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AC CONDUCTIVITY OF AMORPHOUS As-Se-Ag SYSTEM

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Abstract.- A.C. conductivity $\sigma_{ac}$ of amorphous $\text{As}_{40+x}\text{Se}_{60-x}$ and $\text{As}_2\text{Se}_3:\text{Ag}$ has been measured. The $\sigma_{ac}$ of these materials varies as $\omega^s$ in the audio frequency range. At room temperature, the exponent $s$ decreases from 1 to 0.7 with increasing $x$ in $\text{As}_{40+x}\text{Se}_{60-x}$, while $s$ is 0.7 irrespective of Ag content in $\text{As}_2\text{Se}_3:\text{Ag}$. With decreasing temperature, $s$ in $\text{As}_2\text{Se}_3:\text{Ag}$ increases. Defect states due to Ag additives are considered to be directly concerned with $\sigma_{ac}$.

1. INTRODUCTION
The ac conductivity $\sigma_{ac}$ of amorphous semiconductors normally exhibits a frequency dependence as $\sigma_{ac} = \omega^s$ with $0.7 \leq s \leq 1$. The $\sigma_{ac}$ has been considered to be caused by phonon-assisted hopping between localized states near the Fermi level [1]. Recently, Elliott [2,3] investigates the classical hopping between charged dangling defect states [4,5] in chalcogenide glasses. We have reported how Ag additive influences the electrical and optical properties [6,7]. The present paper aims to investigate the effect of Ag additive in amorphous $\text{As}_2\text{Se}_3$ on ac conduction. Since the introduced Ag atoms seem to combine with selenium atoms, the bulk $\text{As}_2\text{Se}_3$ becomes arsenic rich. We measured $\sigma_{ac}$ of arsenic rich samples to investigate the effect of the destruction of short range order due to the deviation from the stoichiometric composition.

2. EXPERIMENTAL PROCEDURE
Amorphous $\text{As}_2\text{Se}_3:\text{Ag}$ was prepared by synthesizing $\text{As}(6N)$, $\text{Se}(6N)$ and $\text{Ag}(5N)$. Silver concentration in the range from 0.025 to 0.5 at% was examined. $\text{As}_{40+x}\text{Se}_{60-x}$ ($x = 0.02, 0.1, 0.2$ and $0.4$) samples were also prepared. The samples used for the measurements were sawn from an ingot in the shape of plate and polished. The size of the samples was typically $20 \times 15 \times 0.3$ mm$^3$. Evaporated gold films were used as electrodes and found to make ohmic contacts with the sample. A guard ring electrode was added on a surface of the samples in order to prevent leakage current. Measurements of frequency dependent conductivity were made with a dielectric loss meter. The $\sigma_{ac}$ is defined according to the relation: $\sigma_{ac} = \sigma_{\omega} - \sigma_{dc}$, where $\sigma_{\omega}$ is the total conductivity observed under ac field and $\sigma_{dc}$ the dc conductivity.

3. RESULTS AND DISCUSSION
3.1. A.C. Conductivity of As-Se-Ag System at Room Temperature
The ac conductivity $\sigma_{ac}$ of amorphous $\text{As}_{40+x}\text{Se}_{60-x}$ and $\text{As}_2\text{Se}_3:\text{Ag}$ at room temperature are shown in Figs.1 and 2, respectively. The $\sigma_{ac}$ of these amorphous materials
The Ag content in As$_{40+x}$Se$_{60-x}$ varies as $\omega^s$ in the frequency range from 100 Hz to 500 kHz. The exponent $s$ decreases from 1 to 0.7 with increasing $x$ in As$_{40+x}$Se$_{60-x}$. In As$_2$Se$_3$:Ag, on the other hand, $s$ has usually a constant value of 0.7 irrespective of Ag content, and the increment of $\sigma_{ac}$ due to the addition of Ag is linearly proportional to Ag content in the concentration range of more than 0.05 at% [7]. Fig.3 shows variation of $s$ with $x$ and Ag content.

If the randomly distributed pair model [1], frequency dependence of $\sigma_{ac}$ can be written approximately as

$$\sigma_{ac} = \left(\frac{r_w}{a}\right)^4 = (B' - \frac{1}{2} \ln \omega)^4,$$  \hspace{1cm} \text{(1)}$$

where $r_w$ is the optimum hopping distance, $a$ the effective radius of localized state's wave function and $B'$ the quantity concerned with $r_w$ and the coefficient of relaxation time [8]. According to this model, the exponent $s$ decreases with
decreasing $r_w$. Since the value of $x$ represents the deviation from stoichiometric composition, the increase of $x$ means the increase of the density of localized defect states. If the carrier hopping between defect states takes place, $r_w$ decreases with increasing $x$. In $\text{As}_2\text{Se}_3:\text{Ag}$, $s$ has a constant value. Accordingly $r_w$ does not much varied with Ag content. If disordered state such as $\text{Ag}_2\text{Se}$ is formed in $\text{As}_2\text{Se}_3:\text{Ag}$ samples [9], two dangling bonds of As are expected to be set up in the neighbourhood of $\text{Ag}_2\text{Se}$ and construct a defect pair. This defect pair is similar to the one proposed by Elliott [2]. It is expected that the distance between two defect states within the pair is restricted in a small range. If the carrier hopping occurs within the defect pair, the experimental results are qualitatively interpreted, where the exponent $s$ has a constant value and the magnitude of $\sigma_{\text{ac}}$ is proportional to Ag content. According to Elliott [2,3], the exponent $s$ is represented as

$$s = 1 - \frac{6kT}{W_M},$$

where $W_M$ is the maximum barrier height which is taken to be equal to the band gap $E_g$ for the case of chalcogenide glasses. However, the barrier height between the defect states around $\text{Ag}_2\text{Se}$ may be smaller than $E_g$. If $W_M$ is about fourth of $E_g$, the value of $s$ becomes 0.7.

The above interpretation is also conducted from the dependence of the magnitude of dc conductivity and its activation energy on $x$ in $\text{As}_{40+x}\text{Se}_{60-x}$ and on Ag content in $\text{As}_2\text{Se}_3:\text{Ag}$, if Ag additives are concerned with dc conductivity [6].

3.2. Temperature Dependence of A.C. Conductivity of $\text{As}_2\text{Se}_3:\text{Ag}$

The ac conductivity $\sigma_{\text{ac}}$ of $\text{As}_2\text{Se}_3:\text{Ag}$ was measured from room temperature down to 77 K. Temperature dependence of total electrical conductivity $\sigma_\omega$ of $\text{As}_{40}\text{Se}_{60}\text{Ag}_{0.5}$ is shown in Fig.4. As seen in this figure, the slope of $\sigma_{\text{ac}} = \sigma_\omega - \sigma_{\text{dc}}$ versus $1/T$ decreases with decreasing temperature and with increasing frequency. As pointed out by Elliott [3], these facts suggest that ac conduction of $\text{As}_2\text{Se}_3:\text{Ag}$ is caused by the classical barrier hopping. It is to be noted, however, that $\sigma_{\text{ac}}$ of undoped $\text{As}_2\text{Se}_3$ is independent of temperature [7].

![Fig.4 Temperature dependence of dc (\(\sigma_{\text{dc}}\)) and total (\(\sigma_\omega\)) conductivity of \(\text{As}_2\text{Se}_3\) containing 0.5 at% Ag.](image-url)
The $\sigma_{ac}$ of $As_{40}Se_{60}Ag_{0.5}$ at different temperatures is shown in Fig.5. Fig.6 shows temperature variation of the exponent $s$ for the samples with various Ag contents. The values of $s$ increase with decreasing temperature. If phonon-assisted hopping mechanism is predominant, $s$ is predicted to decrease with decreasing temperature [10]. In classical hopping mechanism, on the other hand, the temperature dependence of $s$ is satisfied with experimental results as seen in eq.(2). At 77 K, the value of $s$ becomes more than 1. Namely super-linear frequency dependent conductivity is observed. In case that $r_0$ has a small value and states are pairing, it is pointed out by Elliott [11] that super-linear $\sigma_{ac}$ is produced.

By introducing Ag, as mentioned above, $Ag_{2}Se$ may be formed. Defect pairs around $Ag_{2}Se$ are considered to be satisfied with above conditions. In $As_{2}Se_{3}:Ag$, therefore, ac conduction seems to be contributed by the classical barrier hopping between defect states which are closely connected with Ag additives.

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