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The effect of metal cluster formation on THz range spectroscopy

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ABSTRACT

The present paper discusses terahertz (THz) spectroscopy of ion-implanted layer embedded metal particles. MgO (001) single crystals were implanted with −75 KeV Au ions at an ion dose of 1×10^{18} cm^{-2}, later annealed in air at 1100 K for 1 h. After the each process, the transmittance T spectrum ranging from 250 to 2000 nm, and the reflectance R spectrum ranging from 1.2 to 21 THz were measured. The T spectra show the embedded Au particles were formed in the implanted layer. The notion of an effective dielectric function was brought to evaluate the two-phase (Au–MgO) composite material. Consequently, the R THz spectrum analysis showed the dielectric function of the implanted MgO. This dielectric function reveals that dispersion by the optical phonons. The formation of embedded Au clusters in MgO shifted the resonance frequency of the optical phonon, and increased the damping constant. Results show that the THz spectrum reflects the introduced defects and the lattice deformation caused by the ion implantation and Au cluster formation.

1. Introduction

The study on ion implantation into dielectric materials has continued for decades [1–14]. Especially, using ion implantation, to form embedded nano-particles into ionic crystals has recently attracted significant interest. Ion implantation followed by thermal annealing can form embedded nano-particles. The embedded structure enhances the surface plasmon response (SPR) and can generate non-linear optical properties, then its potential is promising for optical and electric devices. The optical properties are responsible for the structure of the ion-implanted crystals such as defects in the crystal lattice as well as the properties of embedded particles such as their size, the volume fraction and the electric conductivity. Evaluation of the lattice structure in ion-implanted layers is therefore crucial.

In this study, THz spectroscopy was applied to the evaluation for ion-implanted ionic crystals with embedded metal particles. The resonance frequency of optical phonon of dielectric materials approximately ranges from THz to tens THz. THz spectroscopy therefore is sensitive to the lattice structure of dielectric materials [15]. Furthermore, THz spectroscopy can obtain the complex dielectric function, which brings us useful information for designing optical and electrical devices; besides, this method is non-destructive.

We have already reported that the defects introduced by ion implantation can be monitored using a THz spectroscopy analysis [16]. In this case the implanted layer however did not contain embedded particles, because ion dose is too low to form embedded particles. Consequently the effect of metal particles was not examined. Therefore this study was carried out in order to clarify the effect of the formation of Au particles in MgO single crystal on the THz range spectroscopy. Results showed that the formation of Au cluster significantly deformed MgO matrix as well as the increase of defects in it.

2. Experimental procedure

Magnesium oxide (MgO) (100) single crystal samples ($20\times20\times0.5$ mm^3) were implanted with −75 KeV Au ions at room temperature, the ion beam density ranged from 2.5 to 6.5 μA/cm^{-2} during the implantation, and the ion dose was 1×10^{18} cm^{-2}. Subsequent to the ion implantation, the sample was annealed in air at 1100 K for 1 h. After the implantation and after the annealing, the optical spectra in two frequency ranges were measured: the transmittance T spectra with a wavelength ranging from 250 to 2000 nm (photon energy: 0.6~5 eV), and the reflectance R spectrum with a wave number ranging from 40 to 700 cm^{-1} (frequency: 1.2–21 THz). The incident angle at the T spectrum measurement was 10°, and that at the R spectrum was 0°.

3. Results

3.1. Spectrum measurements

Fig. 1 shows the T spectra of the Au ion-implanted MgO sample. The spectrum of as-implanted MgO revealed an absorption peak at 2.1 eV. The peak became prominent after the annealing process. This peak indicates that Au clusters are formed in the implanted layer.

Fig. 2 shows the R spectra in THz range of the same Au ion-implanted MgO sample. The ion implantation reduced the reflectance in the range over 12 THz, and the post annealing slightly recovered the reflectance.

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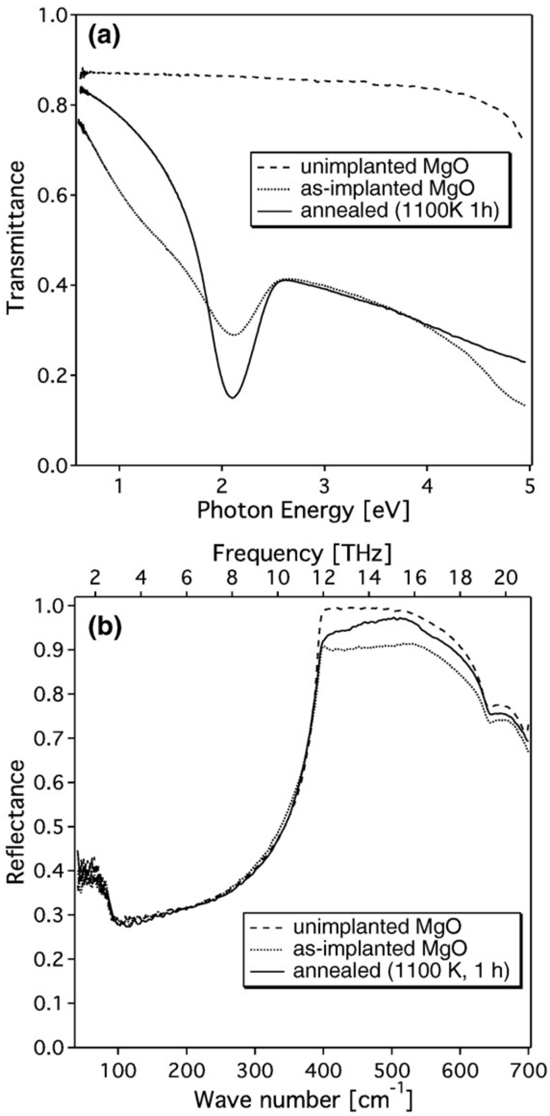

Fig. 1. Optical spectroscopy of Au-implanted MgO, (a) transmittance T spectra (250~2000 nm), (b) reflectance R spectra (40~700 cm^{-1}).

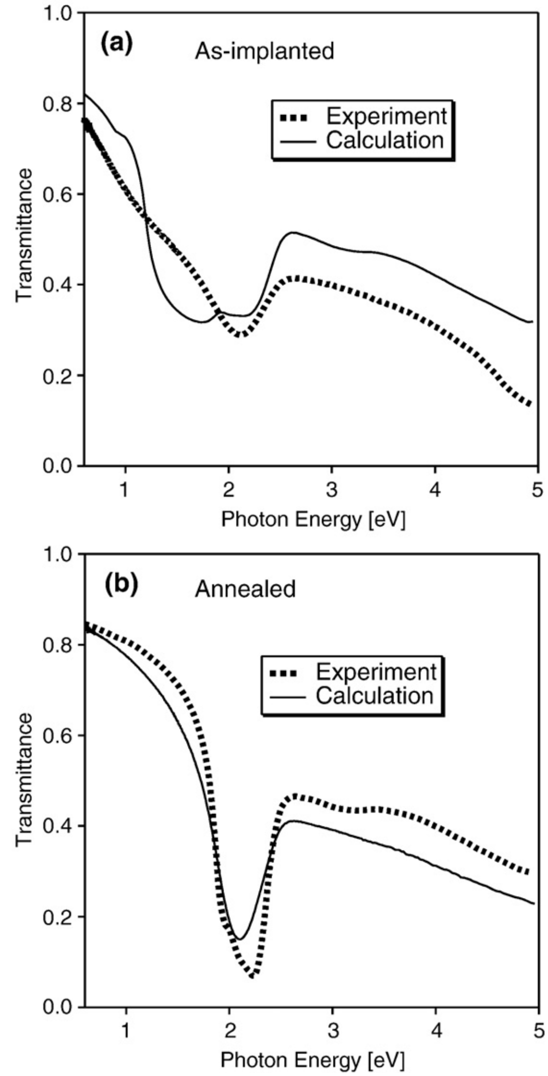

Fig. 3. Fitting result of T spectra, (a) as-implanted (by the Bruggeman formula), (b) annealed at 1100 K for 1 h (by the Maxwell Garnett formula).

3.2. Optical model simulation

To evaluate the lattice structure of Au ion-implanted layer from the spectra, we assumed optical models to obtain the dielectric function. For an ionic crystal such as MgO, the complex dielectric function $\varepsilon(\omega)$ is, based on the classical damping harmonic oscillator model (Lorentian model), expressed as:

$$\varepsilon(\omega) = \varepsilon_b + \sum_j \frac{S_j^2}{\omega_j^2 - \omega^2 - i\gamma_j\omega}, \quad (1)$$

where v_j is the resonance frequency of the jth oscillation mode, S_j is the oscillation strength, and γ_j is the damping constant. On the other hand, for an metallic material such as Au, $\varepsilon(\omega)$ is expressed as:

$$\varepsilon(\omega) = \varepsilon_b + \frac{\omega_P^2}{\omega(\omega - i\gamma)}, \quad (2)$$

where ω_p is plasma frequency. From $\varepsilon(\omega)$, the complex refractive index n is given by $n^2 = \varepsilon$.

The amplitudes r, t of R and T ($R = |r|^2$, $T = |t|^2$) at the boundary between the two layers that have the complex refractive indexes n_a and n_b are given by:

$$r = \frac{n_a - n_b}{n_a + n_b}, \qquad t = \frac{2n_a}{n_a + n_b}. \quad (3)$$

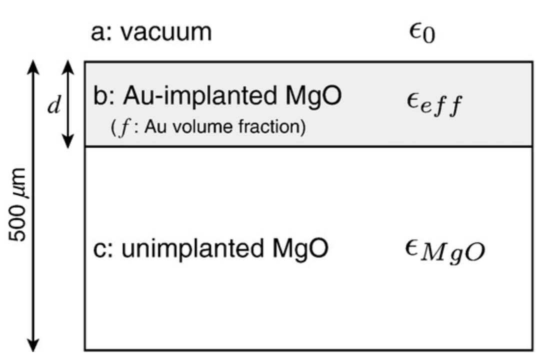

Fig. 2. Schematic of the two-layer model used for spectrum analysis.