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THERMAL AND ELECTRICAL PROPERTIES OF HEAVILY DOPED Ge-Si ALLOYS UP TO 1300°K

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ABSTRACT

The thermal resistivity, Seebeck coefficient, electrical resistivity, and Hall mobility of Ge-Si alloys have been measured throughout the Ge-Si alloy system as functions of impurity concentration in the range 2×10^{18} - $4 \times 10^{20} \text{cm}^{-3}$, and of temperature in the range 300-1300°K. A qualitative interpretation of these properties is given. For power conversion, boron and phosphorus were found to be useful p-type and n-type impurities, respectively because of their high solid solubilities. At 1200°K, the maximum values of the dimensionless figure of merit, zT , were 0.8 for p-type $\text{Ge}_{.15}\text{Si}_{.85}$ alloy doped to $2.1 \times 10^{20} \text{cm}^{-3}$ holes, and 1.0 for n-type $\text{Ge}_{.15}\text{Si}_{.85}$ alloy doped to $2.7 \times 10^{20} \text{cm}^{-3}$ electrons. The maximum overall efficiency of a stable generator operating between 300-1200°K, using the best p-type and n-type materials was computed to be 10%.

I. INTRODUCTION

The recent discovery that the Ge-Si alloys possess a low thermal conductivity at high temperatures¹ has led to an investigation of these alloys for thermoelectric power generation. It was demonstrated that these alloys show a high power generating efficiency.^{1,2} In addition, their high mechanical strength, high melting point, low vapor pressure and resistance to atmospheric oxidation make them attractive for device applications.

This paper describes the detailed investigation of the thermal and electrical properties of Ge-Si alloys as a function of composition, doping impurity additions, and temperature, and the optimization of the material for thermoelectric power generation. The thermal resistivity as a function of composition exhibits a broad maximum near the middle of the alloy system.^{1,3} In addition to high thermal resistivity, however, for power generation, high melting point and large band gap are also required which favor the silicon rich alloys. Hence, on the basis of room temperature measurements taken throughout the Ge-Si alloys system, the $\text{Ge}_{.3}\text{Si}_{.7}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ alloy compositions were selected for the most extensive studies at high temperatures. Power conversion efficiencies were calculated as a function of doping and composition for two temperature ranges of interest.

II. EXPERIMENTAL

A. Materials

Most of the Ge-Si alloys were prepared by a zone leveling technique⁴ under conditions⁵ which avoided inhomogeneity due to constitutional supercooling. A few samples of very high silicon content were prepared by the Czochralski

technique. The Ge-Si alloys consisted of an array of elongated grains oriented in the direction of growth.⁶ Boron was added as a p-type impurity and phosphorus (or arsenic) was added as an n-type impurity. Small amounts of second phase often occurred in the most heavily doped specimens due to the limited solubility of the doping impurities.⁷ The zone leveled ingots, which were 25 cm long, varied less than 0.1 at% per cm in alloy composition and less than 2% per cm in carrier concentration. The alloy composition was obtained from density measurements, using the most recent data⁸ for the variation of density with alloy composition.

B. Measurements

Room temperature measurements were made of the Hall coefficient R_H (at 3600 gauss) and electrical resistivity, ρ , on bridge-shaped specimens, 0.8 mm x 1.7 mm x 12 mm. An ac method was used similar to that described by Donaghue and Eartherly,⁹ but their apparatus was modified to include a phase sensitive detector. The measurements were made at 22 cps. The sensitivity of the apparatus was about 3×10^{-8} V.

Room temperature measurements of thermal conductivity κ and Seebeck coefficient Q were made using an improved version of an absolute thermal conductivity apparatus previously described.¹⁰ The specimens were cut and surface ground to the shape of cubes with 10 mm sides. A film of silicone oil was applied between the sample and heater and sink to reduce thermal contact resistance. The thermal conductivity was corrected for errors due to radiation losses from the heater and the specimen. The former were determined experimentally in the absence of the sample to be about 3%, and the latter were calculated to

be about 1% of the heat conducted through the specimen. The thermocouples were attached to the heater and heat sink rather than to the specimen. This introduced an error in the measurement of the temperature gradient in the specimen due to the thermal contact resistances. These resistances were determined experimentally by measuring specimens of silicon, germanium, and Ge-Si alloys as a function of the length of the specimens. It was found that the contact resistance is independent of alloy composition. The measured sample contact resistance, which consisted of about $0.2 \text{ cm}^2 \text{ degW}^{-1}$ per interface, represented a correction of 3% of the temperature gradient of the specimen. The correction finally applied to the thermal conductivity measurement was only about 1% because of cancellation of the errors.

The measured values of the Seebeck coefficient were converted to absolute values by using the absolute values given in the literature for the reference metals.¹¹

High temperature measurements of the thermal diffusivity, κ/c_p , were made on specimens, 6mm x 6mm x 50mm, using the diffusivity apparatus described in detail by Abeles et al.¹² A thermal sine wave of 3 rpm is propagated along the rod, and the thermal diffusivity is determined from the phase shift and the attenuation of the wave. This method is fast and avoids errors in the thermal conductivity due to radiation losses. At each temperature the Seebeck coefficient and electrical resistivity were measured concurrently. An ac technique¹² which eliminates errors due to spurious emf's was used for measuring the Seebeck coefficient.

Modifications had to be made on the apparatus in order to extend the measurements up to 1300°K. In the improved arrangement shown in Fig. 1 all

nickel parts were replaced by molybdenum in order to eliminate cracking of the heaters. Alloying of the probes and thermocouples with the sample, and their oxidation represented major problems. It was found that niobium does not react with Ge-Si alloys up to very high temperatures and does not oxidize appreciable during the time required for the measurements, provided the pressure was less than 10^{-4} Torr. Therefore, the probes were made of 2 mil diameter niobium wires spot welded to the specimen. To avoid reaction between the thermocouples and the Ge-Si alloy a 5 mil diameter niobium wire was spot welded to the specimen and then the 1.5 mil diameter chromel and alumel wires were spot welded separately to the niobium as shown in Fig. 1. The niobium separates the thermocouples physically from the Ge-Si alloy while at the same time it provides an intimate thermal and electrical contact. The vacuum was improved to give a pressure less than 5×10^{-6} Torr in the sample chamber.

To determine the dependence of the high temperature thermal and electrical properties upon carrier concentration, it is desirable that the carrier concentration of each specimen have a fixed value during the measurements. Arsenic and phosphorus have a limited solid solubility which decreases with decreasing temperature. As a result the thermal and electrical properties of heavily doped n-type alloys change with time during the measurement due to precipitation of arsenic or phosphorus. In order to minimize changes in carrier concentration due to precipitation, most high temperature measurements on heavily doped n-type alloys were performed in the following manner: The samples (specimens 1975, 163 and 162) were annealed at about 1200°K and then quenched to room temperature to give them a large value of n prior to the measurement in the thermal diffusivity

apparatus. The samples were measured, first, over the range 300 to 600°K for which they were in a metastable condition, and then as rapidly as possible at higher temperatures. At 1000°K and above it was assumed that the carrier concentration was close to the thermal equilibrium value.

Precipitation effects have been observed for p-type alloys⁷ with hole concentrations greater than $3 \times 10^{20} \text{ cm}^{-3}$, but only above 1100°K. No precipitation effects were observed during the thermal diffusivity measurements of p-type specimens, however, because these changes were slow compared with the time of measurement.

To convert the thermal diffusivity κ/c_p into the thermal conductivity κ , the specific heat c_p per unit volume at constant pressure is required. Earlier,^{1,3} it was assumed that c_p follows a Debye function and reaches the Dulong-Petit value $c_{DP} = 3R$ per mole at high temperatures. This assumption is incorrect because the specific heat of semiconductors exceeds the Dulong-Petit value at high temperatures as a result of anharmonicity.¹³ Steigmeier and Kudman¹⁴ have shown that it is possible to reduce all available experimental specific heat data on silicon, germanium and III-V compounds to one curve with some spread by plotting c_p/c_{DP} versus T/θ , where θ is the Debye temperature. The specific heat c_p of Ge-Si alloys was derived by fitting a curve through the data given for silicon¹⁵ in Fig. 1 of Ref. 14, and assuming that this curve applies for all Ge-Si alloys. The resulting specific heat for $\text{Ge}_{.3}\text{Si}_{.7}$ alloy is shown in Fig. 2. The curve used for other alloys between the $\text{Ge}_{.3}\text{Si}_{.7}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ compositions differs by less than 1% from the one given in Fig. 2.

The Dulong-Petit specific heats per unit volume, $c_{DP} = 3.312 \times 10^{24} / a^3$ (J $\text{cm}^{-3} \text{ deg}^{-1}$), were computed using the lattice parameter a , in cm, of Ge-Si

alloys.⁸ The Debye temperatures of the alloys were derived from the expression¹⁶ (in cgs units) $\theta = 4.19 \times 10^{-8} a^{-3/2} M^{-1/2} G$, where M is the mean "gram atomic weight" of the alloy, and G is a function of the elastic constants. The value of G was obtained from a linear interpolation between the value for germanium, $G = 1.017$ and the value for silicon, $G = 1.033$.

The assumptions made above for the specific heat of the alloys have since been confirmed experimentally.¹⁷

C. Results

Listed in Table I are the composition, impurity type, and carrier concentration of the samples measured at high temperatures in the thermal diffusivity apparatus. The carrier concentrations given are defined by $n = 1/eR_H$. In Figs. 3, 4 and 5, are shown the temperature dependence of thermal resistivity $1/\kappa$, absolute Seebeck coefficient, Q, and electrical resistivity, ρ , respectively for p-type alloys and in Figs. 6, 7 and 8 are shown the dependence of $1/\kappa$, Q and the Hall mobility, $\mu_H = R_H/\beta$ upon hole concentration, with alloy composition and temperature as parameters. Hall mobility rather than electrical resistivity was plotted as a function of n in order to bring out more clearly the variation of ρ with impurity concentration and alloy composition.

In Figs. 9, 10 and 11, are shown the temperature dependence of $1/\kappa$, Q, and ρ respectively for n-type alloys with the $Ge_{.3}Si_{.7}$ alloy composition and in Figs. 12, 13 and 14 are shown corresponding data for n-type alloys with the compositions $Ge_{.2}Si_{.8}$ and $Ge_{.15}Si_{.85}$. In Figs. 15, 16 and 17 are shown the dependence of $1/\kappa$, Q and μ_H respectively upon electron concentration, with temperature and alloy composition as parameters.

III. DISCUSSION

A. Thermal Conductivity

The thermal conductivity of a semiconductor is the sum of a lattice component, κ_{ℓ} , and an electronic component, κ_{e1} . The electronic component is the sum of the single band and the bipolar contributions. The lattice thermal conductivity is determined by scattering of phonons by phonons, by point defects, and by free carriers.

The curves of $1/\kappa$ for p-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys (Fig. 3) exhibit maxima as a function of temperature because the ambipolar contribution to κ_{e1} offsets the decrease in κ_{ℓ} with increasing temperature. The thermal resistivity at 300°K for p-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys is plotted as a function of carrier concentration in Fig. 6. It is seen that the points measured by the absolute method agree with the diffusivity measurements to within 3%. The curve, which has been drawn through all the points for $\text{Ge}_{.3}\text{Si}_{.7}$ alloy exhibits a maximum at about $3 \times 10^{20} \text{ cm}^{-3}$. The behavior for other alloy compositions is similar. The value of $1/\kappa$ first increases as the impurity concentration increases, due to scattering of phonons by the free carriers. At very high carrier concentrations κ_{e1} becomes appreciable, and the thermal resistivity decreases. With increasing temperature the maximum in thermal resistivity is shifted to lower carrier concentrations because the relative contribution of κ_{e1} is larger.

The dependence of the thermal resistivity of n-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys upon temperature (Fig. 9) and upon carrier concentration (Fig. 15) is qualitatively similar to that for the p-type material, except that the maximum in the curve of $1/\kappa$ versus n has not been reached at $1.5 \times 10^{20} \text{ cm}^{-3}$. This

limit on the carrier concentration was imposed by the limited solid solubility of phosphorus. The increase in $1/\kappa$ with n due to phonon-electron scattering at 300°K is larger for n-type than for p-type alloys. The effect diminishes with increasing temperature. Increases in $1/\kappa$ due to scattering of phonons by free carriers, and the greater effect of electrons than holes in this process have previously been observed in germanium.¹⁸ A detailed analysis of phonon-electron scattering in Ge-Si alloys is given in a separate paper.¹⁹

Figure 18 shows the dependence of thermal resistivity at 300°K upon alloy composition for 'undoped' materials ($n \leq 2 \times 10^{18} \text{ cm}^{-3}$) and materials doped to $1.5 \times 10^{20} \text{ cm}^{-3}$ with boron, arsenic, and phosphorus respectively. The data for the 'undoped' specimens, which were measured in the absolute thermal conductivity apparatus, may include a small photon contribution to the thermal conductivity. The data for the doped specimens were estimated from the data in Figs. 6 and 15. The maximum exhibited by the thermal resistivity of the 'undoped' samples as a function of composition is due to scattering of phonons by point defects.³ Heavily doped samples have a significantly larger thermal resistivity than the undoped samples due to additional scattering of phonons by free carriers as indicated above. It is also seen that the thermal resistivity of phosphorus doped samples is about 5% larger than that of arsenic doped specimens. In both arsenic and phosphorus doped material, phonons may also be scattered by the precipitates. The difference in the thermal resistivity of the arsenic and phosphorus doped alloys may therefore be related to a difference in the nature of the precipitates.

B. Electrical Properties

The Seebeck coefficient (Fig. 4) and electrical resistivity (Fig. 5) of p-type $\text{Ge}_{.3}\text{Si}_{.7}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ alloys increase nearly linearly with temperature. The Hall mobility μ_H varies with temperature approximately as $T^{-0.8}$. The dependence of Q upon $\log n$ for several compositions is shown in Fig. 7. For constant hole concentration at 300°K the Seebeck coefficient increases slightly with increasing silicon content of the alloys. A major part of this effect is probably produced by the change in valence band structure with increasing silicon content in the Ge-Si alloy system.²⁰ No direct comparison will be made with the Seebeck coefficients of silicon and germanium due to the effect of phonon drag in these materials. The Hall mobility, μ_H , at 300°K decreases with increasing hole concentration (Fig. 8), but the change becomes very small above about $2 \times 10^{20} \text{ cm}^{-3}$. Heavily doped p-type Ge-Si alloys have about a factor of 6 smaller Hall mobility than that of lightly doped alloys²¹ due to the large additional ionized impurity scattering.

The dependence of the electrical properties of n-type Ge-Si alloys upon temperature (Figs. 10, 11, 13, and 14) and carrier concentration (Figs. 16 and 17) is qualitatively similar to that of the p-type alloys, except for the following differences: The values of Q and ρ of the n-type specimens #68 and #41 (Figs. 10 and 11), which have relatively small values of carrier concentration, reach a maximum as a function of temperature because of the onset of intrinsic conduction. However, the maxima observed for the heavily doped specimens #1834, #1975, #163, and #162 (Figs. 10, 11, 13, and 14) are not due to intrinsic effects. Rather they are caused by the dissolution of precipitates. For constant carrier concentration the Seebeck coefficient at 300°K is

practically independent of composition in the range $\text{Ge}_{.4}\text{Si}_{.6}$ to $\text{Ge}_{.15}\text{Si}_{.85}$, while that of the $\text{Ge}_{.8}\text{Si}_{.2}$ alloys is about 10% larger, probably due to the change in the numbers of valleys in the conduction band close to this composition.²² The slope of the curve of μ_{H} versus carrier concentration for n-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloy is about twice that of the p-type material. The value of μ_{H} of phosphorus doped $\text{Ge}_{.3}\text{Si}_{.7}$ alloys are about 7% larger than those of arsenic doped alloys at 300°K. A similar effect has previously been observed for Ge and Si doped to high carrier concentrations with As, P, and Sb.²³ A detailed analysis of the electrical properties of Ge, Si and Ge-Si alloys is presented in a separate paper.²⁴

C. Figure of Merit and Efficiency

The temperature dependence of the dimensionless thermoelectric figure of merit, zT , where $z=Q^2/\rho\kappa$, calculated from the experimental data for p-type and n-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys is shown in Figs. 19 and 20, respectively. The shift in the maximum zT of the p-type alloys to progressively higher temperatures for increasing hole concentration reflects primarily the dependence of the quantity Q^2/ρ upon temperature and hole concentration. The temperature dependence of zT for n-type alloy (Fig. 20) is similar, except that the largest values of zT are about 30% larger than the corresponding values for p-type alloy.

The power conversion efficiency, η , of the individual p-type and n-type materials in Table I was calculated for 1-cm cube thermoelements operating over the temperature ranges, 300°-1200°K and 600°-1200°K, using the expression of Cohen and Abeles²⁵ for η of a thermoelectric generator with temperature varying parameters. In the calculation it was assumed that each material was paired with a hypothetical material having properties identical to the measured

values of Q , ρ , and $1/\kappa$ of the real material except that the sign of Q was opposite. For p-type materials, the value of η for each specimen corresponds to a single carrier concentration throughout the thermoelement. Fig. 21 shows the variation of the calculated η with carrier concentration for p-type $\text{Ge}_{.3}\text{Si}_{.7}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ alloys. It can be seen that for the $\text{Ge}_{.3}\text{Si}_{.7}$ composition, the maximum η occurs at 1.8×10^{20} holes cm^{-3} , and that the change in η between $\text{Ge}_{.3}\text{Si}_{.7}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ is small.

For the heavily doped n-type material, the properties change with time because of the temperature dependent solid solubility of the n-type impurity, until the stable condition is reached. Therefore, efficiency calculations of n-type material were made for two cases: (1) for the initial condition using the measured properties and (2) for the stable condition using the estimated equilibrium properties. The resulting values of η are shown in Fig. 21 as a function of the initial carrier concentration. The maximum initial η of $\text{Ge}_{.3}\text{Si}_{.7}$ alloy occurs at 1.5×10^{20} electrons cm^{-3} and thus, phosphorus, which has a higher solid solubility than arsenic, is the most suitable n-type impurity for power generation. It can be seen that the change from initial η to stable η is small for $\text{Ge}_{.3}\text{Si}_{.7}$ alloy and negligible for $\text{Ge}_{.5}\text{Si}_{.85}$ alloy.

For each of the two alloy compositions the measured properties of the best p-type material, boron doped, were then combined with the estimated properties of the best stable n-type material, phosphorus doped, to calculate the actual efficiency of a generator operating over the temperature range 300-1200°K. The resulting efficiencies are 9.9% for a stable $\text{Ge}_{.3}\text{Si}_{.7}$ alloy generator and 10.0% for a stable $\text{Ge}_{.15}\text{Si}_{.85}$ alloy generator.

It is concluded that Ge-Si alloys are competitive in efficiency with the best available high temperature thermoelectric materials.²⁶ Further, the thermal and electrical properties of the n-type and p-type alloys are closely matched, which is a considerable advantage in device design. Most important, however, their chemical stability and mechanical strength at elevated temperatures allows them to be operated without encapsulation both in air and in vacuum without appreciable change in efficiency.

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TABLE I

Properties of the diffusivity specimens

Specimen	Alloy Composition (At % Si)	Impurity	Carrier Concentration (10^{-19}cm^{-3})
7	72.0	B	3.4
42	69.3	B	8.9
1941	71.7	B	18
1970	72.0	B	24
82	71.0	B	35
1241	84.6	B	30
1976	85.3	B	21
68	73.8	As	.22
41	66.8	As	2.3
163	70.2	P	6.7
162	71.3	P	15
1834	79.5	P	14
1975	86.8	P	27

FIGURE CAPTIONS

- Fig. 1 Sample holder: C, ceramic spacer; H, metallized ceramic heater; L, molybdenum heater current leads; Mo, molybdenum; Nb, niobium; S, sample; ST, stainless steel; TC, chromel-alumel thermocouples; W, tungsten.
- Fig. 2 The specific heat of $\text{Ge}_{.3}\text{Si}_{.7}$ alloy as a function of temperature.
- Fig. 3 The thermal resistivity of p-type Ge-Si alloys as a function of temperature. For clarity, lines are drawn only through the data for the $\text{Ge}_{.3}\text{Si}_{.7}$ composition.
- Fig. 4 The absolute Seebeck coefficient of p-type Ge-Si alloys as a function of temperature. For clarity, lines are drawn only through the data for the $\text{Ge}_{.3}\text{Si}_{.7}$ composition.
- Fig. 5 The electrical resistivity of p-type Ge-Si alloys as a function of temperature. For clarity, lines are drawn only through the data for the $\text{Ge}_{.3}\text{Si}_{.7}$ composition.
- Fig. 6 The thermal resistivity of p-type Ge-Si alloys as a function of carrier concentration with temperature and alloy composition as parameters.
- Fig. 7 The absolute Seebeck coefficient of p-type Ge-Si alloys as a function of carrier concentration with temperature and alloy composition as parameters.
- Fig. 8 The Hall mobility of p-type Ge-Si alloys as a function of carrier concentration at 300°K with the alloy composition as a parameter.
- Fig. 9 The thermal resistivity of n-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys as a function of temperature.
- Fig. 10 The absolute Seebeck coefficient of n-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys as a function of temperature.
- Fig. 11 The electrical resistivity of n-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys as a function of temperature.
- Fig. 12 The thermal resistivity of n-type $\text{Ge}_{.2}\text{Si}_{.8}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ alloys as a function of temperature. The numbers indicate the sequence of the measurements. The filled circles represent the initial condition of the samples while the open circles were obtained after partial annealing.

- Fig. 13 The absolute Seebeck coefficient of n-type $\text{Ge}_{.2}\text{Si}_{.8}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ alloys as a function of temperature. The numbers indicate the sequence of the measurements. The filled circles represent the initial condition of the samples while the open circles were obtained after partial annealing.
- Fig. 14 The electrical resistivity of n-type $\text{Ge}_{.2}\text{Si}_{.8}$ and $\text{Ge}_{.15}\text{Si}_{.85}$ alloys as a function of temperature. The numbers indicate the sequence of the measurements. The filled circles represent the initial condition of the samples while the open circles were obtained after partial annealing.
- Fig. 15 The thermal resistivity of n-type Ge-Si alloys as a function of carrier concentration with temperature, alloy composition and doping agent as parameters.
- Fig. 16 The absolute Seebeck coefficient of n-type Ge-Si alloys as a function of carrier concentration with temperature, alloy composition, and doping agent as parameters.
- Fig. 17 The Hall mobility of n-type Ge-Si alloys as a function of carrier concentration at 300°K with alloy composition and doping agent as parameters.
- Fig. 18 The thermal resistivity of undoped and doped, p- and n-type Ge-Si alloys as a function of alloy composition at 300°K.
- Fig. 19 The dimensionless figure of merit zT of p-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys as a function of temperature.
- Fig. 20 The dimensionless figure of merit zT of n-type $\text{Ge}_{.3}\text{Si}_{.7}$ alloys as a function of temperature.
- Fig. 21 The power conversion efficiencies of p-type and n-type Ge-Si alloys as a function of carrier concentration. Hot junction temperature; 1200°K; cold junction temperatures: 300 and 600°K, respectively. The solid symbols shown for the n-type material are estimated values, representing the stable condition, and are plotted for the initial carrier concentration.

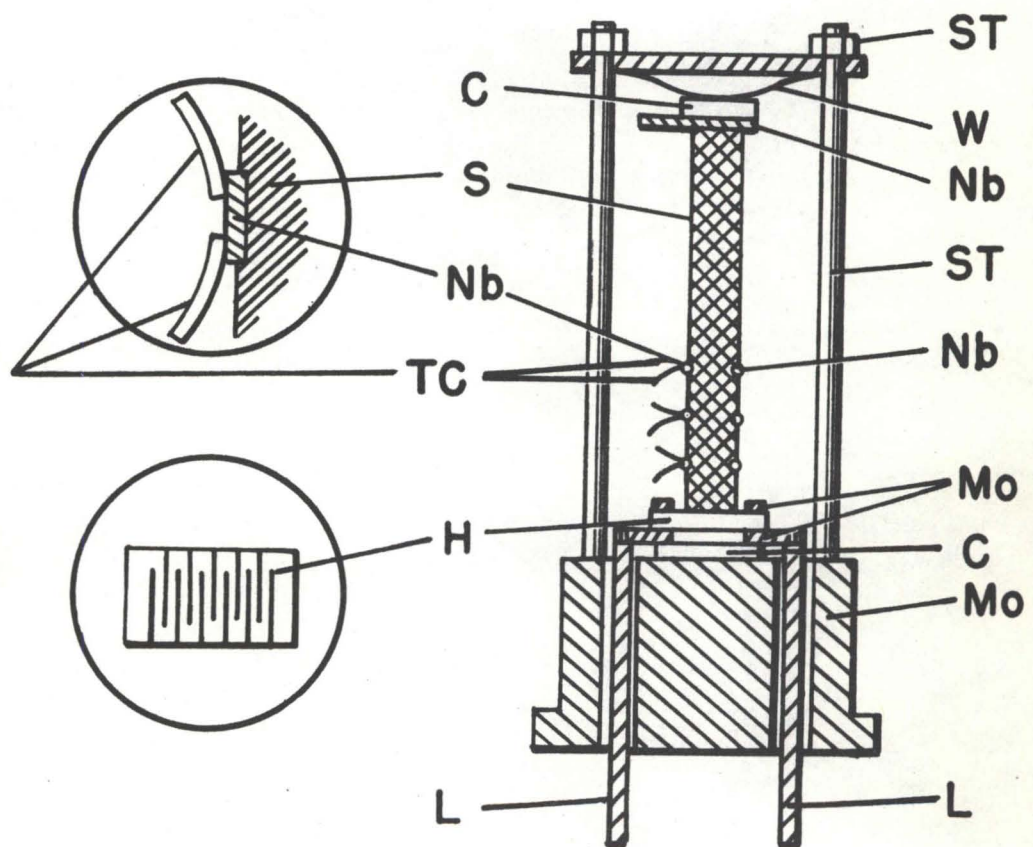


FIG. 1

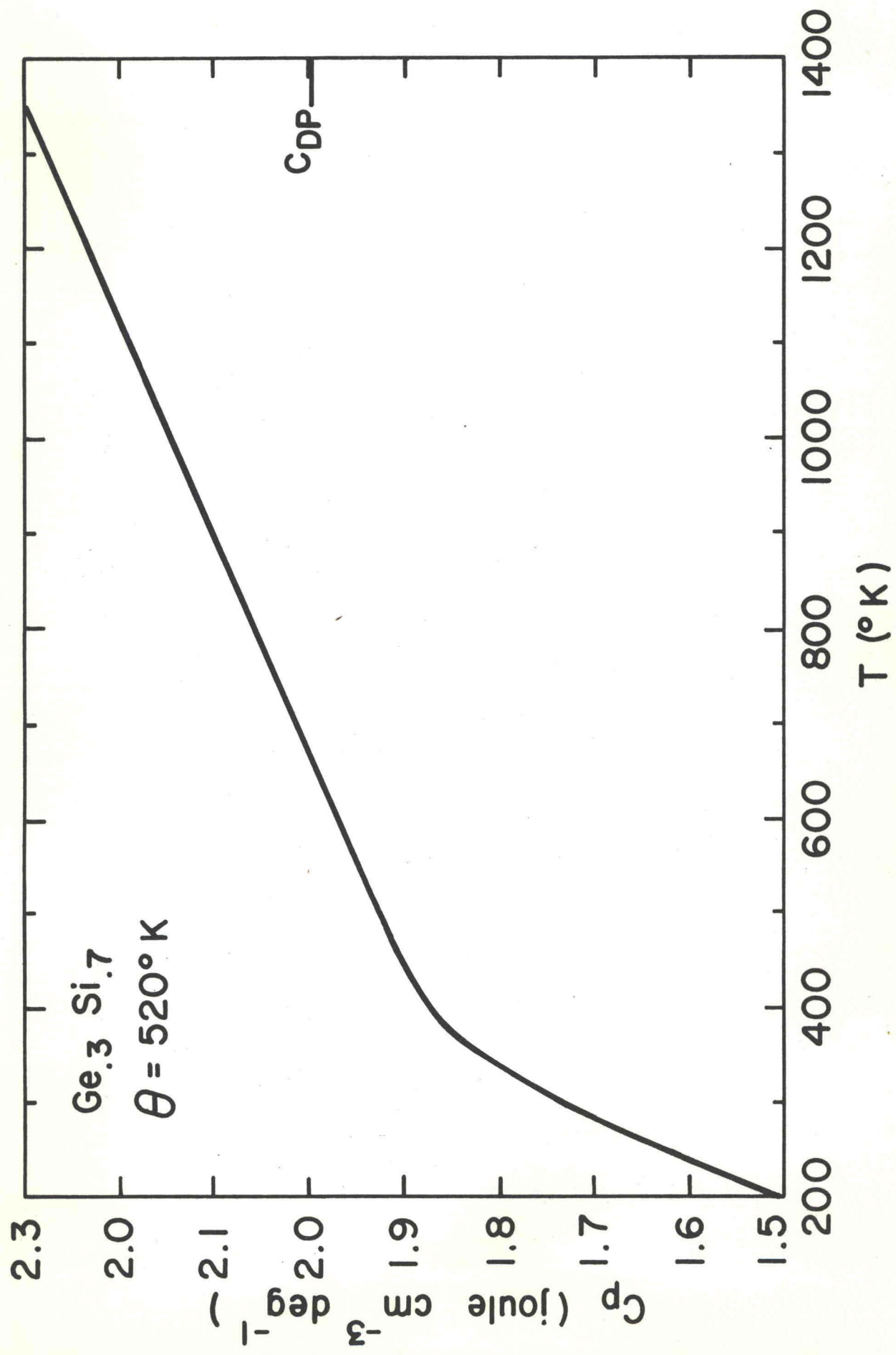
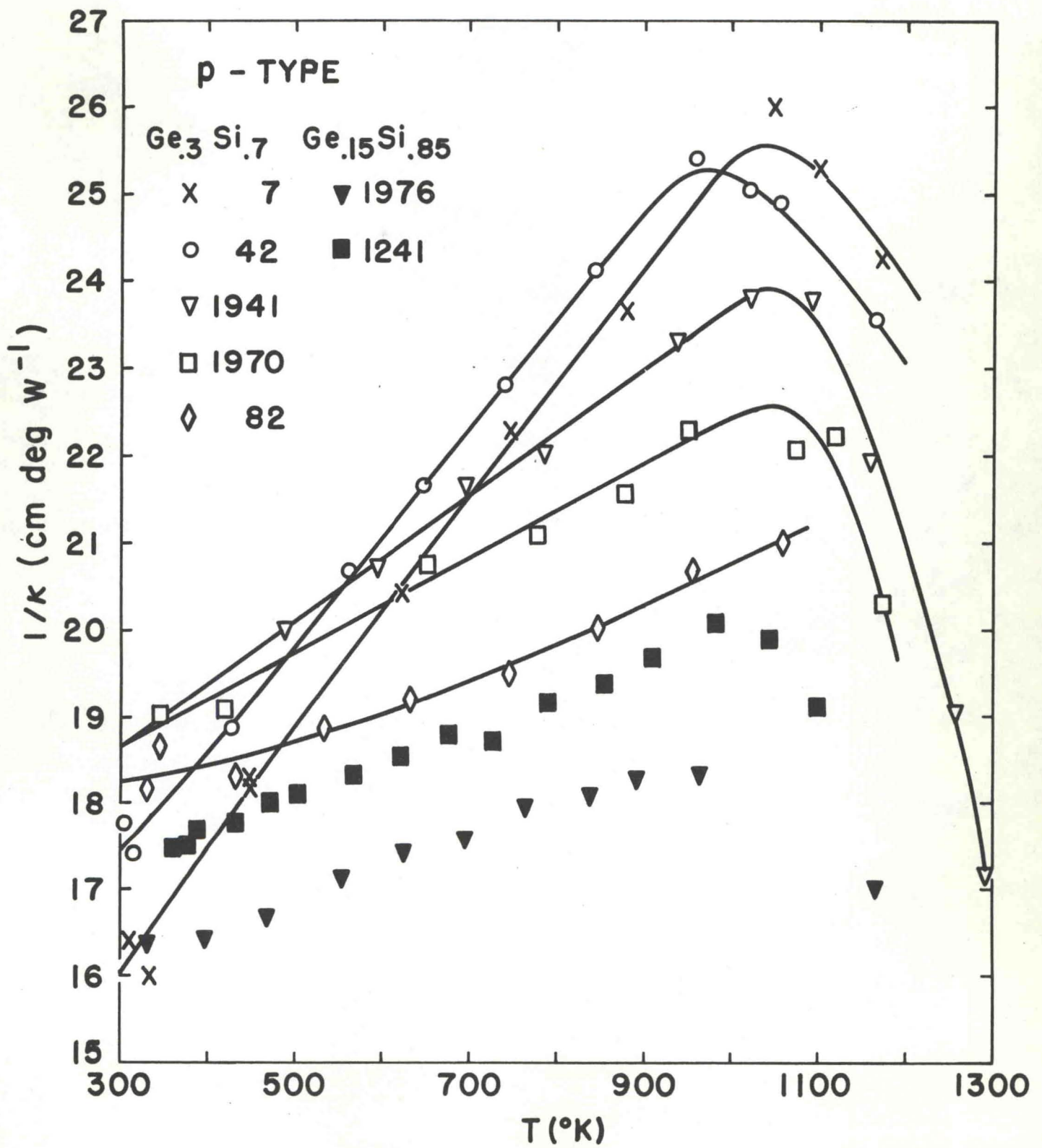


FIG.2



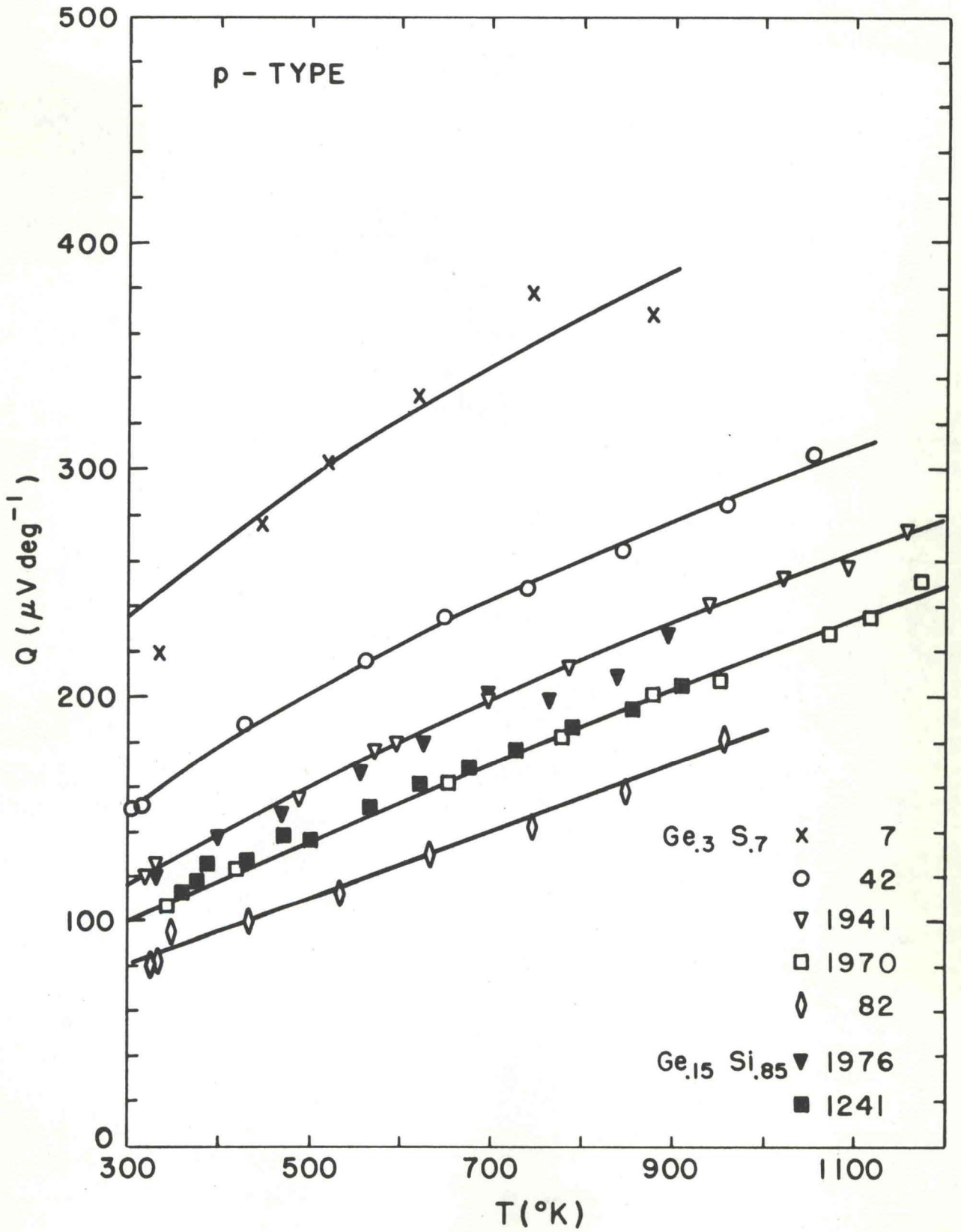


FIG.4

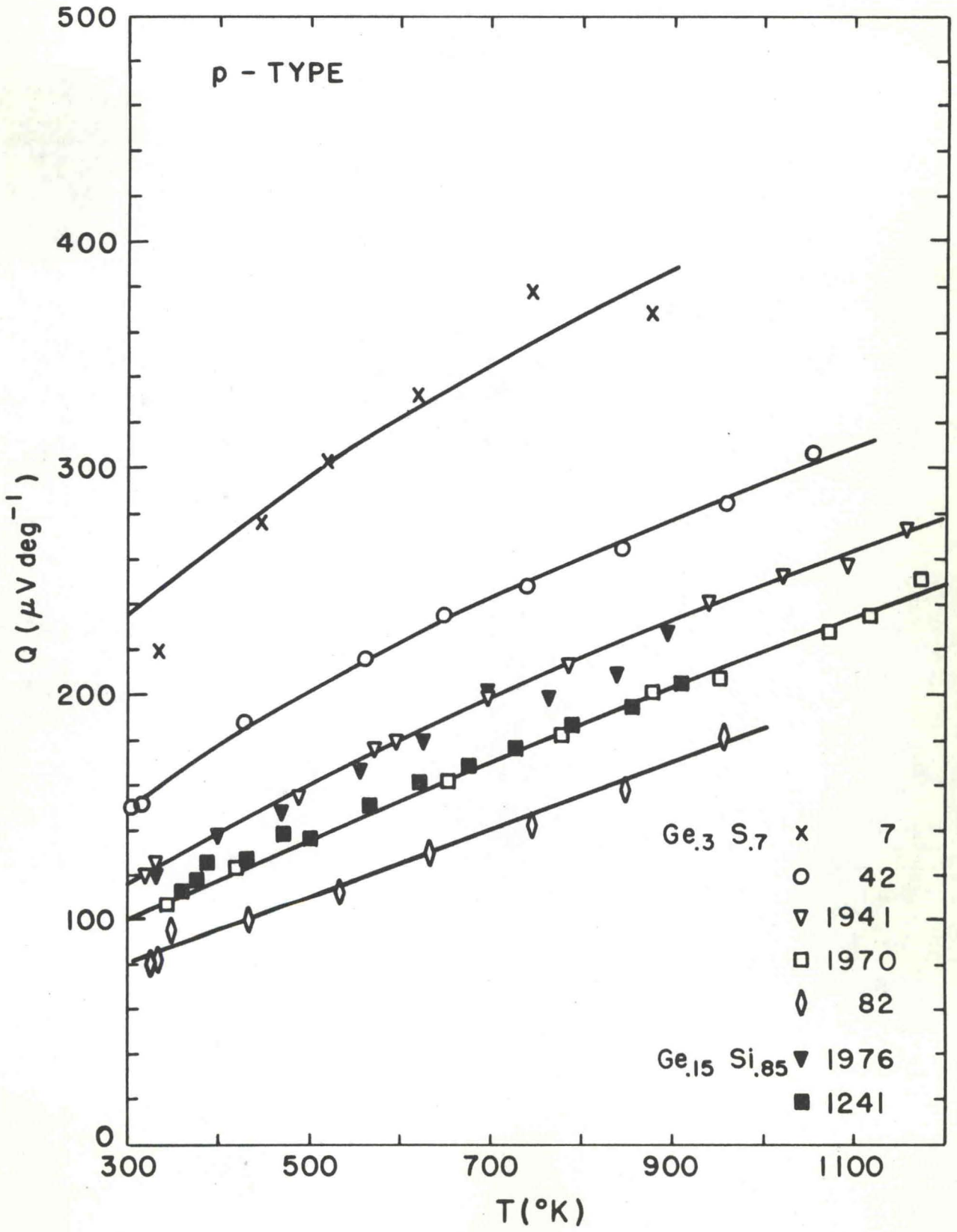
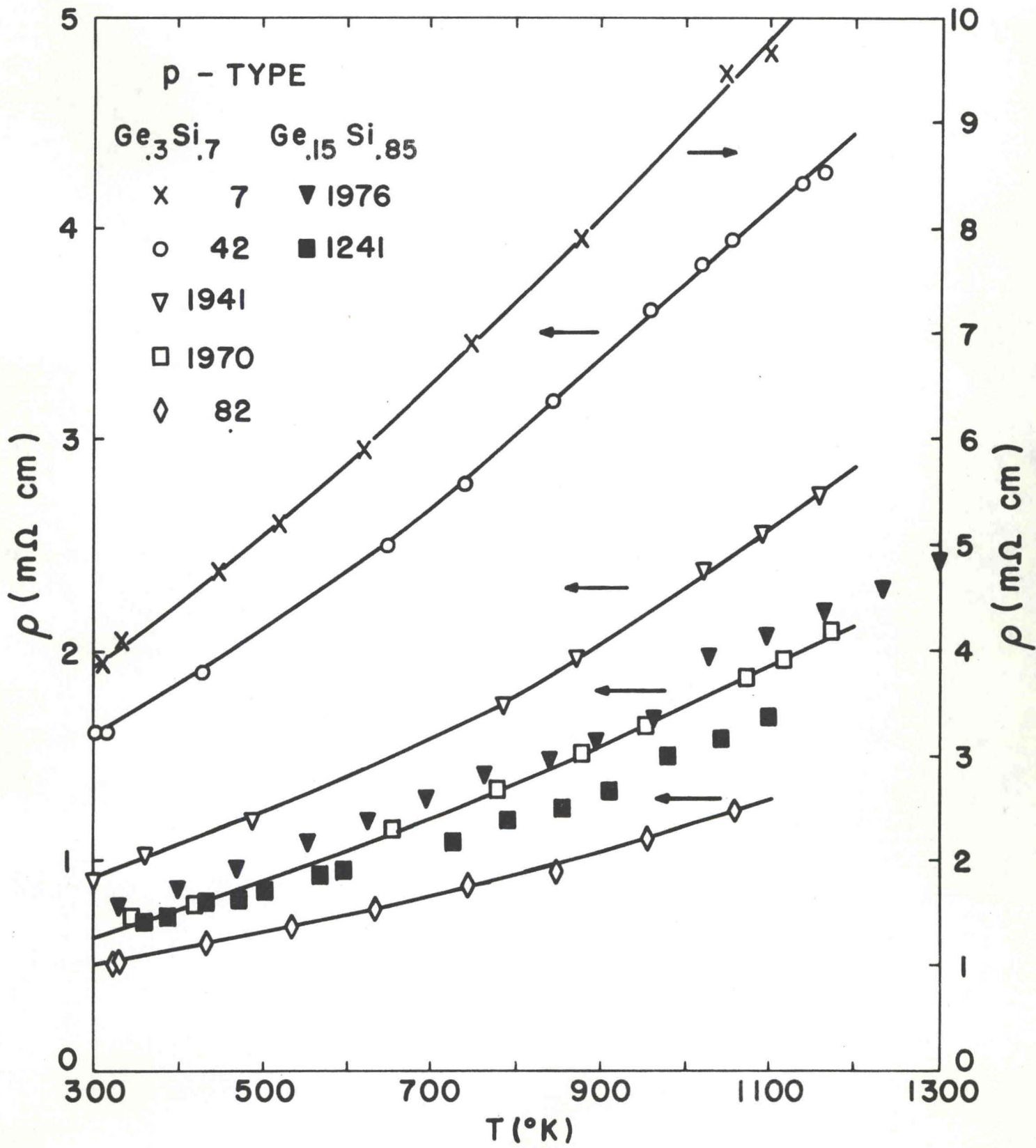


FIG.4



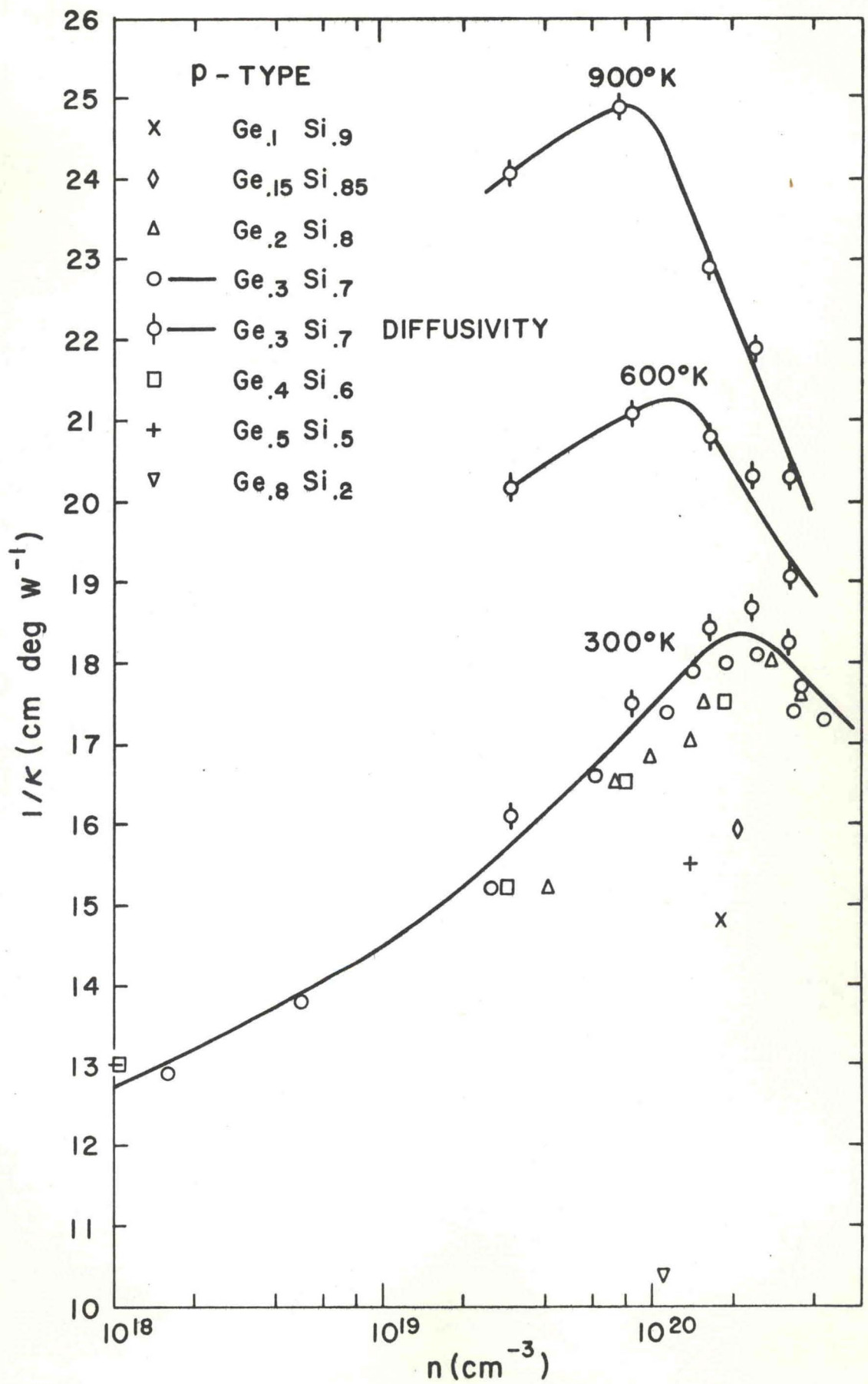


FIG.6

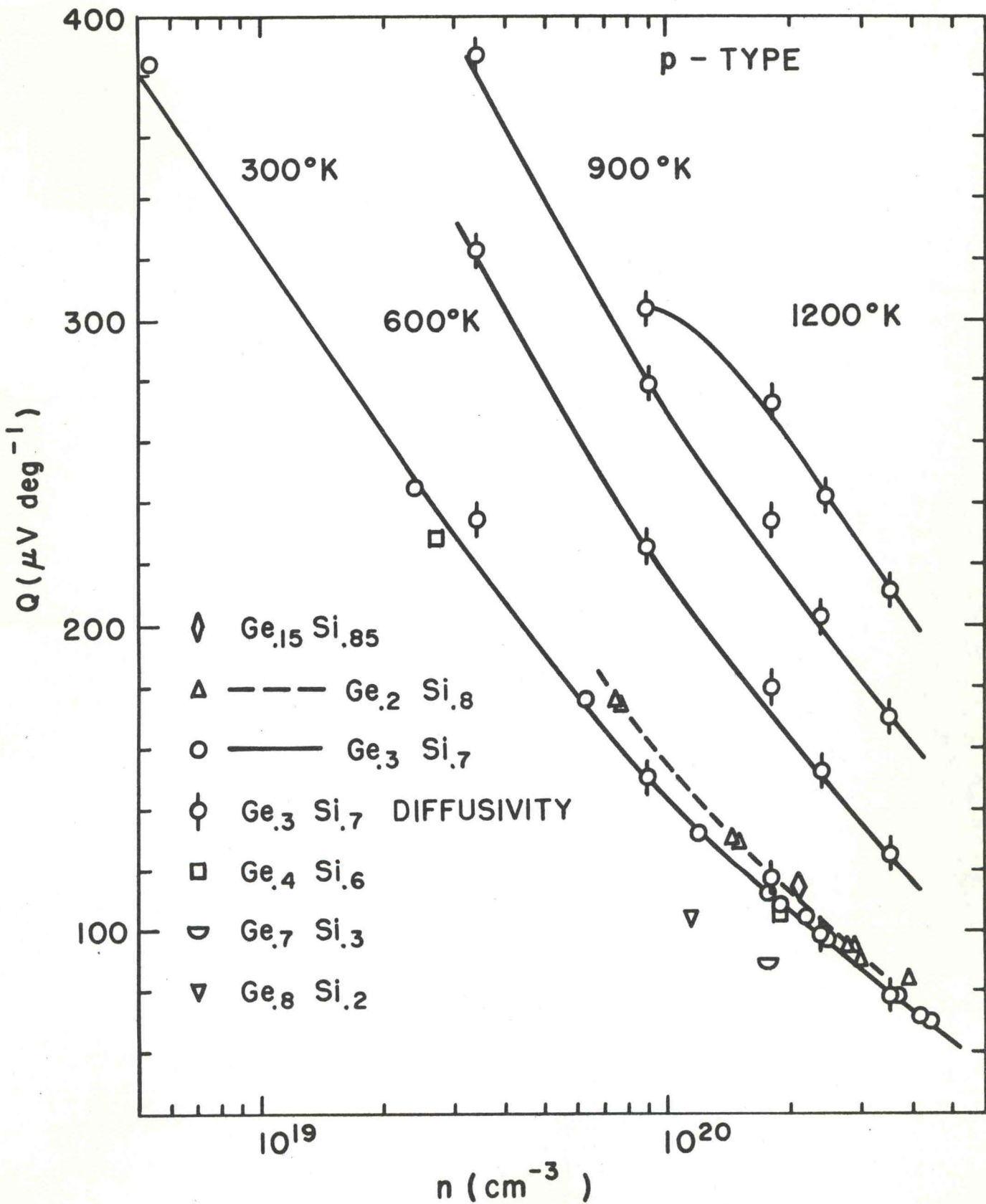


FIG.7

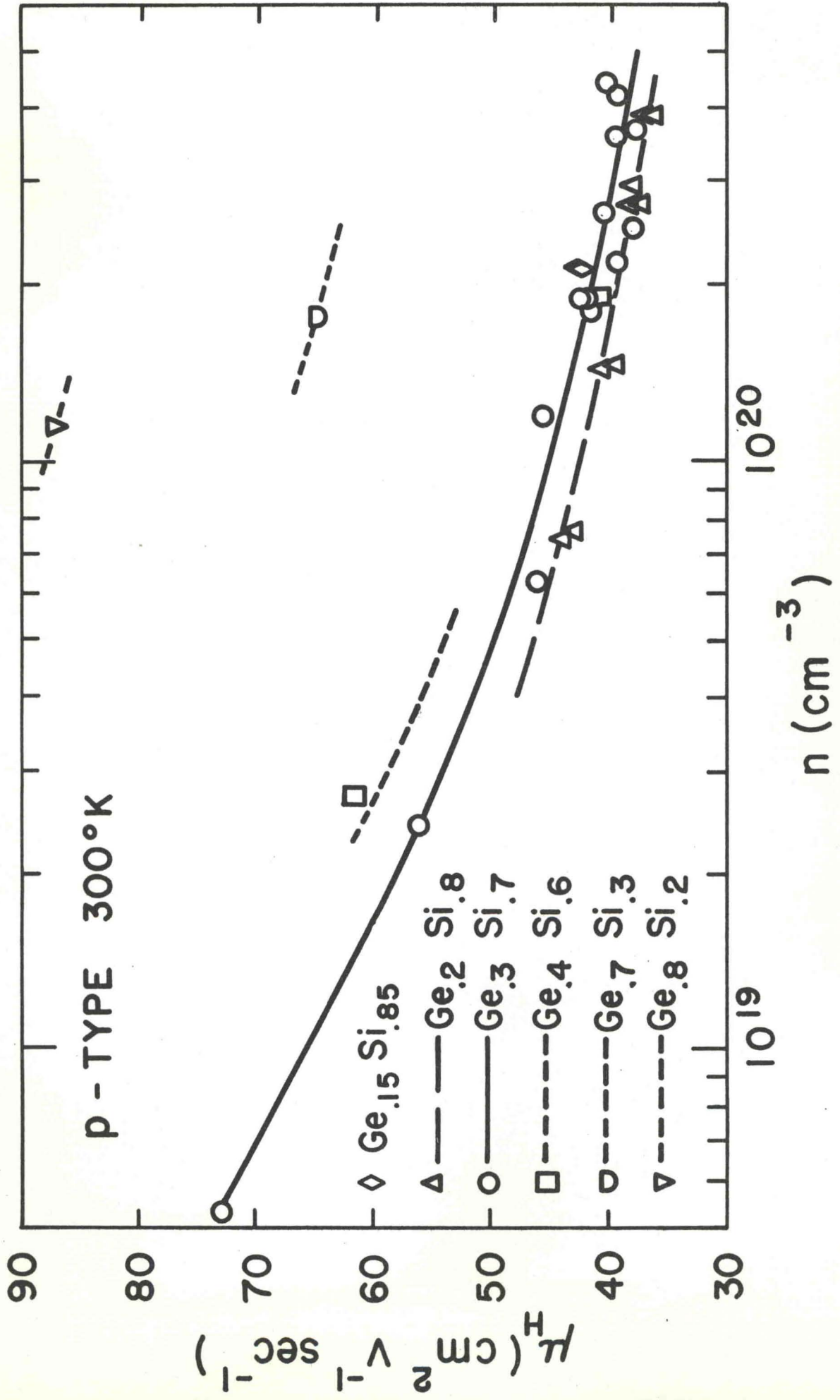


FIG.8

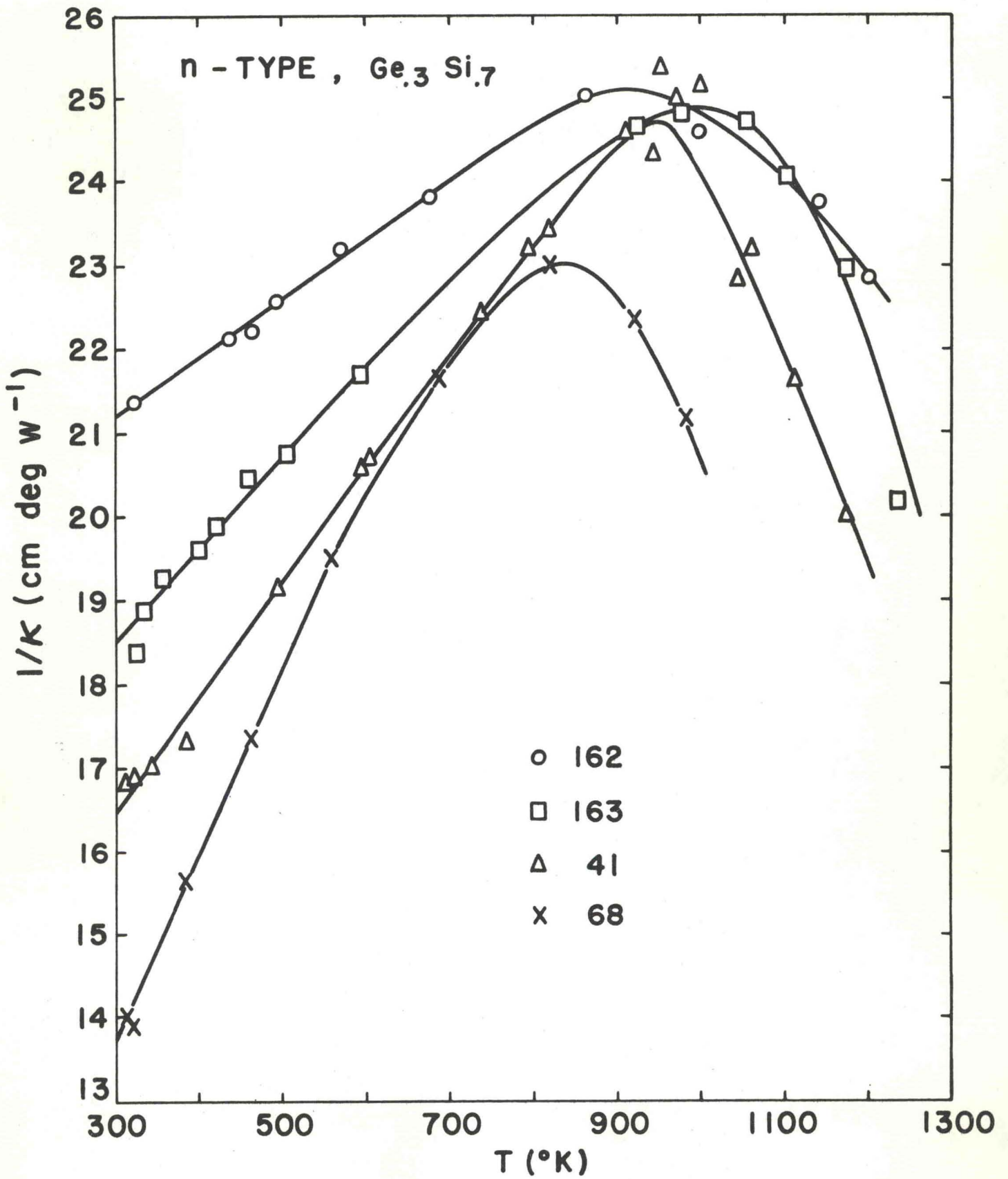


FIG.9

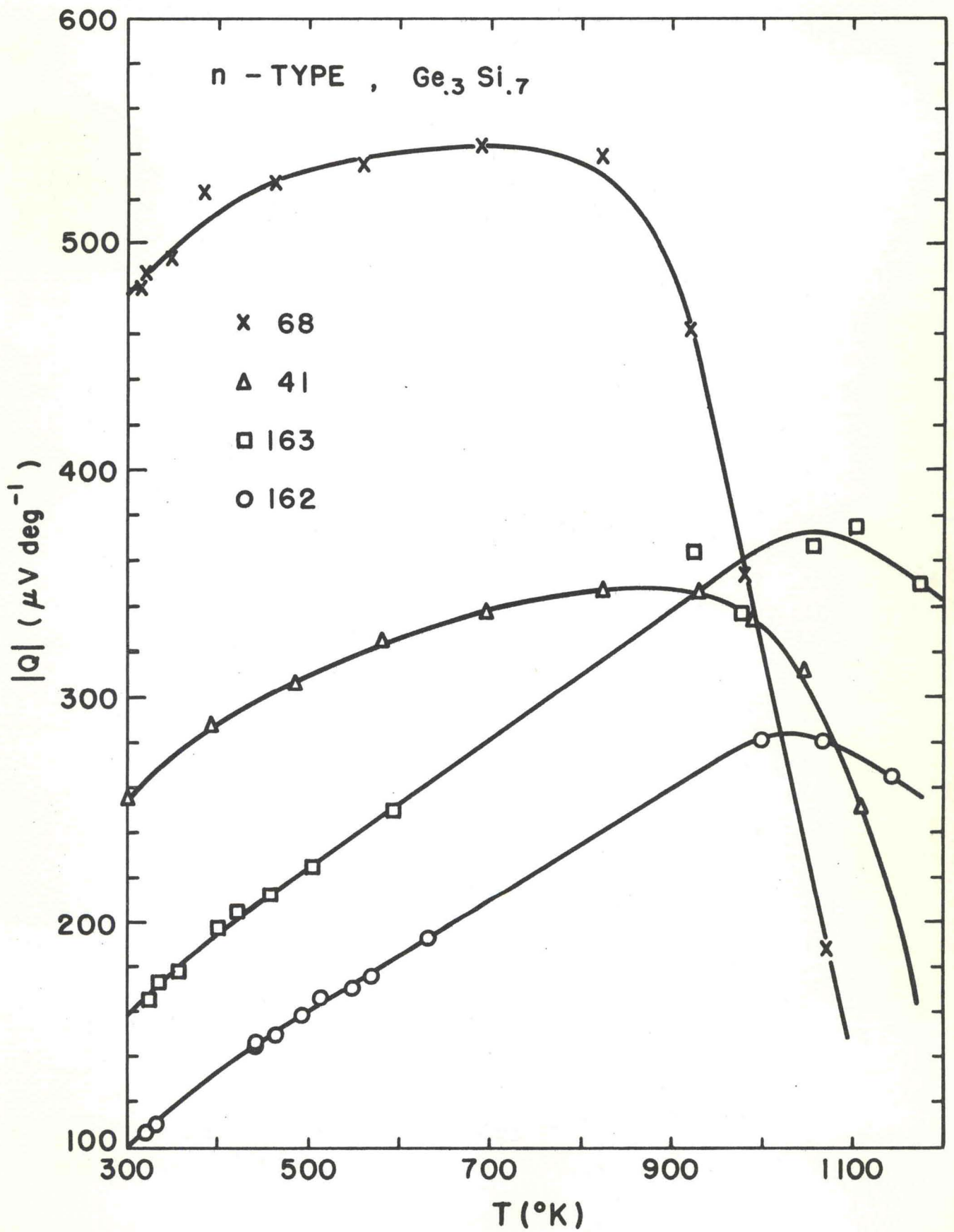


FIG. 10

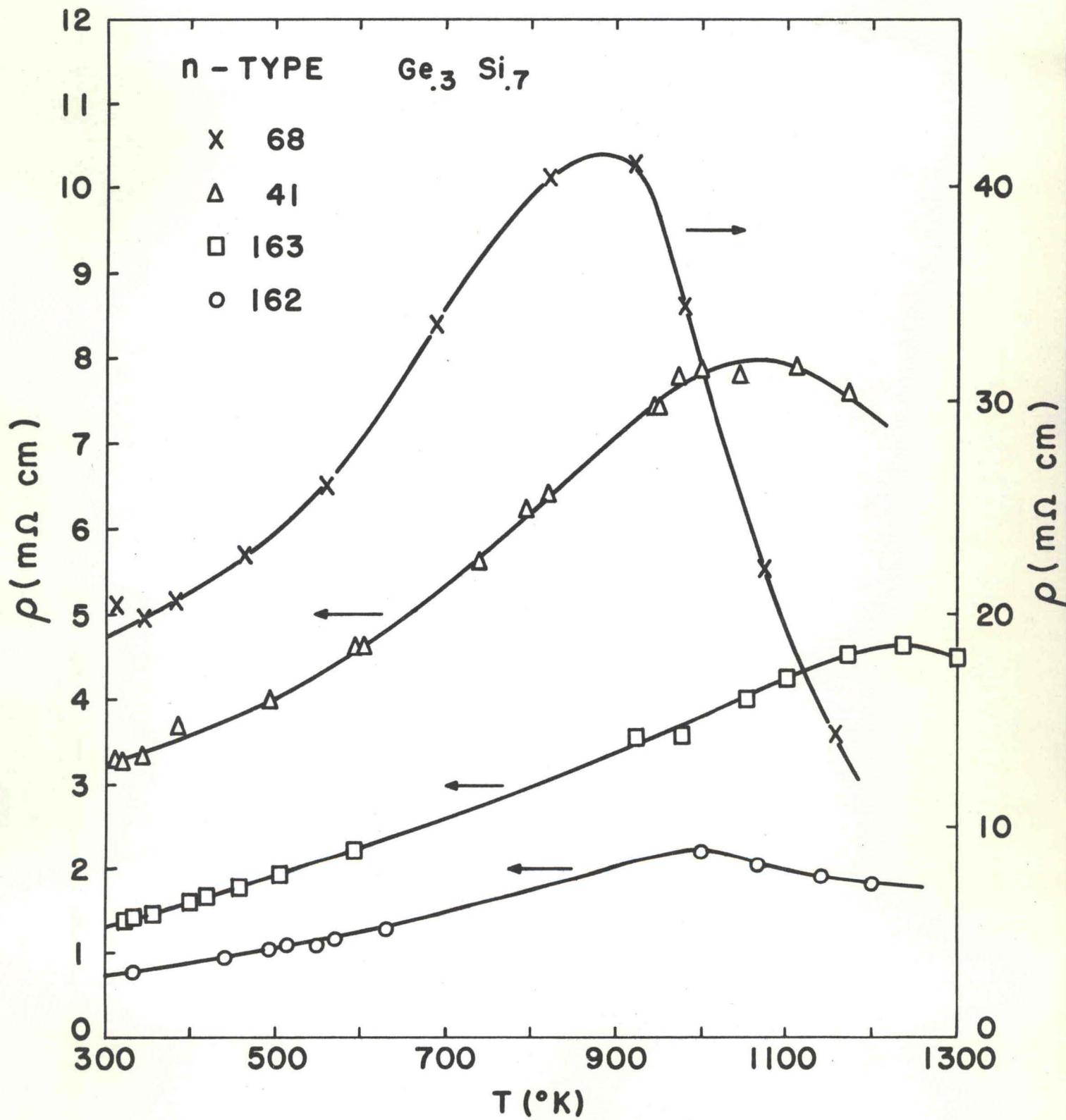


FIG. II

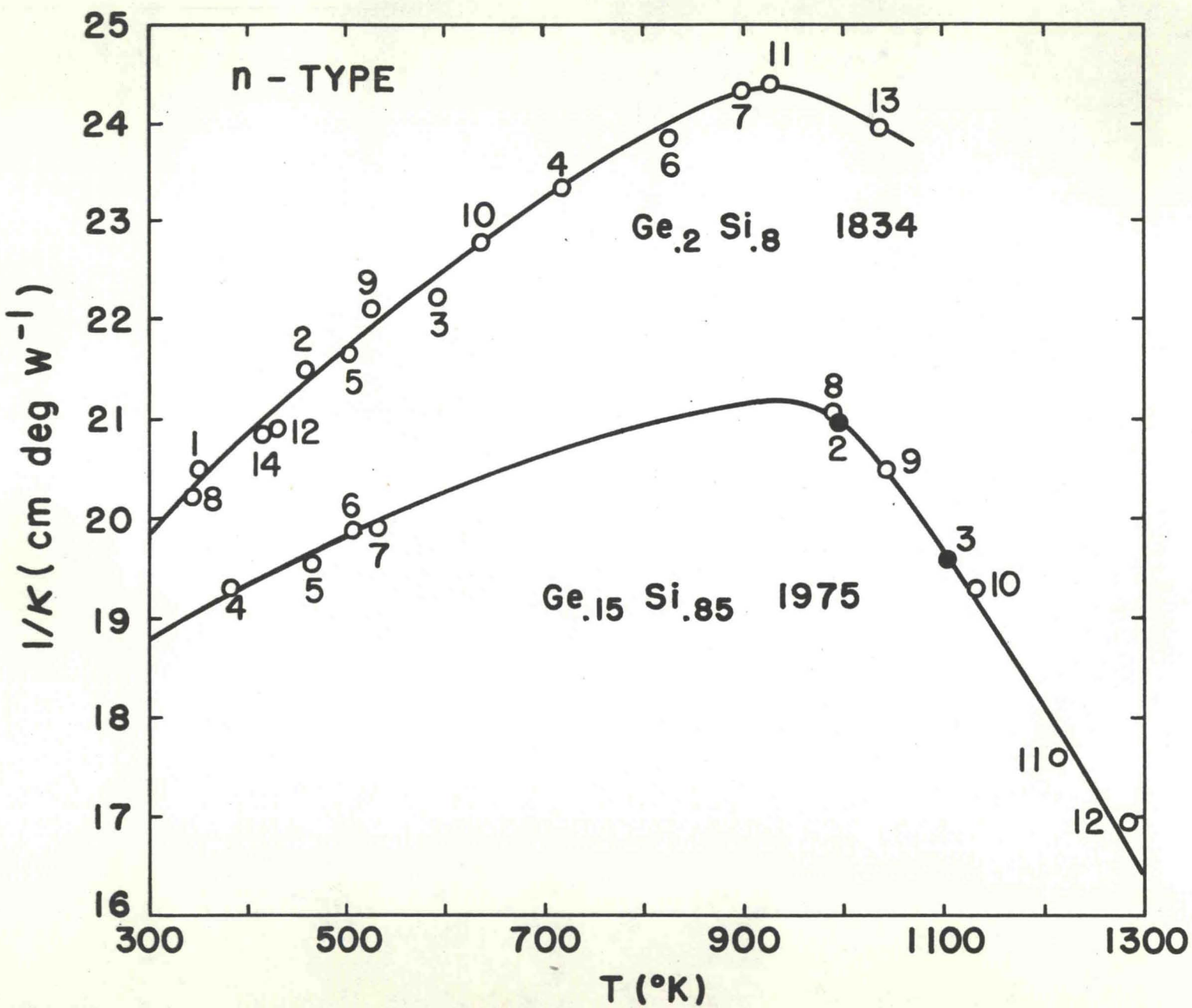


FIG. 12

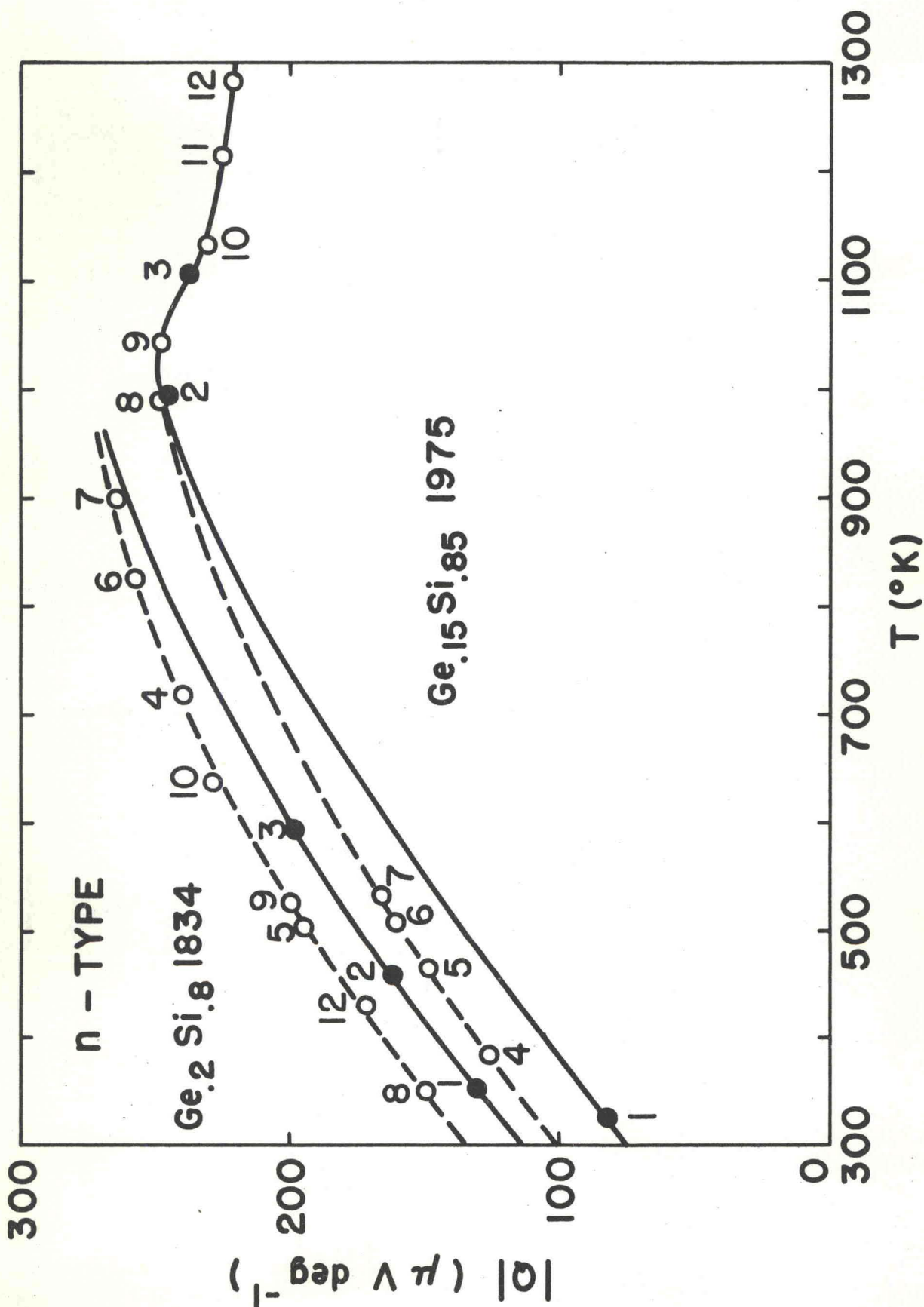


FIG. 13

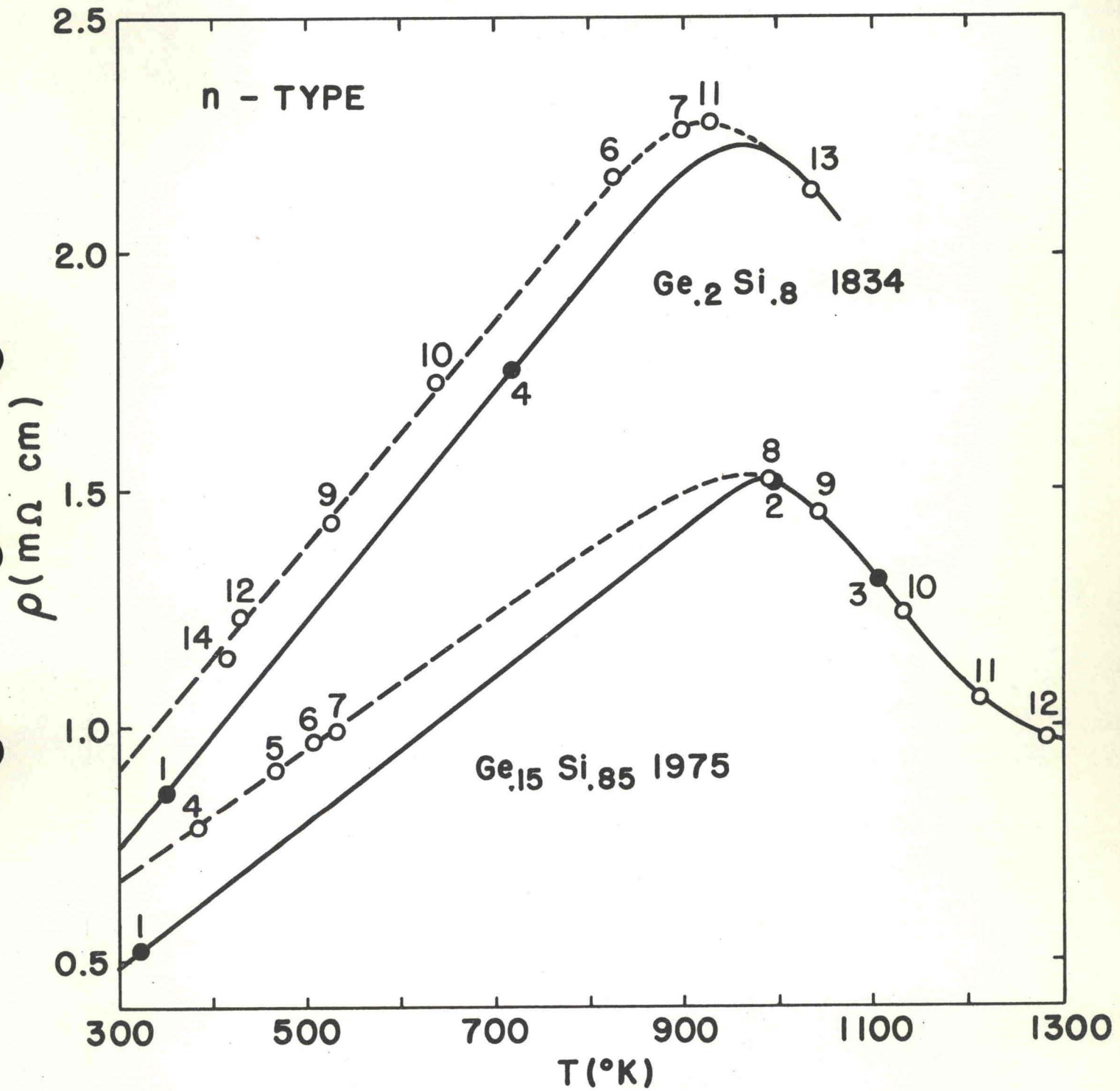


FIG. 14

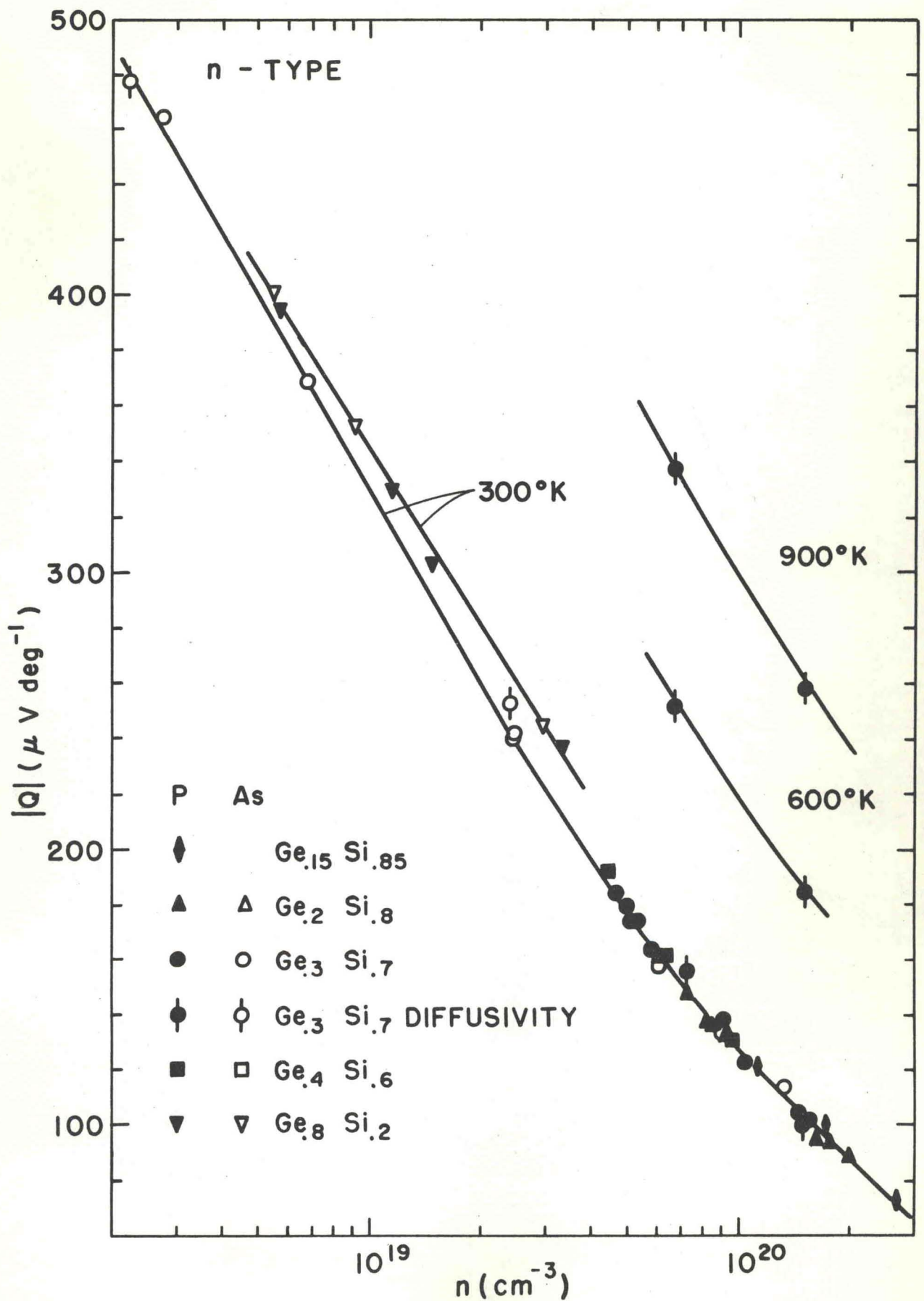


FIG.16

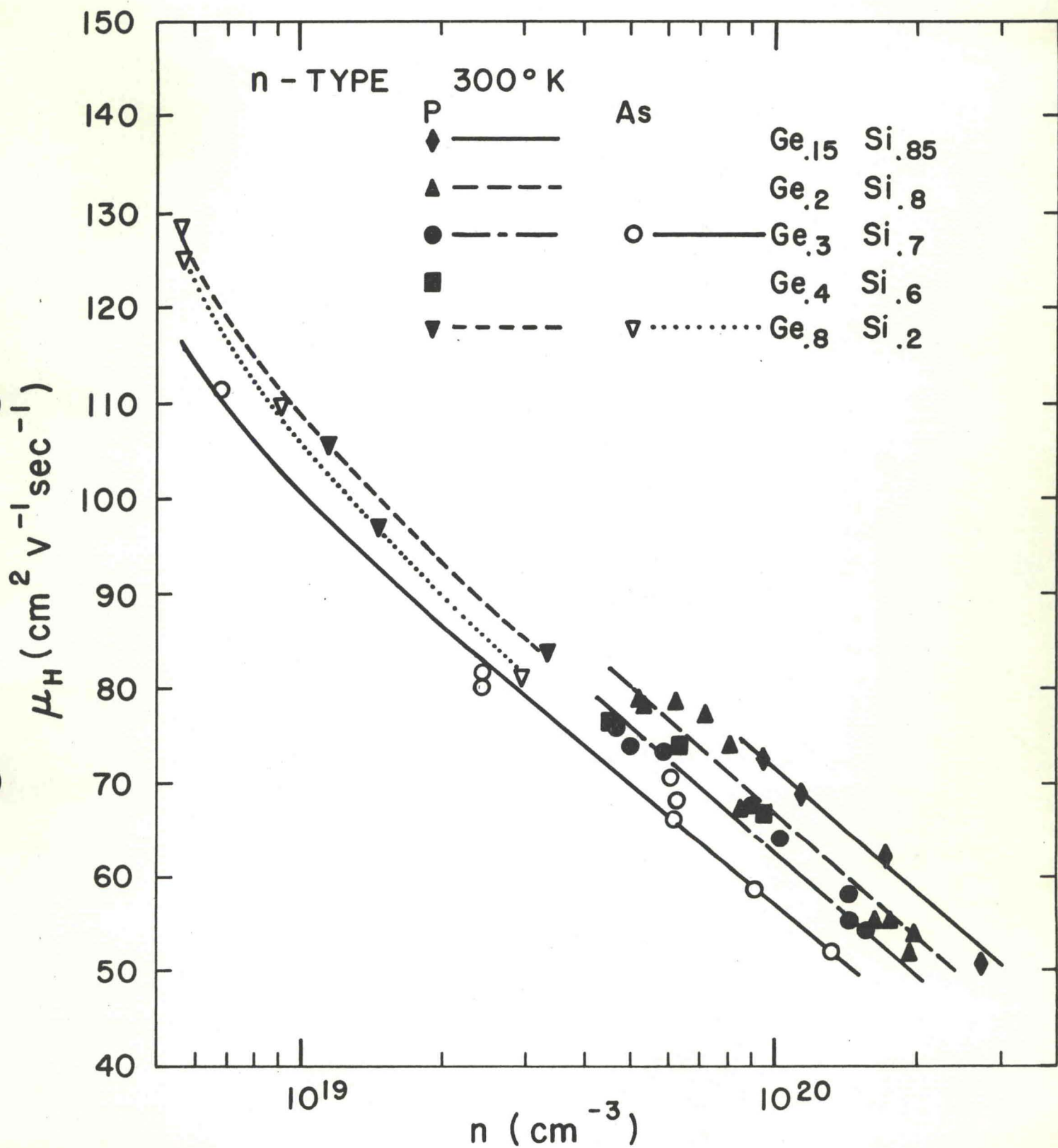


FIG.17

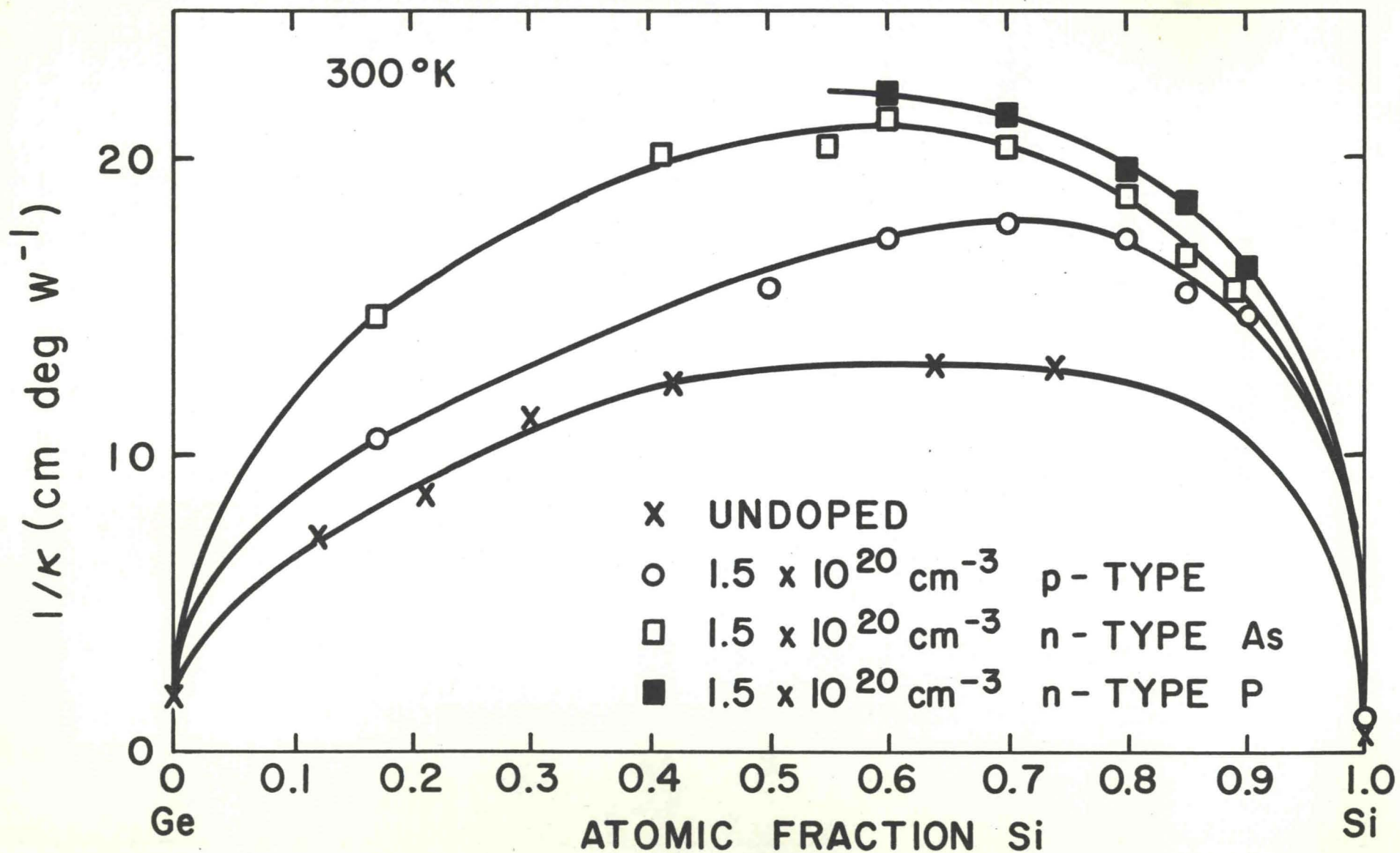


FIG.18

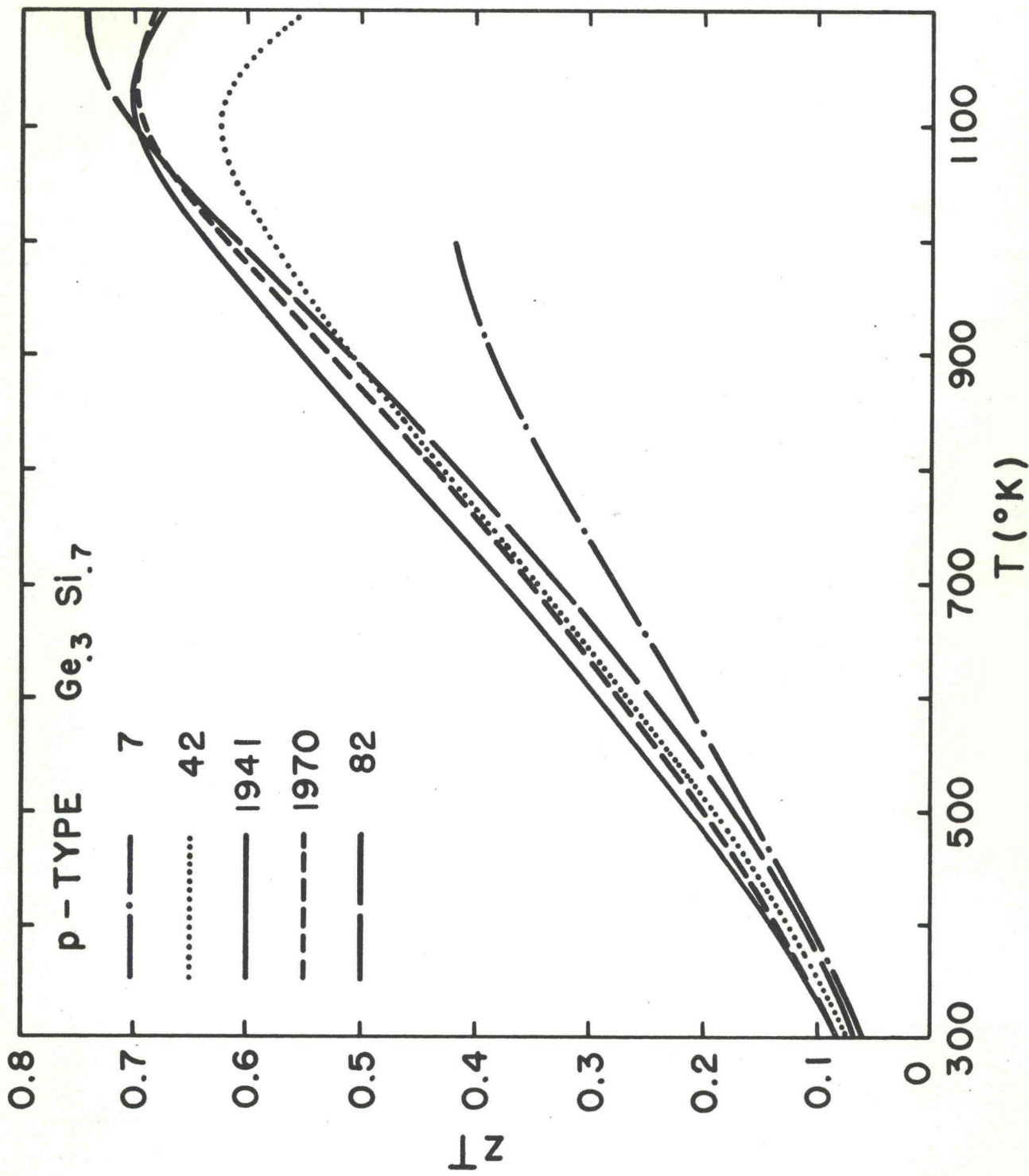


FIG. 19

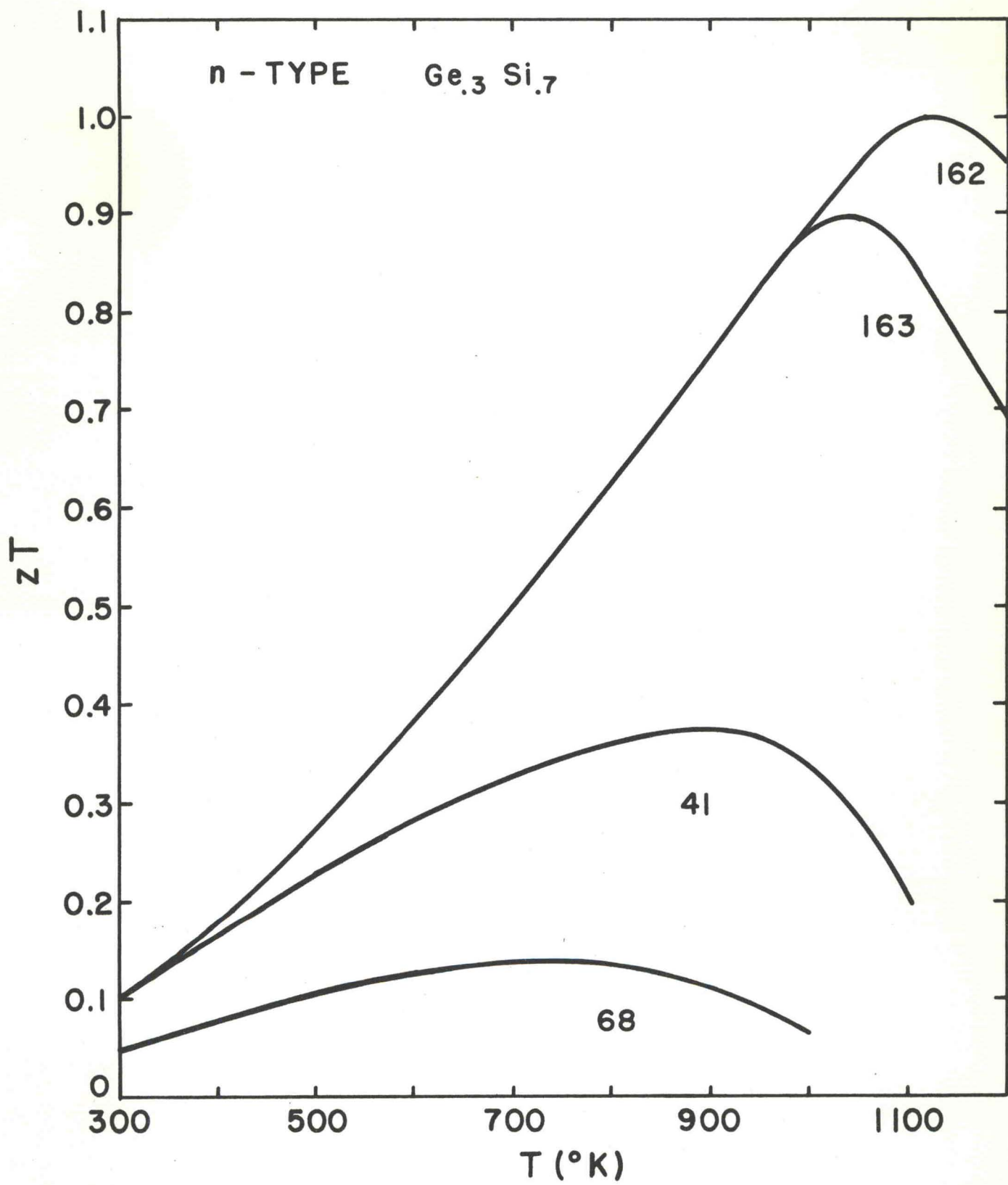


FIG. 20

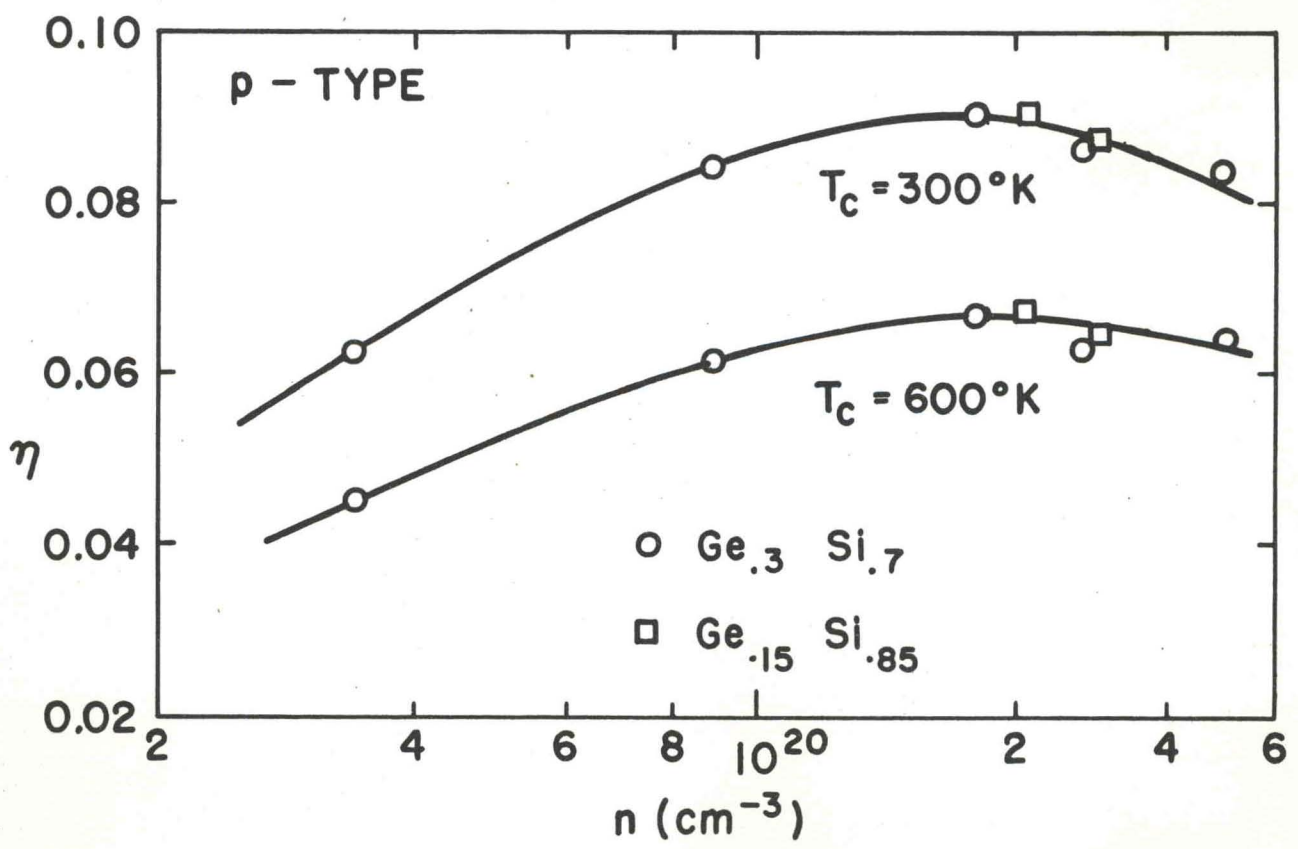
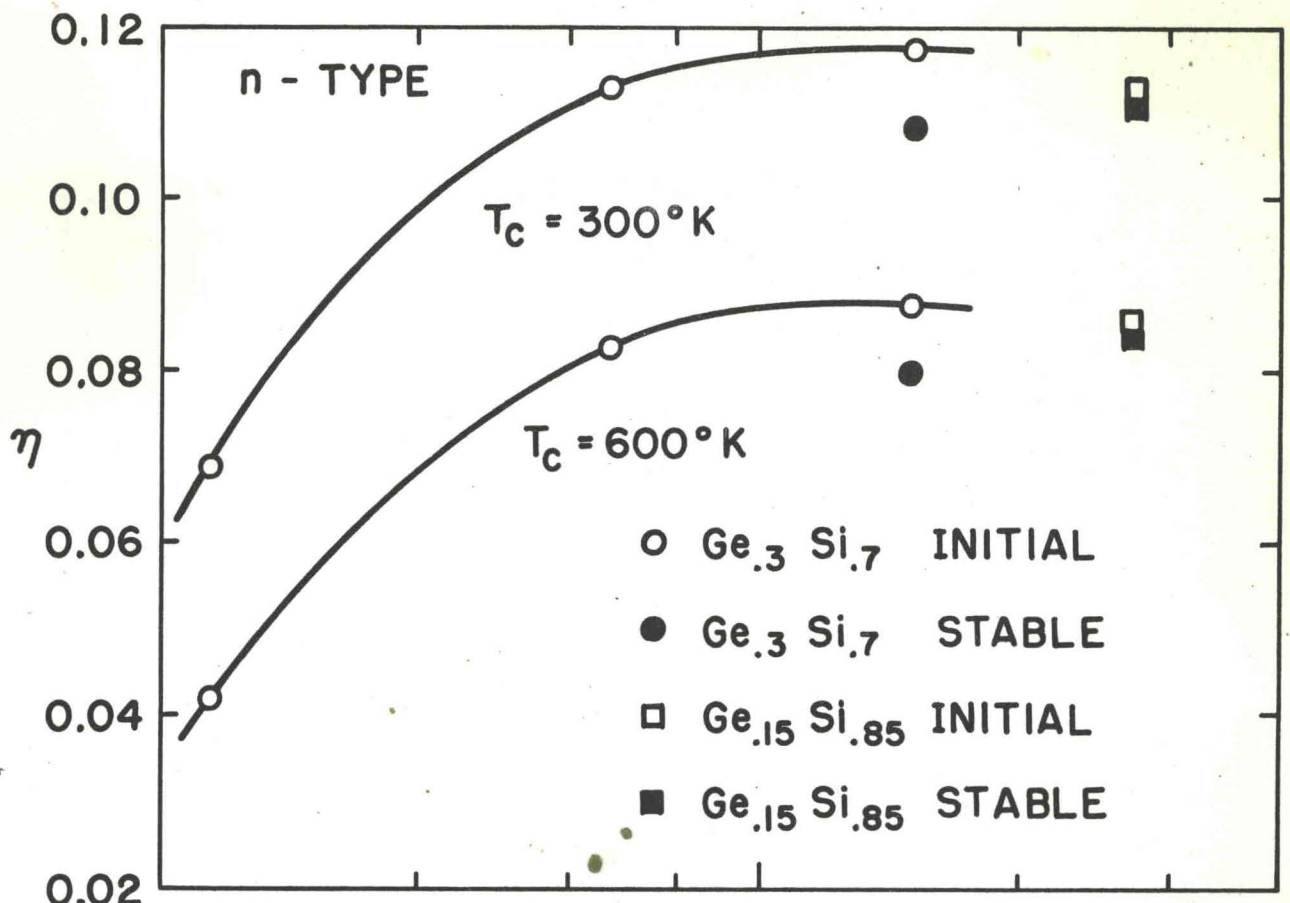


FIG. 21