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Far-Infrared Spectroscopy of the Coulomb Gap in Compensated Semiconductors

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We report on the observation of correlated to random distribution of ionized impurities in strongly compensated n-type Ge as a function of the dopant concentration and temperature.[1] The transition occurs when thermal energy exceeds the Coulombic correlation energy between impurities, i.e., it corresponds to thermal smearing of so-called Coulomb gaps.[2]

Let us first define the problem of our interest by considering a n-type semiconductor with the concentration of hydrogenic donors (N_D) being twice of that of hydrogenic acceptors (N_A); $N_D = 2N_A$. At sufficiently low temperatures, one half of N_D is positively charged ($\frac{1}{2}N_D = N_D^+$) because their bound-electrons are taken away by acceptors. These become negatively charged after accepting electrons ($N_A = N_A^-$). The remaining half of N_D binds electrons so that their charge state is neutral ($\frac{1}{2}N_D = N_D^o$). This system is interesting because the ionized donors (D^+) can modify their distribution with respect to the fixed position of ionized acceptors (A^-) via transferring of electrons between neutral (D^0) and ionized (D^+) donors. Therefore, the distribution of the ionized donors can be either random or correlated depending on the impurity concentration and temperature. For the case of the sufficiently high impurity concentration and low temperatures, the Coulomb gaps have been probed successfully by tunneling experiments.[3-5] However, it is not convenient to perform the tunneling experiment for the low concentration region ($\sim 1 \times 10^{14}$cm^{-3}) in order to probe the possible transition from the random to correlated ionized impurity distributions. Far-infrared spectroscopy of shallow impurities allows for such probing as we demonstrate in this paper. For many years the electric-field broadening of the optical absorption peaks of hydrogenic-impurities has been investigated theoretically by assuming a random distribution of ionized impurity centers.[6] However, it has been pointed out that the ionized impurity distribution at low temperatures is correlated rather than random in order to minimize the total Coulombic energy of the system, i.e., formation of the Coulomb gap takes place.[7] The correlated distribution has been observed in our previous investigation using heavily compensated p-type Ge(Ga;As) samples.[8]

This work probes the n-type Ge:As,Ga with the ionized impurity concentration $N_I < 1 \times 10^{15}$cm^{-3}. The inset of Fig. 1 shows the absorption spectrum of a sample having $N_D = 3.03 \times 10^{13}$cm^{-3}, $N_A = 1.75 \times 10^{13}$cm^{-3}, i.e., $N_I \approx 2N_A = 3.50 \times 10^{13}$ cm^{-3}. It has been recorded at T = 4 K with a resolution of 0.026 cm^{-1} in the wavenumber range between 70 and 110 cm^{-1}. Three distinct peaks correspond to excitations of bound electrons of As in Ge from ground state to 2p$_0$, 2p$_\pm$, and 3p$_\pm$ excited states, respectively. The main frame of Fig. 1 shows from bottom to top the enlargement of the 1s to 2p $_\pm$ transition peaks; the experimental result (open circles), Monte Carlo simulation assuming random distribution of ionized impurities (open squares), and Monte Carlo simulation assuming correlated distribution of impurities (open triangles). The solid curves are Lorentzian fits to each set of data. We focus on the 1s-2p $_\pm$ transition only because its linewidth is broaden solely by the quadrupole interaction among a number of electric field broadening mechanisms such as linear- and second-order Stark effects and quadrupole interactions. Figure 2 shows the full widths at the half maximum (FWHM) vs. N_I at T = 4 K. The experimental data (filled circles) are compared with the theoretical linewidths assuming random (dashed line) and correlated (solid line) distributions of ionized impurities. The excellent agreement between experimentally determined FWHM and the random theory for $N_I < 7.5 \times 10^{13}$ cm^{-3} is a clear evidence for the random distribution of ionized impurities in this low N_I region. When N_I is larger than 7.5×10^{13}

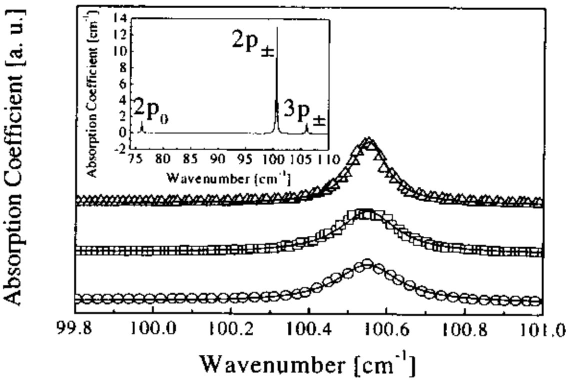

Fig. 1. An inset shows As donor absorption peaks recorded at T =4 K with a sample having $N_I = 3.50 \times 10^{13}$cm^{-3}. The three absorption peaks correspond to the 1s-2p$_0$ (76 cm^{-1}), 1s-2p$_\pm$ (100 cm^{-1}), and 1s-3p$_\pm$ (106 cm^{-1}) transitions. The main frame shows the enlargement of the 1s-2p$_\pm$ (100 cm^{-1}) absorption peak determined experimentally (○), calculated assuming random (□) and correlated (Δ) distributions of ionized impurities using the Monte Carlo method. Solid curves are the best fits to the experimental and calculated points assuming Lorentzian distributions.

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cm^{-3}, the experimental data lie between the estimates of random theory and correlated theory. This implies that the ionized impurity distribution is somewhere between "completely random" and "completely correlated". The Monte Carlo simulation for the correlated distribution shown in Fig. 1 has been performed for $T = 0$ K. However, the measurement was performed at the finite temperature ($T = 4$ K) at which a certain degree of randomization of ionized impurities occurs due to the finite thermal energy. In this case we expect the linewidth to be between the prediction of "completely random" and "completely correlated" assumptions. Figure 3 shows the temperature dependence of the FWHM (■) for a sample having $N_I = 7.8 \times 10^{13}\text{cm}^{-3}$, which is just above the critical concentration $N_{IC} = 7.5 \times 10^{13}\text{cm}^{-3}$ for $T = 4$ K. Fig. 3 shows clearly that the FWHM increases in two steps; the first gradual increase occurs between $T = 5$ and 11 K and the second rapid increase takes place above $T = 14$ K. The second increase at $T > 14$ K is due to thermal ionization of donors as it matches with the increment of N_I (solid curve). The first gradual increase is due to the transition of the ionized impurity distribution from correlated to random, and the two plateaus in FWHM at $T = 2 - 5$ K and $T = 11 - 13$ K represent characteristic FWHM for the two distributions. In order to support our claim that we have observed the transition, we shall estimate the critical temperature (T_c) for the transition. The energy of the Coulomb gap Δ for three dimensions is approximately;[2)]

$$\Delta = e^3 g_0^{1/2} / \kappa^{3/2}, \tag{1}$$

where g_0 is the density of states at the Fermi level of the order

$$g_0 = K N_D \kappa r_D / e^2. \tag{2}$$

K is the compensation ratio and $r_D = (3/4\pi N_D)^{1/3}$ is the distance between donors. Using Eqs. (1) and (2), $\Delta = 0.31$ meV has been obtained for the sample having $N_I = 7.8 \times 10^{13}\text{cm}^{-3}$ in Fig. 3. To first order, we expect T_c to be of the same order as Δ, i.e., $T_c \approx 3.6$ K is what we estimate based on theory. The experimentally found gradual increase starts around 4 K, in very good agreement with the theoretically estimated $T_c \approx 3.6$ K. The inset in Fig. 3 shows the temperature dependence of the FWHM for samples well below N_{IC} and well above N_{IC}. The width of the bottom curve ($N_I = 4.3 \times 10^{13}\text{cm}^{-3}$) remains unchanged because its width is determined solely by the random distribution all the way up to 12 K. The FWHM of the bottom curve ($N_I = 4.3 \times 10^{13}\text{cm}^{-3}$) for the temperature range 2-12K agrees very well with the theoretical prediction of the random theory (the dashed line in Fig. 2). The FWHM of the top curve in the inset ($N_I = 2.26 \times 10^{14}\text{cm}^{-3}$) for the temperature range shown is determined dominantly by the correlated distribution because the donor concentration is high enough for the neighboring ionized impurities to interact with one another. The FWHM increases with the increasing temperature because the partial randomization of the correlated distribution proceeds.

As one can see in the inset of Fig. 3, it takes an extremely sensitive tuning of N_D and N_A in order to observe a clear signature of the transition with two distinct plateaus below the temperatures where ionization takes place. The precise control of both donors and acceptors has been the key for the successful observation of random-to-correlated transition of the ionized impurity distribution in semiconductors.

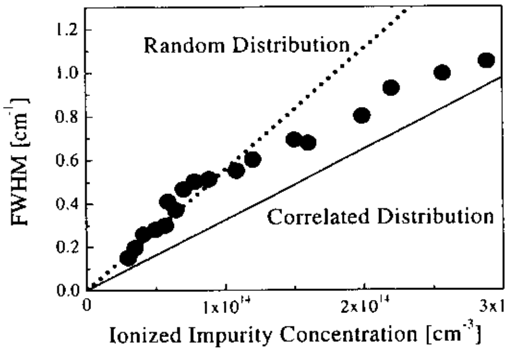

Fig. 2. Experimentally determined FWHM (filled circles) vs. N_I at $T = 4$ K. The dashed line is the prediction based on a random distribution of ions while the solid line is the prediction based on a correlated distribution of ionized impurities at zero temperature.

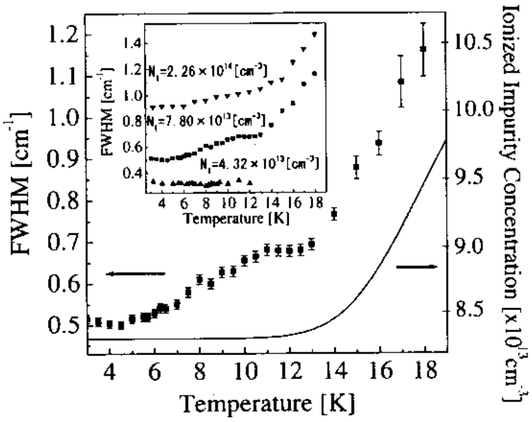

Fig. 3. The main frame shows FWHM vs. temperature for a sample having $N_I = 7.80 \times 10^{13}$ cm^{-3}. The solid curve is the calculated ionized impurity concentration as a function of T for this particular sample. The inset compares FWHM vs. temperature of three samples having $N_I = 4.32 \times 10^{13}$ (▲), 7.80×10^{13} (■), and 2.26×10^{14} cm^{-3} (▼).

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