Molecular-Beam Epitaxial Growth of a Far-Infrared Transparent Electrode for Extrinsic Germanium Photoconductors

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ABSTRACT. We have evaluated the optical and electrical properties of a far-infrared (IR) transparent electrode for extrinsic germanium (Ge) photoconductors at 4 K, which was fabricated by molecular beam epitaxy (MBE). As a far-IR transparent electrode, an aluminum (Al)-doped Ge layer is formed at well-optimized doping concentration and layer thickness in terms of the three requirements: high far-IR transmittance, low-resistivity, and excellent ohmic contact. The Al-doped Ge layer has the far-IR transmittance of >95% within the wavelength range of 40–200 μm, while low-resistivity (∼5 Ω cm) and ohmic contact are ensured at 4 K. We demonstrate the applicability of the MBE technology in fabricating the far-IR transparent electrode satisfying the above requirements.

1. INTRODUCTION

Bulk germanium extrinsic photoconductors (Ge PCs) have been widely used in far-infrared (far-IR) astronomical observations in the 50–120 μm wavelength range (e.g., Haegel et al. 1985). The spectral response of p-type Ge PCs can be extended to longer wavelengths (≈200 μm) by applying a uniaxial compressive stress (Kazanskii et al. 1977). In order to provide high quantum efficiency, broad spectral response, and low cosmic-ray hitting rates compared to those for the bulk Ge PCs, germanium blocked impurity band (Ge BIB) detectors have been developed as next-generation far-IR detectors replacing the bulk Ge PCs (Watson & Huffman 1988; Wu et al. 1991; Watson et al. 1993; Huffman & Casey 1993; Bandaru et al. 2002; Beeman et al. 2007; Kaneda et al. 2011).

BIB detectors require electrodes not with a transverse but with a longitudinal, and thus transparent, configuration to obtain high responsivity, because a far-IR absorbing thin layer (≤10 μm) is formed underneath an electrode. And the construction of a large-format array for both PCs and BIB detectors is indispensable to improve observational efficiency and measurement accuracy. The array, as shown in Figure 1, also needs electrodes with the far-IR transparent configuration. Although such transparent electrodes have been applied to Ge PC arrays, performance of transparent electrodes is not optimized (e.g., Fujiwara et al. 2003).

The transparent electrode requires high far-IR transmittance while ensuring high conductivity and an excellent ohmic contact at temperatures below 4 K. To obtain high far-IR transmittance, a thin layer with a low doping concentration is desirable. On the other hand, a low-resistivity layer equipped with excellent ohmic contact can be achieved by a degenerately doped (n_d ≈ 10^{16}–10^{17} cm⁻³), thick, and epitaxially grown Ge layer. Thus, the doped Ge layer should be epitaxially grown with optimally controlled doping concentration and thickness.

The most commonly used technique to fabricate the transparent electrode is ion implantation on one surface of a device. However, its doping concentration often shows a graded profile. Therefore, the formation of the decreasingly doped layer as a function of implant depth is inevitable. Such a broad doping profile significantly degrades far-IR transmittance due to photon absorption within the thick-implanted layer by free carriers and doped impurities (Hadek et al. 1985; Fujiwara 2000). Nevertheless, there are few investigations in terms of abruptness for the doping profile.

To improve far-IR transmittance, an alternative process technology with precise control of a doping profile, a doping concentration and a layer thickness should be introduced. A promising candidate technology is molecular beam epitaxy (MBE), an ultra—high-vacuum (UHV)-based technique for producing high-quality epitaxial structures with monomolecular layer (monolayer) thickness control. MBE technology allows us to accurately control doping profiles, doping concentrations, and interfaces, thanks to the lower growth rate and temperature compared to other epitaxial growth technologies such as liquid...
phase epitaxy. In this article, we show the properties of the far-IR transparent electrode fabricated by the MBE technology.

2. REQUIREMENTS FOR A FAR-IR TRANSPARENT CONTACT

In this section, we search optimized solutions for the doping concentration \( n_d \) and the layer thickness \( d \), considering the following requirements: high far-IR transmittance, low-resistivity, and excellent ohmic contact. Note that we assume all the dopants are electrically activated; \( n_{ad} = n_d \), where \( n_{ad} \) is activated doping concentration.

2.1. Far-IR Transmittance

The far-IR transmittance \( T_{IR} \) through a sample is defined as \( T_{IR} = I_t / I_0 \), where \( I_0 \) and \( I_t \) are intensities of incident and transmitted far-IR light, respectively. We consider two samples: Sample 1 is a nondoped Ge wafer with a doped Ge layer, while sample 2 is a nondoped Ge wafer without a doped Ge layer. Hereafter, we call the relative transmittance between the two samples \( T_{IR,1}/T_{IR,2} \) as the far-IR transmittance of the doped Ge layer \( T_{IR,d} \).

We require \( T_{IR,d} \geq 0.95 \) at \( T = 4 \) K and \( \lambda = 100 \) \( \mu \)m. To calculate \( T_{IR,d} \), we take into account the free-carrier absorption and photoionization processes in the doped Ge layer with multiple reflection based on Hadek et al. (1985). They presented a theoretical analysis of the far-IR transmittance through ion-implanted Ge samples which took into account free-carrier absorption in the doped Ge layer with its impurity concentration range of \( 10^{16} \leq n_d \leq 10^{19} \) \( \text{cm}^{-3} \). In their analysis, they introduced an absorption parameter \( a \) in the Fresnel coefficients to calculate the transmittance from the vacuum to the nondoped Ge medium through the doped Ge layer and the reflectance from the doped Ge layer surface (see Fig. 4 of Hadek et al. 1985). In our calculation, to include the photoionization process, the absorption parameter is redefined as \( a = \exp(-\alpha d/2 - idn\omega/c) \), where \( \alpha, n, \) and \( c \) are a photoabsorption coefficient, the complex refractive index of Ge, and the speed of light, respectively; Hadek et al. (1985) case corresponds to the case of \( \alpha = 0 \). The photoabsorption coefficient is \( \alpha = \sigma n_d \), where \( \sigma \) is a photoionization cross section of a dopant. The complex refractive index depends on the infrared conductivity \( \sigma(\omega) = \sigma_0/(1 + i\omega\tau) \), where \( \sigma_0 \) and \( \tau \) are the dc conductivity and a single relaxation time, respectively (Hadek et al. 1985).

The layer thickness and doping concentration at \( T_{IR,d} = 0.95 \) are calculated by considering the DC conductivity dependence on impurity concentration at 4 K in Hadek et al. (1985) and \( \sigma \) assumed to be \( 10^{-14} \text{ cm}^{-2} \) (Bratt 1977; Wang et al. 1986). As a result, as can be seen in Figure 2, smaller thickness
and lower doping concentration than those on the solid line are required to achieve $T_{Br,d} > 0.95$.

### 2.2. Resistivity

The requirement for the resistivity is that the resistance of the doped Ge layer ($R_d$) is much lower than that of a Ge PC pixel ($R_{sc}$) for a voltage drop to become negligible at the layer. The requirement is particularly stringent for the large-format array to uniformly apply a detector bias voltage to pixels. Here, we consider an $N \times N$-pixel array that has the same electrode configuration as that in Figure 1; the array has a common doped Ge layer for the pixels. A common metal electrode is formed on the doped Ge layer of pixels on the outermost circumferential sides. At the central pixel of the array, which is farthest from the metal electrode, the fraction of the bias voltage drop $F$ is expressed as $F = R_d/(R_d + R_{sc})$. To obtain $F = 0.01$, the resistivity of the doped Ge layer $\rho_d$ at 4 K should be $\rho_d = 0.02\rho_{ad}/NL$, where $\rho_{ad}$ and $L$ are the resistivity of the Ge PC pixel at 4 K and the pixel size, respectively. The experimental relation between $\rho_d$ and $n_{ad}$ is obtained from Fritzsche & Cuevas (1960). The dashed line in Figure 2 is derived from the above equation on condition that $\rho_d$, $L$, and $N$ are $6 \times 10^8$ $\Omega$ cm (Hiromoto et al. 1989), $5 \times 10^{-2}$ cm, and 128, respectively.

### 2.3. Ohmic Contact

To make an ohmic contact, a contact resistance $R_c$ needs to be sufficiently small compared with $R_{sc}$. Sze & Ng (2007) introduced the characteristic energy $E_{00}$ to categorize a carrier transport process for metal-semiconductor contacts. For p-type Ge with doping concentration of $\geq 10^{16}$ cm$^{-3}$, $E_{00}$ is estimated to be $2 \times 10^{-3}$ eV by applying the hole effective mass of 0.075$m_e$ (Hadek et al. 1985). Under the operation temperature ($T \leq 4$ K) of Ge PCs, $E_{00}$ is much higher than $kT$, which means a pure tunneling process through a Schottky barrier dominates. In this process, higher doping concentration is required. Also, the thickness of the doped Ge layer should be larger than that of the depletion width $W_D$ due to the metal/Ge contact.

As a typical pixel of Ge PCs, $R_{sc}$ is estimated to be $2 \times 10^8 \Omega$ by using $L$ and $\rho_d$ at 4 K. Under the tunneling process, the specific contact resistance ($R_{sc}$) depends strongly on $n_{ad}$, the carrier effective mass $(m^*)$, and the barrier height ($\phi_B$), but virtually independent of temperature (Sze & Ng 2007). Based on $R_{sc}$ for a metal/n-type Ge contact measured by Gallacher et al. (2012) ($R_{sc} \sim 2 \times 10^{-7} \Omega$ cm$^2$ for $n_{ad} = 3 \times 10^{19}$ cm$^{-3}$ and $\phi_B = 0.75$ eV), by applying effective masses of hole and electron ($0.2m_e$, Kahn 1955), we roughly estimate $R_{sc}$ for a metal/p-type Ge contact as

$$R_{sc} \sim 5 \times 10^{-7} \exp\left(\frac{\phi_B}{0.75 \text{ eV}} \sqrt{\frac{3 \times 10^{19} \text{ cm}^{-3}}{n_{ad}}}\right) \Omega \text{cm}^2.$$  (1)

For an Al/Ge contact, since $\phi_B$ is 0.5 eV at 4 K (Thanailakis & Northrop 1973; Sze & Ng 2007), $R_{sc}$ is estimated to be $\leq 5 \times 10^5 \Omega$ cm$^2$ for $n_{ad} \geq 10^{16}$ cm$^{-3}$. Therefore, we obtain $R_c \leq 10^6 \Omega$ by dividing $R_{sc}$ by $L^2$, which is much lower than $R_d$; the condition of $R_c \ll R_d$ is satisfied for $n_{ad} \geq 10^{16}$ cm$^{-3}$.

In the case of $R_c \ll R_d$, which means a voltage drop at the contact is negligible, $W_D$ is simply expressed as $W_D = (2\epsilon_sV_{bi}/qn_{ad})^{1/2}$, where $\epsilon_s$ and $V_{bi}$ (0.5 V at 4 K) are the permittivity of Ge and built-in voltage, respectively. In Figure 2, the dash-dotted line corresponds to the condition of $d = W_D$ for the Al/Ge contact.

From the above three requirements, the enclosed area by the three lines in Figure 2 shows optimized solutions for the fabrication of the far-IR transparent electrode for Ge PCs.

### 3. EXPERIMENTS

#### 3.1. Molecular Beam Epitaxial Growth

Our MBE system is built inside a growth chamber and contains effusion cells for source materials, a substrate holder and heater, a pumping system to achieve UHV ($10^{-10}$–$10^{-11}$ torr), liquid $N_2$ cryopanels, and in situ analysis tools: a reflection high-energy electron diffraction (RHEED) gun and a mass spectrometer. A RHEED pattern on a screen and its brightness are measured by a CCD camera.

The source Ge was prepared from a nondoped Ge ingot with a resistivity of 49–51 $\Omega$ cm, which means that its impurity concentration is much less than the room-temperature intrinsic carrier concentration in Ge ($\sim 10^{13}$ cm$^{-3}$). As for a dopant, since the nondoped Ge shows a p-type conduction below $\sim 100$ K by Hall effect measurements, the dopant is selected from p-type materials for Ge; we chose aluminum (Al, 6N purity). Crucibles

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**Fig. 3.—Depth profile of the Al concentration of sample 1. The detection limit of Al is $\sim 10^{14}$ cm$^{-3}$.**
for Ge and Al are pyrolytic boron nitride (PBN). The effusion cell temperature for Ge is set to be 1250°C at which we observe the Ge beam flux of $1.3 \times 10^{14}$ cm$^{-2}$ s$^{-1}$. Since the fraction of electrical activation of Al in Ge is $\sim 10\%$ for our MBE system ($n_{ad} \sim 0.1n_d$), the target Al doping concentration $n_{Al}$ is $4-5 \times 10^{17}$ cm$^{-3}$ to give wider acceptable range in thickness (see Fig. 2). The effusion cell temperature for Al is set to be 690°C, which corresponds to the Al beam flux of $6 \times 10^9$ cm$^{-2}$ s$^{-1}$.

We prepared a mirror-polished Ge(100) substrate ($10 \times 9$ mm$^2$) with thickness of 300 μm, which was obtained from the nondoped Ge ingot. The substrate was degreased in methanol with ultrasonic for 10 minutes. After degreasing, the substrate was ultrasonically rinsed in deionized water (18 MΩ cm) for 10 minutes to dissolve the native Ge oxide and then (process I) dipped into HF (49% HF:H$_2$O = 1:3) solution for 10 minutes. After this process, (process II) the substrate was blown dry in dry N$_2$ and then exposed to 8 mW cm$^{-2}$ of radiation from a UV Hg arc lamp ($\lambda = 185, 254$ nm) in air for 10 minutes to remove carbon (C) atoms and grow a fresh Ge oxide layer. Then we repeated processes I and II but with duration time of 70 minutes with UV irradiation. The cleaned substrate was immediately introduced through a load lock into a UHV chamber. Prior to MBE growth, we checked carbon coverage of the substrate surface by X-ray photoelectron spectroscopy (XPS) analysis. By measuring intensities of Ge3d and C1s XPS lines, carbon coverage is estimated to be 0.08 monolayers, which corresponds to the value for the HF treatment method in Sun et al. (2006). After the XPS analysis, the substrate was loaded into the MBE chamber. The chamber base pressure was $\sim 10^{-10}$ torr and increased by up to $10^{-9}$ torr. The substrate was heated at 600°C to remove the Ge oxide layer. The heating process was stopped when brightness contrast between a diffraction spot and a diffuse component on the RHEED screen was maximum. Finally, an Al-doped Ge layer was grown on the front side of the substrate for 3 hr. During the growth, when we confirmed the change of a RHEED pattern from 3D to 2D growth mode, the substrate temperature was set to be 500°C from 600°C. The processed sample was cut into two pieces for providing samples for optical and electrical measurements.

3.2. Optical and Electrical Measurements

Far-IR transmittances of samples 1 and 2 were measured at 4 K by using Fourier transform infrared spectroscopy. Geometries of the two samples are summarized in Table 1. To cool them down to 4 K, we installed the samples into a cryostat which contained a wheel to rotate the samples in and out of the beam, a Si bolometer detector, and a preamplifier. Far-IR transmittance measurements for both samples were performed by measuring this transmission relative to that of a blank aperture. The reproducibility of the measurement was $\sim 5\%$.

To measure resistivity of the Al-doped Ge layer formed on the nondoped Ge substrate (sample 1), four-point probe measurements were performed with bias current of 10 μA at 4 K. Since most of carriers in the substrate are trapped into impurity levels and hence the charged impurity atoms are neutralized at 4 K (carrier freeze-out), the resistivity of the substrate is expected to be much higher than that of the Al-doped Ge layer at 4 K. Therefore, measured values can be regarded as the resistivity of the Al-doped Ge layer itself.

The condition that $R_c$ is much lower than $R_d$ should be satisfied to make an ohmic contact. If not ($R_c \approx R_d$), current-voltage ($I - V$) curves show Schottky diode behavior. In order to investigate $I - V$ curves, we prepared sample 3, an Al layer with thickness of 0.2 μm was formed on the Al-doped Ge layer (see Table 1). The Al and Al-doped Ge layers were then electrically split into two parts by chemical etching. By using the two metal contact pads, we measured $I - V$ curves for sample 3 with the four-terminal method at temperatures between 5 and 20 K.

4. RESULTS AND DISCUSSIONS

Figure 3 shows the depth profile of Al concentration for sample 1, which was obtained from a secondary ion mass spectrometry analysis. The detection limit of the Al concentration is about $10^{14}$ cm$^{-3}$. As can be seen in Figure 3, the thickness of the Al-doped Ge layer is 0.22 μm. The Al doping concentration of $\sim 4 \times 10^{17}$ cm$^{-3}$ is within the range of the target values, the variation of which is 20% (3σ) in the depth range of 0.02 to 0.18 μm. Activated doping concentration is measured by a spread resistance analysis at 300 K and is $\sim 10\%$ of the

| Sample | Measurement | Ge substrate (mm$^2$) | Al-doped Ge layer (mm$^2$) | Al layer (mm$^2$)$^3$ |
|--------|-------------|----------------------|---------------------------|----------------------|
| 1      | OM, EM$^a$  | $10 \times 5 \times 0.3$ mm$^2$ | $10 \times 5 \times 0.0002$ mm$^3$ | None |
| 2      | OM         | $10 \times 5 \times 0.3$ mm$^2$ | None | None |
| 3      | EM         | $10 \times 4 \times 0.3$ mm$^2$ | $4 \times 4 \times 0.0002$ mm$^3$ | $4 \times 4 \times 0.0002$ mm$^3$ |

$^a$Optical measurement.

$^b$Electrical measurement.

$^c$Sizes of the Al-doped Ge and Al layers for sample 3 are those of two split electrodes.
Al doping concentration. The large variation around the depth of 0.2 μm seems to be caused by the change in the substrate temperature from 600°C to 500°C; higher substrate temperature increases the rate of Al re-evaporation from the substrate surface and thus decreases its concentration. The local peak of the Al concentration at the depth of 0.21–0.23 μm indicates the growth interface.

At the growth interface, the profile shows an exponential decrease with the scale length of \(2 \times 10^{−3}\) μm, which is about an order of magnitude lower than that obtained from the ion-implantation technology (Fujiwara 2000). In our MBE system, we demonstrated high controllability for both doping concentration and profile. In Figure 2, the obtained parameter set for the Al-doped Ge layer is shown as the filled star. Polarized Raman spectroscopy in the \(z(x, x)z\) scattering configuration was applied to investigate the crystal structure of the Al-doped Ge layer. For Ge(100), if \(x, y,\) and \(z\) axes are set to be [100], [010], and [001], respectively, the Raman line intensity should be minimum at the angle of 0° between the [100] axis and the light polarization and maximum at the angles of ±45°. Figure 4 shows dependence of the Raman line intensity on the angle for the Al-doped Ge layer (filled circle) and the nondoped Ge substrate (open circle). The results confirm that the Al-doped Ge layer is epitaxially grown on the substrate.

Figure 5a shows the far-IR transmittance of the Al-doped Ge layer at 4 K, which is obtained by dividing \(T_{IR}\) of sample 1 by that of sample 2, as shown in Figure 5b. Measured data distribute around \(T_{IR,d} = 1.0\) (40 ≤ \(\lambda\) ≤ 200 μm), whereas those for ion-implanted samples show \(T_{IR,d} = 0.6–0.7\) (e.g. Fujiwara 2000). Thus, there is no significant far-IR absorption in the Al-doped Ge layer. In Figure 5b, \(T_{IR}\) values of both samples are 0.51 at \(\lambda = 100\) μm. By using the refractive index of 3.7 at 4 K, which is estimated by measuring the interference fringes around \(\lambda = 100\) μm, transmittance of the samples is calculated to be 0.50, which is in good agreement with the measured value.

The resistivity of the Al-doped Ge layer is 5 ± 3 Ω cm at 4 K, which is consistent with that of p-type Ge with \(n_{ad} \sim 4 \times 10^{16}\) cm\(^{-3}\) (Fritzsche & Cuevas 1960). Although the resistivity of the Al-doped Ge layer is much higher than that of an ion-implanted layer (∼10\(^{-2}\) Ω cm, Fujiwara 2000), it meets the resistivity requirement of \(\rho_t \leq 38\) Ω cm for \(d = 0.2\) μm.

At temperatures lower than ∼10 K, obtained \(I − V\) curves in Figure 6a show symmetry around \(V = 0\) mV. In particular, in the inset that shows the \(I − V\) curves within the range of \(|V| \leq 400\) mV, they show excellent symmetry and a linear dependence, as indicated by the best-fit linear regression lines. The \(I − V\) curves exhibit that a linear ohmic dependence at lower voltage is followed by nonlinear increase in slope at higher voltage. These properties are typical \(I − V\) curves obtained for pure Ge crystals (∼10\(^{11}\) cm\(^{-3}\), Teitsworth & Westervelt 1986).
To further confirm that the $I - V$ curves come from the electrical characteristic of the nondoped Ge, we investigated the relation between the current and temperature. Under low temperatures, the nondoped Ge is known to show p-type conduction that is attributed to residual gallium (Ga). Since Ga acceptors in Ge have the binding energy $E_{\text{Ga}}$ of 10.8 meV, the current is expected to increase with temperature in proportion to $\exp(-E_{\text{Ga}}/kT)$. Figure 7 shows the dependence of the current on the inverse of temperature at $V = 100$ mV. At higher temperatures, the current exponentially increases with $T^{-1}$, as indicated by the dashed line. Therefore, the result shows that the electrical characteristics are dominated by those of the nondoped Ge, implying that an excellent ohmic contact ($R_c \ll R_d$) is realized below $T \sim 10$ K.

However, $I - V$ curves within the temperature range of 10–20 K clearly show asymmetry around $V = 0$ mV, as seen in Figure 6b. Because the resistivity of the nondoped Ge exponentially decreases as temperature increases from 4 K to 10 K, the ohmic contact condition for an electrode seems to be broken ($R_c \approx R_d$). In this condition, $I - V$ curves should be asymmetric. Here, we regard an equivalent circuit for one of the Al/Ge contacts as a circuit that consists of a Schottky diode and a resistance connected in parallel; the equivalent circuit for the other Al/Ge contact is considered to be a resistance. The theoretical $I - V$ curve expected from the equivalent circuit model shows the dashed line in Figure 6b and fits the measured data. The result means one of the Al/Ge contacts is not the ohmic contact condition at $T \gtrsim 10$ K.

From the fact that $R_c \approx R_d$ at $T \sim 10$ K, we can roughly estimate the specific contact resistance. By using the measured resistivity ($\sim 10 \Omega$ cm) of the nondoped Ge at 10 K and considering the structure geometry of sample 3, $R_d(\approx R_c)$ is roughly estimated to be $\sim 1$ Ω. Thus, the specific contact resistance is $\sim 10^{-1} \Omega \text{cm}^2$ by multiplying $R_c$ by the area of the metal electrode (0.16 cm$^2$). Under the tunneling process through a Schottky barrier ($kT \ll E_{\text{Ga}}$), the specific contact resistance is less sensitive to temperature (Sze & Ng 2007). Thus, $R_{\text{sc}} \approx 10^{-1} \Omega \text{cm}^2$ can also be applied at temperatures below 10 K and is roughly in agreement with the expected value of $R_{\text{sc}}(\sim 5 \times 10^{-2} \Omega \text{cm}^2)$ obtained from equation (1) with $n_{\text{ad}} = 4 \times 10^{16} \text{cm}^{-3}$ and $\phi_B = 0.5$ eV.

5. CONCLUSIONS

As a far-IR transparent electrode for Ge PCs, the Al-doped Ge layer is epitaxially formed on the nondoped Ge substrate by using the MBE technology. The activated doping concentration and layer thickness of the Al-doped Ge layer are $4 \times 10^{16} \text{cm}^{-3}$ and 0.2 μm, respectively, which are within the range of...
optimized solutions for the three requirements: high far-IR transmittance, low-resistivity, and ohmic contact. We have evaluated the optical and electrical properties of the Al-doped Ge layer at 4 K. We obtained the far-IR transmittance of greater than 95% within the wavelength range of 40–200 μm, while resistivity was low enough (5 Ω cm) compared to the requirement value of 38 Ω cm for d = 0.2 μm. We also confirmed ohmic contact between the Al-doped Ge layer and the Al electrode. We demonstrated that the MBE technology is very applicable in fabricating an excellent far-IR transparent electrode for Ge PCs.

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