, 2, and 4 decrease monotonically with the time slices. While this may be due to the increase in the lattice temperature T_L caused by the Joule heating of the sequential electric pulses, a further investigation of the tem-

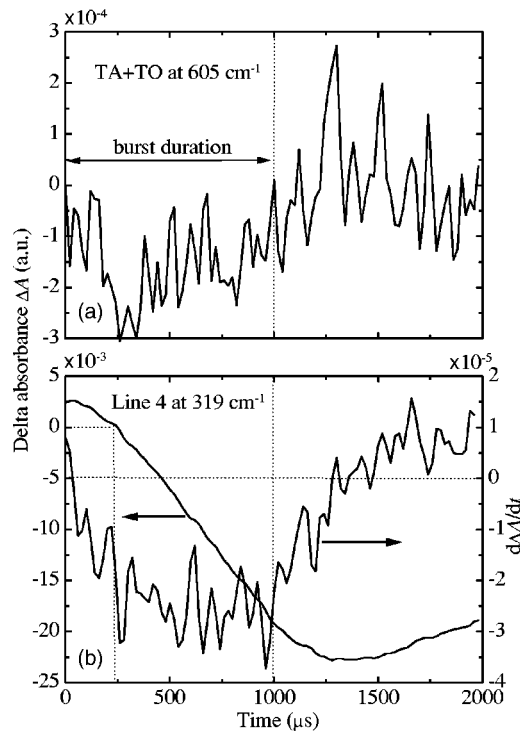


FIG. 3. The delta absorbance (a) for the multiphonon (TA+TO) process at 605 cm^{-1} and (b) for line 4 at 319 cm^{-1} as functions of time. Also shown in panel (b) is the derivative of the delta absorbance for line 4 with respect to time. The data were taken from spectra C at $T_0=45\text{ K}$, $T^*=53\text{ K}$, and $E=1250\text{ V/cm}$.

perature variation with time slices rules out the simple interpretation. In fact, T_L drops rather than rises during the first time slices, since there are a small amount of free holes available to generate Joule heating. This is also evident from Fig. 3(a) where the temporal variation of the delta absorbance at 605 cm^{-1} was recorded from spectra C. The absorption at 605 cm^{-1} has been known as the lattice absorption of bulk Si associated with the process of absorption of a photon and emission of two phonons (TA+TO).²⁰ Such a multiphonon absorption increases with T_L rising. With the aid of the temperature dependence of the absorption at 605 cm^{-1} , we can read from Fig. 3(a), in spite of the small signal-to-noise ratio, that the lattice temperature for spectra C still dropped during the initial $300\text{ }\mu\text{s}$ (corresponding to the first 15 time slices) but then rose until the temporal point approximately at $1250\text{ }\mu\text{s}$ which is $250\text{ }\mu\text{s}$ after the end of the bursts. The lag of the T_L variation to the switching of electric-field heating is obviously caused by the lower heat generation rate compared with the rate of heat drained by the cryostat. We expect similar temperature variations for spectra A and B although a significant variation of the absorption at 605 cm^{-1} was not observed due to the low temperature ($T_0/T^*=20\text{ K}/27\text{ K}$ and $24\text{ K}/27\text{ K}$ for spectra A and B, respectively).²⁰ On the basis of the preceding argument, we suggest that the variation of the $p_{3/2}$ absorptions are not simply dominated by the temperature variation. The decrease in the $p_{3/2}$ absorptions must result mainly from continuous depopulation of the holes in the ground state by the sequential electric-field pulses, accompanied with accumulation of holes in the excited states in the duration between two near-

est pulses. After an electric-field pulse passing, a majority of the excess free holes in the absence of electric fields are captured from continuous band states to localized excited impurity states in a short time and then would cascade to the ground state if the next electric pulse came a sufficiently long time later.²¹ However, the duration between two nearest pulses ($20\text{ }\mu\text{s}$) appears not long enough for the holes to complete a transition from the excited to the ground states; otherwise, the system would return to a thermal-equilibrium state before the next pulse came and, in this case, the absorption strength would simply be a function of temperature only. We conclude that there must exist long-lived (LL) excited states in B-doped silicon so that the hole accumulation in these states occurs in the duration between two nearest pulses. The existence of such long-lived states has also been experimentally confirmed by Lehto and Proctor¹⁹ and more recently by Pokrovskii *et al.*²² A small fraction of holes in the ground state as well as a majority of the holes in the excited states (including the LL states and other excited states) are then released to the band by the application of the next electric pulse. As a consequence, the ground state population N_G drops continuously with the time slices while the concentration p of free holes increases. (The values are average over the time resolution $\Delta\tau$.) Because of the high capture rate of holes from the continuous band states to the excited states, the excited state population N_{LX} is expected to be strongly related with p ; that is, N_{LX} also increases continuously with the time slices. The depopulation of the ground state is more enhanced for a higher electric field and also at a higher temperature, as can be seen from the three sets of absorption spectra A, B, and C. This is because a higher electric field can reduce the breakdown delay and excite more holes from the ground state in the short pulse duration.¹⁹ The efficiency of the impact excitation depends also upon the concentration of initial free holes in the continuous band states.¹⁹ At a higher temperature or at a latter time slice where there are more initial free holes, the population N_G decreases and N_{LX} increases more rapidly. This explains the more noticeable decrease in delta absorbance of lines 1, 2, and 4 at latter time slices and at a higher temperature (for spectra C). With the picture described, it is not difficult to understand the variations of ΔA_4 and $d\Delta A_4/dt$ with time, as shown in Fig. 3(b). The absorption ΔA_4 first slightly increases (with positive $d\Delta A_4/dt$) because the effect of T_L dropping dominates over that of the accumulation of LL holes which then becomes dominant. The heating is more and more important with the time since more and more free holes are available for generating heat. At $250\text{ }\mu\text{s}$ when the heat generated is balanced with that drained out, T_L reaches the minimum, as can be seen from Fig. 3(a). After this time, T_L then becomes increasing, enhancing the decrease of ΔA_4 as can be seen from Fig. 3(b). A very recent observation of THz electroluminescence from B-doped Si can also serve as an additional evidence for the existence of the long-lived excited states and the depopulation of the ground state with successive electric pulses.²³

Now let us turn our attention to the $p_{1/2}$ lines in Fig. 2 (see also Fig. 4 for clarity), which are associated with the electric-dipole transitions from the ground state to the reso-

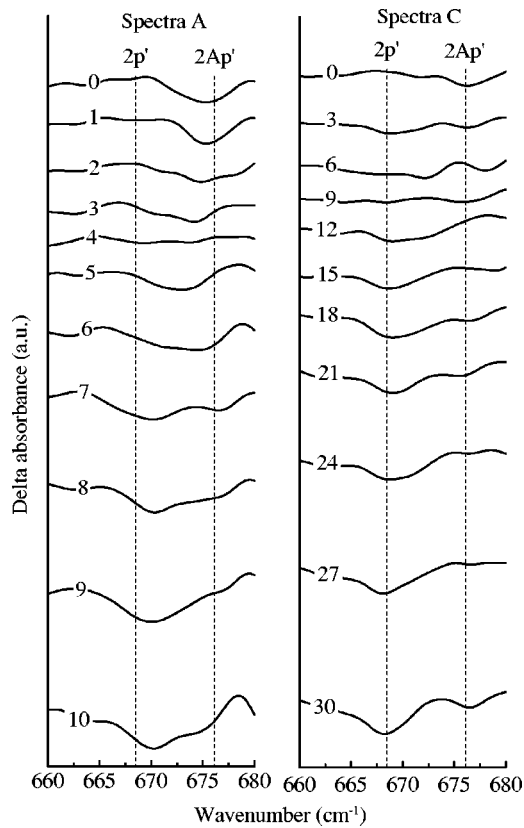


FIG. 4. The electroabsorption spectra in the range of large wave number. The left panel is taken from spectra A and the right panel is taken from spectra C in Fig. 2.

nant states. The $2p'$ line at 668 cm^{-1} and the $3p'$ line at 693 cm^{-1} have been observed for B-doped Si in the absence of electric fields,^{2,3,5} while the line marked $2Ap'$ at 676 cm^{-1} has never been reported. According to the calculated results of Buczko and Bassani,⁸ there are two energy levels close in energy to the first resonant state Γ_6^- ; one is the fourfold degenerate level Γ_8^- located at 0.08 meV (in electronic energy) below Γ_6^- and the other is a twofold degenerate level Γ_7^+ at 0.95 meV below Γ_6^- . The line $2Ap'$ has the wave number associated with the transition from the ground states (Γ_8^+) to the Γ_7^+ states, which is nearly forbidden in the absence of electric fields due to the same parity of the envelope functions of the Γ_8^+ and Γ_7^+ states. (Note that the total wave functions lack a definite parity since the system lacks the inversion symmetry.) However, under an electric field applied along the [001] direction, the symmetry of the system reduces from the T_d group to the D_{2d} group. Consequently, the three energy levels of the resonant states $\Gamma_6^- + \Gamma_7^+ + \Gamma_8^-$ are hybridized together and split into four energy levels $2\Gamma_6 + 2\Gamma_7$ (Γ_6 and Γ_7 being the irreducible representations of D_{2d}) in a sufficiently strong electric field. Based on the estimation $\langle r \rangle eE > 1.5\text{ meV}$, where $\langle r \rangle > 150\text{ \AA}$ is the mean radius of the resonant states⁸ and e the electronic charge, we expect a considerable mixing of the resonant states by the external fields. In this situation, the selection rule is no longer valid and the variation of the $2Ap'$ absorption becomes observable, as Fig. 2 shows. In contrast to the $p_{3/2}$ absorptions, the $p_{1/2}$ absorptions are smaller at almost all the time slices than the reference absorption measured in the absence of electric

fields because of the nonpositive delta absorbance of the $p_{1/2}$ lines. This means that the T_L dropping and the electric fields at the initial time slices do not cause a decrease in the hole population of the resonant states. Instead, the population in the resonant states increases since the negative delta absorbance of the $2Ap'$ line cannot be explained simply by the decrease in the ground state population N_G . As has been described previously, N_G decreases during the first time slice because of the application of the electric pulses. It is an important finding that the negative delta absorbance moves from $2Ap'$ to $2p'$ with the time slices. We attribute the phenomenon to a transfer of the excess captured holes from the high-energy resonant state to the low-energy one or probably to the $p_{3/2}$ band states through resonant escape. The negative delta absorbance of the $2p'$ line at latter time slices can be explained by the decrease in the ground state population or the increase in the population of the first resonant state. Such relaxation processes involving the resonant states appear complicated and require further experimental and theoretical investigations for identification.

IV. CONCLUSION

In conclusion, we have observed the variations of the hole populations in the ground, resonant, and localized excited states for B-doped Si under trains of electric-field pulses by the technique of TR far-infrared electroabsorption spectroscopy. The ground state population decreases monotonically with the successive short electric pulses because of the breakdown delay and the hole accumulation in the long-lived excited states. We have also observed the $2Ap'$ absorption line at 676 cm^{-1} which is believed to be caused by the electric-field induced mixing of the resonant states. The population of excess holes in the high-energy resonant state has been observed. As time goes on, the holes may transfer to the low-energy state or escape to the continuous band states.

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