Neutral-impurity scattering in isotopically engineered Ge

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Neutral-impurity scattering of electrons and holes at low temperatures has been studied in isotopically engineered Ge single crystals. Use of the neutron transmutation doping technique provides the necessary dopant uniformity and low compensation. We find excellent agreement between the lowtemperature experimental mobility and phase-shift calculations for the hydrogen atom scaled to the impurity atoms in semiconductors.

I. INTRODUCTION

The low-temperature mobility of free carriers in semiconductors is mainly determined by ionized- and neutral-impurity scattering. The ionized-impurity scattering mechanism has been extensively studied,¹ and various aspects of this process are now quite well understood. Scattering by neutral impurities is much less effective than by ionized centers, i.e., its contribution is significant only in crystals with low compensation and at very low temperatures where most of the free carriers are frozen on the impurity sites.

Good examples of devices in which the neutralimpurity scattering mechanism becomes important are extrinsic semiconductors for infrared photoconductors (Ge:Ga, Ge:Zn, Ge:Be, Ge:Cu, Si:B, etc.²) that are widely used by astrophysicists³ and condensed-matter scientists.⁴ Because photoconductor materials are typically nominally uncompensated and run at low temperatures ($T<10$ K), understanding of the mobility dominated by neutralimpurity scattering is crucial for the development and modeling of these infrared detectors.

Theoretical research on neutral-impurity scattering has Theoretical research on neutral-impurity scattering has
been active since 1950 .⁵⁻¹¹ Following the progress of the theory, a few experimental studies probing the nature of neutral-impurity scattering were published. $12-15$ However, for these experiments it was necessary to introduce adjustable parameters to obtain good agreement between the experimental results and theory. Consequently, there still exists no solid experimental verification for the theoretical models of the neutral-impurity scattering rate.

In order to clearly discern the effects of neutral impurities on the carrier mobility, one has to reduce or eliminate the much more efficient ionized-impurity scattering. This can be realized only in uncompensated materials. Furthermore, since the standard scattering models assume a random distribution of scattering centers, it is very important that the impurities are uniformly distributed in the samples. These conditions are not easily achievable with standard doping techniques, where considerable compensation and/or nonuniform impurity distributions are frequently observed.

In this paper, in order to satisfy the very strict materials requirements for our neutral-impurity scattering experiment, we have applied the neutron transmutation doping (NTD) technique¹⁶ to Ge samples of controlled 70 Ge and 74 Ge isotopic compositions. Such studies are now possible due to the availability of highly enriched Ge isotopes.^{17,18} Using NTD on isotopically controlled Ge samples we can control both the acceptor (Ga) and donor (As) concentrations. The NTD method also leads to very uniform impurity distributions down to the atomic level.¹⁹ Ge crystals prepared in this way provide an ideal system to study neutral-impurity scattering.

The main objective of this paper is to directly compare experimentally measured Hall mobilities with theoretical mobilities calculated without any adjustable parameters.

II. THEORY

A. Neutral impurity scattering

In 1950 Erginsoy⁵ realized that the neutral-impurity scattering cross sections in semiconductors can be de-

rived by taking electron-hydrogen $(e^- -H)$ scattering cross sections in free space and modifying them to semiconductors by scaling the electron mass and the dielectric constant. Unfortunately the exact e^- -H cross sections in free space were not available to Erginsoy at the time. He took approximate e^- -H results of Massey and Moiseiwitsch²⁰ and calculated the inverse relaxation time τ^{-1} , the scattering rate, for neutral-impurity scattering:⁵

$$
\tau_{\text{neutral}}^{-1} = \frac{20\kappa N_N \hbar^3}{m^*^2 e^2} \,, \tag{1}
$$

where κ is the dielectric constant, e is the electron charge, N_N is the neutral-impurity concentration, and m^* is the electron effective mass. Equation (1) can be considered only as a first-order approximation because the prefactor 20 is an empirically determined constant and only the lowest-order s partial wave is taken into account in the phase-shift calculation. Anselm⁶ and Sclar⁷ later attempted to improve Erginsoy's calculation by including an overcharged H^- state, which might become important at low temperatures. Sclar also derived an expressio which relates τ^{-1} in semiconductors to the phase shifts of e^- -H scattering:²¹

$$
\tau_{\text{neutral}}^{-1} = 2\pi N_N v \frac{2}{k^2} \sum_{l=0}^{\infty} (l+1) \sin^2(\delta_l - \delta_{l+1}), \qquad (2)
$$

where v is the velocity of an incident electron, k is the wave number, and δ_l is the *l*th partial phase shift. Nearly exact phase-shift calculations of e^- -H scattering were finally performed in 1961 by Temkin and Lamkin²² and Schwartz. 23 These calculations included scattering for singlet and triplet states. Blagosklonskaya et al .⁸ scaled these results to semiconductors and obtained cross sections that turned out to be appropriate only when the incident electron energy was less than $\frac{1}{100}$ of the binding energy of a scattering center. McGill and Baron⁹ used the results of Sclar [Eq. (2)] in the form

$$
\tau_{\text{neutral}}^{-1} = \frac{4\pi N_N \hbar e^2}{2\kappa m^* E_B} \sum_{l=0}^{\infty} \frac{(l+1)}{4w^{1/2}} [3 \sin^2(\delta_l^- - \delta_{l+1}^-)] + \sin^2(\delta_l^+ - \delta_{l+1}^+)] , \qquad (3)
$$

where E_B is the binding energy of the scattering centers, $w \equiv E/E_B$ where E is the incident electron energy, and δ_l^+ and δ_l^- are the *l*th partial phase shift for the singlet and triplet states, respectively. By inserting δ_l^+ and $\delta_l^ (l=0-2)$ of e^- -H scattering calculated by Temkin and Lamkin²² into Eq. (3), McGill and Baron graphically showed the accurate τ^{-1} as a function of w for neutral impurity scattering in semiconductors.⁹ The result of McGill and Baron has been considered as an appropriate model for neutral-impurity scattering in semiconductors and has been discussed in detail in many standard textbooks.²⁴ Later Meyer and Bartoli reevaluated the problem and provided an analytical expression that is essentially the same as the graphical solution of McGill and Baron but covering a wider incident-electron energy range: 10

$$
\tau_{\text{neutral}}^{-1} = \frac{A(w)\kappa N_N \hbar^3}{m_H^*^2 e^2} \,, \tag{4a}
$$

with

$$
A(w) = \frac{35.2}{w^{1/2}} \frac{(1+e^{-50w})(1+80.6w+23.7w^2)}{(1+41.3w+133w^2)} \times \left[\frac{1}{w} \ln(1+w) - \frac{(1+0.5w-1.7w^2)}{(1+w)^3} \right].
$$
 (4b)

Here m_H^* is the hydrogenic effective mass given by

$$
m_{\rm H}^* = \frac{E_B \kappa^2 m_0}{E_{\rm H}} \quad , \tag{5}
$$

where m_0 is the electron rest mass and $E_H = 13.6$ eV is the binding energy of hydrogen. In their original treatment Meyer and Bartoli used square-conductivity effective mass m_{con}^{*2} instead of m_H^{*2} in the denominator of Eq. (4a). Two effective masses in the denominator of Eq. (4a) come from two different origins: one is the mass of an incident electron and the other is the mass of a bound electron at the scattering center. For this reason it may look appropriate to use m_{con}^* times m_H^* instead of m_H^{*2} . However, because we are adopting the results of e^- -H scattering in free space in which the masses of incident and bound electrons are the same, we must keep the two masses in the denominator the same in order for our scaling to semiconductors to be valid. Therefore appropriate parameters to use are $m_H^{\ast 2}$ for scaling of the Bohr radius in Eq. (4a) and $w = E/E_B$ for the scaling of the incident electron energy in Eq. (4b). One should also be careful with the choice of E_B for this calculation. All previous studies cited above employed experimentally determined E_{R} , which included contributions from central cores of impurities. Because we are only interested in the Bohr radius of scattering centers, a theoretically calculated E_B of a perfect hydrogenic impurity without any central-cell corrections should be used. This approach is appropriate since the central-cell potential is highly localized and therefore, although it can affect the binding energy, it does not significantly change the size of the neutral impurity, which is mainly determined by the long-range Coulomb interactions.

B. Total-mobility calculation for doped Ge at low temperatures

In our total-mobility calculation we employ a standard relaxation-time approximation. This approach is valid because we are limiting ourselves to low temperatures $(T < 25$ K) where the inelastic optical-phonon deformation-potential scattering is negligible. Three scattering mechanisms are considered: neutral-impurity, ionized-impurity, and acoustic-phonon deformationpotential scattering.

The neutral-impurity scattering contribution is calculated using both Eqs. (1) and (4) so we can compare the models of Erginsoy and Meyer and Bartoli to our experiment. The concentration of neutral-impurity centers as a function of temperature $N_N(T)$ in each sample is given by

TABLE I. Parameters used in the total-mobility calculations.

	$Ge:As$ $(n-type)$	$Ge:Ga(p-type)$
к	16	
m_{con}^*	0.12m ₀	0.28m ₀
B_{ac} (Ref. 28)	1.08×10^{10} g ^{3/2} K ^{-3/2}	9.50×10^8 g ^{3/2} K ^{-3/2}
E_R (theoretical)	12.5 meV $(Ref. 29)$	11.2 meV $(Ref. 30)$

$$
N_N(T) = N_{\rm MJ} - N_{\rm MN} - n(T) \tag{6}
$$

where N_{MJ} , N_{MN} , and $n(T)$ are the majority-impurity, minority-impurity, and free-carrier concentrations, respectively.

For the ionized-impurity scattering, we employ the Brooks-Herring expression,^{25,26}

$$
\tau_{\text{ion}}^{-1} = \frac{\pi N_I e^4 (k_B T)^{-3/2} x^{-3/2}}{(2m_{\text{con}}^*)^{1/2} \kappa^2} \times \left[\ln \left(1 + \frac{4x}{a} \right) - \frac{4x/a}{1 + 4x/a} \right],
$$
\nwhere $a = \frac{2\pi \hbar^2 e^2 n}{m^* \kappa k_B^2 T^2}$, (7)

 $x = E/k_BT$ (E is the incident electron energy), m_{con}^* is the average conductivity effective mass, and N_I is the ionized-impurity concentration. The temperaturedependent N_I in each sample is given by

$$
N_I(T) = n(T) + 2N_{MN} \tag{8}
$$

For the acoustic-phonon deformation-potential scatter $ing, ²⁷$

$$
\tau_{\rm ac}^1 = B_{\rm ac} (m_{\rm con}^* T)^{3/2} x^{1/2} , \qquad (9)
$$

where the constant B_{ac} has well-established values for *n*and p-type Ge as shown in Table I.

Having found τ^{-1} of all three scattering mechanism we calculate an average $\langle \tau \rangle$ using the Maxwell-Boltzman integration:

$$
\langle \tau \rangle = \frac{4}{3\sqrt{\pi}} \int_0^\infty \frac{x^{3/2} \exp(-x)}{\tau_{\rm ac}^{-1} + \tau_{\rm ion}^{-1} + \tau_{\rm neutral}^{-1}} dx \quad . \tag{10}
$$

Finally the total mobility
$$
\mu_{\text{tot}}
$$
 is then given by
\n
$$
\mu_{\text{tot}} = e \langle \tau \rangle / m_{\text{con}}^* \ . \tag{11}
$$

All the parameters required for the mobility calculations are well known in Ge (Table I). The only unknown material parameters at this point are sample-dependent N_{MJ} , N_{MN} , and $n(T)$ in Eqs. (6) and (8). However, as is shown later, all three parameters can be determined precisely for each sample by performing variabletemperature Hall-effect measurements. Consequently all mobility calculations are performed without any adjustable or scaling parameters.

III. EXPERIMENT

Our growth method of isotopically enriched 74 Ge and 70 Ge crystals has been described in detail in Ref. 31. The electrically active residual-impurity concentration in the Ge crystals before NTD is typically $\sim 10^{12}$ cm⁻³. Wafers were cut from each ingot and thermal-neutron irradiated at the University of Missouri Research Reactor facility. After the irradiation, the 74 Ge wafers became As doped, i.e., *n*-type due to the ⁷⁴Ge + $n \rightarrow$ ⁷⁵Ge \rightarrow ⁷⁵As β ⁻ decay reaction, while the ⁷⁰Ge wafers turned *p*-type due to the 70 Ge + $n \rightarrow {}^{71}$ Ge $\rightarrow {}^{71}$ Ga electron-capture reaction. Unavoidable fast-neutron radiation damage was removed by thermal annealing at 650°C for 10 sec in a N_2 atmosphere. A series of 74 Ge:As and 70 Ge:Ga wafers of netcarrier concentrations in the range $10^{14} - 10^{16}$ cm⁻³ and less than 1% compensation were produced in this manner. A detailed description of neutron-transmutation doping of 70 Ge and 74 Ge crystals are given in Ref. 32. Hall and resistivity measurements were performed in order to determine the free-carrier concentration and the mobility as a function of temperature. Disk-shaped (\sim 6 mm diameter, ~ 0.5 mm thick) samples with the van der Pauw contact configuration were used in all measurements. Phosphorus and boron were implanted to NTD 74 Ge:As and NTD 70 Ge:Ga samples, respectively, for the formation of Ohmic contacts. A magnetic induction of 3000 G was used in all Hall measurements.

IV. RESULTS AND DISCUSSION

Our first experimental step is the determination of $n(T)$, N_{MJ} , and N_{MN} in each sample performing Halleffect measurements. Figures 1(a) and l(b) show the temperature-dependent free-carrier concentrations $n(T)$ in four ${}^{74}Ge:As$ and two ${}^{70}Ge:Ga$ samples, respectively. The experimental curves are fitted with the following standard semiconductor statistics, which describes the temperature dependence of the free-carrier concentration in semiconductors doped by shallow majority impurities $N_{\rm MJ}$ and compensated by minority impurities $N_{\rm MN}$.³³

$$
n(T) = 2(N_{\rm MJ} - N_{\rm MN}) / \{ [1 + (N_{\rm MN}/gN_B) \exp(E_{\rm MJ}/k_B T)] + \sqrt{[1 + (N_{\rm MN}gN_B) \exp(E_{\rm MJ}/k_B T)]^2 + (4/gN_B)(N_{\rm MJ} - N_{\rm MN}) \exp(E_{\rm MJ}/k_B T)} \},
$$
(12)

FIG. 1. Temperature dependence of freecarrier concentration in (a) four ⁷⁴Ge:As and (b) two ⁷⁰Ge:Ga samples:⁷⁴Ge:As-1 (\bullet), ⁷⁴Ge:As – 2 (●), ⁷⁴Ge:As – 3 (□), ⁷⁴Ge:As – 4 (■), ⁷⁰Ge:Ga – 1 (△), and ⁷⁰Ge:Ga – 2 (▲).

FIG. 2. Experimentally measured carrier mobility in (a) four 74 Ge:As and (b) two 70 Ge:Ga samples. The symbols representing different samples are the same as those in Fig. $\mathbf{1}$.

FIG. 3. Data points represent experimentally measured carrier mobility in (a) four ⁷⁴Ge:As and (b) two ⁷⁰Ge:Ga samples. The symbols representing different samples are the same as those in Fig. 1. For a direct comparison theoretically calculated mobility using Erginsoy's model (broken line) and the model of Meyer and Bartoli (solid line) is shown for each sample. The contributions of the different scattering mechanisms to the total mobility of the ${}^{70}Ge:Ga-1$ sample are shown in the upper half of (b).

TABLE II. N_{MJ} and N_{MN} obtained from the Hall curve fittings.

Sample	$N_{\rm MJ}$ (cm ⁻³)	N_{MN} (cm ⁻³)
74 Ge:As – 1	4.5×10^{14}	2.5×10^{12}
74 Ge:As -2	1.8×10^{15}	6.0×10^{12}
$^{74}Ge:As-3$	2.9×10^{15}	2.1×10^{13}
$^{74}Ge:As-4$	1.5×10^{16}	4.2×10^{13}
70 Ge:Ga -1	3.2×10^{14}	1.0×10^{12}
70 Ge:Ga -2	8.8×10^{14}	5.1×10^{12}

where $n(T)$ is the free-electron (hole) concentration, N_{MI} and N_{MN} are the majority- and minority-impurity concentration, respectively, N_B is the effective conduction (valance-) band density of states, $g = \frac{1}{2}$ (g = 4) is the spin (valance-) degeneracy for a donor (acceptor), and E_{MJ} are the experimentally determined ionization energies: 14 and 11.07 meV for As and Ga, respectively. All fits we obtained using Eq. (12) are very good (see Fig. 1) and $N_{\rm MJ}$ and $N_{\rm MN}$ are accurately determined for each sample as shown in Table II.

Figures 2(a) and 2(b) show the temperature-dependent Hall mobility for all samples in the temperature range $T=6-300$ K. We have demonstrated in our recent paper that the mobility in the high- $(T>80 \text{ K})$, intermediate- $(T=20-80 \text{ K})$, and low- $(T<20 \text{ K})$ temperature regimes is dominated by phonon, ionized-impurity, and neutralimpurity scattering, respectively.³⁴

We now turn our attention to the low-temperature regime where mobilities are dominated by neutral-impurity scattering. Figures 3(a) and 3(b) show a direct comparison of our experimental results with theoretical totalmobihty curves calculated using the procedure described in Sec. IIB. For each sample two theoretical totalmobility curves are calculated: one using Eq. (1) (Erginsoy) and the other using Eq. (4) (Meyer and Bartoli). A strikingly good agreement was obtained between the experimental and theoretical mobilities calculated with the model of Meyer and Bartoli for all samples. In the temperature range of interest $(T < 25$ K), the condition

 μ B > 1 of the high magnetic-induction limit is met for all samples, i.e., the Hall factor r_H is unity. Thus our data points in Fig. 3 should represent the drift mobility μ_d . The contribution of hopping conduction to the measured mobility is negligible because, as seen in Fig. 1, there is no deviation between the experimental points and the fitted curves for all samples even at the lowest T . For completeness, we have also calculated the Hall factor r_H using $r_H = \langle \tau^2 \rangle / \langle \tau \rangle^2$. As a result we have found that r_H was in the range 1.05-1.¹ for all data points shown in Fig. 3. Therefore, even if our Hall results were affected by r_H , the 5-10% upward shift of each curve calculated with the theory of Meyer and Bartoli would still be in good agreement with our experimental results, while it would lead to further deviation from Erginsoy's theory. Also shown in Fig. 3(b} are the contributions of diFerent scattering mechanisms to the total mobility in 70 Ge:Ga – 1 which contains the least amount of Ga impurities as seen in Table II. Even with this small amount of the neutral-impurity concentration, neutral Ga becomes the dominant scattering center below 16 K. Mobilities in other samples with higher Ga and As concentrations are dominated by neutral-impurity scattering up to higher temperatures. For example, neutralimpurity scattering dominates up to $T \sim 21$ K in highly doped 74 Ge:As -4.

In order to demonstrate the importance of the homogeneous dopant distribution, we have performed the same study on samples cut from Ge:Ga crystals grown by the conventional Czochralski (Cz) method. Compared to our NTD 74 Ge:As and NTD 70 Ge:Ga, we expect these samples to have less homogeneous Ga impurity distributions since Ga impurities were introduced to Ge melt during the crystal growth. We have measured four Cz Ge:Ga samples cut from four different ingots. All samples had $[Ga] \sim 1.5 \times 10^{14}$ cm⁻³ and $\tilde{N}_{MN} \sim 2 \times 10^{11}$ cm⁻³. Among these four samples, mobilities of only two samples had a fair agreement with the theoretically calculated mobility. However, mobilities in two other samples as shown in Figs. 4(a) and 4(b) substantially deviate from the theory. These observed deviations of the measured mobility from the theoretical calculations are most likely

FIG. 4. (a) and (b) A direct comparison of theoretically calculated mobilities to experimentally measured mobilities in two conventional Ge:Ga samples.

due to inhomogeneous Ga impurity distributions in melt-doped Ge. Only the use of the neutrontransmutation-doped semiconductors with randomly distributed dopants allows for an accurate test of the neutral impurity-scattering models.

V. SUMMARY AND CONCLUSIONS

We have performed a detailed study of neutralimpurity scattering by measuring the free-carrier mobility in Ge:As and Ge:Ga samples. The application of the neutron-transmutation doping technique to isotopically enriched 74 Ge and 70 Ge crystals allowed preparation of samples with controlled compensation and truly randomly distributed dopants. We have shown that the lowtemperature mobility is accurately described by the phase-shift-based scattering model of hydrogen atoms scaled to hydrogenic impurities in semiconductors.

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