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Stark broadening of impurity absorption lines by inhomogeneous electric fields in highly compensated germanium

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Stark broadening of Zeeman absorption lines caused by inhomogeneous electric fields in highly compensated Ge has been studied by means of far-infrared magneto-optical absorption spectroscopy measurements. A number of transmutation-doped Ge single crystals with a systematically varying compensation ratio were employed. The broadening of the full width at half maximum (FWHM) of an absorption line of the Ga acceptor is studied as a function of excitation light intensity with above-band-gap energy. The FWHM increases with decreasing intensity of the band-edge light excitation. Observation of the theoretically predicted $\frac{4}{3}$-power law of Stark broadening, due to ionized impurities, is reported. The line broadening originates in the Stark effect, due to inhomogeneous electric fields caused by the random distribution of ionized impurities. In order to understand the mechanism for the line broadening in detail, a numerical approach based on a Monte Carlo simulation has been performed. The results of this simulation show that the inhomogeneity of the field distribution becomes larger with increasing concentration of ionized impurities. The simulation based on a perfectly random distribution for an initial impurity arrangement gives a fairly good agreement with the experimental results. We conclude that the distribution of impurities in transmutation-doped Ge samples is close to random. [S0163-1829(96)00423-7]

I. INTRODUCTION

Highly compensated semiconductors have attracted considerable attention.[1–3] These semiconductors are interesting because the electronic properties of impurities are strongly affected by the Stark effect originating in the spatially varying internal electric fields caused by the ionized acceptors and donors. For a highly compensated p-type semiconductor, all donors are ionized in thermal equilibrium at low temperature. The concentration of ionized acceptors is the same as that of the ionized donors N_D, and the concentration of neutral acceptors is N_A-N_D, where N_A is the concentration of the acceptors. Only the neutral acceptors show Zeeman absorption in the far-infrared region.

Electrons and holes photoexcited across the band gap are captured efficiently by ionized impurities. A dominant relaxation process subsequent to the impurity neutralization is donor-acceptor (D-A) recombination.[4] The recombination probability of D-A recombination is closely related to the distance between a donor and an acceptor. The relaxation time increases to several seconds for samples with low-impurity concentrations. As a result of recombination, impurities are ionized, resulting in electric fields. Since the distribution of ionized impurities is, in general, inhomogeneous, the induced electric fields are similarly inhomogeneous. These electric fields give rise to Stark shifts of the energy levels of neutral impurities. A hole bound to a neutral acceptor experiences the sum of Coulomb interaction with all other ionized impurities. These internal electric fields affect the position and the full width at half maximum (FWHM) of the Zeeman absorption line.

Absorption spectra of shallow impurities in semiconductors have been investigated extensively by several authors.[5–7] Line broadening is caused by phonons, wave function overlap, strain, and electric fields. Phonon broadening is basically a lifetime effect, due to electron-phonon interaction between bound carriers and phonons.[8,9] Concentration broadening is due to overlap of wave functions between bound states.[10,11] With increasing impurity concentration, the wave functions of the ground and bound excited states of impurity atoms begin to overlap, leading to broadening. Strain broadening is caused by the presence of imperfections, which can be electrically active and inactive[12] impurities, dislocations, and precipitates.

Finally, Stark broadening, the subject in this study, has its origin in internal electric fields induced by ionized impurities. Investigation of external and internal electric-field effects in boron-doped Si have been carried out in detail.[1,2] The observed Stark broadening is attributed, in this case, to an unresolved partial lifting of the degeneracy of bound excited states of acceptors. In uncompensated boron-doped Si at temperature higher than 50 K, neutral boron absorption lines are affected by the screened Coulomb fields of thermally ionized impurities. In compensated boron- and phosphorous-doped Si, the compensation effects at low temperature are attributed to the unscreened Coulomb fields of ionized impurities. In a study of the internal Stark effect in highly compensated Ge by Ohyama,[3] the widths of impurity absorption lines were investigated through controlling the number of ionized impurities by changing the intensity of the band-edge light excitation. It was found that the observed broadening of In acceptor lines can be attributed to unresolved splitting caused by the Coulomb fields of ionized impurities.

Larsen *et al.* have calculated the linewidths and compared their findings with experimental results obtained with GaAs.[13–15] They also discussed the inhomogeneous broadening of Zeeman absorption lines of shallow donors.

Effects of the internal electric fields on a hydrogenic impurity state can be considered in analogy with the hydrogenic model. The dependence of the linewidth on the concentration of randomly distributed ionized impurities has been given by Stoneham.[16] In the first-order Stark effect and second-order Stark effect, the line broadening Δ is in proportion to $N_i^{2/3}$ and $N_i^{4/3}$, respectively, where N_i is the ionized impurity concentration. Moreover, at low concentrations a field-gradient effect plays an important role in the broadening of spectral lines. This effect is due to the interaction between the electric quadrupole moment of a neutral impurity and the field-gradient induced by ionized impurities. The line broadening Δ is proportional to N_i. In all these cases, it is assumed that the distribution of impurities in a semiconductor is completely random.

We have studied the far-infrared magneto-optical absorption in highly compensated Ge under band-gap photoexcitation (band-edge light excitation). Transmutation-doped Ge samples were employed. The compensation in these samples varied with their isotopic composition. The linewidth of Zeeman absorption was measured for a range of ionized impurity concentrations. The concentration of ionized impurities N_i was controlled by the band-edge light intensity. The dependence of the linewidth on N_i will be discussed and compared with the result of Stoneham. Our results are in agreement with the theoretically predicted $\frac{4}{3}$-power law between Stark broadening and N_i.

For a more detailed discussion of the concentration dependence of ionized impurities, we have carried out a Monte Carlo simulation to investigate this broadening mechanism numerically and compared the results of this simulation with our experimental findings obtained in highly compensated semiconductors. We find that the broadening of the linewidth depends on the spatial distribution of impurities. We can show that the line broadening analysis can yield information on the initial distribution of impurities in a semiconductor.

II. EXPERIMENTAL PROCEDURES

Our far-infrared (FIR) laser was a discharge-type pulse laser with a repetition rate of 30 Hz, using H_2O and D_2O vapor at pressures between 1 and 2×10^{-1} Torr. Two wavelengths, 119 and 84 μm were employed. In addition, an optically pumped laser was employed to investigate the frequency dependence of the line broadening. The FIR light was detected by a Sb-doped Ge photoconductive detector.

The applied magnetic field of up to 9 T was produced by a vertical-type superconducting solenoid. All the measurements were carried out in the Faraday configuration. The temperature was varied between 1.8 and 30 K and was monitored by a carbon-glass thermometer.

For the band-edge light optical excitation measurements, a xenon flash lamp or a tungsten incandescent lamp were employed. The repetition rate of the xenon flash lamp was 15 Hz and it was synchronized with every other FIR laser pulse. The intensity of excitation light was controlled by neutral density filters. A glass filter, which cuts light with energy larger than the band gap was also employed. The absorbance is derived from the following equation:

TABLE I. Isotopically controlled, neutron-transmutation doped Ge samples.

Sample	Type	N_D (cm^{-3}) As	N_A (cm^{-3}) Ga	Compensation ratio (K)
o	*p*		9.0×10^{14}	~0
a	*p*	6.00×10^{14}	7.30×10^{15}	0.082
b	*p*	1.32×10^{15}	3.27×10^{15}	0.40
c	*p*	1.39×10^{15}	2.83×10^{15}	0.49
d	*p*	1.47×10^{15}	2.42×10^{15}	0.61
e	*p*	1.54×10^{15}	2.02×10^{15}	0.76

$$\text{absorbance}=\ln(I_0/I), \tag{1}$$

where I and I_0 are the intensities of the transmitted FIR light with and without band-edge light.

Instead of band-edge light, electric-field excitation was also employed. This method was mainly used to investigate the effect of uniform electric fields on the line broadening.

Five *p*-type Ge samples doped by the neutron-transmutation-doping (NTD) method[17–19] were employed. As donors and Ga acceptors were found by the NTD process. By adjusting the relative concentrations of the ^{70}Ge and ^{74}Ge isotopes, variously compensated Ge samples can be prepared with NTD. The compensation ratio for a *p*-type semiconductor is defined as

$$K=\frac{N_D}{N_A}. \tag{2}$$

The values of K for the employed samples ranged from 0.082 to 0.76 as shown in Table I. The shape of samples was always elliptical. The typical sample size was $7\times5\times1$ mm^3. The flat faces were normal to the $\langle100\rangle$ crystallographic axis and the magnetic field was parallel to this axis.

III. EXPERIMENTAL RESULTS

A. Band-edge light intensity dependence

Figure 1 shows the FIR magneto-optical absorption spectrum for sample *b*, with $K=0.40$. Under band-edge light excitation, some absorption lines of the Ga acceptors appear in the spectrum. However, the intensity of these absorption lines is considerably smaller without band-edge light, because N_D donors and acceptors are ionized in the equilibrium state (''dark'' state). We will focus on a sharp absorption line that appears near 2.3 T. This line has been identified previously to be the Zeeman absorption of Ga acceptors associated with Landau levels.[20] We find that the Zeeman absorption line without band-edge light is much broader and weaker than that with band-edge light. Figure 2 shows the dependence of the integrated intensity of the above-mentioned absorption line on the time after a band-edge light pulse excitation of sample *c*. After a few seconds, the sample

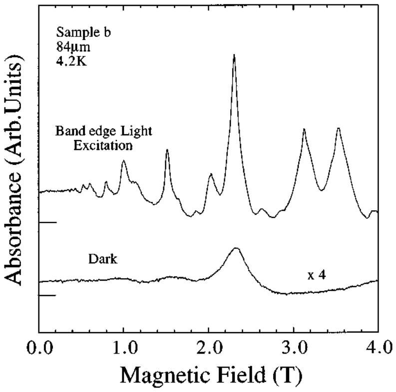

FIG. 1. Far-infrared magneto-optical absorption spectrum for the 84-μm laser line in sample b ($K=0.40$), with and without band-edge light. The sharp line near 2.3 T, without the band-edge light is broader and weaker than that under strong band-edge light.

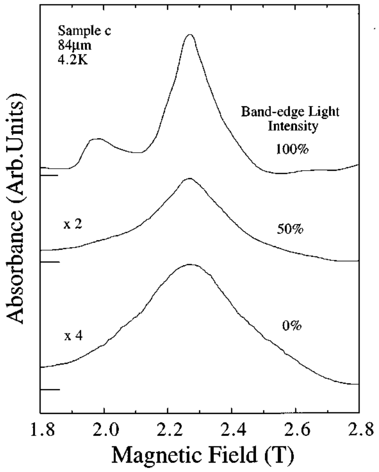

FIG. 3. The absorption lines of sample c ($K=0.49$) for three band-edge light intensities as 0, 50, and 100 %. With decreasing the band-edge light intensity, the FWHM of the absorption line increases.

returns to equilibrium. This slow decay arises from donor-acceptor (D-A) recombinations.

Zeeman absorption lines in sample c with $K=0.49$ under band-edge light excitation with relative intensity of 100, 50, and 0% are shown in Fig. 3. With decreasing band-edge light intensity, the FWHM of the line becomes much broader. Results for sample c are similar to those obtained with sample b. Here, the absorption line observed at 1.95 T is due to As donors.

The dependence of the FWHM for sample c on the band-edge light intensity is shown in Fig. 4. When the excitation light intensity increases to above 50% of maximum, the FWHM approaches a constant value Δ_0, which corresponds to the FWHM observed in the strong band-edge light intensity limit. This saturated value Δ_0 arises from broadening mechanisms other than the Stark effect, i.e., phonon broadening, concentration broadening, strain broadening, etc. Because all broadening processes are independent from one another, we find a Stark broadened FWHM by subtracting Δ_0 from experimentally observed FWHM Δ. Hereafter, we focus on the Stark broadening quantity $\Delta - \Delta_0$.

Figure 5 shows the dependence of the FWHM on the concentration N_i of ionized impurities. The concentration of ionized impurities is estimated from the integrated intensity of the absorption lines. It is found that the Stark broadening follows a $N_i^{4/3}$ power law when the concentration of ionized impurities is higher than 10^{15} cm^{-3}. Accordingly, our result must be identified with the second-order Stark effect predicted by the inhomogeneous broadening theory.[16]

Figure 6 shows the dependence of the FWHM of the absorption line of samples, which have different compensation ratios of 0.082, 0.49, 0.61, and 0.76. The ionized impurity concentration dependence of the line broadening for all samples closely follows a $N_i^{4/3}$ power law for $N_i < 10^{15}$ cm^{-3}. For $N_i > 10^{15}$ cm^{-3} a deviation from $N_i^{4/3}$ power law is observed.

B. Magnetic field and external electric-field dependence

Figure 7 shows two series of linewidths as a function of the concentration of the ionized impurities obtained for 2.3 and 5.5 T. There is a clear magnetic-field dependence of the absolute value of the FWHM, but both follow the $N_i^{4/3}$ power laws.

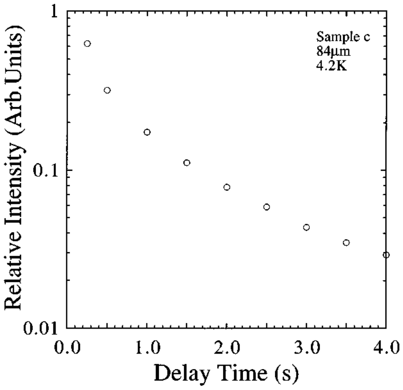

FIG. 2. Time variation of the integrated intensity for the absorption line at 4.2 K. This integrated intensity is normalized to one at time zero. The long decay time is due to the slow donor-acceptor recombination.

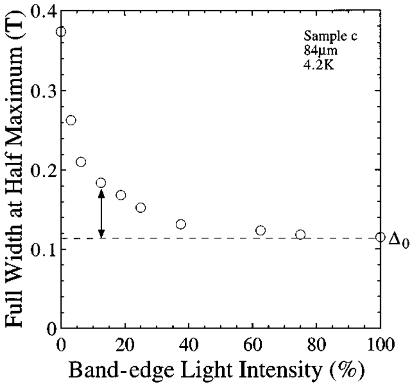

FIG. 4. The FWHM of absorption line for sample c for various band-edge light intensities. When the band-edge light intensity increases above 50%, the FWHM decreases and approaches to a constant value Δ_0.

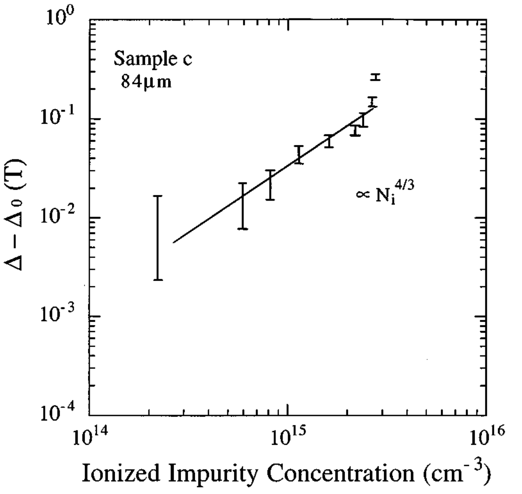

FIG. 5. The dependence of the FWHM of absorption line for sample *c* on the concentration of the ionized impurities. The Stark broadening quantity $\Delta - \Delta_0$ is shown. The variation of the line broadening shows the $N_i^{4/3}$ power law (solid line), which was predicted by Stoneham.

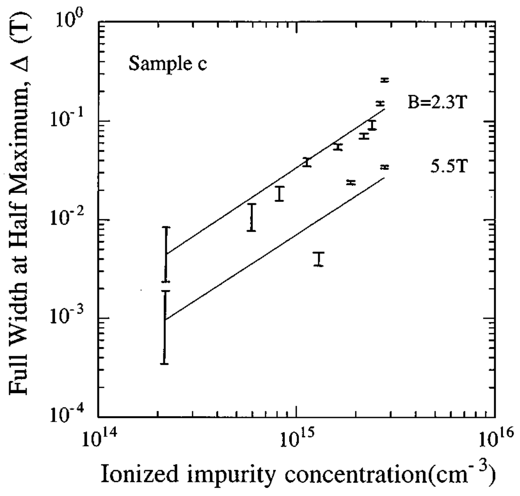

FIG. 7. The dependence of the FWHM of the absorption line for sample *c* on the concentration of ionized impurities under magnetic fields 2.3 and 5.5 T. The variation of the FWHM at 5.5 T is smaller than that at 2.3 T, but both data show the $N_i^{4/3}$ power law (solid line).

When the magnetic field is applied, the wave function of a hydrogenic impurity is compressed in a plane normal to the direction of the magnetic field. We introduce the quantity γ given by

$$\gamma = \frac{\hbar \omega_c}{2\mathcal{R}^*}, \tag{3}$$

where $\mathcal{R}^*$ is the effective Rydberg energy. For the magnetic field of 2.3 and 5.5 T, the value of γ are estimated to be 0.267 and 0.645, respectively. The shrinkage of the wave function means that the effective Bohr radius becomes smaller, and that the separation between the energy levels becomes larger. Thus, the Stark broadening based on the second-order perturbation theory is expected to decrease as the magnetic field increases. Based on the theory of Kogan and Lifshits,[12] one expects that the second-order Stark broadening is proportional to $(N_i a_B^3)^{4/3}$, where a_B is the Bohr radius. Thus, the dependence of the FWHM on the concentration of ionized impurities in a magnetic field does not change functionally, but the magnitude of the line broadening depends on the magnetic field.

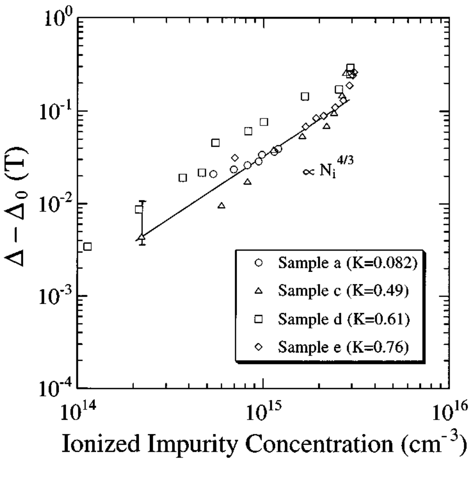

FIG. 6. The dependence of the FWHM of absorption line of samples with various compensation ratios: 0.082, 0.49, 0.61, and 0.76, respectively. The solid line is obtained by the theoretical calculation, but the origin is arbitrary. The maximum error is shown by the bar. The variation of line broadening in all samples shows a $N_i^{4/3}$ power law (solid line).

When an external electric field is applied to sample *o* up to 100 V/cm, no changes in the FWHM of the absorption line are observed. In this case, there are two possible contributions to the broadening of the FWHM, due to the external electric field. One is the effect of the electric-field on the electronic energy levels and the other is on the ionization of neutral donor. Up to 100 V/cm, both contributions to the broadening, due to external electric field, are much weaker than that due to the internal electric fields discussed in this section. This result is in reasonable agreement with our estimate of the strength of the internal electric field.

IV. MONTE CARLO SIMULATION AND DISCUSSION

Monte Carlo simulation were carried out in order to analyze the dependence of the FWHM of the absorption line on the band-edge light intensities.[21] Specifically, the dependence of the internal electric fields on the distribution of the ionized impurities was studied. In the first stage, the calculation was carried out for the case of a completely random distribution of ionized impurities in which there is no spatial correlation for the position of each impurity. The procedure is briefly summarized in the following. First, the initial configuration of impurities was set. A three-dimensional system with $(3000)^3$ lattice points was employed. In this space, 1730 acceptors and 850 donors were randomly distributed. Second, the electric field at the position of every neutral impurity was calculated. Third, the spatial dependence of the total electric field was calculated. Finally, the standard deviation σ of the electric field was obtained. Moreover, this calculational process was repeated for various concentration

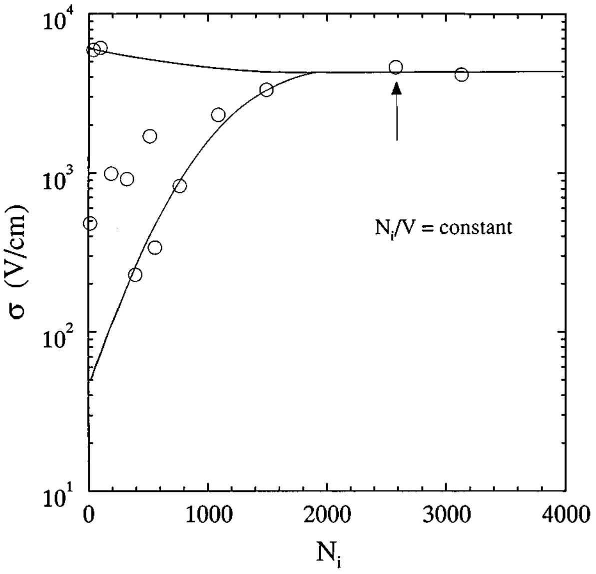

FIG. 8. The standard deviation σ in the distribution of the electric field for various system sizes, L, under the condition that N_i/V is a constant. The notation N_i and V $(=L^3)$ denote the concentration of ionized impurities and volume of the system, respectively. Two solid lines show minimum and maximum σ. The difference between the solid lines is caused by the fluctuations in the computational results. The standard deviation saturates at a system size above $N_i = 2000$. The value denoted by an arrow was employed in the following calculations.

ratios of ionized to total impurities.

Figure 8 shows σ of the electric field as a function of system size L under the condition that N_i/V is a constant, with N_i and V $(=L^3)$ being the number of ionized impurities and the volume of this system, respectively. It appears that σ saturates with system size at a certain value. If the system size is small, neutral impurities located near the surface of the system are not negligible. Accordingly, the fluctuation for σ is considerably large. For large system size, the influence of the impurities near the surface becomes relatively weaker. Since this figure shows clear saturation of σ at the system size above $N_i = 2000$, surface effects can be ignored and the system yields accurate results in the Monte Carlo calculation. The value denoted by an arrow $[(3000)^3$ lattice points$]$ was employed in the following.

The distribution amplitude of the internal electric fields at the position of a neutral impurity for various concentrations of ionized impurities is shown as a function of the electric field in Fig. 9. N denotes the concentration of neutral impurities. The distribution amplitude of the electric fields are plotted for relative ionized impurity concentrations equal to 30%, 70%, and 100% of the total impurity concentration. Each curve is obtained by smoothing a histogram with a deviation of 5%. With increasing concentration of the ionized impurities, the distribution of the internal electric fields shifts toward a higher field and it also becomes much larger.

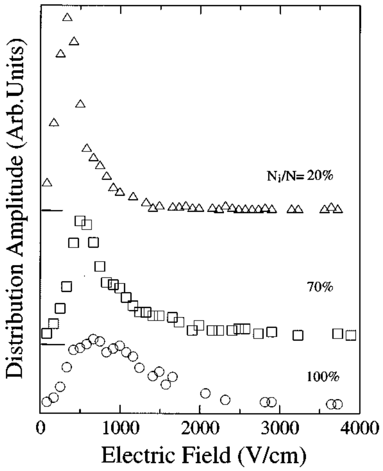

FIG. 9. The distribution amplitude of electric fields at neutral impurity sites for various concentrations of ionized impurities. N denotes the total impurity concentration. With increasing ionized impurity concentration, the distribution shifts towards the higher field and becomes broader.

The FWHM of the Zeeman absorption lines is approximately proportional to σ of the electric-field distribution. Thus, it is important to examine the concentration dependence of the distribution. Figure 10 shows the standard deviation in the distribution of electric-field squared σ^2 for two cases of initial configurations, random and correlated. N_0 denotes a normalization factor. In a random configuration, it is found that σ is proportional to the density of ionized impurities to the four-third power law.

The Holtzmark function has been employed for various problems, such as the distribution of electric fields induced by charged particles in plasma[22] and ionized impurities in semiconductors.[13,14,23] This function can be applied only in the case of a random distribution of ionized impurities and is expressed as a function of electric field E as follows:[24]

$$F(E) = \frac{2}{\pi E}\int_0^\infty x \sin x \exp[-(E_H/E)^{3/2}x^{3/2}]dx, \quad (4)$$

with the electric field E_H defined by

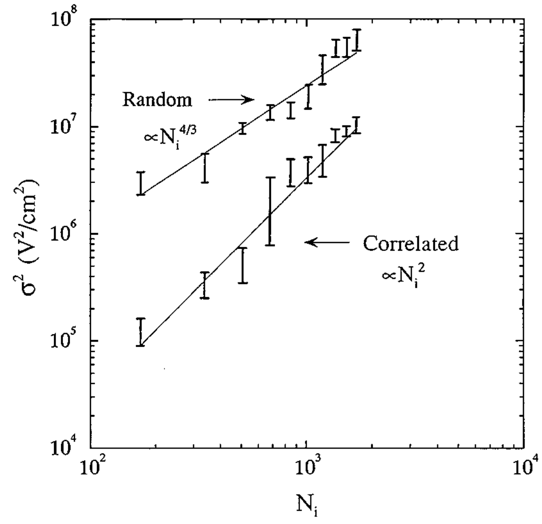

FIG. 10. The standard deviation in the distribution of the electric field squared in two cases of initial configurations, which are random and correlated with dipole pairs, being in proportion to $N_i^{4/3}$ and N_i^2, respectively. Solid lines show calculational results.

$$E_H = 2.6\frac{e}{4\pi\varepsilon_0\varepsilon}N_i^{2/3}. \quad (5)$$

Actually, it is found that the extent of the distribution in the second-order Stark effect obtained by using this function is proportional to $N_i^{4/3}$. The analytical results obtained by Larsen are in excellent agreement with the results of the Monte Carlo simulation. This fact supports the validity of the Monte Carlo simulation. These results are also in good agreement with the experimental results. Accordingly, we conclude that the second-order Stark effect strongly contributes to the observed line broadening in the Ge NTD samples. It is expected that perfectly random distributions of ionized impurities are formed, because of the completely random distribution of the various Ge isotopes.[17–19]

Larsen[13,14] pointed out the possibility of weak pairing between a donor and an acceptor. From this, it is expected that the FWHM of an absorption line obtained by experiment[15] will be smaller than that for his calculation based on a random distribution. Indeed, the FWHM of the absorption line obtained experimentally is well explained by the correlated, i.e., weak pairing model.

In order to apply this analysis to samples from crystals grown by the Czochralski method and doped in the melt, the relation between various distributions of ionized impurities and the line broadening are discussed. In particular, the concentration dependence of σ in the distribution of the electric field for various distributions of ionized impurities is calculated in the following. As an extreme case, we consider the situation in which the distance between an ionized donor and an ionized acceptor corresponds to the effective Bohr radius ($\sim$40 Å) of a donor or an acceptor in Ge. Such a spatially correlated pair of an ionized donor and an ionized acceptor causes a dipole. The orientation of the dipole is assumed to be completely random. In this case, it is found that the variation of σ is proportional to a N_i^2 power law, as shown in Fig. 10. This variation can be qualitatively explained by a simple nearest-neighbor approximation.[14] Our experimental results clearly do not support such a close-pair distribution of ionized donors and acceptors.

V. CONCLUSION

The broadening of a Zeeman absorption line of Ga acceptors in highly compensated Ge has been investigated systematically. The FWHM of the Zeeman absorption line increases as the intensity of band-edge light decreases. It is concluded that the line broadening is caused by a Stark effect, due to inhomogeneous electric fields induced by the ionized impurities. The experimental results are qualitatively in agreement with the theory by Stoneham,[16] based on randomly distributed ionized impurities and they are dominated by the second-order Stark effect.

In practice, the spatial distribution of impurities in semiconductors cannot be determined unambiguously. The distribution of the strength of the electric-field induced by ionized impurities in the presence of a spatial correlation between impurities cannot be deduced analytically. In order to analyze the relationship between the inhomogeneous electric field and the distribution of ionized impurities, a Monte Carlo simulation was carried out.

The simulation assuming a random distribution of ionized impurities yields values, that are in good agreement with our experimental results. From this simulation, it is concluded that the distribution of ionized impurities in transmutation-doped Ge must be random. Moreover, the spatially correlated distribution of ionized impurities has been investigated in the Monte Carlo simulation. Especially, in the case of a system of correlated *D*-*A* dipole pairs, the distribution of electric fields does not obey the $N_i^{4/3}$ power law. However, it is found that the broadening of the spatially correlated distribution in this case is proportional to N_i^2. For melt doped samples, studies like the ones presented here could find spatially correlated distributions of impurities.

Recently, inhomogeneous line broadening of dopants in a quantum well has been analyzed theoretically.[25,26] In such a low-dimensional system, it is expected that interesting phenomena concerning the distribution of impurities will occur.

From the scientific as well as technical point of view, the understanding of the distribution of impurities remain very important. The Monte Carlo method developed for this study appears to be useful for determining the initial distribution of impurities in semiconductors. In the future, line broadening due to the internal Stark effect for various distributions of ionized impurities will be understood in detail and investigated experimentally for a variety of compensated semiconductors.

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