Temperature and carrier-density dependence of electron-hole scattering in silicon investigated by optical-pump terahertz-probe spectroscopy

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We measured the optical conductivity $\tilde{\sigma}(\omega)$ spectra of photodoped silicon by optical-pump terahertz-probe spectroscopy and analyzed them with a two-carrier Drude model. Taking into account the values of electron (hole)-phonon scattering rates previously reported in chemically doped silicon, we evaluated the electron-hole scattering rates γ_{e-h} . From 293 to 90 K, the magnitudes and temperature dependence of γ_{e-h} were successfully reproduced by a theoretical model including the effects of Rutherford scattering, Coulomb screening, and Pauli exclusion. This suggests that these three factors dominate electron-hole scattering processes in silicon. Below 90 K, γ_{e-h} becomes larger than that of the theoretical curve, which is attributable to a prolongation of the relaxation time of hot carriers.

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Carrier dynamics in electron-hole systems has been one of the central issues in the physics of semiconductors [1-7], not only for the emergence of interesting physical phenomena such as the exciton-Mott transition [1,2] and Bose-Einstein condensation of excitons [3], but also for the development of modern optoelectronic devices. In conventional inorganic semiconductors, the scattering rates of the free carriers and energy intervals of exciton levels (or exciton binding energies) fall in the terahertz frequency region. Recent developments in terahertz time-domain spectroscopy (THz-TDS) have enabled us to observe the dynamics of photogenerated carriers and/or excitons. For example, in silicon, the exciton binding energy and energy splitting between 1s and 2p excitons are 14.7 meV (\sim 3.5 THz) and 10 meV (\sim 2.4 THz), respectively, so that induced absorption due to excitons as well as Drudetype responses of photogenerated carriers can be directly detected by a combination of the pump-probe (PP) method and THz-TDS [3,6,7]. Using such an approach, the exciton-Mott transition has been indeed demonstrated [3,4].

When we consider applications of semiconductors to optoelectronic devices, it is significant to elucidate the scattering mechanisms of photocarriers. A mobility of carriers is determined by a scattering rate γ via the simple equation $\mu = e/m^*\gamma$, where m^* and e are an effective mass and the elementary charge, respectively. For chemically doped carriers, scatterings are known to consist of two processes: carrier-phonon scatterings and carrier-ionized-impurity scatterings [8]. On the other hand, for photocarriers, electron-hole scatterings would play significant roles in addition to carrier-phonon scatterings [9–13]. However, experimental studies clarifying the mechanism of electron-hole scatterings are lacking.

In the present study, in order to clarify the mechanisms of electron-hole scattering in semiconductors, we applied optical-pump terahertz-probe spectroscopy on silicon, which is a typical indirect-gap semiconductor [Fig. 1(a)] [14], and determined its complex optical conductivity $\tilde{\sigma}(\omega)$ spectra due to photocarriers. From the analyses of $\tilde{\sigma}(\omega)$ spectra with a twocarrier Drude model, we evaluated the electron-hole scattering rate γ_{e-h} at various temperatures and photocarrier densities. By comparing the temperature dependence of γ_{e-h} with that deduced from a theoretical model including the effects of Rutherford scatterings, the screening of Coulomb interactions, and Pauli exclusion [9–12], we discuss the mechanism of electron-hole scatterings in silicon.

As a sample, we used a nondoped silicon single crystal with the thickness of 1 mm, whose resistivity is larger than $1000 \,\Omega$ cm at 293 K. For optical-pump terahertz-probe spectroscopy, we utilized as the light source a Ti:sapphire regenerative amplifier with a pulse energy of 2.4 mJ, a central photon energy of 1.55 eV, a pulse duration of 25 fs, and a repetition rate of 1 kHz. The output from the amplifier was divided into two beams. One was used for the generation of pump pulses (1.77-2.53 eV) using an optical parametric amplifier. The other was used for the generation and detection of THz probe pulses. We employed air-plasma-induced terahertz radiation in order to obtain broadband (0.8-7.2 THz) pulses [15,16]. Temporal wave forms of THz probe pulses transmitted through the sample were measured by electro-optical (EO) sampling with a 300- μ m-thick GaP crystal, in which the detectable probe range is 0.8-7.2 THz. In order to obtain $\tilde{\sigma}(\omega)$ spectra, we alternately measured wave forms of transmitted THz pulses without a pump pulse E(t) [Fig. 1(b)] and with a pump pulse $E(t) + \Delta E(t)$ [Fig. 1(c)] by using two optical choppers. We extracted $\tilde{\sigma}(\omega)$ using the following equation [17–19],

$$\tilde{\sigma}(\omega) = -\frac{\epsilon_0 c (n_0 + 1)}{L} \frac{\Delta E(\omega)}{\tilde{E}(\omega)},$$
(1)

where ϵ_0 is the vacuum permittivity, *c* the velocity of light, n_0 the refractive index of silicon in the terahertz frequency region $(n_0 = 3.4)$, and *L* the penetration depth of pump pulses. *L* is obtained from the absorption-coefficient (α) spectra [20,21]. $\tilde{E}(\omega)$ and $\Delta \tilde{E}(\omega)$ are complex Fourier transforms of E(t) and $\Delta E(t)$, respectively. Typical wave forms of E(t) and $E(t) + \Delta E(t)$ and their relative Fourier components $[|\Delta \tilde{E}(\omega)/\tilde{E}(\omega)|$ and $\Delta \phi(\omega)$] are presented in Figs. 1(d) and 1(e), respectively. The delay time t_d of a THz probe pulse [the maximum of |E(t)|] relative to a pump pulse was adjusted by changing the path length of the pump pulse.

Figure 2(a) shows $\tilde{\sigma}(\omega)$ spectra at 293 K, which were obtained by a 1.77 eV pump (the excitation density $x_{\rm ph} = 1.3 \times 10^2 \,\mu \text{J/cm}^2$ [22]) at $t_{\rm d} = 2$ ps, when a pump pulse and a THz pulse are completely separated in the time domain. To



FIG. 1. (Color online) (a) Band structure of silicon [14]. (b), (c) Schematics of optical-pump terahertz-probe spectroscopy (b) without and (c) with a pump pulse. (d) Terahertz electric-field wave forms with $[E(t) + \Delta E(t)]$ and without [E(t)] a pump pulse. (e) Transient complex transmissivity $[|\Delta \tilde{E}(\omega)/\tilde{E}(\omega)|$ and $\Delta \phi(\omega)]$ spectra due to photocarriers.

analyze $\tilde{\sigma}(\omega)$ spectra, we adopt a two-carrier Drude model,

$$\tilde{\sigma}(\omega) = ne^2 \left(\frac{1}{m_{\rm e}} \frac{1}{\gamma_{\rm e} - i\omega} + \frac{1}{m_{\rm h}} \frac{1}{\gamma_{\rm h} - i\omega} \right), \qquad (2)$$

where n is carrier density of the electron or hole, $m_{\rm e}$ (m_h) is the effective mass of the electron (hole), and γ_e (γ_h) is the scattering rate of the electron (hole). γ_e (γ_h) is the sum of an electron-hole scattering and a carrier-phonon scattering according to the Mathiesen's law, $\gamma_e = \gamma_{e-h} + \gamma_{e-p}$ ($\gamma_h =$ $\gamma_{e-h} + \gamma_{h-p}$), where γ_{e-p} (γ_{h-p}) is an electron (hole)-phonon scattering rate. From mobility values of chemically doped silicon with low carrier concentrations of 1.0×10^{14} cm⁻³ for an electron and $1.0 \times 10^{15} \text{ cm}^{-3}$ for a hole at room temperature [8], we deduced γ_{e-p} and γ_{h-p} using the general formula $\mu =$ $e/m^*\gamma$. For the effective masses, we used literature values, $m_{\rm e} = 0.26m_0$ and $m_{\rm h} = 0.37m_0$ (m_0 is the free electron mass) [23]. Thus, we fit Eq. (2) to the experimental $\tilde{\sigma}(\omega)$ data with two fitting parameters, n and γ_{e-h} . We neglected scatterings between the same kinds of carriers (electron-electron and hole-hole scatterings). These scatterings do not contribute to the relaxation of currents, because the momentum is conserved during the scattering processes [12,13]. In Fig. 2(a), dashed lines are fitted curves with $\gamma_{e-h}/2\pi = 3.9$ THz and n = 2.6×10^{17} cm⁻³, which reproduce well the $\tilde{\sigma}(\omega)$ spectra. The responses of electrons and holes are shown by different colors. From the values of γ_{e-h} , the mobilities of the electrons and holes are deduced to be $\mu_e = e/m_e\gamma_e = 2.3 \times 10^2 \text{ cm}^2/\text{V} \text{ s}$ and $\mu_{\rm h} = e/m_{\rm h}\gamma_{\rm h} = 1.4 \times 10^2 \text{ cm}^2/\text{V} \text{ s}$, respectively.

In the analyses presented above, we did not take into account the spatial diffusions of photocarriers, which should give rise to errors in the evaluated values of *n* and γ_{e-h} . To investigate the carrier diffusion effects, we measured the excitation-photon-energy (E_{ex}) dependence of γ_{e-h} at 293 K

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FIG. 2. (Color online) (a) $\operatorname{Re}(\tilde{\sigma})$ and $\operatorname{Im}(\tilde{\sigma})$ spectra in silicon measured with a 1.77 eV pump at 293 K. Dashed lines indicate fitting curves. The responses of electrons and holes are shown by shades. (b) Carrier-density dependence of γ_{e-h} . Penetration depth for each pump energy is shown in the inset [20]. (c) $\operatorname{Re}(\tilde{\sigma})$ and $\operatorname{Im}(\tilde{\sigma})$ spectra for different pump photon densities. Crossing points correspond to $\bar{\gamma}/2\pi$. Dashed lines are fitting curves. (d) Carrier-density dependence of γ_{e-h} . Electron-ionized-impurity scattering γ_{e-D} in *n*-type silicon is shown by the solid line [8].

and $t_d = 2$ ps, which is shown in Fig. 2(b). γ_{e-h} changes depending on E_{ex} , which is attributable to the presence of carrier diffusions. With an increase of E_{ex} , *L* becomes shorter [the inset of Fig. 2(b)] [20], leading to the enhancement of carrier diffusion effects. However, the 1.65 and 1.77 eV excitations give the same γ_{e-h} values at the same *n* values, so that carrier diffusions can be neglected for these energies at $t_d = 2$ ps. Thus, we selected $E_{ex} = 1.77$ eV and $t_d = 2$ ps for the measurements of $\tilde{\sigma}(\omega)$ [24].

Next, we discuss the carrier-density dependence of $\tilde{\sigma}(\omega)$ in more detail. Figure 2(c) shows $\tilde{\sigma}(\omega)$ spectra at $t_d = 2$ ps for three carrier densities, $n = 2.6 \times 10^{17}$, 1.3×10^{17} , and 5.2×10^{16} cm⁻³. With an increase of the carrier density, the crossing point of the real and imaginary parts which corresponds to the average carrier scattering rate $\bar{\gamma}/2\pi$ shifts to a higher energy. Figure 2(d) shows the carrier-density dependence of γ_{e-h} . In the low carrier-density region, γ_{e-h} increases linearly with carrier density, reflecting the increase in the encounter rate of an electron and a hole. This trend is consistent with the previous result reported by Hendry *et al.* [13]. In the high carrier-density region, γ_{e-h} deviates from the linear dependence and tends to saturate, which is attributable to the screening of

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FIG. 3. (Color online) (a) Temperature dependence of $\tilde{\sigma}(\omega)$ for the 1.77 eV pump ($n = 2.0 \times 10^{17} \text{ cm}^{-3}$). Dashed lines indicate fitting curves. (b) Temperature dependence of the penetration depth *L* at 1.77 eV [21]. (c) Temperature dependence of γ_{e-p} and γ_{h-p} (see text) [26,27]. Dashed lines are extrapolated curves. (d) Temperature dependence of γ_{e-h} .

the Coulomb interaction between the electron and hole due to other carriers. Such a behavior is consistent with the theory reported by Combescot et al. [9,10]. The electron-density (i.e., dopant-density) dependence of the electron-ionized-impurity scattering rate γ_{e-D} in phosphorus-doped *n*-type silicon is also shown by a solid line in Fig. 2(d) [8]. γ_{e-D} is smaller than a half of γ_{e-h} . In the case of chemically doped samples, ionized impurities are fixed at lattice points and cannot move around. This causes a decrease in the encounter rate between the electron (hole) and donor (acceptor) compared to the case of photogenerated electrons and holes, both of which are mobile. Thus, the scattering rate between the electron (hole) and donor (acceptor) is smaller than that between photogenerated electrons and holes. As a result, at the same carrier density, the mobilities of the photocarriers become smaller than those of carriers injected by elemental substitutions [25].

To investigate the mechanism for electron-hole scatterings, which is the main subject of the present study, we measured the temperature dependence of γ_{e-h} . In this experiment, we adjusted x_{ph} so that the carrier density was constant (n = 2.0×10^{17} cm⁻³) at all temperatures, taking into account the temperature dependence of the penetration depth L [Fig. 3(b)] [21]. Figure 3(a) shows $\tilde{\sigma}(\omega)$ spectra at three typical temperatures, 270, 120, and 30 K. $\bar{\gamma}/2\pi$ increases with decreasing temperature. To determine γ_{e-h} , we calculated γ_{e-p} and γ_{h-p} using the temperature dependence of the mobilities in n-type silicon with a dopant density of 1.2×10^{14} cm⁻³ [26] and in *p*-type silicon with a dopant density of 4.5×10^{14} cm⁻³ [27], which are shown in Fig. 3(c). The dashed lines are freehand extrapolations. The error of the extrapolation does not affect the analysis because γ_{e-p} and γ_{h-p} are almost equal to zero in this temperature region. Using these values, we obtained the



FIG. 4. (Color online) (a) Schematics of (i) Rutherford scatterings, (ii) screening of Coulomb interactions at low temperatures (low *T*) and high temperatures (high *T*), and (iii) Pauli exclusion at T = 0 K and $0 < T \ll T_F$. At T = 0 K, electron-hole scatterings are prohibited. (b) Open circles show the temperature dependence of γ_{e-h} shown in Fig. 3(d). Solid circles show theoretical values obtained from Eqs. (6) and (7). Values in classical and quantum limits are also shown by dotted lines.

temperature dependence of γ_{e-h} [Fig. 3(d)], which increases monotonically as the temperature decreases.

To interpret the temperature (T) dependence of γ_{e-h} , we refer to the theoretical study reported by Combescot et al., in which the scattering mechanism was discussed in two limiting cases: $T \gg T_F$ (classical limit) and $T \ll T_F$ (quantum limit). Here, $T_{\rm F}$ is the Fermi temperature $[T_{\rm F} = \hbar^2/2mk_{\rm B}(\frac{3\pi^2 n}{6})^{2/3}]$, where \hbar is the reduced Planck constant and $k_{\rm B}$ is the Boltzmann constant. In the classical limit, the carriers are regarded as classical particles and γ_{e-h} is dominated by Rutherford scatterings caused by Coulomb interactions and the screening of Coulomb interactions. The Rutherford scattering is a general mechanism of the scattering between charged particles, which is proportional to $T^{-3/2}$ [9]. With decreasing temperature, a particle is easily captured by an oppositely charged carrier due to the reduction of its velocity [Fig. 4(a)(i)], so that γ_{e-h} increases. At low temperatures, however, the screening effect is enhanced by carrier-velocity reductions and rather suppresses γ_{e-h} as $\gamma_{e-h} \propto \ln T$ [Fig. 4(a)(ii)]. Taking these two mechanisms into account, the temperature dependence of γ_{e-h} can be calculated by the Boltzmann approach [9,10] as

$$\gamma_{\text{e-h}} = \frac{16}{9\pi^{3/2}} k_{\text{B}} T_{\text{Ryd}} \left(\frac{T_{\text{F}}}{T}\right)^{3/2} \ln\left(\frac{T^2}{T_{\text{Ryd}}^{1/2} T_{\text{F}}^{3/2}}\right).$$
(3)

Here, $k_{\rm B}T_{\rm Ryd}(=me^4/2\hbar^2\epsilon^2)$ corresponds to the exciton binding energy, which scales the Coulomb interaction between an electron and a hole. $m = (\frac{1}{m_{\rm e}} + \frac{1}{m_{\rm h}})^{-1}$ and ϵ is the static permittivity of silicon. The calculated curve in our experimental condition [Fig. 3(d)] is shown by the red dotted line in Fig. 4(b) [28].

In the quantum limit ($T \ll T_F$), carriers degenerate into the bottom of the band (Fermi degeneracy). Due to the energy conservation law, the interaction between an electron and a hole is reduced and γ_{e-h} goes to zero at T = 0 K [Fig. 4(a)(iii)]. For $0 < T \ll T_F$, carriers around the Fermi surface can be excited and scatter each other. The number of these carriers is proportional to T, so that $\gamma_{e-h} \propto T^2$ [Fig. 4(a)(iii)]. As a result, γ_{e-h} is written by the following equation [11],

$$\gamma_{\text{e-h}} = \frac{1}{6} k_{\text{B}} T_{\text{Ryd}} \left(\frac{T}{T_{\text{F}}} \right)^2 \left(x \tan^{-1} x - \frac{x^2}{1 + x^2} \right) \pi \nu^{4/3} \left(\frac{m_{\text{e}}^2 m_{\text{h}}^2}{m^4} \right),$$
(4)

$$x = \frac{\nu^{1/6} \pi^{1/2} m^{1/2}}{\left(\nu m_{\rm e} + \nu^{1/3} m_{\rm h}\right)^{1/2}} \left(\frac{T_{\rm F}}{T_{\rm Ryd}}\right)^{1/4},\tag{5}$$

where ν is the conduction-band degeneracy and is equal to 6 in silicon [8]. The temperature dependence of γ_{e-h} calculated by Eq. (4) for $0 < T \ll T_F$ is denoted by the blue dotted line in Fig. 4(b).

Around $T \sim T_{\rm F}$, both classical and quantum effects should be considered. Sernelius calculated $\gamma_{\rm e-h}$ in this intermediate region by utilizing the generalized Drude model and Kubo formula [11,12,29]. According to his calculation,

$$\sigma(0) = \frac{e^2}{\eta_{\text{e-h}}} = \frac{ne^2}{m\gamma_{\text{e-h}}},$$
(6)

$$\eta_{\text{e-h}} = \frac{6\hbar^2\beta}{n^2} \int_0^\infty \frac{d\omega}{2\pi} \frac{1}{\sinh^2\frac{\hbar\beta\omega}{2}} \\ \times \int \frac{d\boldsymbol{q}}{(2\pi)^3} q_{\text{u}}^2 \frac{\text{Im}\alpha_{\text{e}}(\boldsymbol{q},\omega)\text{Im}\alpha_{\text{h}}(\boldsymbol{q},\omega)}{|1+6\alpha_{\text{e}}(\boldsymbol{q},\omega)+\alpha_{\text{h}}(\boldsymbol{q},\omega)|^2}, \qquad (7)$$

where $\beta = 1/k_{\rm B}T$ and $\alpha_{\rm e}$ ($\alpha_{\rm h}$) is the polarizability of electron (hole) obtained by a random phase approximation [30]. q and $q_{\rm u}$ are the momentum and its projection on the direction of the electric field, respectively. Only the imaginary part of the polarizability can be obtained analytically, so that we calculated the real part with the use of the Kramers-Kronig relation and obtained the temperature dependence of $\gamma_{\rm e-h}$ from Eqs. (6) and (7) for our experimental condition (n = 2.0×10^{17} cm⁻³), which is shown by solid circles in Fig. 4(b). This curve has a peak at around $T \sim T_{\rm F}$ and coincides with the

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curves of classical and quantum limits for $T \gg T_{\rm F}^{\rm h}$ (the Fermi temperature of holes) and $T \ll T_{\rm F}^{\rm e}$ (the Fermi temperature of electrons), respectively [31].

In Fig. 4(b), we showed again the experimental γ_{e-h} data of Fig. 3(d) by open circles. For $T \gg T_F^h$, the absolute values of γ_{e-h} as well as their temperature dependence are well reproduced by the theoretical curve. This demonstrates that electron-hole scattering is explained by the above-mentioned three mechanisms. Below 60 K, the theoretical curve turns downward, reflecting the quantum effect, but the experimental γ_{e-h} does not show such a decrease.

A possible origin for the deviation of the experimental γ_{e-h} from the theoretical one is an excess energy of the photocarriers, since $E_{ex}(1.77 \text{ eV})$ was 0.65 eV larger than the indirect gap of silicon (1.12 eV) [Fig. 1(a)]. The previous two-photon photoemission spectroscopy revealed that at 296 and 90 K, photocarriers are rapidly relaxed and thermally equilibrated to phonons at $t_d = 2$ ps [32,33]. Judging from these results, we can consider that the increase of temperature at that time is very small (<5 K) above 90 K. This is the reason why the γ_{e-h} values evaluated at $t_d = 2$ ps are in accord with the theoretical values at $T \gg T_{\rm F}^{\rm F}$.

On the other hand, it was suggested from optical-pump terahertz-probe spectroscopy that it takes several hundred picoseconds for the carriers to relax below 60 K [5]. Such a long relaxation time is attributable to small populations of phonons. Therefore, it is reasonable to consider that below 60 K, photocarriers at $t_d = 2$ ps have finite kinetic energies or equivalently high temperatures. This would be the reason why the experimental data are larger than theoretical ones.

In summary, we measured optical conductivity $\tilde{\sigma}(\omega)$ spectra due to photocarriers in nondoped silicon by optical-pump terahertz-probe spectroscopy. By analyzing $\tilde{\sigma}(\omega)$ spectra with the two-carrier Drude model, electron-hole scattering rates γ_{e-h} were evaluated. From 293 to 90 K, the absolute values and temperature dependence of γ_{e-h} were well reproduced by the theoretical curve, in which Rutherford scattering, Coulomb screening, and Pauli exclusion were taken into account. This demonstrates that these three factors can explain electron-hole scattering processes in silicon.

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